ON THE OPTICAL PROPERTIES OF ROUGH SURFACES AND THIN FILMS

M. M. WIND

ON THE OPTICAL PROPERTIES OF ROUGH SURFACES AND THIN FILMS

ON THE OPTICAL PROPERTIES OF ROUGH SURFACES AND THIN FILMS

PROEFSCHRIFT

TER VERKRIJGING VAN DE GRAAD VAN DOCTOR AAN DE RIJKSUNIVERSITEIT TE LEIDEN, OP GEZAG VAN DE RECTOR MAGNIFICUS DR. J. J. M. BEENAKKER, HOOGLERAAR IN DE FACULTEIT DER WISKUNDE EN NATUURWETENSCHAPPEN, VOLGENS BESLUIT VAN HET COLLEGE VAN DEKANEN TE VERDEDIGEN OP DONDERDAG 22 JANUARI 1987 TE KLOKKE 15.15 UUR

DOOR

MAURITS MATTHIJS WIND

GEBOREN TE DEN HAAG IN 1958

DRUKKERIJ DE KEMPENAER - OEGSTGEEST - 1987

Leden van de promotiecommissie:

Promotores:	Prof. Dr. D. Bedeaux
	Prof. Dr. P. Mazur
Co-promotor:	Dr. J. Vlieger
Referent:	Dr. R. Greef
Overige leden:	Prof. Dr. J.M.J. van Leeuwen
	Prof. Dr. H.J. Hilhorst
	Prof. Dr. G. Frossati

Aan mijn ouders

Aan Lap

CONTENTS

OUTLINE

It is well known that the amplitudes of the light reflected and transmitted by a perfectly flat substrate are given by the Fresnel formulae. The effects of a plane parallel film covering the flat substrate can be incorporated without difficulty. The systems one encounters in practice, however, are usually more complicated. For example the substrate can be rough. Furthermore the thickness of the film may vary along the substrate. Both the roughness of the substrate and the varying thickness of the film modify the Fresnel formulae considerably. In this thesis we consider two special cases in detail. The first case concerns a continuous film of varying thickness on a rough surface. The second case concerns a discontinuous film consisting of identical truncated spherical particles on a flat substrate.

The theory for the continuous film of varying thickness on a rough substrate is developed in chapter II. Both the roughness and the thickness of the film are supposed to be statistically homogeneous and isotropic along the surface. The amplitude of the roughness as well as the thickness of the film are furthermore assumed to be small compared to the wavelength of the incident light. The theory is based on this fact and uses an expansion in these small parameters. A crucial assumption is furthermore that the normal on the curved surfaces of the film and the substrate has a direction which is everywhere close to its average direction. The optical properties are described by a small number of electromagnetic constitutive coefficients. Formulae for these coefficients are derived in terms of the height-height correlation functions of the upper and lower surfaces of the film, and its average thickness. The reflectance, transmittance and the ellipsometric coefficient of the system are expressed in terms of the constitutive coefficients, for arbitrary angles of incidence.

In chapter III we apply this theory to a rough surface that is characterized by a Gaussian height-height correlation function with a correlation length much larger than the wavelength of light. The reflectance, transmittance and ellipsometric coefficient are calculated in this limit and the results are compared with those obtained by Ohlídal and co-workers in the same limit. It is proved that the difference in results found is a direct consequence of the neglect of the influence of the local curvature of the rough surface on the local electric field by these authors. This, as is shown in this chapter, is an inconsistent approximation in a theory in which all contributions to the second order in the height are systematically taken into account.

In order to understand the optical properties of a discontinuous film consisting of truncated spheres we develop in chapter IV a general method for the calculation of the polarizability of a truncated spherical particle on a substrate, extending a method first developed by Berreman. We assume that the particle is much smaller than the wavelength of light, so that we can neglect retardation and use static dipole (and multipole) fields. The relation between the polarizability and the optical properties of an assembly of such particles is discussed. The method is applied to the special cases of a sphere, hemisphere and thin spherical cap on a substrate. For the latter case agreement is found between the theories developed in chapter II and chapter IV of this thesis.

In chapter V we return to the general case of a truncated sphere on a substrate. We derive a complete set of recurrence relations by means of which approximate values of the polarizability can be obtained within every desired degree of accuracy. We apply the method to the calculation of the normal and parallel polarizabilities of a truncated spherical gold particle on a sapphire substrate in the optical region. We then calculate for perpendicularly incident light in the optical region the transmittance of a system of such gold particles in a square lattice on a sapphire substrate. The results are found to be in good agreement with values experimentally obtained by Niklasson and Craighead.

OPTICAL PROPERTIES OF THIN FILMS ON ROUGH SURFACES

1. Introduction

Research of surfaces and thin films has grown strongly in the last decades. On the one hand this was a consequence of the improvement in high vacuum techniques. On the other hand the knowledge of the properties of these systems is very important for many practical applications. The results of this research have e.g. been used for the development of solar cells, anti-reflection coatings and magnetic memory devices. One of the methods to investigate the properties of surfaces is by making use of light. Generally, one measures the ellipsometric coefficient, the transmittance or reflectance. Furthermore, one can perform light scattering experiments. It was especially the introduction of the laser that was responsible for the fast growing interest in optical measuring methods.

More recently, also the theoretical research of surfaces of solids and liquids has been started. The optical properties are affected by the roughness of these surfaces, as well as by the presence of thin films (oxid layers on metals, contamination of fluids, etc.). For the calculation of the influence of surface roughness on these properties one often uses Kirchhoff's theory of diffraction. The form of the electromagnetic field on the surface is then postulated (see e.g. ref. 1). The film is usually assumed to be homogeneous and isotropic (dielectric constant ϵ).

An alternative theory, appropriate for the description of the optical properties of both thin films and rough boundaries, has been developed by Albano, Bedeaux and Vlieger[2]). In this method a finite boundary layer between two dielectric media is replaced by a fictitious interface, situated somewhere within the original layer and separating directly the two media. If the electromagnetic fields are extended analytically to this new interface, they become in general discontinuous. The discontinuities of these fields can then be expressed in terms of fluctuating polarization and magnetization densities, located at the interface, by means of a set of boundary conditions. These surface polarization and magnetization densities, in their turn, can be related to the extended electromagnetic fields on both sides of the interface by means of fluctuating surface susceptibilities. This general method is applied in section 2 to a boundary layer, consisting of a thin isotropic film on top of a rough surface, and the calculation is carried out up to second order in the film thickness and surface roughness over the wavelength of light. It is assumed that the fictitious interface is flat and the x–y-plane is chosen to coincide with it. The idea to replace a boundary layer by an equivalent system with surface densities (or currents) originates from Kröger and Kretschmann[3]), who gave exact expressions for the surface polarization and magnetization densities in the case of a rough surface.

For the optical properties, which have to be calculated in the present paper, it is sufficient to know the average electro-magnetic fields (the bulk fields). These fields are related to the average surface polarization and magnetization densities $\boldsymbol{P}^s$ and $\boldsymbol{M}^s$ by a set of boundary conditions, whereas $\boldsymbol{P}^s$ and $\boldsymbol{M}^s$ can be expressed in terms of the fields by a set of constitutive equations. It will be shown in section 4 that the latter equation has the form

$$\begin{aligned} \boldsymbol{P}^s &= \sum_\nu \boldsymbol{\xi}^\nu \cdot (E_x^\nu, E_y^\nu, D_z^\nu) - \frac{1}{2c}\sum_\nu \tau^\nu \hat{\boldsymbol{z}} \wedge \frac{\partial}{\partial t}\boldsymbol{H}^\nu , \\ \boldsymbol{M}^s &= -\frac{1}{2c}\sum_\nu \tau^\nu \hat{\boldsymbol{z}} \wedge \frac{\partial}{\partial t}\boldsymbol{E}^\nu , \end{aligned} \tag{1.1}$$

where $\hat{\boldsymbol{z}} = (0, 0, 1)$ is the normal on the dividing surface and c is the velocity of light. The coefficients $\tau^\nu(\nu = +$ or $-)$ couple $\boldsymbol{P}^s$ to the time derivative of the bulk magnetic fields $\boldsymbol{H}^\nu$ on both sides ($\nu = +$ or $-$) of the dividing surface, and $\boldsymbol{M}^s$ to the same derivatives of the bulk electric fields $\boldsymbol{E}^\nu$. The interfacial dielectric susceptibility tensors $\boldsymbol{\xi}^\nu$ couple $\boldsymbol{P}^s$ to the parallel components of the bulk electric fields $\boldsymbol{E}^\nu$ and the normal components of the bulk displacement fields $\boldsymbol{D}^\nu$. In general τ^ν and $\boldsymbol{\xi}^\nu$ are functions of frequency ω and wave-vector $\boldsymbol{k}_\parallel \equiv (k_x, k_y, 0)$ parallel to the x–y-plane. Because of the rotational symmetry around the z-axis and translational symmetry, $\boldsymbol{\xi}^\nu$ is an isotropic tensor of the general form

$$\begin{aligned} \boldsymbol{\xi}^\nu(\boldsymbol{k}_\parallel, \omega) = {}& \tfrac{1}{2}\gamma_l^\nu(k_\parallel, \omega)\hat{\boldsymbol{k}}_\parallel\hat{\boldsymbol{k}}_\parallel + \tfrac{1}{2}\gamma_{tr}^\nu(k_\parallel, \omega)(1 - \hat{\boldsymbol{k}}_\parallel\hat{\boldsymbol{k}}_\parallel - \hat{\boldsymbol{z}}\hat{\boldsymbol{z}}) \\ & + \tfrac{1}{2}\beta^\nu(k_\parallel, \omega)\hat{\boldsymbol{z}}\hat{\boldsymbol{z}} - \tfrac{1}{2}\mathrm{i}\delta^\nu(k_\parallel, \omega)\boldsymbol{k}_\parallel\hat{\boldsymbol{z}} + \tfrac{1}{2}\mathrm{i}\eta^\nu(k_\parallel, \omega)\hat{\boldsymbol{z}}\boldsymbol{k}_\parallel , \end{aligned} \tag{1.2}$$

where $k_\parallel \equiv |\boldsymbol{k}_\parallel|$ and $\hat{\boldsymbol{k}}_\parallel \equiv \boldsymbol{k}_\parallel/k_\parallel$. (The subscripts l and tr of γ^ν stand for longitudinal and transversal).

In section 4 we shall derive general expressions for the surface constitutive coefficients τ^v, γ_l^v, γ_{lr}^v, β^v, δ^v and η^v in terms of the four height–height correlation functions of the upper and lower surfaces of the film, and its average thickness. In section 5 the transmittance, reflectance and ellipsometric coefficient, as well as the energy loss due to absorption within the film and scattering at its boundaries, are expressed in terms of these constitutive coefficients for light polarized parallel (p) and normal (s) to the plane of incidence and for arbitrary angles of incidence.

2. Description of a thin film on a rough surface by means of fluctuating surface susceptibilities

We consider a thin isotropic film (solid or liquid, average thickness $d \ll$ wavelength λ of incident light), with dielectric constant ϵ, covering the rough surface of a substrate (solid or liquid) with dielectric constant ϵ^+ (see fig. 1). The instantaneous position of this surface is given by*

$$z = f^+(r_\parallel, t), \tag{2.1}$$

where $r_\parallel \equiv (x, y, 0)$. The upper surface of the film, i.e. the interface between the film and the ambient (dielectric constant ϵ^-), is also rough and characterised by the equation

$$z = f^-(r_\parallel, t). \tag{2.2}$$

The substrate is supposed to fill the whole region of space where $z > f^+(r_\parallel, t)$, whereas the ambient fills the region where $z < f^-(r_\parallel, t)$. Both dielectric constants ϵ^+ and ϵ may generally be complex, whereas ϵ^- is real and usually $\simeq 1$ (ambient is vacuum or vapour). The averages of $f^+(r_\parallel, t)$ and $f^-(r_\parallel, t)$ are assumed to be constant and given by

$$\langle f^+(r_\parallel, t)\rangle = d, \qquad \langle f^-(r_\parallel, t)\rangle = 0, \tag{2.3}$$

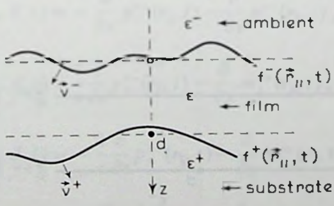


Fig. 1. Cross section of thin film on rough surface.

* For liquids $f^\pm$ will be time dependent. For solids the time dependence of $f^\pm$ may be neglected.

where d is the average film thickness. The symbol $\langle \ldots \rangle$ stands for some ensemble average. The special form of this ensemble, which will strongly depend on the type of system under consideration, need not to be given here.

It is important to remark that, since the functions $z = f^+(\boldsymbol{r}_\parallel, t)$ and $z = f^-(\boldsymbol{r}_\parallel, t)$ may *partly coincide*, the results of the present paper will in principle apply to both continuous and discontinuous films. It will be assumed that the functions $z = f^\pm(\boldsymbol{r}_\parallel, t)$ are single valued and differentiable. The normals on these surfaces in the points $(x, y, z = f^\pm(\boldsymbol{r}_\parallel, t)) = (\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t))$ are given by

$$\boldsymbol{\nu}^\pm(\boldsymbol{r}_\parallel, t) \equiv \left(-\frac{\partial f^\pm}{\partial x}, -\frac{\partial f^\pm}{\partial y}, 1\right)\left[\left(\frac{\partial f^\pm}{\partial x}\right)^2 + \left(\frac{\partial f^\pm}{\partial y}\right)^2 + 1\right]^{-1/2}. \tag{2.4}$$

The boundary conditions for the electromagnetic fields $\boldsymbol{e}$ and $\boldsymbol{b}$ and the displacement fields $\boldsymbol{d}$ and $\boldsymbol{h}$ at the interfaces are given by

$$\begin{aligned}
&\boldsymbol{\nu}^\pm(\boldsymbol{r}_\parallel, t)\cdot[\boldsymbol{d}^\pm(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t) - \boldsymbol{d}(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t)] = 0\,,\\
&[\mathbf{1} - \boldsymbol{\nu}^\pm(\boldsymbol{r}_\parallel, t)\boldsymbol{\nu}^\pm(\boldsymbol{r}_\parallel, t)]\cdot[\boldsymbol{e}^\pm(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t) - \boldsymbol{e}(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t)] = 0\,,\\
&\boldsymbol{\nu}^\pm(\boldsymbol{r}_\parallel, t)\cdot[\boldsymbol{b}^\pm(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t) - \boldsymbol{b}(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t)] = 0\,,\\
&[\mathbf{1} - \boldsymbol{\nu}^\pm(\boldsymbol{r}_\parallel, t)\boldsymbol{\nu}^\pm(\boldsymbol{r}_\parallel, t)]\cdot[\boldsymbol{h}^\pm(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t) - \boldsymbol{h}(\boldsymbol{r}_\parallel, f^\pm(\boldsymbol{r}_\parallel, t), t)] = 0\,.
\end{aligned} \tag{2.5}$$

The fields with superscripts + or − are the fields in the substrate and in the ambient respectively, whereas the fields without superscripts are those within the film.

In a previous paper[4]) we have seen, using an analysis first given by Kröger and Kretschmann[3]), that rough interfaces between dielectrics can be replaced by flat interfaces by introducing equivalent fluctuating surface polarization and magnetization densities. Generalizing this procedure to the present case, we may replace the system of fig. 1 by the system drawn in fig. 2: a plane parallel film of thickness d and with dielectric constant ϵ between two media with dielectric constants ϵ^+ and ϵ^-. When the electromagnetic fields are analytically extended from the interfaces $z = f^-(\boldsymbol{r}_\parallel, t)$ and $z = f^+(\boldsymbol{r}_\parallel, t)$ (in fig. 1) to the new interfaces

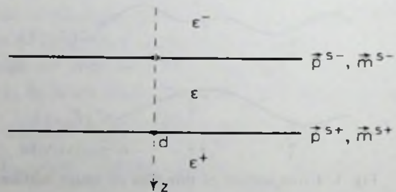


Fig. 2. Description of the system with plane parallel film and two surface polarization and magnetization densities.

$z=0$ and $z=d$ (in fig. 2) respectively, the z components of $\boldsymbol{d}$ and $\boldsymbol{b}$ and the x and y components of $\boldsymbol{e}$ and $\boldsymbol{h}$ are no longer continuous at these new interfaces. The discontinuities of these fields can, however, be related to equivalent fluctuating surface polarization and magnetization densities $\boldsymbol{p}^{s-}$, $\boldsymbol{m}^{s-}$ and $\boldsymbol{p}^{s+}$, $\boldsymbol{m}^{s+}$ in the planes $z=0$ and $z=d$:

$$
\begin{aligned}
&e_x(\boldsymbol{r}_\parallel,0,t)-e_x^-(\boldsymbol{r}_\parallel,0,t)=-\frac{1}{c}\frac{\partial}{\partial t}m_y^{s-}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial x}p_z^{s-}(\boldsymbol{r}_\parallel,t),\\
&e_y(\boldsymbol{r}_\parallel,0,t)-e_y^-(\boldsymbol{r}_\parallel,0,t)=\frac{1}{c}\frac{\partial}{\partial t}m_x^{s-}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}p_z^{s-}(\boldsymbol{r}_\parallel,t),\\
&d_z(\boldsymbol{r}_\parallel,0,t)-d_z^-(\boldsymbol{r}_\parallel,0,t)=-\frac{\partial}{\partial x}p_x^{s-}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}p_y^{s-}(\boldsymbol{r}_\parallel,t),\\
&h_x(\boldsymbol{r}_\parallel,0,t)-h_x^-(\boldsymbol{r}_\parallel,0,t)=\frac{1}{c}\frac{\partial}{\partial t}p_y^{s-}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial x}m_z^{s-}(\boldsymbol{r}_\parallel,t),\\
&h_y(\boldsymbol{r}_\parallel,0,t)-h_y^-(\boldsymbol{r}_\parallel,0,t)=-\frac{1}{c}\frac{\partial}{\partial t}p_x^{s-}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}m_z^{s-}(\boldsymbol{r}_\parallel,t),\\
&b_z(\boldsymbol{r}_\parallel,0,t)-b_z^-(\boldsymbol{r}_\parallel,0,t)=-\frac{\partial}{\partial x}m_x^{s-}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}m_y^{s-}(\boldsymbol{r}_\parallel,t)
\end{aligned}
\tag{2.6}
$$

and

$$
\begin{aligned}
&e_x^+(\boldsymbol{r}_\parallel,d,t)-e_x(\boldsymbol{r}_\parallel,d,t)=-\frac{1}{c}\frac{\partial}{\partial t}m_y^{s+}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial x}p_z^{s+}(\boldsymbol{r}_\parallel,t),\\
&e_y^+(\boldsymbol{r}_\parallel,d,t)-e_y(\boldsymbol{r}_\parallel,d,t)=\frac{1}{c}\frac{\partial}{\partial t}m_x^{s+}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}p_z^{s+}(\boldsymbol{r}_\parallel,t),\\
&d_z^+(\boldsymbol{r}_\parallel,d,t)-d_z(\boldsymbol{r}_\parallel,d,t)=-\frac{\partial}{\partial x}p_x^{s+}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}p_y^{s+}(\boldsymbol{r}_\parallel,t),\\
&h_x^+(\boldsymbol{r}_\parallel,d,t)-h_x(\boldsymbol{r}_\parallel,d,t)=\frac{1}{c}\frac{\partial}{\partial t}p_y^{s+}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial x}m_z^{s+}(\boldsymbol{r}_\parallel,t),\\
&h_y^+(\boldsymbol{r}_\parallel,d,t)-h_y(\boldsymbol{r}_\parallel,d,t)=-\frac{1}{c}\frac{\partial}{\partial t}p_x^{s+}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}m_z^{s+}(\boldsymbol{r}_\parallel,t),\\
&b_z^+(\boldsymbol{r}_\parallel,d,t)-b_z(\boldsymbol{r}_\parallel,d,t)=-\frac{\partial}{\partial x}m_x^{s+}(\boldsymbol{r}_\parallel,t)-\frac{\partial}{\partial y}m_y^{s+}(\boldsymbol{r}_\parallel,t)
\end{aligned}
\tag{2.7}
$$

(cf. eq. (1.1) of ref. 4). The densities at the right-hand sides of these equations are related to the fields by (cf. eqs. (2.8)–(2.11) of ref. 4)

$$p^{s-}(r_\parallel, t) = -\int_0^{f^-(r_\parallel,t)} [(d_x, d_y, -e_z)(r,t) - (d_x^-, d_y^-, -e_z^-)(r,t)]\,\mathrm{d}z\,,$$

$$m^{s-}(r_\parallel, t) = -\int_0^{f^-(r_\parallel,t)} [(b_x, b_y, -h_z)(r,t) - (b_x^-, b_y^-, -h_z^-)(r,t)]\,\mathrm{d}z\,, \tag{2.8}$$

$$p^{s+}(r_\parallel, t) = -\int_d^{f^+(r_\parallel,t)} [(d_x^+, d_y^+, -e_z^+)(r,t) - (d_x, d_y, -e_z)(r,t)]\,\mathrm{d}z\,,$$

$$m^{s+}(r_\parallel, t) = -\int_d^{f^+(r_\parallel,t)} [(b_x^+, b_y^+, -h_z^+)(r,t) - (b_x, b_y, -h_z)(r,t)]\,\mathrm{d}z\,. \tag{2.9}$$

We can furthermore replace the system described above, with a plane parallel film and two fluctuating polarization and magnetization densities p^{s+}, m^{s+} and p^{s-}, m^{s-}, by the system, drawn in fig. 3, with only *one* equivalent surface polarization and magnetization density $p^s(r_\parallel, t)$ and $m^s(r_\parallel, t)$ in the x–y-plane. The densities p^s and m^s must be chosen such that the electromagnetic fields for $z<0$ and $z>d$ in fig. 3 are identical with those in fig. 2 for the same z-values. The parallel components p_x^s and p_y^s of this surface polarization density are found in the following way: the difference of $d_z^+(r_\parallel, d, t)$ and $d_z^-(r_\parallel, 0, t)$ in fig. 2 is given by

$$d_z^+(r_\parallel, d, t) - d_z^-(r_\parallel, 0, t) = -\frac{\partial}{\partial x} p_x^{s+}(r_\parallel, t) - \frac{\partial}{\partial y} p_y^{s+}(r_\parallel, t)$$
$$-\frac{\partial}{\partial x} p_x^{s-}(r_\parallel, t) - \frac{\partial}{\partial y} p_y^{s-}(r_\parallel, t) + d_z(r_\parallel, d, t) - d_z(r_\parallel, 0, t)\,, \tag{2.10}$$

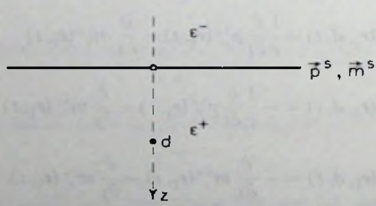


Fig. 3. Description of the system with one polarization and magnetization density.

where we have used the third boundary conditions in eqs.(2.6) and (2.7). Now it follows with Maxwell's equation div $\boldsymbol{d} = 0$ that

$$d_z(\boldsymbol{r}_\parallel, d, t) - d_z(\boldsymbol{r}_\parallel, 0, t) = \int_0^d \frac{\partial}{\partial z} d_z(\boldsymbol{r}, t)\,\mathrm{d}z$$

$$= -\frac{\partial}{\partial x}\int_0^d d_x(\boldsymbol{r}, t)\,\mathrm{d}z - \frac{\partial}{\partial y}\int_0^d d_y(\boldsymbol{r}, t)\,\mathrm{d}z\,, \tag{2.11}$$

so that eq. (2.10) can be written as

$$d_z^+(\boldsymbol{r}_\parallel, d, t) - d_z^-(\boldsymbol{r}_\parallel, 0, t) = -\frac{\partial}{\partial x}\Bigg[p_x^{s+}(\boldsymbol{r}_\parallel, t) + p_x^{s-}(\boldsymbol{r}_\parallel, t)$$

$$+ \int_0^d d_x(\boldsymbol{r}, t)\,\mathrm{d}z\Bigg] - \frac{\partial}{\partial y}\Bigg[p_y^{s+}(\boldsymbol{r}_\parallel, t) + p_y^{s-}(\boldsymbol{r}_\parallel, t) + \int_0^d d_y(\boldsymbol{r}, t)\,\mathrm{d}z\Bigg]. \tag{2.12}$$

Since $d_z^+(\boldsymbol{r}, t)$ for $z \geqslant d$ and $d_z^-(\boldsymbol{r}, t)$ for $z \leqslant 0$ must be the same in the systems drawn in figs. 2 and 3, the relation (2.12) must also hold for the latter system. The field $d_z^+(\boldsymbol{r}, t)$ can again analytically be extended to the x–y-plane and we find

$$d_z^+(\boldsymbol{r}_\parallel, 0, t) = d_z^+(\boldsymbol{r}_\parallel, d, t) - \int_0^d \frac{\partial}{\partial z} d_z^+(\boldsymbol{r}, t)\,\mathrm{d}z$$

$$= d_z^+(\boldsymbol{r}_\parallel, d, t) + \frac{\partial}{\partial x}\int_0^d d_x^+(\boldsymbol{r}, t)\,\mathrm{d}z + \frac{\partial}{\partial y}\int_0^d d_y^+(\boldsymbol{r}, t)\,\mathrm{d}z\,, \tag{2.13}$$

where we have used the Maxwell equation div $\boldsymbol{d}^+ = 0$. From the last two equations we obtain the following boundary conditions for d_z at $z = 0$ for the system drawn in fig. 3:

$$d_z^+(\boldsymbol{r}_\parallel, 0, t) - d_z^-(\boldsymbol{r}_\parallel, 0, t) = -\frac{\partial}{\partial x}\Bigg[p_x^{s+}(\boldsymbol{r}_\parallel, t)$$

$$+ p_x^{s-}(\boldsymbol{r}_\parallel, t) + \int_0^d d_x(\boldsymbol{r}, t)\,\mathrm{d}z - \int_0^d d_x^+(\boldsymbol{r}, t)\,\mathrm{d}z\Bigg]$$

$$-\frac{\partial}{\partial y}\Bigg[p_y^{s+}(\boldsymbol{r}_\parallel, t) + p_y^{s-}(\boldsymbol{r}_\parallel, t) + \int_0^d d_y(\boldsymbol{r}, t)\,\mathrm{d}z - \int_0^d d_y^+(\boldsymbol{r}, t)\,\mathrm{d}z\Bigg]. \tag{2.14}$$

Since the boundary conditions in $z = 0$ are given by (see eq. (1.1) of ref. 4)

$$
\begin{aligned}
e_x^+(r_\|, 0, t) - e_x^-(r_\|, 0, t) &= -\frac{1}{c}\frac{\partial}{\partial t} m_y^s(r_\|, t) - \frac{\partial}{\partial x} p_z^s(r_\|, t)\,, \\
e_y^+(r_\|, 0, t) - e_y^-(r_\|, 0, t) &= \frac{1}{c}\frac{\partial}{\partial t} m_x^s(r_\|, t) - \frac{\partial}{\partial y} p_z^s(r_\|, t)\,, \\
d_z^+(r_\|, 0, t) - d_z^-(r_\|, 0, t) &= -\frac{\partial}{\partial x} p_x^s(r_\|, t) - \frac{\partial}{\partial y} p_y^s(r_\|, t)\,, \\
h_x^+(r_\|, 0, t) - h_x^-(r_\|, 0, t) &= \frac{1}{c}\frac{\partial}{\partial t} p_y^s(r_\|, t) - \frac{\partial}{\partial x} m_z^s(r_\|, t)\,, \\
h_y^+(r_\|, 0, t) - h_y^-(r_\|, 0, t) &= -\frac{1}{c}\frac{\partial}{\partial t} p_x^s(r_\|, t) - \frac{\partial}{\partial y} m_z^s(r_\|, t)\,, \\
b_z^+(r_\|, 0, t) - b_z^-(r_\|, 0, t) &= -\frac{\partial}{\partial x} m_x^s(r_\|, t) - \frac{\partial}{\partial y} m_y^s(r_\|, t)\,,
\end{aligned}
\tag{2.15}
$$

we find for the parallel components of the surface polarization density $\boldsymbol{p}^s$ in $z = 0$

$$
\begin{aligned}
p_x^s(r_\|, t) &= p_x^{s+}(r_\|, t) + p_x^{s-}(r_\|, t) + \int_0^d d_x(r, t)\,\mathrm{d}z - \int_0^d d_x^+(r, t)\,\mathrm{d}z \\
&= -\int_0^{f^-(r_\|, t)} [d_x(r, t) - d_x^-(r, t)]\,\mathrm{d}z - \int_0^{f^+(r_\|, t)} [d_x^+(r, t) - d_x(r, t)]\,\mathrm{d}z\,, \\
p_y^s(r_\|, t) &= p_y^{s+}(r_\|, t) + p_y^{s-}(r_\|, t) + \int_0^d d_y(r, t)\,\mathrm{d}z - \int_0^d d_y^+(r, t)\,\mathrm{d}z \\
&= -\int_0^{f^-(r_\|, t)} [d_y(r, t) - d_y^-(r, t)]\,\mathrm{d}z - \int_0^{f^+(r_\|, t)} [d_y^+(r, t) - d_y(r, t)]\,\mathrm{d}z\,,
\end{aligned}
\tag{2.16}
$$

where we have used (2.8) and (2.9). The normal component of $\boldsymbol{p}^s$ and the parallel and normal components of the surface magnetization density $\boldsymbol{m}^s$ are obtained in an analogous way. One finds, using the other Maxwell equations $\operatorname{rot}\boldsymbol{e} = -(1/c)\partial\boldsymbol{b}/\partial t$, $\operatorname{rot}\boldsymbol{h} = (1/c)\partial\boldsymbol{d}/\partial t$ and $\operatorname{div}\boldsymbol{b} = 0$,

$$
p_z^s(r_\|, t) = \int_0^{f^-(r_\|, t)} [e_z(r, t) - e_z^-(r, t)]\,\mathrm{d}z + \int_0^{f^-(r_\|, t)} [e_z^+(r, t) - e_z(r, t)]\,\mathrm{d}z\,,
$$

$$m_x^s(r_\parallel, t) = -\int_0^{f^-(r_\parallel,t)} [b_x(r,t) - b_x^-(r,t)]\,dz - \int_0^{f^+(r_\parallel,t)} [b_x^+(r,t) - b_x(r,t)]\,dz\,,$$

$$m_y^s(r_\parallel, t) = -\int_0^{f^-(r_\parallel,t)} [b_y(r,t) - b_y^-(r,t)]\,dz - \int_0^{f^+(r_\parallel,t)} [b_y^+(r,t) - b_y(r,t)]\,dz\,, \tag{2.17}$$

$$m_z^s(r_\parallel, t) = \int_0^{f^-(r_\parallel,t)} [h_z(r,t) - h_z^-(r,t)]\,dz + \int_0^{f^+(r_\parallel,t)} [h_z^+(r,t) - h_z^-(r,t)]\,dz\,.$$

The equations (2.16) and (2.17) can alternatively be written as

$$p_x^s(r_\parallel, t) = \int_0^{f^-(r_\parallel,t)} [\epsilon^- e_x^-(r,t) - \epsilon e_x(r,t)]\,dz + \int_0^{f^+(r_\parallel,t)} [\epsilon e_x(r,t) - \epsilon^+ e_x^+(r,t)]\,dz\,,$$

$$p_y^s(r_\parallel, t) = \int_0^{f^-(r_\parallel,t)} [\epsilon^- e_y^-(r,t) - \epsilon e_y(r,t)]\,dz + \int_0^{f^+(r_\parallel,t)} [\epsilon e_y(r,t) - \epsilon^+ e_y^+(r,t)]\,dz\,,$$

$$p_z^s(r_\parallel, t) = -\int_0^{f^-(r_\parallel,t)} \left[\frac{1}{\epsilon^-} d_z^-(r,t) - \frac{1}{\epsilon} d_z(r,t)\right] dz$$

$$-\int_0^{f^+(r_\parallel,t)} \left[\frac{1}{\epsilon} d_z(r,t) - \frac{1}{\epsilon^+} d_z^+(r,t)\right] dz\,, \tag{2.18}$$

$$m_x^s(r_\parallel, t) = \int_0^{f^-(r_\parallel,t)} [h_x^-(r,t) - h_x(r,t)]\,dz + \int_0^{f^+(r_\parallel,t)} [h_x(r,t) - h_x^+(r,t)]\,dz\,,$$

$$m_y^s(r_\parallel, t) = \int_0^{f^-(r_\parallel,t)} [h_y^-(r,t) - h_y(r,t)]\,dz + \int_0^{f^+(r_\parallel,t)} [h_y(r,t) - h_y^+(r,t)]\,dz\,,$$

$$m_z^s(r_\parallel, t) = -\int_0^{f^-(r_\parallel,t)} [b_z^-(r,t) - b_z(r,t)]\,dz - \int_0^{f^+(r_\parallel,t)} [b_z(r,t) - b_z^+(r,t)]\,dz\,,$$

where we have used the relations $\boldsymbol{d}^\pm = \epsilon^\pm \boldsymbol{e}^\pm$, $\boldsymbol{d} = \epsilon \boldsymbol{e}$, $\boldsymbol{h}^\pm = \boldsymbol{b}^\pm$ and $\boldsymbol{h} = \boldsymbol{b}$, i.e. the magnetic permeability is assumed to be equal to unity everywhere in space.

In principle, eq. (2.18) gives the values of $\boldsymbol{p}^s(r_\parallel, t)$ and $\boldsymbol{m}^s(r_\parallel, t)$ exactly to all orders in $f^{\pm}(\boldsymbol{r}_{\|}, t)$. In the following, however, we want to know the quantities $\boldsymbol{p}^{s}$ and $\boldsymbol{m}^{s}$ only up to the second order in $f^{\pm}$. We therefore develop the integrands at the right-hand sides of eq. (2.18) into Taylor series of z around $z = 0$ and integrate over z. For $p_x^s(\boldsymbol{r}_{\|}, t)$ we then obtain up to second order

$$\begin{aligned} p_x^s(\boldsymbol{r}_{\|}, t) &= [\epsilon^- e_x^-(\boldsymbol{r}_{\|}, 0, t) - \epsilon e_x(\boldsymbol{r}_{\|}, 0, t)] f^-(\boldsymbol{r}_{\|}, t) + [\epsilon e_x(\boldsymbol{r}_{\|}, 0, t) \\ &\quad - \epsilon^+ e_x^+(\boldsymbol{r}_{\|}, 0, t)] f^+(\boldsymbol{r}_{\|}, t) - \frac{1}{2c}\frac{\partial}{\partial t}[\epsilon^- h_y^-(\boldsymbol{r}_{\|}, 0, t) - \epsilon h_y(\boldsymbol{r}_{\|}, 0, t)] \\ &\quad \times \{f^-(\boldsymbol{r}_{\|}, t)\}^2 - \frac{1}{2c}\frac{\partial}{\partial t}[\epsilon h_y(\boldsymbol{r}_{\|}, 0, t) - \epsilon^+ h_y^+(\boldsymbol{r}_{\|}, 0, t)]\{f^+(\boldsymbol{r}_{\|}, t)\}^2 \\ &\quad + \frac{1}{2}\frac{\partial}{\partial x}[d_z^-(\boldsymbol{r}_{\|}, 0, t) - d_z(\boldsymbol{r}_{\|}, 0, t)\{f^-(\boldsymbol{r}_{\|}, t)\}^2 \\ &\quad + \frac{1}{2}\frac{\partial}{\partial x}[d_z(\boldsymbol{r}_{\|}, 0, t) - d_z^+(\boldsymbol{r}_{\|}, 0, t)]\{f^+(\boldsymbol{r}_{\|}, t)\}^2 , \end{aligned} \tag{2.19}$$

where we have used Maxwell's equation $\operatorname{rot} \boldsymbol{e} = -(1/c)\partial \boldsymbol{b}/\partial t$ and $\boldsymbol{d}^{\pm} = \epsilon^{\pm}\boldsymbol{e}^{\pm}$, $\boldsymbol{d} = \epsilon \boldsymbol{e}$, $\boldsymbol{h}^{\pm} = \boldsymbol{b}^{\pm}$, $\boldsymbol{h} = \boldsymbol{b}$. At the right-hand side of eq. (2.19) we can now eliminate the fields $\boldsymbol{e}$, $\boldsymbol{d}$ and $\boldsymbol{h}$ by using the boundary conditions eqs. (2.6) and (2.7), together with the expressions eqs. (2.8) and (2.9) for the surface polarization and magnetization densities $\boldsymbol{p}^{s\pm}$ and $\boldsymbol{m}^{s\pm}$. To *first* order in $f^{\pm}(\boldsymbol{r}_{\|}, t)$ one then obtains

$$\begin{aligned} e_x(\boldsymbol{r}_{\|}, 0, t) - e_x^{\pm}(\boldsymbol{r}_{\|}, 0, t) &= \left(\frac{\epsilon - \epsilon^{\pm}}{\epsilon\epsilon^{\pm}}\right)\frac{\partial}{\partial x}[f^{\pm}(\boldsymbol{r}_{\|}, t) d_z^{\pm}(\boldsymbol{r}_{\|}, 0, t)] , \\ e_y(\boldsymbol{r}_{\|}, 0, t) - e_y^{\pm}(\boldsymbol{r}_{\|}, 0, t) &= \left(\frac{\epsilon - \epsilon^{\pm}}{\epsilon\epsilon^{\pm}}\right)\frac{\partial}{\partial y}[f^{\pm}(\boldsymbol{r}_{\|}, t) d_z^{\pm}(\boldsymbol{r}_{\|}, 0, t)] , \\ d_z(\boldsymbol{r}_{\|}, 0, t) - d_z^{\pm}(\boldsymbol{r}_{\|}, 0, t) &= (\epsilon - \epsilon^{\pm})\frac{\partial}{\partial x}[f^{\pm}(\boldsymbol{r}_{\|}, t) e_x^{\pm}(\boldsymbol{r}_{\|}, 0, t)] \\ &\quad + (\epsilon - \epsilon^{\pm})\frac{\partial}{\partial y}[f^{\pm}(\boldsymbol{r}_{\|}, t) e_y^{\pm}(\boldsymbol{r}_{\|}, 0, t)] , \\ h_x(\boldsymbol{r}_{\|}, 0, t) - h_x^{\pm}(\boldsymbol{r}_{\|}, 0, t) &= -(\epsilon - \epsilon^{\pm})\frac{1}{c}\frac{\partial}{\partial t}[f^{\pm}(\boldsymbol{r}_{\|}, t) e_y^{\pm}(\boldsymbol{r}_{\|}, 0, t)] , \\ h_y(\boldsymbol{r}_{\|}, 0, t) - h_y^{\pm}(\boldsymbol{r}_{\|}, 0, t) &= (\epsilon - e^{\pm})\frac{1}{c}\frac{\partial}{\partial t}[f^{\pm}(\boldsymbol{r}_{\|}, t) e_x^{\pm}(\boldsymbol{r}_{\|}, 0, t)] , \\ b_z(\boldsymbol{r}_{\|}, 0, t) - b_z^{\pm}(\boldsymbol{r}_{\|}, 0, t) &= 0 . \end{aligned} \tag{2.20}$$

In the relations with the plus signs we have analytically extended the fields from $z = d$ (in eq. (2.7)) to $z = 0$. If we use the first order relations (2.20) to eliminate e_x, b_y and d_z at the right-hand side of eq. (2.19), which is at least of the first order in $f^{\pm}(\boldsymbol{r}_{\|}, t)$, we find the following result for $p_x^s(\boldsymbol{r}_{\|}, t)$, correct up to the second order in $f^{\pm}(\boldsymbol{r}_{\|}, t)$:

$$\begin{aligned}
p_x^s(\boldsymbol{r}_{\|}, t) = & -(\epsilon - \epsilon^-) f^-(\boldsymbol{r}_{\|}, t) e_x^-(\boldsymbol{r}_{\|}, 0, t) + (\epsilon - \epsilon^+) f^+(\boldsymbol{r}_{\|}, t) e_x^+(\boldsymbol{r}_{\|}, 0, t) \\
& - \left(\frac{\epsilon - \epsilon^-}{\epsilon^-}\right) f^-(\boldsymbol{r}_{\|}, t) \frac{\partial}{\partial x} [f^-(\boldsymbol{r}_{\|}, t) d_z^-(\boldsymbol{r}_{\|}, 0, t)] \\
& + \left(\frac{\epsilon - \epsilon^+}{\epsilon^+}\right) f^+(\boldsymbol{r}_{\|}, t) \frac{\partial}{\partial x} [f^+(\boldsymbol{r}_{\|}, t) d_z^+(\boldsymbol{r}_{\|}, 0, t)] \\
& + \frac{1}{2c} (\epsilon - \epsilon^-) \{f^-(\boldsymbol{r}_{\|}, t)\}^2 \frac{\partial}{\partial t} h_y^-(\boldsymbol{r}_{\|}, 0, t) \\
& - \frac{1}{2c} (\epsilon - \epsilon^+) \{f^+(\boldsymbol{r}_{\|}, t)\}^2 \frac{\partial}{\partial t} h_y^+(\boldsymbol{r}_{\|}, 0, t) \,.
\end{aligned} \tag{2.21}$$

In a completely analogous way we can find second order expressions for p_y^s, p_z^s and the three components of $\boldsymbol{m}^s$ from eq. (2.18), together with the Maxwell equations and the relations eq. (2.20). By introducing the fields*

$$\boldsymbol{n}_e \equiv (e_x, e_y, d_z) \qquad \text{and} \qquad \boldsymbol{n}_m \equiv (h_x, h_y, b_z) \tag{2.22}$$

and the matrices ($\nu = +$ or $-$)

$$\xi_b^{(1)\nu}(\boldsymbol{r}_{\|}, t) \equiv \nu(\epsilon - \epsilon^\nu) f^\nu(\boldsymbol{r}_{\|}, t) \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \dfrac{1}{\epsilon\epsilon^\nu} \end{pmatrix}, \tag{2.23}$$

$$\xi_b^{(2)\nu}(\boldsymbol{r}_{\|}, t) \equiv \nu(\epsilon - \epsilon^\nu) \{f^\nu(\boldsymbol{r}_{\|}, t)\}^2 \begin{pmatrix} 0 & 0 & \dfrac{1}{\epsilon^\nu}\dfrac{\partial}{\partial x} \\ 0 & 0 & \dfrac{1}{\epsilon^\nu}\dfrac{\partial}{\partial y} \\ -\dfrac{1}{\epsilon}\dfrac{\partial}{\partial x} & -\dfrac{1}{\epsilon}\dfrac{\partial}{\partial y} & 0 \end{pmatrix}, \tag{2.24}$$

$$\xi_b^{(3)\nu}(\boldsymbol{r}_{\|}, t) \equiv \tfrac{1}{2}\nu(\epsilon - \epsilon^\nu) \begin{pmatrix} 0 & 0 & \dfrac{1}{\epsilon^\nu}\dfrac{\partial}{\partial x} \\ 0 & 0 & \dfrac{1}{\epsilon^\nu}\dfrac{\partial}{\partial y} \\ -\dfrac{1}{\epsilon}\dfrac{\partial}{\partial x} & -\dfrac{1}{\epsilon}\dfrac{\partial}{\partial y} & 0 \end{pmatrix} \{f^\nu(\boldsymbol{r}_{\|}, t)\}^2 \tag{2.25}$$

* In the next section we shall see why it is convenient to introduce the fields $\boldsymbol{n}_e$ and $\boldsymbol{n}_m$, instead of $\boldsymbol{e}$, $\boldsymbol{d}$, $\boldsymbol{h}$ or $\boldsymbol{b}$.

and

$$\xi_b^{(4)\nu}(r_\parallel, t) \equiv -\tfrac{1}{2}\nu(\epsilon - \epsilon^\nu)\{f^\nu(r_\parallel, t)\}^2 \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{2.26}$$

one then obtains the following result:

$$\begin{aligned} p^s(r_\parallel, t) &= \sum_{i=1}^{3} \sum_\nu \xi_b^{(i)\nu}(r_\parallel, t)\cdot n_e^\nu(r_\parallel, 0, t) + \sum_\nu \xi_b^{(4)\nu}(r_\parallel, t)\cdot \frac{1}{c}\frac{\partial}{\partial t} n_m^\nu(r_\parallel, 0, t), \\ m^s(r_\parallel, t) &= \sum_\nu \xi_b^{(4)\nu}(r_\parallel, t)\cdot \frac{1}{c}\frac{\partial}{\partial t} n_e^\nu(r_\parallel, 0, t), \end{aligned} \tag{2.27}$$

where Σ_ν denotes summation over $\nu = +$ and $\nu = -$. The quantities $\xi_b^{(i)\nu}(r_\parallel, t)$ are the fluctuating surface susceptibilities, describing in the equivalent surface polarization and magnetization formalism the thin film on a rough surface up to second order in $f^\nu(r_\parallel, t)$. One should note that for $i = 1, 3$ and 4 these coefficients are ordinary functions of $r_\parallel$ and t, but for $i = 2$ they also contain differential operators $\partial/\partial x$ and $\partial/\partial y$, describing the spatial dispersion of the fields along the x–y-plane. The subscript b stands for bare, denoting that we have here unaveraged fluctuating quantities.

3. Maxwell theory in the presence of induced surface polarization and magnetization densities

In the last section we have seen that a thin film on a rough surface could be described by equivalent surface polarization and magnetization densities $p^s(r_\parallel, t)$ and $m^s(r_\parallel, t)$. The fluctuating electric and magnetic fields e and b and the displacement fields d and h satisfy Maxwell's equations,

$$\begin{aligned} \operatorname{rot} e &= -\frac{1}{c}\frac{\partial}{\partial t} b, \qquad \operatorname{div} d = 0, \\ \operatorname{rot} h &= \frac{1}{c}\frac{\partial}{\partial t} d, \qquad \operatorname{div} b = 0. \end{aligned} \tag{3.1}$$

The induced polarization and magnetization densities satisfy

$$p = d - e \qquad \text{and} \qquad m = b - h. \tag{3.2}$$

From eqs. (3.1) and (3.2) we find the wave equations for e and h,

$$\begin{aligned} \left(\operatorname{rot}\operatorname{rot} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right) e &= -\frac{1}{c^2}\frac{\partial^2}{\partial t^2} p - \frac{1}{c}\frac{\partial}{\partial t}\operatorname{rot} m, \\ \left(\operatorname{rot}\operatorname{rot} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right) h &= -\frac{1}{c^2}\frac{\partial^2}{\partial t^2} m + \frac{1}{c}\frac{\partial}{\partial t}\operatorname{rot} p. \end{aligned} \tag{3.3}$$

Defining the Fourier transform of a field $a(\boldsymbol{r}, t)$ by

$$a(\boldsymbol{k}, \omega) = \int d\boldsymbol{r}\, dt\, e^{-i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)} a(\boldsymbol{r}, t), \tag{3.4}$$

the wave equation (3.3) may be written as

$$\begin{aligned} \left[\boldsymbol{kk} - k^2 + \left(\frac{\omega}{c}\right)^2\right]\cdot \boldsymbol{e} &= -\left(\frac{\omega}{c}\right)^2 \boldsymbol{p} + \frac{\omega}{c}\boldsymbol{k} \wedge \boldsymbol{m}, \\ \left[\boldsymbol{kk} - k^2 + \left(\frac{\omega}{c}\right)^2\right]\cdot \boldsymbol{h} &= -\left(\frac{\omega}{c}\right)^2 \boldsymbol{m} - \frac{\omega}{c}\boldsymbol{k} \wedge \boldsymbol{p}. \end{aligned} \tag{3.5}$$

The formal retarded solution of these wave equations is

$$\begin{aligned} \boldsymbol{e} &= \boldsymbol{E}_{\mathrm{v}} - \boldsymbol{F}\cdot\left[\boldsymbol{p} - \left(\frac{\omega}{c}\right)^{-1} \boldsymbol{k} \wedge \boldsymbol{m}\right], \\ \boldsymbol{h} &= \boldsymbol{H}_{\mathrm{v}} - \boldsymbol{F}\cdot\left[\boldsymbol{m} + \left(\frac{\omega}{c}\right)^{-1} \boldsymbol{k} \wedge \boldsymbol{p}\right]. \end{aligned} \tag{3.6}$$

Here $\boldsymbol{E}_{\mathrm{v}}$ and $\boldsymbol{H}_{\mathrm{v}}$ are the vacuum fields. $\boldsymbol{F}$ is the retarded dipole propagator, which is diagonal in $\boldsymbol{k}, \omega$ representation, with diagonal elements given by

$$\begin{aligned} \boldsymbol{F}(\boldsymbol{k}, \omega) &\equiv \left(\frac{\omega}{c}\right)^2 \left[\boldsymbol{kk} - k^2 + \left(\frac{\omega}{c} + i0\right)^2\right]^{-1} \\ &= \left[k^2 - \left(\frac{\omega}{c} + i0\right)^2\right]^{-1} \left[\boldsymbol{kk} - \left(\frac{\omega}{c}\right)^2\right]. \end{aligned} \tag{3.7}$$

Here i0 is an infinitesimally small positive imaginary number.

In the regions occupied by the media with dielectric constants ϵ^+ and ϵ^-, one has

$$\boldsymbol{p}^{\pm}(\boldsymbol{r}, t) = (\epsilon^{\pm} - 1)\boldsymbol{e}(\boldsymbol{r}, t) \qquad \text{and} \qquad \boldsymbol{m}^{\pm}(\boldsymbol{r}, t) = 0, \tag{3.8}$$

since the magnetic permeability in both media was assumed to be unity. Due to the surface roughness and the presence of the film, one has a fluctuating excess polarization and magnetization density in this region of space[2]). As we have seen in section 2, these excess polarization and magnetization densities can be replaced[2]) by equivalent polarization and magnetization densities $\boldsymbol{p}^{\mathrm{s}}(\boldsymbol{r}_{\parallel}, t)\delta(z)$ and $\boldsymbol{m}^{\mathrm{s}}(\boldsymbol{r}_{\parallel}, t)\delta(z)$, where $\boldsymbol{p}^{\mathrm{s}}$ and $\boldsymbol{m}^{\mathrm{s}}$ are given by eq. (2.27). The total polarization and magnetization densities are therefore given by

$$\begin{aligned} \boldsymbol{p}(\boldsymbol{r}, t) &= \boldsymbol{p}^-(\boldsymbol{r}, t)\theta(-z) + \boldsymbol{p}^{\mathrm{s}}(\boldsymbol{r}_{\parallel}, t)\delta(z) + \boldsymbol{p}^+(\boldsymbol{r}, t)\theta(z), \\ \boldsymbol{m}(\boldsymbol{r}, t) &= \boldsymbol{m}^{\mathrm{s}}(\boldsymbol{r}_{\parallel}, t)\delta(z), \end{aligned} \tag{3.9}$$

where $\theta(z)$ is the Heaviside function.

As was discussed in refs. 5 and 2, the singular nature of $\boldsymbol{p}$ and $\boldsymbol{m}$ implies that d_x, d_y, e_z, b_x, b_y and h_z are also singular at $z = 0$. However, e_x, e_y, d_z, h_x, h_y and b_z are *not* singular in $z = 0$. These fields only have discontinuities at $z = 0$ which are given by eq. (2.15). In our analysis it is most convenient to use the non-singular fields, which were already defined in section 2, eq. (2.22). In order to write the formal solution of Maxwell's equations in terms of these non-singular fields, we define

$$\boldsymbol{K}(\boldsymbol{k}, \omega) \equiv \boldsymbol{F}(\boldsymbol{k}, \omega) - \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \tag{3.10}$$

$$\boldsymbol{L}(\boldsymbol{k}, \omega) \equiv \left(\frac{\omega}{c}\right)^{-1} \boldsymbol{F}(\boldsymbol{k}, \omega) \cdot \begin{pmatrix} 0 & -k_z & k_y \\ k_z & 0 & -k_x \\ -k_y & k_x & 0 \end{pmatrix}. \tag{3.11}$$

Eq. (3.6) then yields

$$\begin{aligned} \boldsymbol{n}_e &= \boldsymbol{E}_v - \boldsymbol{K} \cdot \boldsymbol{p} + \boldsymbol{L} \cdot \boldsymbol{m}, \\ \boldsymbol{n}_m &= \boldsymbol{H}_v - \boldsymbol{K} \cdot \boldsymbol{m} - \boldsymbol{L} \cdot \boldsymbol{p}. \end{aligned} \tag{3.12}$$

If $\boldsymbol{p}^s = \boldsymbol{m}^s = 0$, one finds from this equation the fields reflected and transmitted by the flat interface between the + and − medium,

$$\begin{aligned} \boldsymbol{N}_e^{(0)} &= \boldsymbol{E}_v - \boldsymbol{K} \cdot \boldsymbol{P}_0, \\ \boldsymbol{N}_m^{(0)} &= \boldsymbol{H}_v - \boldsymbol{L} \cdot \boldsymbol{P}_0, \end{aligned} \tag{3.13}$$

where

$$\boldsymbol{P}_0(\boldsymbol{r}, t) = \xi_0(z) \cdot \boldsymbol{N}_e^{(0)}(\boldsymbol{r}, t) \tag{3.14}$$

and

$$\xi_0(z) = \xi^- \theta(-z) + \xi^+ \theta(z), \quad \text{with } \xi^v = (\epsilon^v - 1) \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \dfrac{1}{\epsilon^v} \end{pmatrix}. \tag{3.15}$$

Substituting eq. (3.14) into eq. (3.13) one finds*

$$\begin{aligned} \boldsymbol{N}_e^{(0)} &= (1 + \boldsymbol{K} \cdot \xi_0)^{-1} \cdot \boldsymbol{E}_v, \\ \boldsymbol{N}_m^{(0)} &= \boldsymbol{H}_v - \boldsymbol{L} \cdot \xi_0 \cdot (1 + \boldsymbol{K} \cdot \xi_0)^{-1} \cdot \boldsymbol{E}_v. \end{aligned} \tag{3.16}$$

In order to find the expressions for the fields $\boldsymbol{n}_e$ and $\boldsymbol{n}_m$ in the general case when

* The symbol $(\ )^{-1}$ means that one has the inverse operator.

p^s and m^s are unequal to zero, we write eq. (3.9) in the following form:

$$\begin{aligned} p(r, t) &= \xi_0(z)\cdot n_e(r, t) + p^s(r_\parallel, t)\delta(z)\,, \\ m(r, t) &= m^s(r_\parallel, t)\delta(z)\,. \end{aligned} \tag{3.17}$$

Using this equation, together with eq. (3.16), one may write for eq. (3.12)

$$\begin{aligned} n_e &= N_e^{(0)} - K_0\cdot p^s\delta(z) + L_0\cdot m^s\delta(z)\,, \\ n_m &= N_m^{(0)} - L\cdot(1-\xi_0\cdot K_0)\cdot p^s\delta(z) - (K + L\cdot\xi_0\cdot L_0)\cdot m^s\delta(z)\,, \end{aligned} \tag{3.18}$$

where we have introduced the following propagators:

$$\begin{aligned} K_0 &\equiv (1 + K\cdot\xi_0)^{-1}\cdot K\,, \\ L_0 &\equiv (1 + K\cdot\xi_0)^{-1}\cdot L\,. \end{aligned} \tag{3.19}$$

In eq. (3.18) $K_0\cdot p^s\delta(z)$ is a short-hand notation for

$$\int K_0(r_\parallel, z, t\,|\,r'_\parallel, z', t')\cdot p^s(r'_\parallel, t')\delta(z')\,\mathrm{d}r'_\parallel\,\mathrm{d}z'\,\mathrm{d}t'$$

in coordinate representation, etc. Eqs. (3.18) with (3.19) give the fields n_e and n_m due to the surface polarization and magnetization densities p^s and m^s.

4. Surface constitutive coefficients

In this section we shall derive expressions for the surface constitutive coefficients, describing the optical behaviour of the thin film on the rough surface. These coefficients can be found be expressing the average surface polarization and magnetization densities

$$P^s \equiv \langle p^s\rangle \qquad \text{and} \qquad M^s \equiv \langle m^s\rangle \tag{4.1}$$

in terms of the average fields

$$N_e \equiv \langle n_e\rangle \qquad \text{and} \qquad N_m \equiv \langle n_m\rangle\,. \tag{4.2}$$

It follows from eqs. (2.27) and (4.1) that

$$\begin{aligned} P^s(r_\parallel, t) &= \sum_{i=1}^{3}\sum_{\nu}\langle\xi_b^{(i)\nu}(r_\parallel, t)\cdot n_e^\nu(r_\parallel, 0, t)\rangle + \sum_{\nu}\left\langle\xi_b^{(4)\nu}(r_\parallel, t)\cdot\frac{1}{c}\frac{\partial}{\partial t}n_m^\nu(r_\parallel, 0, t)\right\rangle, \\ M^s(r_\parallel, t) &= \sum_{\nu}\left\langle\xi_b^{(4)\nu}(r_\parallel, t)\cdot\frac{1}{c}\frac{\partial}{\partial t}n_e^\nu(r_\parallel, 0, t)\right\rangle. \end{aligned} \tag{4.3}$$

Since $\xi_b^{(i)\nu}(r_\parallel, t)$ is, for $i = 2$, 3 and 4, of second order in $f^\nu(r_\parallel, t)$ (according to eqs. (2.24)–(2.26)), we can replace n_e and n_m by $N_e^{(0)}$ and $N_m^{(0)}$ in the corresponding terms in eq.(4.3), as follows from eq. (3.18). Furthermore, since $\xi_b^{(1)\nu}(\boldsymbol{r}_\parallel, t)$ is of first order in $f^\nu(\boldsymbol{r}_\parallel, t)$ (according to eq. (2.23)), we can replace $\boldsymbol{n}_e$ in the term with $i = 1$ in eq. (4.3) by $\boldsymbol{N}_e^{(0)} - \boldsymbol{K}_0 \cdot \boldsymbol{p}^s\delta(z)$, as follows to the first order from eq. (3.18):

$$\begin{aligned}
\boldsymbol{n}_e^\nu(\boldsymbol{r}_\parallel, 0, t) &= \boldsymbol{N}_e^{(0)\nu}(\boldsymbol{r}_\parallel, 0, t) - \int \boldsymbol{K}_0(\boldsymbol{r}_\parallel, z = \nu 0, t \mid \boldsymbol{r}'_\parallel, z', t') \\
&\qquad \cdot \boldsymbol{p}^s(\boldsymbol{r}'_\parallel, t')\delta(z')\, d\boldsymbol{r}'_\parallel\, dz'\, dt' \\
&= \boldsymbol{N}_e^{(0)\nu}(\boldsymbol{r}_\parallel, 0, t) - \int \boldsymbol{K}_0(\boldsymbol{r}_\parallel, z = \nu 0, t \mid \boldsymbol{r}'_\parallel, z' = 0, t') \cdot \boldsymbol{p}^s(\boldsymbol{r}'_\parallel, t')\, d\boldsymbol{r}'_\parallel\, dt' \\
&= \boldsymbol{N}_e^{(0)\nu}(\boldsymbol{r}_\parallel, 0, t) - \sum_{\nu'} \int \boldsymbol{K}_0(\boldsymbol{r}_\parallel, z = \nu 0, t \mid \boldsymbol{r}'_\parallel, z' = 0, t') \cdot \xi_b^{(1)\nu'}(\boldsymbol{r}'_\parallel, t') \\
&\qquad \cdot \boldsymbol{N}_e^{(0)\nu'}(\boldsymbol{r}'_\parallel, 0, t')\, d\boldsymbol{r}'_\parallel\, dt' ,
\end{aligned} \tag{4.4}$$

where we have used again eqs. (2.27) and (3.18). Up to second order eq. (4.3) therefore becomes

$$\begin{aligned}
\boldsymbol{P}^s(\boldsymbol{r}_\parallel, t) &= \sum_{i=1}^{3} \sum_\nu \langle \xi_b^{(i)\nu}(\boldsymbol{r}_\parallel, t)\rangle \cdot \boldsymbol{N}_e^{(0)\nu}(\boldsymbol{r}_\parallel, 0, t) \\
&\quad - \sum_\nu \sum_{\nu'} \int \langle \xi_b^{(1)\nu}(\boldsymbol{r}_\parallel, t) \cdot \boldsymbol{K}_0(\boldsymbol{r}_\parallel, z = \nu 0, t \mid \boldsymbol{r}'_\parallel, z' = 0, t') \cdot \xi_b^{(1)\nu'}(\boldsymbol{r}'_\parallel, t')\rangle \\
&\quad \cdot \boldsymbol{N}_e^{(0)\nu'}(\boldsymbol{r}'_\parallel, 0, t')\, d\boldsymbol{r}'_\parallel\, dt' + \sum_\nu \langle \xi_b^{(4)\nu}(\boldsymbol{r}_\parallel, t)\rangle \cdot \frac{1}{c}\frac{\partial}{\partial t} \boldsymbol{N}_m^{(0)\nu}(\boldsymbol{r}_\parallel, 0, t) ,
\end{aligned} \tag{4.5}$$

$$\boldsymbol{M}^s(\boldsymbol{r}_\parallel, t) = \sum_\nu \langle \xi_b^{(4)}(\boldsymbol{r}_\parallel, t)\rangle \cdot \frac{1}{c}\frac{\partial}{\partial t} \boldsymbol{N}_e^{(0)\nu}(\boldsymbol{r}_\parallel, 0, t) ,$$

or briefly

$$\begin{aligned}
\boldsymbol{P}^s &= \sum_{i=1}^{3} \sum_\nu \langle \xi_b^{(i)\nu}\rangle \cdot \boldsymbol{N}_e^{(0)\nu} - \sum_\nu \sum_{\nu'} \langle \xi_b^{(1)\nu} \cdot \boldsymbol{K}_0 \cdot \xi_b^{(1)\nu'}\rangle \cdot \boldsymbol{N}_e^{(0)\nu'} + \sum_\nu \langle \xi_b^{(4)\nu}\rangle \cdot \frac{1}{c}\frac{\partial}{\partial t} \boldsymbol{N}_m^{(0)\nu} , \\
\boldsymbol{M}^s &= \sum_\nu \langle \xi_b^{(4)\nu}\rangle \cdot \frac{1}{c}\frac{\partial}{\partial t} \boldsymbol{N}_e^{(0)\nu} .
\end{aligned} \tag{4.6}$$

Now it follows from eqs. (4.2) and (4.4) that, up to first order,

$$\begin{aligned}
\boldsymbol{N}_e^{(0)\nu}(\boldsymbol{r}_\parallel, 0, t) &= \boldsymbol{N}_e^\nu(\boldsymbol{r}_\parallel, 0, t) + \sum_{\nu'} \int \boldsymbol{K}_0(\boldsymbol{r}_\parallel, z = \nu 0, t \mid \boldsymbol{r}'_\parallel, z' = 0, t') \\
&\qquad \cdot \langle \xi_b^{(1)\nu'}(\boldsymbol{r}'_\parallel, t')\rangle \cdot \boldsymbol{N}_e^{\nu'}(\boldsymbol{r}'_\parallel, 0, t')\, d\boldsymbol{r}'_\parallel\, dt' ,
\end{aligned} \tag{4.7}$$

which may be used to replace $\boldsymbol{N}_e^{(0)\nu}$ by $\boldsymbol{N}_e^\nu$ in the first order term $\Sigma_\nu \langle \xi_b^{(1)\nu}\rangle \cdot \boldsymbol{N}_e^{(0)\nu}$ at the right-hand side of eq. (4.5) (or (4.6)). In all other terms in this equation we may replace $N_e^{(0)\nu}$ and $N_m^{(0)\nu}$ simply by N_e^ν and N_m^ν, since these terms are all of second order. We can then write for eq. (4.6)[9]

$$\boldsymbol{P}^s = \sum_{i=1}^{3} \sum_{\nu} \langle \xi_b^{(i)\nu} \rangle \cdot \boldsymbol{N}_e^\nu - \sum_{\nu} \sum_{\nu'} \langle (\xi_b^{(1)\nu} - \langle \xi_b^{(1)\nu} \rangle) \cdot \boldsymbol{K}_0 \cdot (\xi_b^{(1)\nu'} - \langle \xi_b^{(1)\nu'} \rangle) \rangle \cdot \boldsymbol{N}_e^{\nu'}$$
$$+ \sum_{\nu} \langle \xi_b^{(4)\nu} \rangle \cdot \frac{1}{c} \frac{\partial}{\partial t} \boldsymbol{N}_m^\nu , \tag{4.8}$$
$$\boldsymbol{M}^s = \sum_{\nu} \langle \xi_b^{(4)\nu} \rangle \cdot \frac{1}{c} \frac{\partial}{\partial t} \boldsymbol{N}_e^\nu .$$

Defining

$$\xi_{se}^\nu = \sum_{i=1}^{3} \langle \xi_b^{(i)\nu} \rangle - \sum_{\nu'} \langle (\xi_b^{(1)\nu'} - \langle \xi_b^{(1)\nu'} \rangle) \cdot \boldsymbol{K}_0 \cdot (\xi_b^{(1)\nu} - \langle \xi_b^{(1)\nu} \rangle) \rangle ,$$
$$\xi_{sm}^\nu = \langle \xi_b^{(4)\nu} \rangle , \tag{4.9}$$

eq. (4.8) can be written in the form

$$\boldsymbol{P}^s = \sum_{\nu} \left(\xi_{se}^\nu \cdot \boldsymbol{N}_e^\nu + \xi_{sm}^\nu \cdot \frac{1}{c} \frac{\partial}{\partial t} \boldsymbol{N}_m^\nu \right),$$
$$\boldsymbol{M}^s = \sum_{\nu} \xi_{sm}^\nu \cdot \frac{1}{c} \frac{\partial}{\partial t} \boldsymbol{N}_e^\nu . \tag{4.10}$$

Comparing the last equation with eq. (1.1), we see that eq. (4.9) will provide expressions for the surface constitutive coefficients describing the dielectric (optical) behaviour of the thin film on a rough surface.

It follows with eq. (2.23) that

$$\langle \xi_b^{(1)\nu} \rangle = \nu(\epsilon - \epsilon^\nu) \langle f^\nu \rangle \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \dfrac{1}{\epsilon\epsilon^\nu} \end{pmatrix}, \tag{4.11}$$

where $\langle f^\nu \rangle$ is given by eq. (2.3). Furthermore, it follows with eqs. (2.24) and (2.26) that

$$\langle \xi_b^{(2)\nu} \rangle = \nu(\epsilon - \epsilon^\nu)\{\langle f^\nu \rangle^2 + \langle (\Delta f^\nu)^2 \rangle\} \begin{pmatrix} 0 & 0 & \dfrac{1}{\epsilon^\nu} \dfrac{\partial}{\partial x} \\ 0 & 0 & \dfrac{1}{\epsilon^\nu} \dfrac{\partial}{\partial y} \\ -\dfrac{1}{\epsilon} \dfrac{\partial}{\partial x} & -\dfrac{1}{\epsilon} \dfrac{\partial}{\partial y} & 0 \end{pmatrix}, \tag{4.12}$$

$$\langle \xi_b^{(4)\nu} \rangle = -\tfrac{1}{2}\nu(\epsilon - \epsilon^\nu)\{\langle f^\nu \rangle^2 + \langle (\Delta f^\nu)^2 \rangle\} \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{4.13}$$

where we have introduced the functions

$$\Delta f^\nu(\mathbf{r}_\parallel, t) \equiv f^\nu(\mathbf{r}_\parallel, t) - \langle f^\nu \rangle, \tag{4.14}$$

which describe the deviations of the surfaces $f^\nu(\mathbf{r}_\parallel, t)$ with respect to their averages. We now consider $\langle \xi_b^{(3)\nu} \rangle$, which according to eq. (2.25) will depend on $\langle \partial (f^\nu)^2/\partial x \rangle$ and $\langle \partial (f^\nu)^2/\partial y \rangle$. Since

$$\begin{aligned} \left\langle \frac{\partial}{\partial x}\{f^\nu(\mathbf{r}_\parallel, t)\}^2 \right\rangle &= \frac{\partial}{\partial x}\langle \{f^\nu(\mathbf{r}_\parallel, t)\}^2 \rangle, \\ \left\langle \frac{\partial}{\partial y}\{f^\nu(\mathbf{r}_\parallel, t)\}^2 \right\rangle &= \frac{\partial}{\partial y}\langle \{f^\nu(\mathbf{r}_\parallel, t)\}^2 \rangle, \end{aligned} \tag{4.15}$$

and since it follows from translational invariance in the x–y-plane that $\langle \{f^\nu(\mathbf{r}_\parallel, t)\}^2 \rangle$ does not depend on x and y, one finds that the averages, eq. (4.15) vanish, so that

$$\langle \xi_b^{(3)\nu} \rangle = 0 . \tag{4.16}$$

We now define the following four correlation functions for the deviations $\Delta f^\nu(\mathbf{r}_\parallel, t)$ of the positions of the two surfaces $f^\nu(\mathbf{r}_\parallel, t)$ from their averages $\langle f^\nu \rangle$:

$$S^{\nu\nu'}(\mathbf{r}_\parallel - \mathbf{r}'_\parallel, t - t') \equiv \langle \Delta f^\nu(\mathbf{r}_\parallel, t) \Delta f^{\nu'}(\mathbf{r}'_\parallel, t') \rangle , \tag{4.17}$$

where we have used translational invariance in the x–y-plane and stationarity. In terms of these correlation functions we can write for the $\mathbf{K}_0$ dependent term in eq. (4.9), using eqs. (2.23), (4.11) and (4.14),

$$\begin{aligned} &\sum_{\nu'} \langle (\xi_b^{(1)\nu'}(\mathbf{r}_\parallel, t) - \langle \xi_b^{(1)\nu'} \rangle) \cdot \mathbf{K}_0(\mathbf{r}_\parallel, z = \nu' 0, t \,|\, \mathbf{r}'_\parallel, z' = 0, t') \\ &\quad \cdot (\xi_b^{(1)\nu}(\mathbf{r}'_\parallel, t') - \langle \xi_b^{(1)\nu} \rangle) \rangle = \sum_{\nu'} \nu' \nu (\epsilon - \epsilon^{\nu'})(\epsilon - \epsilon^\nu) \\ &\quad \times S^{\nu'\nu}(\mathbf{r}_\parallel - \mathbf{r}'_\parallel, t - t') \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \dfrac{1}{\epsilon \epsilon^{\nu'}} \end{pmatrix} \\ &\quad \cdot \mathbf{K}_0(\mathbf{r}_\parallel, z = \nu' 0, t \,|\, \mathbf{r}'_\parallel, z' = 0, t') \cdot \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \dfrac{1}{\epsilon \epsilon^{\nu}} \end{pmatrix}. \end{aligned} \tag{4.18}$$

Since[6])

$$\boldsymbol{K}_0(\boldsymbol{r}_\parallel, z, t \mid \boldsymbol{r}'_\parallel, z', t') = \boldsymbol{K}_0(\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel, z, t - t' \mid z'), \tag{4.19}$$

we find that the expression eq. (4.18) depends on $\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel$ and $t - t'$ and therefore becomes diagonal after Fourier transformation, i.e. proportional to $\delta(\boldsymbol{k}_\parallel - \boldsymbol{k}'_\parallel)\delta(\omega - \omega')$

$$\sum_{\nu'} \langle (\xi_b^{(1)\nu'} - \langle \xi_b^{(1)\nu'} \rangle) \cdot \boldsymbol{K}_0 \cdot (\xi_b^{(1)\nu} - \langle \xi_b^{(1)\nu} \rangle) \rangle (\boldsymbol{k}_\parallel, \omega \mid \boldsymbol{k}'_\parallel, \omega')$$

$$= \delta(\boldsymbol{k}_\parallel - \boldsymbol{k}'_\parallel)\delta(\omega - \omega') \sum_{\nu'} \nu' \nu (\epsilon - \epsilon^{\nu'})(\epsilon - \epsilon^{\nu}) \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \frac{1}{\epsilon\epsilon^{\nu'}} \end{pmatrix}$$

$$\cdot \int \mathrm{d}\boldsymbol{k}''_\parallel \, \mathrm{d}\omega'' S^{\nu'\nu}(\boldsymbol{k}_\parallel - \boldsymbol{k}''_\parallel, \omega - \omega'') \boldsymbol{K}_0(\boldsymbol{k}''_\parallel, z = \nu' 0, \omega'' \mid z' = 0)$$

$$\cdot \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \frac{1}{\epsilon\epsilon^{\nu}} \end{pmatrix}, \tag{4.20}$$

where $S^{\nu'\nu}(\boldsymbol{k}_\parallel, \omega)$ is the Fourier transform of $S^{\nu'\nu}(\boldsymbol{r}_\parallel, t)$, eq. (4.17), and $\boldsymbol{K}_0(\boldsymbol{k}_\parallel, z, \omega \mid z')$ the (partial) Fourier transform of $\boldsymbol{K}_0(\boldsymbol{r}_\parallel, z, t \mid z')$, eq. (4.19). The other terms at the right-hand sides of eq. (4.9) are also diagonal in $\boldsymbol{k}_\parallel, \omega$ representation and are easily found using eqs. (4.11), (4.12) and (4.13). One then obtains for eq. (4.9) in $\boldsymbol{k}_\parallel$, ω representation

$$\xi^{\nu}_{\mathrm{e,m}}(\boldsymbol{k}_\parallel, \omega \mid \boldsymbol{k}'_\parallel, \omega') = \xi^{\nu}_{\mathrm{e,m}}(\boldsymbol{k}_\parallel, \omega)(2\pi)^3 \delta(\boldsymbol{k}_\parallel - \boldsymbol{k}'_\parallel)\delta(\omega - \omega'), \tag{4.21}$$

with

$$\xi^{\nu}_{\mathrm{e}}(\boldsymbol{k}_\parallel, \omega) = \nu(\epsilon - \epsilon^{\nu}) \langle f^{\nu} \rangle \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \frac{1}{\epsilon\epsilon^{\nu}} \end{pmatrix}$$

$$+ \mathrm{i}\nu(\epsilon - \epsilon^{\nu}) \{ \langle f^{\nu} \rangle^2 + \langle (\Delta f^{\nu})^2 \rangle \} \begin{pmatrix} 0 & 0 & \frac{1}{\epsilon^{\nu}} k_x \\ 0 & 0 & \frac{1}{\epsilon^{\nu}} k_y \\ -\frac{1}{\epsilon} k_x & -\frac{1}{\epsilon} k_y & 0 \end{pmatrix}$$

$$-\sum_{\nu'} \nu'\nu(\epsilon - \epsilon^{\nu'})(\epsilon - \epsilon^{\nu}) \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \frac{1}{\epsilon\epsilon^{\nu'}} \end{pmatrix} \tag{4.22}$$

$$\cdot (2\pi)^{-3} \int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega' S^{\nu'\nu}(\boldsymbol{k}_{\parallel} - \boldsymbol{k}'_{\parallel}, \omega - \omega') \boldsymbol{K}_0(\boldsymbol{k}'_{\parallel}, z = \nu' 0, \omega' \,|\, z' = 0)$$

$$\cdot \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \frac{1}{\epsilon\epsilon^{\nu}} \end{pmatrix},$$

$$\xi^{\nu}_{\mathrm{m}}(\boldsymbol{k}_{\parallel}, \omega) = -\tfrac{1}{2}\nu(\epsilon - \epsilon^{\nu})\{\langle f^{\nu}\rangle^2 + \langle(\Delta f^{\nu})^2\rangle\} \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

It follows with eq. (4.17) that

$$\langle(\Delta f^{\nu})^2\rangle = S^{\nu\nu}(\boldsymbol{r}_{\parallel} = 0, t = 0) = (2\pi)^{-3} \int \mathrm{d}\boldsymbol{k}_{\parallel}\, \mathrm{d}\omega S^{\nu\nu}(\boldsymbol{k}_{\parallel}, \omega) . \tag{4.23}$$

In ref. 7, eq. (2.2), we have found that

$$\boldsymbol{K}_0(\boldsymbol{k}_{\parallel}, z = \nu 0, \omega \,|\, z' = 0) = -\mathrm{i} \frac{k_{\perp}^{+} k_{\perp}^{-}}{\epsilon^{+} k_{\perp}^{-} + \epsilon^{-} k_{\perp}^{+}} \hat{\boldsymbol{k}}_{\parallel}\hat{\boldsymbol{k}}_{\parallel}$$
$$- \frac{\mathrm{i}(\omega/c)^2}{k_{\perp}^{+} + k_{\perp}^{-}} (1 - \hat{\boldsymbol{k}}_{\parallel}\hat{\boldsymbol{k}}_{\parallel} - \hat{\boldsymbol{z}}\hat{\boldsymbol{z}}) - \mathrm{i} \frac{\epsilon^{+}\epsilon^{-} k_{\parallel}^{2}}{\epsilon^{+} k_{\perp}^{-} + \epsilon^{-} k_{\perp}^{+}} \hat{\boldsymbol{z}}\hat{\boldsymbol{z}}$$
$$+ \frac{\mathrm{i}\nu}{\epsilon^{+} k_{\perp}^{-} + \epsilon^{-} k_{\perp}^{+}} (\epsilon^{-\nu} k_{\perp}^{\nu} \boldsymbol{k}_{\parallel}\hat{\boldsymbol{z}} + \epsilon^{\nu} k_{\perp}^{-\nu} \hat{\boldsymbol{z}}\boldsymbol{k}_{\parallel}) , \tag{4.24}$$

where $k_{\perp}^{\nu} \equiv \{\epsilon^{\nu}(\omega/c)^2 - k_{\parallel}^2\}^{1/2}$, $\hat{\boldsymbol{k}}_{\parallel} = \boldsymbol{k}_{\parallel}/k_{\parallel}$, with $k_{\parallel} = |\boldsymbol{k}_{\parallel}|$ and $\hat{\boldsymbol{z}} = (0, 0, 1)$. Using this expression in eq. (4.22) we find that one can write

$$\xi^{\nu}_{\mathrm{e}}(\boldsymbol{k}_{\parallel}, \omega) = \tfrac{1}{2}\gamma^{\nu}_{\mathrm{l}}(\boldsymbol{k}_{\parallel}, \omega)\hat{\boldsymbol{k}}_{\parallel}\hat{\boldsymbol{k}}_{\parallel} + \tfrac{1}{2}\gamma^{\nu}_{\mathrm{tr}}(1 - \hat{\boldsymbol{k}}_{\parallel}\hat{\boldsymbol{k}}_{\parallel} - \hat{\boldsymbol{z}}\hat{\boldsymbol{z}})$$
$$+ \tfrac{1}{2}\beta^{\nu}(\boldsymbol{k}_{\parallel}, \omega)\hat{\boldsymbol{z}}\hat{\boldsymbol{z}} - \tfrac{1}{2}\mathrm{i}\delta^{\nu}(\boldsymbol{k}_{\parallel}, \omega)\boldsymbol{k}_{\parallel}\hat{\boldsymbol{z}} + \tfrac{1}{2}\mathrm{i}\eta^{\nu}(\boldsymbol{k}_{\parallel}, \omega)\hat{\boldsymbol{z}}\boldsymbol{k}_{\parallel} \tag{4.25}$$

and (cf. eq. (1.1))

$$\xi^{\nu}_{\mathrm{m}}(\boldsymbol{k}_{\parallel}, \omega) = \tfrac{1}{2}\tau^{\nu} \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{4.26}$$

where

$$\gamma^{\nu}_{\parallel}((\boldsymbol{k}_{\parallel}, \omega) = 2\nu(\epsilon - \epsilon^{\nu})\langle f^{\nu}\rangle + 2\mathrm{i}(2\pi)^{-3}\sum_{\nu'} \nu'\nu(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'}) \int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega'$$

$$\times\ S^{\nu'\nu}(\boldsymbol{k}_{\parallel} - \boldsymbol{k}'_{\parallel}, \omega - \omega')\left\{\cos^2\phi\, \frac{q'^{+}_{\perp} q'^{-}_{\perp}}{\epsilon^{+} q'^{-}_{\perp} + \epsilon^{-} q'^{+}_{\perp}}\right.$$

$$\left. + \sin^2\phi\, \frac{(\omega'/c)^2}{q'^{+}_{\perp} + q'^{-}_{\perp}}\right\}, \tag{4.27}$$

$$\gamma^{\nu}_{\perp}(\boldsymbol{k}_{\parallel}, \omega) = 2\nu(\epsilon - \epsilon^{\nu})\langle f^{\nu}\rangle + 2\mathrm{i}(2\pi)^{-3}\sum_{\nu'} \nu'\nu(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'}) \int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega'$$

$$\times\ S^{\nu'\nu}(\boldsymbol{k}_{\parallel} - \boldsymbol{k}'_{\parallel}, \omega - \omega')\left\{\sin^2\phi\, \frac{q'^{+}_{\perp} q'^{-}_{\perp}}{\epsilon^{+} q'^{-}_{\perp} + \epsilon^{-} q'^{+}_{\perp}}\right.$$

$$\left. + \cos^2\phi\, \frac{(\omega'/c)^2}{q'^{+}_{\perp} + q'^{-}_{\perp}}\right\}, \tag{4.28}$$

$$\beta^{\nu}(\boldsymbol{k}_{\parallel}, \omega) = 2\nu\, \frac{\epsilon - \epsilon^{\nu}}{\epsilon\epsilon^{\nu}}\, \langle f^{\nu}\rangle + 2\mathrm{i}(2\pi)^{-3} \sum_{\nu'} \nu'\nu\, \frac{(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'})\epsilon^{+}\epsilon^{-}}{\epsilon^{\nu}\epsilon^{\nu'}\epsilon^{2}}$$

$$\times \int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega'\, S^{\nu'\nu}(\boldsymbol{k}_{\parallel} - \boldsymbol{k}'_{\parallel}, \omega - \omega')\, \frac{k'^{2}_{\parallel}}{\epsilon^{+} q'^{-}_{\perp} + \epsilon^{-} q'^{+}_{\perp}}\,. \tag{4.29}$$

$$\delta^{\nu}(\boldsymbol{k}_{\parallel}, \omega) = -2\nu\left(\frac{\epsilon - \epsilon^{\nu}}{\epsilon^{\nu}}\right)\left\{\langle f^{\nu}\rangle^2 + (2\pi)^{-3}\int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega'\, S^{\nu\nu}(\boldsymbol{k}'_{\parallel}, \omega')\right\}$$

$$+\ 2\nu(2\pi)^{-3}k_{\parallel}^{-1}\sum_{\nu'} \frac{(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'})\epsilon^{-\nu'}}{\epsilon\epsilon^{\nu}}$$

$$\times \int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega'\, S^{\nu'\nu}(\boldsymbol{k}_{\parallel} - \boldsymbol{k}'_{\parallel}, \omega - \omega')$$

$$\times\ k'_{\parallel}\cos\phi\, \frac{q'^{\nu'}_{\perp}}{\epsilon^{+} q'^{-}_{\perp} + \epsilon^{-} q'^{+}_{\perp}}\,, \tag{4.30}$$

$$\eta^{\nu}(\boldsymbol{k}_{\parallel}, \omega) = -2\nu\left(\frac{\epsilon - \epsilon^{\nu}}{\epsilon}\right)\left\{\langle f^{\nu}\rangle^2 + (2\pi)^{-3}\int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega'\, S^{\nu\nu}(\boldsymbol{k}'_{\parallel}, \omega')\right\}$$

$$-\ 2\nu(2\pi)^{-3}k_{\parallel}^{-1}\sum_{\nu'} \frac{(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'})}{\epsilon} \int \mathrm{d}\boldsymbol{k}'_{\parallel}\, \mathrm{d}\omega'\, S^{\nu'\nu}(\boldsymbol{k}_{\parallel} - \boldsymbol{k}'_{\parallel}, \omega - \omega')$$

$$\times\ k'_{\parallel}\cos\phi\, \frac{q'^{-\nu'}_{\perp}}{\epsilon^{+} q'^{-}_{\perp} + \epsilon^{-} q'^{+}_{\perp}} \tag{4.31}$$

and

$$\tau^{\nu}(\boldsymbol{k}_{\|}, \omega) = \tau^{\nu} = -\nu(\epsilon - \epsilon^{\nu})\left\{\langle f^{\nu}\rangle^2 + (2\pi)^{-3}\int \mathrm{d}\boldsymbol{k}'_{\|}\,\mathrm{d}\omega' S^{\nu\nu}(\boldsymbol{k}'_{\|}, \omega')\right\}, \tag{4.32}$$

with $q_{\perp}'^{\nu} \equiv \{\epsilon^{\nu}(\omega'/c)^2 - k_{\|}'^2\}^{1/2}$ and ϕ the angle between $\boldsymbol{k}_{\|}$ and $\boldsymbol{k}'_{\|}$ ($\hat{\boldsymbol{k}}_{\|}\cdot\hat{\boldsymbol{k}}'_{\|} \equiv \cos\phi$). (Note that $\langle f^-\rangle = 0$).

The coefficients* $\gamma_{\mathrm{l}}^{\nu}$, $\gamma_{\mathrm{tr}}^{\nu}$, β^{ν}, δ^{ν} and η^{ν} can in principle be evaluated, using the formulae eqs. (4.27)–(4.32), if the 4 correlation function $S^{\nu\nu'}(\boldsymbol{k}_{\|}, \omega)$ are known as well as the average film thickness $d = \langle f^+\rangle$ and the dielectric constants ϵ^{ν} and ϵ. In practice $S^{\nu\nu'}(\boldsymbol{k}_{\|}, \omega)$ may be replaced by $S^{\nu\nu'}(\boldsymbol{k}_{\|})\cdot 2\pi\delta(\omega)$. For solids this is evident since $S^{\nu\nu'}(\boldsymbol{r}_{\|}, t) = S^{\nu\nu'}(\boldsymbol{r}_{\|})$. Since for fluids the typical velocities of the interfaces are much smaller than the velocity of light, one may also use the equilibrium correlation function in eqs. (4.27)–(4.32). We shall furthermore assume rotational symmetry of these correlation functions around the z-axis, so that $S^{\nu\nu'}(\boldsymbol{r}_{\|}) = S^{\nu\nu'}(|\boldsymbol{r}_{\|}|)$ and therefore $S^{\nu\nu'}(\boldsymbol{k}_{\|}) = S^{\nu\nu'}(|\boldsymbol{k}_{\|}|)$. We may therefore put

$$S^{\nu\nu'}(\boldsymbol{k}_{\|}, \omega) = S^{\nu\nu'}(|\boldsymbol{k}_{\|}|)\cdot 2\pi\delta(\omega) \tag{4.33}$$

in eqs. (4.27)–(4.32). Introducing planar polar coordinates $k'_{\|}$ and ϕ (for $\boldsymbol{k}'_{\|}$), one can then write for eqs. (4.27)–(4.32), after integration over ω',

$$\begin{aligned} \gamma_{\mathrm{l}}^{\nu}(\boldsymbol{k}_{\|}, \omega) &= 2\nu(\epsilon - \epsilon^{\nu})\langle f^{\nu}\rangle + 2\mathrm{i}(2\pi)^{-2}\sum_{\nu'} \nu'\nu(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'}) \\ &\quad \times \int_0^{\infty} \mathrm{d}k'_{\|}\int_0^{2\pi} \mathrm{d}\phi\, k'_{\|} S^{\nu'\nu}(\kappa_{\|})\left\{\cos^2\phi\,\frac{q_{\perp}^{+}q_{\perp}^{-}}{\epsilon^{+}q_{\perp}^{-} + \epsilon^{-}q_{\perp}^{+}} + \sin^2\phi\,\frac{(\omega/c)^2}{q_{\perp}^{+} + q_{\perp}^{-}}\right\} \\ &\equiv 2\nu(\epsilon - \epsilon^{\nu})\langle f^{\nu}\rangle + \gamma_{\mathrm{l}}^{(\mathrm{c})\nu}(\boldsymbol{k}_{\|}, \omega), \end{aligned} \tag{4.34}$$

$$\begin{aligned} \gamma_{\mathrm{tr}}^{\nu}(\boldsymbol{k}_{\|}, \omega) &= 2\nu(\epsilon - \epsilon^{\nu})\langle f^{\nu}\rangle + 2\mathrm{i}(2\pi)^{-2}\sum_{\nu'} \nu'\nu(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'}) \\ &\quad \times \int_0^{\infty} \mathrm{d}k'_{\|}\int_0^{2\pi} \mathrm{d}\phi\, k'_{\|} S^{\nu'\nu}(\kappa_{\|})\left\{\sin^2\phi\,\frac{q_{\perp}^{+}q_{\perp}^{-}}{\epsilon^{+}q_{\perp}^{-} + \epsilon^{-}q_{\perp}^{+}} + \cos^2\phi\,\frac{(\omega/c)^2}{q_{\perp}^{+} + q_{\perp}^{-}}\right\} \\ &\equiv 2\nu(\epsilon - \epsilon^{\nu})\langle f^{\nu}\rangle + \gamma_{\mathrm{tr}}^{(\mathrm{c})\nu}(\boldsymbol{k}_{\|}, \omega), \end{aligned} \tag{4.35}$$

$$\begin{aligned} \beta^{\nu}(\boldsymbol{k}_{\|}, \omega) &= 2\nu\frac{\epsilon - \epsilon^{\nu}}{\epsilon\epsilon^{\nu}}\langle f^{\nu}\rangle + 2\mathrm{i}(2\pi)^{-2}\sum_{\nu'} \nu'\nu\,\frac{(\epsilon - \epsilon^{\nu})(\epsilon - \epsilon^{\nu'})\epsilon^{+}\epsilon^{-}}{\epsilon^{\nu}\epsilon^{\nu'}\epsilon^2} \\ &\quad \times \int_0^{\infty} \mathrm{d}k'_{\|}\int_0^{2\pi} \mathrm{d}\phi\, k_{\|}'^3 S^{\nu'\nu}(\kappa_{\|})\frac{1}{\epsilon^{+}q_{\perp}^{-} + \epsilon^{-}q_{\perp}^{+}} \\ &\equiv 2\nu\frac{\epsilon - \epsilon^{\nu}}{\epsilon\epsilon^{\nu}}\langle f^{\nu}\rangle + \beta^{(\mathrm{c})\nu}(\boldsymbol{k}_{\|}, \omega), \end{aligned} \tag{4.36}$$

* The subscripts l and tr of γ^{ν} stand for longitudinal and transversal (see also section 1).

$$\delta^{\nu}(k_{\|},\omega) = -2\nu\left(\frac{\epsilon-\epsilon^{\nu}}{\epsilon^{\nu}}\right)\left\{\langle f^{\nu}\rangle^2 + (2\pi)^{-1}\int_0^{\infty} \mathrm{d}k'_{\|}k'_{\|}S^{\nu\nu}(k'_{\|})\right\}$$

$$+ 2\nu(2\pi)^{-2}k_{\|}^{-1}\sum_{\nu'}\frac{(\epsilon-\epsilon^{\nu})(\epsilon-\epsilon^{\nu'})\epsilon^{-\nu'}}{\epsilon\epsilon^{\nu}}\int_0^{\infty}\mathrm{d}k'_{\|}\int_0^{2\pi}\mathrm{d}\phi k'^2_{\|}\cos\phi S^{\nu'\nu}(\kappa_{\|})$$

$$\times\frac{q^{\nu'}_{\perp}}{\epsilon^{+}q^{-}_{\perp}+\epsilon^{-}q^{+}_{\perp}} \equiv -2\nu\left(\frac{\epsilon-\epsilon^{\nu}}{\epsilon^{\nu}}\right)\langle f^{\nu}\rangle^2 + \delta^{(c)\nu}(k_{\|},\omega)\,, \tag{4.37}$$

$$\eta^{\nu}(k_{\|},\omega) = -2\nu\left(\frac{\epsilon-\epsilon^{\nu}}{\epsilon}\right)\left\{\langle f^{\nu}\rangle^2 + (2\pi)^{-1}\int_0^{\infty} \mathrm{d}k'_{\|}k'_{\|}S^{\nu\nu}(k'_{\|})\right\}$$

$$- 2\nu(2\pi)^{-2}k_{\|}^{-1}\sum_{\nu'}\frac{(\epsilon-\epsilon^{\nu})(\epsilon-\epsilon^{\nu'})}{\epsilon}\int_0^{\infty}\mathrm{d}k'_{\|}\int_0^{2\pi}\mathrm{d}\phi k'^2_{\|}\cos\phi S^{\nu'\nu}(\kappa_{\|})$$

$$\times\frac{q^{-\nu'}_{\perp}}{\epsilon^{+}q^{-}_{\perp}+\epsilon^{-}q^{+}_{\perp}} \equiv -2\nu\left(\frac{\epsilon-\epsilon^{\nu}}{\epsilon}\right)\langle f^{\nu}\rangle^2 + \eta^{(c)\nu}(k_{\|},\omega) \tag{4.38}$$

and

$$\tau^{\nu} = -\nu(\epsilon-\epsilon^{\nu})\left\{\langle f^{\nu}\rangle^2 + (2\pi)^{-1}\int_0^{\infty} \mathrm{d}k'_{\|}k'_{\|}S^{\nu\nu}(k'_{\|})\right\}$$

$$\equiv -\nu(\epsilon-\epsilon^{\nu})\langle f^{(\nu)}\rangle^2 + \tau^{(c)\nu}\,, \tag{4.39}$$

with

$$\kappa_{\|} \equiv (k_{\|}^2 + k'^2_{\|} - 2k_{\|}k'_{\|}\cos\phi)^{1/2} \quad \text{and} \quad q^{\nu}_{\perp} \equiv \{\epsilon^{\nu}(\omega/c)^2 - k'^2_{\|}\}^{1/2}.$$

We find that the coefficients $\gamma^{\nu}_{\|}$, γ^{ν}_{tr}, β^{ν}, δ^{ν} and η^{ν} are functions of $k_{\|}$ and ω, so that $\xi^{\nu}_{\text{sc}}(k_{\|},\omega)$, eq. (4.25), is now exactly of the form expected on symmetry grounds, i.e. rotational symmetry around the z-axis and translational symmetry (see eq. (1.2)).

We see that the constitutive coefficients $\gamma^{\nu}_{\|}$, γ^{ν}_{tr}, β^{ν}, δ^{ν}, η^{ν} and τ^{ν} are the sums of two contributions: one from the fact that there is material in the form of a thin film (with average thickness d) on the rough surface, and the other ($\gamma^{(c)\nu}_{\|}$, $\gamma^{(c)\nu}_{\text{tr}}$, $\beta^{(c)\nu}$, $\delta^{(c)\nu}$, $\eta^{(c)\nu}$ and $\tau^{(c)\nu}$) from the correlations of the roughnesses of the upper and lower surface of the film. For a further evaluation of the integrals in eqs. (4.34)–(4.39) one needs explicit expressions for the four correlation function $S^{\nu\nu'}(k_{\|})$. Such calculations will be given in a future publication.

We finally want to remark that, by choosing $\epsilon = \epsilon^{+}$ and calling $f^{-} = f$ in eqs. (4.34)–(4.39), one must obtain the same results for $\gamma_{\|}(k_{\|},\omega) \equiv \frac{1}{2}\Sigma_{\nu}\gamma^{\nu}_{\|}(k_{\|},\omega)$,

$\gamma_{\mathrm{u}}(k_\parallel,\omega)\equiv\frac{1}{2}\Sigma_\nu\gamma_{\mathrm{u}}^\nu(k_\parallel,\omega)$, $\beta(k_\parallel,\omega)\equiv\frac{1}{2}\Sigma_\nu\beta(k_\parallel,\omega)$ and $\tau\equiv\frac{1}{2}\Sigma_\nu\tau^\nu$ as in ref. 7, eq. (1.4), whereas $\delta(k_\parallel,\omega)\equiv\frac{1}{2}\Sigma_\nu\delta^\nu(k_\parallel,\omega)$ and $\eta(k_\parallel,\omega)\equiv\frac{1}{2}\Sigma_\nu\eta^\nu(k_\parallel,\omega)$ must appear to be equal, and also given by eq. (1.4) of that reference. For γ_{l}, γ_{u}, β and τ this is immediately clear. One also easily proves the equality of δ and η. By considering a suitable linear combination of δ and η one then proves the equivalence with the expression given for $\delta(k_\parallel,\omega)$ in eq. (1.4) of ref. 7.

5. Reflectance, transmittance and ellipsometric coefficient

In this section we shall study how the optical properties of a surface are changed by roughness and the presence of a thin film. To this end we shall calculate the influence of the constitutive coefficients γ_{l}^ν, γ_{u}^ν, β^ν, δ^ν, η^ν and τ^ν, eqs. (4.34)–(4.39), on the reflection and transmission amplitudes of a light wave. We shall first consider the case where the polarization of the incident-beam is normal to the plane of incidence, which is chosen to be the x–z-plane. The incident fields are then given by

$$\begin{aligned}
&\boldsymbol{E}_{\mathrm{i}}(\boldsymbol{r},t)=(0,1,0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_{\mathrm{i}}\cdot\boldsymbol{r}-\omega t)}\,,\\
&\boldsymbol{B}_{\mathrm{i}}(\boldsymbol{r},t)=n^-(-\cos\theta_{\mathrm{i}},0,\sin\theta_{\mathrm{i}})\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_{\mathrm{i}}\cdot\boldsymbol{r}-\omega t)}\,,\quad \text{for } z<0\,,
\end{aligned}\tag{5.1}$$

where $n^-=\sqrt{\epsilon^-}$ is the refractive index, ω the frequency and $\boldsymbol{k}_{\mathrm{i}}=n^-\omega/c(\sin\theta_{\mathrm{i}},0,\cos\theta_{\mathrm{i}})$ the wave-vector of the incident light, whereas θ_{i} is the angle of incidence. The amplitude has been set equal to unity, which obviously does not affect the values of the optical quantities, to be calculated. The reflected and transmitted fields are given by

$$\begin{aligned}
&\boldsymbol{E}_{\mathrm{r}}(\boldsymbol{r},t)=r_{\mathrm{s}}(0,1,0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_{\mathrm{r}}\cdot\boldsymbol{r}-\omega t)}\,,\\
&\boldsymbol{B}_{\mathrm{r}}(\boldsymbol{r},t)=n^-r_{\mathrm{s}}(\cos\theta_{\mathrm{r}},0,\sin\theta_{\mathrm{r}})\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_{\mathrm{r}}\cdot\boldsymbol{r}-\omega t)}\,,\quad \text{for } z<0\,,
\end{aligned}\tag{5.2}$$

and

$$\begin{aligned}
&\boldsymbol{E}_{\mathrm{t}}(\boldsymbol{r},t)=t_{\mathrm{s}}(0,1,0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_{\mathrm{t}}\cdot\boldsymbol{r}-\omega t)}\,,\\
&\boldsymbol{B}_{\mathrm{t}}(\boldsymbol{r},t)=n^+t_{\mathrm{s}}(-\cos\theta_{\mathrm{t}},0,\sin\theta_{\mathrm{t}})\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_{\mathrm{t}}\cdot\boldsymbol{r}-\omega t)}\,,\quad \text{for } z>0\,,
\end{aligned}\tag{5.3}$$

where $n^+=\sqrt{\epsilon^+}$, $\boldsymbol{k}_{\mathrm{r}}=n^-\omega/c(\sin\theta_{\mathrm{r}},0,-\cos\theta_{\mathrm{r}})$ is the wave-vector of the reflected wave (θ_{r} = angle of reflection) and $\boldsymbol{k}_{\mathrm{t}}=n^+\omega/c(\sin\theta_{\mathrm{t}},0,\cos\theta_{\mathrm{t}})$ the wave-vector of the transmitted wave (θ_{t} = angle of transmission). Only in the case of real n^+ the angle θ_{t} is real. The case of complex n^+ will be considered below after eq. (5.9).

Substituting

$$\boldsymbol{N}_\epsilon^-=\lim_{z\uparrow0}(E_{\mathrm{i},x}+E_{\mathrm{r},x},\,E_{\mathrm{i},y}+E_{\mathrm{r},y},\,\epsilon^-E_{\mathrm{i},z}+\epsilon^-E_{\mathrm{r},z})\,,$$

$$N_e^+ = \lim_{z\downarrow 0}(E_{t,x}, E_{t,y}, \epsilon^+ E_{t,z})\,, \tag{5.4}$$

$$N_m^- = \lim_{z\uparrow 0}(\boldsymbol{B}_i + \boldsymbol{B}_r),\ N_m^+ = \lim_{z\downarrow 0} \boldsymbol{B}_t\,.$$

into the right-hand sides of the constitutive equations (4.10), we obtain, with eqs. (4.25), (4.26) and (5.1)–(5.3), the surface polarization and magnetization densities $\boldsymbol{P}^s$ and $\boldsymbol{M}^s$, induced by the plane wave. These densities can then be substituted into the right-hand sides of the equations for the discontinuities of the fields

$$\begin{aligned}
N_{e,x}^+ - N_{e,x}^- &= -\frac{1}{c}\frac{\partial}{\partial t} M_y^s - \frac{\partial}{\partial x} P_x^s\,,\\
N_{e,y}^+ - N_{e,y}^- &= \frac{1}{c}\frac{\partial}{\partial t} M_x^s - \frac{\partial}{\partial y} P_z^s\,,\\
N_{e,z}^+ - N_{e,z}^- &= -\frac{\partial}{\partial x} P_x^s - \frac{\partial}{\partial y} P_y^s\,,\\
N_{m,x}^+ - N_{m,x}^- &= \frac{1}{c}\frac{\partial}{\partial t} P_y^s - \frac{\partial}{\partial x} M_z^s\,,\\
N_{m,y}^+ - N_{m,y}^- &= -\frac{1}{c}\frac{\partial}{\partial t} P_x^s - \frac{\partial}{\partial y} M_z^s\,,\\
N_{m,z}^+ - N_{m,z}^- &= -\frac{\partial}{\partial x} M_x^s - \frac{\partial}{\partial y} M_y^s\,,
\end{aligned} \tag{5.5}$$

which follow from eq. (2.15) together with eqs. (2.22), (4.1) and (4.2). Using again eq. (5.4), with eqs. (5.1)–(5.3), in the left-hand sides of eq. (5.5), one obtains a set of two independent equations to determine r_s, t_s, θ_r and θ_t. It then follows that θ_r and θ_t are given as usual by Snell's law

$$\theta_r = \theta_i \equiv \theta \qquad \text{and} \qquad n^+ \sin\theta_t = n^- \sin\theta_i \tag{5.6}$$

and are consequently not affected by the constitutive coefficients $\gamma_{\parallel}^v$, γ_{tt}^v, β^v, δ^v, η^v and τ^v. For r_s and t_s one finds, up to second order in the average film thickness and surface roughness,

$$\begin{aligned}
r_s = r_s^0\{&1 + 2\mathrm{i}(\omega/c)\gamma_{\text{tt}} n^-(\epsilon^- - \epsilon^+)^{-1}\cos\theta\\
&+ 4(\omega/c)^2 \tau n^- n^+(\epsilon^- - \epsilon^+)^{-1}\cos\theta\cos\theta_t\\
&- 2(\omega/c)^2\gamma_{\text{tt}}^2(\epsilon^- - \epsilon^+)^{-1} n^- \cos\theta(n^-\cos\theta + n^+\cos\theta_t)^{-1}\},\\
t_s = t_s^0\{&1 + \mathrm{i}(\omega/c)\gamma_{\text{tt}}(n^-\cos\theta + n^+\cos\theta_t)^{-1}\\
&- (\omega/c)^2\tau(n^-\cos\theta - n^+\cos\theta_t)(n^-\cos\theta + n^+\cos\theta_t)^{-1}\\
&- (\omega/c)^2\gamma_{\text{tt}}^2(n^-\cos\theta + n^+\cos\theta_t)^{-2}\}\,,
\end{aligned} \tag{5.7}$$

with the Fresnel amplitudes

$$r_s^0 \equiv (n^- \cos\theta - n^+ \cos\theta_t)(n^- \cos\theta + n^+ \cos\theta_t)^{-1}\,,$$
$$t_s^0 \equiv 2n^- \cos\theta(n^- \cos\theta + n^+ \cos\theta_t)^{-1} \tag{5.8}$$

and $\tau \equiv \frac{1}{2}(\tau^+ + \tau^-)$ and $\gamma_{tr} \equiv \frac{1}{2}(\gamma_{tr}^+ + \gamma_{tr}^-)$, (see end of section 4).

For complex refractive index n^+ the angle of transmittance θ_t is no longer real and $n^+ \cos\theta_t$ has to be replaced by $u + \mathrm{i}v$ in the above formulae,

$$n^+ \cos\theta_t = u + \mathrm{i}v\,. \tag{5.9}$$

From eqs. (5.6) and (5.9) one then obtains

$$\epsilon^+ = n^{+2} = n^{+2}\sin^2\theta_t + n^{+2}\cos^2\theta_t = n^{-2}\sin^2\theta + (u + \mathrm{i}v)^2$$
$$= \epsilon^- \sin^2\theta + u^2 - v^2 + 2\mathrm{i}uv\,. \tag{5.10}$$

Introducing

$$\rho^+ \equiv \mathrm{Re}\,\epsilon^+\,, \qquad \sigma^+ \equiv \mathrm{Im}\,\epsilon^+\,, \tag{5.11}$$

we get

$$\rho^+ = \epsilon^- \sin^2\theta + u^2 - v^2\,, \qquad \sigma^+ = 2uv\,, \tag{5.12}$$

so that

$$u = \tfrac{1}{2}\sqrt{2}\sqrt{(\rho^+ - \epsilon^- \sin^2\theta) + \sqrt{(\rho^2 - \epsilon^- \sin^2\theta)^2 + \sigma^{+2}}}\,,$$
$$v = \tfrac{1}{2}\sqrt{2}\sqrt{-(\rho^+ - \epsilon^- \sin^2\theta) + \sqrt{(\rho^+ - \epsilon^- \sin^2\theta)^2 + \sigma^{+2}}}\,. \tag{5.13}$$

Next we consider the case, where the incident wave is polarized parallel to the plane of incidence. Then the incident, reflected and transmitted fields are given by

$$\boldsymbol{E}_i(\boldsymbol{r}, t) = (\cos\theta_i, 0, -\sin\theta_i)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i\cdot\boldsymbol{r} - \omega t)}\,,$$
$$\boldsymbol{B}_i(\boldsymbol{r}, t) = n^-(0, 1, 0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i\cdot\boldsymbol{r} - \omega t)}\,, \quad \text{for } z < 0\,, \tag{5.14}$$

$$\boldsymbol{E}_r(\boldsymbol{r}, t) = r_p(-\cos\theta_r, 0, -\sin\theta_r)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_r\cdot\boldsymbol{r} - \omega t)}\,,$$
$$\boldsymbol{B}_r(\boldsymbol{r}, t) = n^- r_p(0, 1, 0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_r\cdot\boldsymbol{r} - \omega t)}\,, \quad \text{for } z < 0\,, \tag{5.15}$$

$$\boldsymbol{E}_t(\boldsymbol{r}, t) = t_p(\cos\theta_t, 0, -\sin\theta_t)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_t\cdot\boldsymbol{r} - \omega t)}\,,$$
$$\boldsymbol{B}_t(\boldsymbol{r}, t) = n^+ t_p(0, 1, 0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_t\cdot\boldsymbol{r} - \omega t)}\,, \quad \text{for } z > 0\,, \tag{5.16}$$

where the angles and wave-vectors are identical to those for the case of s-polarized light. Proceeding along the same lines as above, we again find the relation eq. (5.6), whereas the amplitudes r_p and t_p are, up to second order in the film thickness and surface roughness, given by

$$
\begin{aligned}
r_{\mathrm{p}} = r_{\mathrm{p}}^{0}\{ & 1 + 2\mathrm{i}(\omega/c)\gamma_{\mathrm{l}} n^{-} \cos\theta \cos^{2}\theta_{\mathrm{t}} (\epsilon^{-} \cos^{2}\theta_{\mathrm{t}} - \epsilon^{+} \cos^{2}\theta)^{-1} \\
& - 2\mathrm{i}(\omega/c)\beta n^{-}\epsilon^{+}\epsilon^{-} \cos\theta \sin^{2}\theta (\epsilon^{-} \cos^{2}\theta_{\mathrm{t}} - \epsilon^{+} \cos^{2}\theta)^{-1} \\
& + 4(\omega/c)^{2}\tau n^{+} n^{-} \cos\theta \cos\theta_{\mathrm{t}} (\epsilon^{-} \cos^{2}\theta_{\mathrm{t}} - \epsilon^{+} \cos^{2}\theta)^{-1} \\
& - 2(\omega/c)^{2}(\delta + \eta) n^{+} n^{-} \epsilon^{-} \cos\theta \cos\theta_{\mathrm{t}} \sin^{2}\theta (\epsilon^{-} \cos^{2}\theta_{\mathrm{t}} - \epsilon^{+} \cos^{2}\theta)^{-1} \\
& - 2(\omega/c)^{2}\gamma_{\mathrm{l}}^{2} n^{-} \cos^{2}\theta \cos^{3}\theta_{\mathrm{t}} (\epsilon^{-} \cos^{2}\theta_{\mathrm{t}} - \epsilon^{+} \cos^{2}\theta)^{-1} \\
& \times (n^{-} \cos\theta_{\mathrm{t}} + n^{-} \cos\theta)^{-1} \\
& + 2(\omega/c)^{2}\beta^{2} n^{+} \epsilon^{+} (\epsilon^{-})^{3} \sin^{4}\theta \cos\theta (\epsilon^{-} \cos^{2}\theta_{\mathrm{t}} - \epsilon^{+} \cos^{2}\theta)^{-1} \\
& \times (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-1} \\
& - 2(\omega/c)^{2}\gamma_{\mathrm{l}}\beta n^{+} n^{-} \epsilon^{-} \cos\theta \cos\theta_{\mathrm{t}} \sin^{2}\theta (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-2}\}, \\
t_{\mathrm{p}} = t_{\mathrm{p}}^{0}\{ & 1 + \mathrm{i}(\omega/c)\gamma_{\mathrm{l}} \cos\theta \cos\theta_{\mathrm{t}} (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-1} \\
& + \mathrm{i}(\omega/c)\beta n^{+} n^{-} \epsilon^{-} \sin^{2}\theta (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-1} \\
& - (\omega/c)^{2}\tau (n^{-} \cos\theta_{\mathrm{t}} - n^{+} \cos\theta)(n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-1} \\
& - (\omega/c)^{2}\delta\epsilon^{-} n^{+} \sin^{2}\theta \cos\theta (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-1} \\
& + (\omega/c)^{2}\eta\epsilon^{-} n^{-} \sin^{2}\theta \cos\theta_{\mathrm{t}} (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-1} \\
& - (\omega/c)^{2}\gamma_{\mathrm{l}}^{2} \cos^{2}\theta \cos^{2}\theta_{\mathrm{t}} (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-2} \\
& - (\omega/c)^{2}\beta^{2}\epsilon^{+}(\epsilon^{-})^{3} \sin^{4}\theta (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-2} \\
& - 2(\omega/c)^{2}\gamma_{\mathrm{l}}\beta n^{+} n^{-} \epsilon^{-} \sin^{2}\theta \cos\theta \cos\theta_{\mathrm{t}} (n^{-} \cos\theta_{\mathrm{t}} + n^{+} \cos\theta)^{-2}\},
\end{aligned}
\tag{5.17}
$$

with the Fresnel amplitudes

$$
\begin{aligned}
r_{\mathrm{p}}^{0} &\equiv (n^{+} \cos\theta - n^{-} \cos\theta_{\mathrm{t}})(n^{+} \cos\theta + n^{-} \cos\theta_{\mathrm{t}})^{-1}, \\
t_{\mathrm{p}}^{0} &\equiv 2n^{-} \cos\theta (n^{+} \cos\theta + n^{-} \cos\theta_{\mathrm{t}})^{-1}.
\end{aligned}
\tag{5.18}
$$

One should note that only the symmetric combinations γ_{l}, γ_{tt}, β, δ, η and τ (see end of section 4) appear in the amplitudes r_{s}, t_{s}, eq. (5.7) and r_{p}, t_{p}, eq. (5.17).

If we use eqs. (4.34)–(4.39), the expressions eqs. (5.7) and (5.17) can be written as

$$
\begin{aligned}
r_{\mathrm{s}} = r_{\mathrm{s}}^{0}(d) + r_{\mathrm{s}}^{0}\{ & 2\mathrm{i}(\omega/c)\gamma_{\mathrm{tt}}^{(\mathrm{c})} n^{-} (\epsilon^{-} - \epsilon^{+})^{-1} \cos\theta \\
& + 4(\omega/c)^{2}\tau^{(\mathrm{c})} n^{-} n^{+} (\epsilon^{-} - \epsilon^{+})^{-1} \cos\theta \cos\theta_{\mathrm{t}}\}, \\
t_{\mathrm{s}} = t_{\mathrm{s}}^{0}(d) + t_{\mathrm{s}}^{0}\{ & \mathrm{i}(\omega/c)\gamma_{\mathrm{tt}}^{(\mathrm{c})} (n^{-} \cos\theta + n^{+} \cos\theta_{\mathrm{t}})^{-1} \\
& - (\omega/c)^{2}\tau^{(\mathrm{c})} (n^{-} \cos\theta - n^{+} \cos\theta_{\mathrm{t}})(n^{-} \cos\theta + n^{+} \cos\theta_{\mathrm{t}})^{-1}\}
\end{aligned}
\tag{5.19}
$$

and

$$
\begin{aligned}
r_{\mathrm{p}} = r_{\mathrm{p}}^0(d) + r_{\mathrm{p}}^0\{&2\mathrm{i}(\omega/c)\gamma_{\mathrm{l}}^{(\mathrm{c})}n^- \cos\theta \cos^2\theta_{\mathrm{t}}(\epsilon^- \cos^2\theta_{\mathrm{t}} - \epsilon^+ \cos^2\theta)^{-1} \\
&- 2\mathrm{i}(\omega/c)\beta^{(\mathrm{c})}n^-\epsilon^+\epsilon^- \cos\theta \sin^2\theta(\epsilon^- \cos^2\theta_{\mathrm{t}} - \epsilon^+ \cos^2\theta)^{-1} \\
&+ 4(\omega/c)^2(\tau^{(\mathrm{c})} - \delta^{(\mathrm{c})}\epsilon^- \sin^2\theta)n^+n^- \cos\theta \cos\theta_{\mathrm{t}} \\
&\times (\epsilon^- \cos^2\theta_{\mathrm{t}} - \epsilon^+ \cos^2\theta)^{-1}\}\,, \\
t_{\mathrm{p}} = t_{\mathrm{p}}^0(d) + t_{\mathrm{p}}^0\{&\mathrm{i}(\omega/c)\gamma_{\mathrm{l}}^{(\mathrm{c})} \cos\theta \cos\theta_{\mathrm{t}}(n^- \cos\theta_{\mathrm{t}} + n^+ \cos\theta)^{-1} \\
&+ \mathrm{i}(\omega/c)\beta^{(\mathrm{c})}n^+n^-\epsilon^- \sin^2\theta(n^- \cos\theta_{\mathrm{t}} + n^+ \cos\theta)^{-1} \\
&- (\omega/c)^2(\tau^{(\mathrm{c})} - \delta^{(\mathrm{c})}\epsilon^- \sin^2\theta)(n^- \cos\theta_{\mathrm{t}} - n^+ \cos\theta) \\
&\times (n^- \cos\theta_{\mathrm{t}} + n^+ \cos\theta)^{-1}\}\,,
\end{aligned} \tag{5.20}
$$

where we have used the fact that the coefficients $\gamma_{\mathrm{l}}^{(\mathrm{c})}$, $\gamma_{\mathrm{tr}}^{(\mathrm{c})}$, $\beta^{(\mathrm{c})}$, $\delta^{(\mathrm{c})}$ and $\tau^{(\mathrm{c})}$ are of second order in the surface roughness and that $\delta^{(\mathrm{c})} = \eta^{(\mathrm{c})}$. The latter equality can be proved in a completely analogous way as was indicated in the last paragraph of section 4 for a rough surface without film. The quantities $r_{\mathrm{s}}^0(d)$, $t_{\mathrm{s}}^0(d)$, $r_{\mathrm{p}}^0(d)$ and $t_{\mathrm{p}}^0(d)$ denote the reflection and transmission amplitudes for a plane parallel film of thickness d. One can easily check that one has here second order approximations of the exact expressions for these amplitudes, given e.g. in ref. 8.

The reflectance and transmittance can be found by dividing the normal component of the Poynting-vector of the reflected and the transmitted beam by the normal component of the Poynting-vector of the incident beam. In this way one obtains

$$
\begin{aligned}
&R_{\mathrm{s}} = |r_{\mathrm{s}}|^2\,, \qquad R_{\mathrm{p}} = |r_{\mathrm{p}}|^2\,, \\
&T_{\mathrm{s}} = |t_{\mathrm{s}}|^2 \frac{u}{n^- \cos\theta}\, \mathrm{e}^{-2v(\omega/c)z}\,, \qquad T_{\mathrm{p}} = |t_{\mathrm{p}}|^2 \frac{(u\rho^+ + v\sigma^+)}{|\epsilon^+|n^- \cos\theta}\, \mathrm{e}^{-2v(\omega/c)z}\,.
\end{aligned} \tag{5.21}
$$

The attenuation of the transmitted waves is caused by the absorption in the substrate. The loss of energy from the beams, by absorption in the film and by scattering by the surfaces, is given by

$$
Q_{\mathrm{s,p}} \equiv 1 - R_{\mathrm{s,p}} - T_{\mathrm{s,p}}\,, \tag{5.22}
$$

where the value of z in the transmittance has to be set equal to d, the average position of the interface between the film and the substrate (see fig. 1).

The ellipsometric coefficient is defined by

$$
r \equiv r_{\mathrm{p}}/r_{\mathrm{s}} \equiv \rho \exp \mathrm{i}\Delta\,. \tag{5.23}
$$

Substituting the expressions, eqs. (5.19) and (5.20), for the amplitudes r_{s}, t_{s}, r_{p} and t_{p} into eqs. (5.21), (5.22) and (5.23), one obtains for the reflectances R_{s} and R_{p}, the transmittances T_{s} and T_{p}, the energy losses Q_{s} and Q_{p}, and the ellipsometric coefficient r,

$$R_s = R_s^0(d) - 4(\omega/c)n^- \cos\theta\{(n^-\cos\theta + u)^2 + v^2\}^{-2}$$
$$\times [(\epsilon^- - \rho^+)\,\mathrm{Im}\,\gamma_{tt}^{(c)} + \sigma^+\,\mathrm{Re}\,\gamma_{tt}^{(c)}$$
$$- 2(\omega/c)\{(\epsilon^- u - \rho^+ u - \sigma^+ v)\,\mathrm{Re}\,\tau^{(c)} - (\epsilon^- v - \rho^+ v + \sigma^+ u)\,\mathrm{Im}\,\tau^{(c)}\}], \tag{5.24}$$

$$T_s = T_s^0(d) - 8(\omega/c)n^- \cos\theta\{(n^-\cos\theta + u)^2 + v^2\}^{-1} u\, e^{-2v(\omega/c)z}$$
$$\times [(n^-\cos\theta + u)\,\mathrm{Im}\,\gamma_{tt}^{(c)} - v\,\mathrm{Re}\,\gamma_{tt}^{(c)}$$
$$+ (\omega/c)\{(\epsilon^- - \rho^+ - 2v^2)\,\mathrm{Re}\,\tau^{(c)} + 2n^-\cos\theta\, v\,\mathrm{Im}\,\tau^{(c)}\}], \tag{5.25}$$

$$Q_s = Q_s^0(d) + 4(\omega/c)n^-\cos\theta\{(n^-\cos\theta + u)^2 + v^2\}^{-1}$$
$$\times \{\mathrm{Im}\,\gamma_{tt}^{(c)} + 2(\omega/c)v\,\mathrm{Im}\,\tau^{(c)}\}, \tag{5.26}$$

$$R_p = R_p^0(d) - 4(\omega/c)n^-\cos\theta\{(n^- u + \rho^+\cos\theta)^2 + (n^- v + \sigma^+\cos\theta)^2\}^{-2}$$
$$\times [\{\epsilon^-(u^2+v^2)^2 - \cos^2\theta(u^2 - v^2)(\rho^{+2} - \sigma^{+2})$$
$$- 4\cos^2\theta\, uv\rho^+\sigma^+\}\,\mathrm{Im}\,\gamma_l^{(c)} + 2\cos^2\theta\{(u^2 - v^2)\rho^+\sigma^+$$
$$- uv(\rho^{+2} - \sigma^{+2})\}\,\mathrm{Re}\,\gamma_l^{(c)} + \epsilon^-\sin^2\theta\{(\rho^{+2} + \sigma^{+2})^2\cos^2\theta$$
$$- \epsilon^-(\rho^{+2} - \sigma^{+2})(u^2 - v^2) - 4\epsilon^- uv\rho^+\sigma^+\}\,\mathrm{Im}\,\beta^{(c)}$$
$$+ 2\epsilon^-\sin^2\theta\{(\rho^{+2} - \sigma^{+2})uv - (u^2 - v^2)\rho^+\sigma^+\}\,\mathrm{Re}\,\beta^{(c)}$$
$$+ 2(\omega/c)\{\epsilon^-(u^2+v^2) - (\rho^{+2} + \sigma^{+2})\cos^2\theta\}(u\rho^+ + v\sigma^+)$$
$$\times \mathrm{Re}(\epsilon^-\sin^2\theta\,\delta^{(c)} - \tau^{(c)}) + 2(\omega/c)\{\epsilon^-(u^2+v^2)$$
$$+ (\rho^{+2} + \sigma^{+2})\cos^2\theta\}(v\rho^+ - u\sigma^+)\,\mathrm{Im}(\epsilon^-\sin^2\theta\,\delta^{(c)} - \tau^{(c)})], \tag{5.27}$$

$$T_p = T_p^0(d) - 8(\omega/c)n^-\cos\theta\{(n^- u + \rho^+\cos\theta)^2 + (n^- v + \sigma^+\cos\theta)^2\}^{-2}$$
$$\times (u\rho^+ + v\sigma^+)\, e^{-2v(\omega/c)z}[\cos\theta\{n^-(u^2+v^2)$$
$$+ \cos\theta(u\rho^+ + v\sigma^+)\}\,\mathrm{Im}\,\gamma_l^{(c)} + \cos^2\theta(v\rho^+ - u\sigma^+)\,\mathrm{Re}\,\gamma_l^{(c)}$$
$$+ n^-\epsilon^-\sin^2\theta\{\cos\theta(\rho^{+2} + \sigma^{+2}) + n^-(u\rho^+ + v\sigma^+)\}\,\mathrm{Im}\,\beta^{(c)}$$
$$- (\epsilon^-)^2\sin^2\theta(v\rho^+ - u\sigma^+)\,\mathrm{Re}\,\beta^{(c)} - (\omega/c)n^-\cos\theta$$
$$\times \{\epsilon^-(u^2+v^2) - \cos^2\theta(\rho^{+2} + \sigma^{+2})\}\,\mathrm{Re}(\epsilon^-\sin^2\theta\,\delta^{(c)} - \tau^{(c)})$$
$$+ 2(\omega/c)n^-\cos\theta(v\rho^+ - u\sigma^+)\,\mathrm{Im}(\epsilon^-\sin^2\theta\,\delta^{(c)} - \tau^{(c)})], \tag{5.28}$$

$$Q_p = Q_p^{(0)}(d) + 4(\omega/c)n^-\cos\theta\{(n^- u + \rho^+\cos\theta)^2 + (n^- v + \sigma^+\cos\theta)^2\}^{-1}$$
$$\times \{(u^2+v^2)\,\mathrm{Im}\,\gamma_l^{(c)} + \epsilon^-\sin^2\theta(\rho^{+2} + \sigma^{+2})\,\mathrm{Im}\,\beta^{(c)}$$
$$+ 2(\omega/c)(v\rho^+ - u\sigma^+)\,\mathrm{Im}(\epsilon^-\sin^2\theta\,\delta^{(c)} - \tau^{(c)})\}, \tag{5.29}$$

$$r = r^0(d) - 2(\omega/c)n^-\cos\theta(n^-\cos\theta + w)(n^-\cos\theta - w)^{-1}$$
$$\times (n^- w + \epsilon^+\cos\theta)^{-2}[\mathrm{i}w^2\gamma_l^{(c)} - \mathrm{i}(\epsilon^- - \epsilon^+)^{-1}(\epsilon^- w^2 - \epsilon^{+2}\cos^2\theta)\gamma_{tt}^{(c)}$$
$$- \mathrm{i}\epsilon^-\epsilon^{+2}\sin^2\theta\,\beta^{(c)} - 2(\omega/c)\sin^2\theta\, w\{\epsilon^-\epsilon^+\delta^{(c)} - (\epsilon^- + \epsilon^+)\tau^{(c)}\}], \tag{5.30}$$

where $w \equiv u + iv$. The first terms at the right-hand sides of eqs. (5.24)–(5.30) denote the values of the various quantities for a parallel film of thickness d. They are found by substitution of the values of $r^0_{s,p}(d)$ and $t^0_{s,p}(d)$ into the right-hand sides of eqs. (5.21)–(5.23), retaining only terms up to second order in d. The other terms at the right-hand sides of eqs. (5.24)–(5.30) describe the influence of surface roughness on $R_{s,p}$, $T_{s,p}$, $Q_{s,p}$, and r by means of the coefficients $\gamma^{(c)}_{1}$, $\gamma^{(c)}_{tr}$, $\beta^{(c)}$, $\delta^{(c)}$ and $\tau^{(c)}$. As we have seen in section 4, the latter quantities can in principle be evaluated, if the four height–height correlation functions $S^{vv'}$, eq. (4.17), are known.

References

1) I. Ohlidal and F. Lukeš, Opt. Acta **19** (1972) 817.
2) A.M. Albano, D. Bedeaux and J. Vlieger, Physica **102A** (1980) 105.
3) E. Kröger and E. Kretschmann, Z. Phys. **237** (1970) 1.
4) J. Vlieger and D. Bedeaux, Physica **85A** (1976) 389.
5) D. Bedeaux and J. Vlieger, Physica **67** (1973) 55.
6) J. Vlieger and D. Bedeaux, Thin Solid Films **69** (1980) 107.
7) B.J.A. Zielińska, D. Bedeaux and J. Vlieger, Physica **117A** (1983) 28.
8) M. Born and E. Wolf, Principles of Optics (Pergamon, Oxford, 1970) pp. 627–633.

9) Note that in the above derivation of the average surface polarization and magnetization densities we have systematically replaced, up to second order in the amplitudes of the surface roughness and the average film thickness, fluctuating fields by average fields. In doing so, local field effects are taken into account only in an approximate way, cf. reference 6 for a detailed discussion of this point. One can demonstrate that this procedure is valid when the direction of the normal on the curved surfaces is everywhere close to its average direction. For random rough surfaces this means that the theory will be applicable when the amplitude of the surface roughness is much smaller than the correlation length of the roughness. For discontinuous films, however, the approach will in general not be useful.

OPTICAL PROPERTIES OF THIN FILMS ON ROUGH SURFACES

II. LONG CORRELATION LENGTH LIMIT; COMPARISON WITH EARLIER WORK

1. Introduction

In a previous paper[1]), hereafter referred to as I, we have developed a theory of the optical properties of thin films on rough (i.e. non-planar) surfaces. This was done by applying a general method of Albano, Bedeaux and Vlieger[2]) to describe the electromagnetic properties of a finite boundary layer between two dielectric media by singular surface polarization and magnetization densities situated at a fictitious interface somewhere within this layer. These densities were calculated, up to second order in the film thickness and surface roughness over the wavelength of light, for a thin isotropic film covering the rough surface of a substrate (see paper I, fig. 1). The polarization and magnetization densities fluctuate due to the surface roughness of the film and the substrate. For the calculation of the optical properties, as the reflectance, transmittance and ellipsometric coefficient, it is sufficient to know the average electromagnetic fields. These fields are related to the average surface polarization and magnetization densities $\boldsymbol{P}^s$ and $\boldsymbol{M}^s$ by a set of boundary conditions, whereas $\boldsymbol{P}^s$ and $\boldsymbol{M}^s$ are expressed in terms of the fields by a set of constitutive equations. Formulae for the constitutive coefficients, appearing in these equations, were derived in paper I in terms of the height–height correlation functions of the upper and lower surfaces of the film, and its average thickness. Finally the reflectance, transmittance and ellipsometric coefficient were expressed in terms of the constitutive coefficients, for arbitrary angles of incidence.

In the present paper the above theory is applied in the case that the correlation length of the surface roughness is much larger than the wavelength of the light. In that case the surface roughness correlation functions simplify considerably. Since we will consider so-called identical films (the upper and lower surface of the film are identical), only one correlation function is left. With this auto-correlation function we calculate in section 2 the various surface constitutive coefficients and subsequently the reflectance, transmittance and ellipsometric coefficient of the film. These calculations are performed up to second order in the film thickness and surface roughness over the wavelength of light and the correlation length of the rough surface of the film.

As results we find that the calculated reflectance and transmittance of the film are the same as obtained by Ohlídal, Navrátil and Lukeš[3]) (for normally incident light) if we neglect rather small quadratic terms in the surface roughness over the correlation length. In the expression for the ellipsometic coefficient, however, these are the only second order terms contributing, and it is therefore important to compare our result for the ellipsometric coefficient with that obtained earlier by other authors. It appears that our result is different from that obtained by Ohlídal and Lukeš[4]), using the Helmholtz–Kirchhoff integral, but also from the result obtained by the same authors[5]), using the Stratton–Chu–Silver integral. In the first case these authors obtain a contribution to the ellipsometric coefficient from surface roughness, which does not vanish for normally incident light, as it should do for isotropic surfaces. So, in fact, we only have to compare our result for the ellipsometric coefficient with that of ref. 5. It is then found that our result is fundamentally different from that of Ohlídal and Lukeš[5]): according to our theory the effect of surface roughness on the ellipsometric coefficient changes sign when the light beam is reversed, which is not the case according to Ohlídal and Lukeš[5]). In principle, this could be tested by experiment. Ohlídal and Lukeš[4]) have performed an ellipsometric experiment on a rough silicon crystal, covered by a thin SiO_2-film, however only with light incident from the ambient side. It seems to be a pure coincidence that for this system our formulae give, within a few per cents, the same results as theirs.

In section 3 we show that the difference in results for the ellipsometric coefficient obtained by Ohlídal and Lukeš[4,5]) and in the present paper is due to an inconsistency in the assumptions made by those authors in order to calculate the electromagnetic field on a rough surface. To this end we first prove, by a direct calculation of this field, up to second order in the surface roughness that it differs from the field postulated by Ohlídal and Lukeš[4,5]) according to the so-called tangent plane approximation. We furthermore prove that this latter field can be obtained by neglecting the curvature of the rough surface in our calculations. We show, however, that this is an inconsistent approximation in the second order calculation of the field.

We know from the calculations of Ohlídal and Lukeš that the Helmholtz–Kirchhoff integral[4]) leads to a different result for the ellipsometric coefficient as the Stratton–Chu–Silver integral[5]), if in both cases the electromagnetic field on the rough surface of the film is calculated by means of the tangent plane approximation. In section 4 we prove that, if the correct second order field, calculated in section 3, is used, both the Helmholtz–Kirchhoff and the Stratton–Chu–Silver integral lead to the same far field and to our expression for the ellipsometric coefficient of a thin film on a rough surface, as calculated in section 2.

2. Long correlation length limit of surface roughness; comparison with earlier work

We consider a thin isotropic solid film, covering the rough surface of a solid substrate. This is a special case of that treated in paper I, where the functions $f^{\nu}(r_{\parallel}) = f^{\nu}(x, y)$, describing the positions of the upper ($\nu = -$) and lower ($\nu = +$) surface of the film, are independent of time (see paper I, section 2). The four correlation functions for the deviations $\Delta f^{\nu}(r_{\parallel})$ of these functions from their averages $\langle f^{\nu} \rangle$,

$$S^{\nu\nu'}(r_{\parallel} - r'_{\parallel}) \equiv \langle \Delta f^{\nu}(r_{\parallel}) \Delta f^{\nu'}(r'_{\parallel}) \rangle \tag{2.1}$$

(cf. eq. (I.4.17)), will be assumed to be differentiable functions, which can be developed into Taylor series around $r_{\parallel} - r'_{\parallel} = 0$.

For solid films one usually supposes that these correlation functions are Gaussian:

$$S^{\nu\nu'}(r_{\parallel} - r'_{\parallel}) = \sigma^2_{\nu\nu'} \exp(-|r_{\parallel} - r'_{\parallel}|^2 / l^2_{\nu\nu'}) , \tag{2.2}$$

where $\sigma_{\nu\nu'} \equiv \langle \Delta f^{\nu} \Delta f^{\nu'} \rangle^{1/2}$ characterize the surface roughness and $l_{\nu\nu'}$ are four correlation lengths. For *large* values of $l_{\nu\nu'}$ one may write for these functions

$$S^{\nu\nu'}(r_{\parallel} - r'_{\parallel}) \simeq \sigma^2_{\nu\nu'}(1 - |r_{\parallel} - r'_{\parallel}|^2 / l^2_{\nu\nu'}) . \tag{2.3}$$

It is, however, clear that, up to second order, every correlation function of the type described above, which is rotationally symmetric in the x–y plane, may be written in the form eq. (2.3). (Note that the auto-correlation function for a fluid–fluid interface, eq. (3.40) of ref. 6, does not satisfy this condition.)

In the following we shall, for simplicity's sake, only consider the case of identical films. Then all four correlation functions are the same:

$$S^{\nu\nu'} = S \tag{2.4}$$

and we obtain, in the long correlation length limit ($l \gg$ wavelength λ)

$$S(r_\parallel) = \sigma^2(1 - r_\parallel^2/l^2)\,. \tag{2.5}$$

Fourier transformation of this auto-correlation function gives

$$S(k_\parallel) \simeq (2\pi)^2\sigma^2\{\delta(k_\parallel) + l^{-2}\delta''(k_x)\delta(k_y) + l^{-2}\delta(k_x)\delta''(k_y)\}\,, \tag{2.6}$$

where δ is the Dirac δ-function and δ'' denotes the second derivative of this function.

Substituting the expression eq. (2.6) for the correlation functions $S^{\nu\nu'}(k_\parallel, \omega) = S^{\nu\nu'}(k_\parallel)2\pi\delta(\omega)$ into the right-hand sides of eqs. (I.4.27)–(I.4.32), we can evaluate the constitutive coefficients $\gamma_\mathrm{l}^{(c)\nu}$, $\gamma_\mathrm{tr}^{(c)\nu}$, $\beta^{(c)\nu}$, $\delta^{(c)\nu}$, $\eta^{(c)\nu}$ and $\tau^{(c)\nu}$, defined by eqs. (I.4.34)–(I.4.39). In the expressions for the reflectance, transmittance and ellipsometric coefficient only the symmetric combinations $\gamma_\mathrm{l}^{(c)} = \frac{1}{2}\Sigma_\nu\,\gamma_\mathrm{l}^{(c)\nu}$, $\gamma_\mathrm{tr}^{(c)} = \frac{1}{2}\Sigma_\nu\,\gamma_\mathrm{tr}^{(c)\nu}$, $\beta^{(c)} = \frac{1}{2}\Sigma_\nu\,\beta^{(c)\nu}$, $\delta^{(c)} = \frac{1}{2}\Sigma_\nu\,\delta^{(c)\nu} = \eta^{(c)}$ and $\tau^{(c)} = \frac{1}{2}\Sigma_\nu\,\tau^{(c)\nu}$ appear, and only as functions of $k_\parallel = (n^- \sin\theta\ \omega/c, 0)$ and ω, where θ is the angle of incidence of the light beam, ω its frequency, c the velocity of light in vacuum and n^- the refractive index of the ambient. One obtains:

$$\begin{aligned}\gamma_\mathrm{l}^{(c)} = {} & \mathrm{i}(\epsilon^+ - \epsilon^-)^2\sigma^2(c/\omega)[(\omega/c)^2\cos\theta\cos\theta_\mathrm{t}(n^+\cos\theta + n^-\cos\theta_\mathrm{t})^{-1} \\ & + l^{-2}\sin^2\theta(n^+\cos\theta + n^-\cos\theta_\mathrm{t})^{-3}(\epsilon^+n^+n^-\cos^3\theta\cos^3\theta_\mathrm{t})^{-1} \\ & \times\{-n^-n^+\epsilon^-\cos^6\theta - 3(\epsilon^-)^2\cos^5\theta\cos\theta_\mathrm{t} - 2n^+n^-(\epsilon^- - \epsilon^+)\cos^4\theta\cos^2\theta_\mathrm{t} \\ & + 2(4\epsilon^+\epsilon^- - (\epsilon^+)^2 - (\epsilon^-)^2)\cos^3\theta\cos^3\theta_\mathrm{t} + 2n^+n^-(\epsilon^- - \epsilon^+)\cos^2\theta\cos^4\theta_\mathrm{t} \\ & - 3(\epsilon^+)^2\cos\theta\cos^5\theta_\mathrm{t} - n^-n^+\epsilon^+\cos^6\theta_\mathrm{t}\}]\,,\end{aligned} \tag{2.7}$$

$$\begin{aligned}\gamma_\mathrm{tr}^{(c)} = {} & \mathrm{i}(\epsilon^+ - \epsilon^-)^2\sigma^2(c/\omega)[(\omega/c)^2(n^-\cos\theta + n^+\cos\theta_\mathrm{t})^{-1} \\ & + l^{-2}\sin^2\theta(\epsilon^+n^+n^-\cos^3\theta\cos^3\theta_\mathrm{t})^{-1} \\ & \times\{2n^+n^-\cos^2\theta\cos^2\theta_\mathrm{t}(n^+\cos\theta + n^-\cos\theta_\mathrm{t})^{-1} \\ & + (\epsilon^+\cos^2\theta_\mathrm{t} + \epsilon^-\cos^2\theta + n^+n^-\cos\theta\cos\theta_\mathrm{t}) \\ & \times(n^-\cos\theta + n^+\cos\theta_\mathrm{t})^{-1}\}]\,,\end{aligned} \tag{2.8}$$

$$\beta^{(c)} = \mathrm{i}(\epsilon^+ - \epsilon^-)^2(\epsilon^+\epsilon^-)^{-1}\sigma^2(c/\omega)$$
$$\times \{[(\omega/c)^2 n^- \sin^2\theta(n^+)^{-1}(n^+\cos\theta + n^-\cos\theta_t)^{-1}$$
$$+ l^{-2}(n^+n^-)^{-1}(n^+\cos\theta + n^-\cos\theta_t)^{-1}$$
$$\times \{4 + 6\sin^2\theta(\epsilon^+n^+\cos\theta_t + \epsilon^-n^-\cos\theta)$$
$$\times (n^+\cos\theta + n^-\cos\theta_t)^{-1}(\epsilon^+\cos\theta\cos\theta_t)^{-1}$$
$$+ \sin^4\theta(n^+\cos\theta + n^-\cos\theta_t)^{-2}(\epsilon^+)^{-2}(\cos\theta\cos\theta_t)^{-3}$$
$$\times (n^+n^-(\epsilon^-)^2\cos^4\theta + 3(\epsilon^-\cos\theta)^3\cos\theta_t$$
$$+ 4n^+n^-\epsilon^+\epsilon^-\cos^2\theta\cos^2\theta_t + 3(\epsilon^+\cos\theta_t)^3\cos\theta$$
$$+ n^+n^-(\epsilon^+)^2\cos^4\theta_t)\}] , \tag{2.9}$$

$$\delta^{(c)} = (\epsilon^+ - \epsilon^-)\sigma^2(c/\omega)^2$$
$$\times [(\omega/c)^2(n^-\cos\theta + n^+\cos\theta_t)(n^+\cos\theta + n^-\cos\theta_t)^{-1}(n^+n^-)^{-1}$$
$$+ l^{-2}(\epsilon^+ - \epsilon^-)^2(n^+\cos\theta + n^-\cos\theta_t)^{-3}(n^+n^-\cos\theta\cos\theta_t)^{-3}$$
$$\times (n^+\cos^3\theta + 3n^-\cos^2\theta\cos\theta_t + 3n^+\cos\theta\cos^2\theta_t + n^-\cos^3\theta_t)] = \eta^{(c)} , \tag{2.10}$$

$$\tau^{(c)} = \tfrac{1}{2}(\epsilon^+ - \epsilon^-)\sigma^2 , \tag{2.11}$$

where θ_t is the angle of transmittance, n^+ the refractive index of the substrate, whereas $\epsilon^+ = (n^+)^2$ and $\epsilon^- = (n^-)^2$ are the dielectric constants of the substrate and the ambient, respectively. For complex refractive index n^+ this angle of transmittance is no longer real and is then given by eqs. (I.5.9)–(I.5.13). To simplify calculations, however, we shall assume here that n^+ is real. Substituting expressions (2.7)–(2.11) into the right-hand sides of eqs. (I.5.24)–(I.5.30), with $u = n^+\cos\theta_t$, $v = 0$, $\rho^+ = \epsilon^+$ and $\sigma^+ = \mathrm{Im}\,\epsilon^+ = 0$, we find, up to second order in the average film thickness d and the surface roughness σ over the wavelength λ and the correlation length l,

$$R_s = R_s^0(d)[1 - 4\epsilon^-(\omega/c)^2\sigma^2\cos^2\theta + 4(\epsilon^+ - \epsilon^-)l^{-2}\sigma^2\sin^2\theta\cos^{-2}\theta(n^+\cos\theta_t)^{-3}$$
$$\times \{2n^+n^-\cos^2\theta\cos^2\theta_t(n^+\cos\theta + n^-\cos\theta_t)^{-1}$$
$$+ (\epsilon^+\cos^2\theta_t + \epsilon^-\cos^2\theta + n^+n^-\cos\theta\cos\theta_t)(n^-\cos\theta + n^+\cos\theta_t)^{-1}\}] , \tag{2.12}$$

$$T_s = T_s^0(d)[1 - (\omega/c)^2\sigma^2(n^- \cos\theta - n^+ \cos\theta_t)^2$$
$$- 2(\epsilon^+ - \epsilon^-)^2 l^{-2}\sigma^2 \sin^2\theta$$
$$\times (n^- \cos^3\theta)^{-1}(n^+ \cos\theta_t)^{-3}(n^- \cos\theta + n^+ \cos\theta_t)^{-1}$$
$$\times \{2n^+n^- \cos^2\theta \cos^2\theta_t(n^+ \cos\theta + n^- \cos\theta_t)^{-1}$$
$$+ (\epsilon^+ \cos^2\theta_t + \epsilon^- \cos^2\theta + n^+n^- \cos\theta \cos\theta_t)(n^- \cos\theta + n^+ \cos\theta_t)^{-1}\}],$$
$$(2.13)$$

$$Q_s = Q_s^0(d) + 4\sigma^2 n^- \cos\theta(n^- \cos\theta - n^+ \cos\theta_t)^2[(\omega/c)^2(n^- \cos\theta + n^+ \cos\theta_t)^{-1}$$
$$+ l^{-2} \sin^2\theta(n^- \cos^3\theta)^{-1}(n^+ \cos\theta_t)^{-3}$$
$$\times \{2n^+n^- \cos^2\theta \cos^2\theta_t(n^+ \cos\theta + n^- \cos\theta_t)^{-1}$$
$$+ (\epsilon^+ \cos^2\theta_t + \epsilon^- \cos^2\theta + n^+n^- \cos\theta \cos\theta_t)(n^- \cos\theta + n^+ \cos\theta_t)^{-1}\}],$$
$$(2.14)$$

$$R_p = R_p^0(d)[1 - 4\epsilon^-(\omega/c)^2\sigma^2 \cos^2\theta + (\epsilon^+ - \epsilon^-)l^{-2}\sigma^{-2} \sin^2\theta\, f(\theta, \theta_t, \epsilon^+, \epsilon^-)],$$
$$(2.15)$$

$$T_p = T_p^0(d)[1 - (\omega/c)^2(n^- \cos\theta - n^+ \cos\theta_t)^2\sigma^2$$
$$+ (\epsilon^+ - \epsilon^-)^2 l^{-2}\sigma^2 \sin^2\theta\, g(\theta, \theta_t, \epsilon^+, \epsilon^-)], \qquad (2.16)$$

$$Q_p = Q_p^0(d) + 4(\epsilon^+ - \epsilon^-)^2(\omega/c)^2\sigma^2 n^- \cos\theta$$
$$\times (n^+ \cos\theta \cos^3\theta_t + n^- \sin^4\theta)(n^+)^{-1}(n^- \cos\theta_t + n^+ \cos\theta)^{-3}$$
$$+ (\epsilon^+ - \epsilon^-)^2 l^{-2}\sigma^2 \sin^2\theta\, h(\theta, \theta_t, \epsilon^+, \epsilon^-), \qquad (2.17)$$

$$r = r^0(d)[1 + 2l^{-2}\sigma^2(\epsilon^+ - \epsilon^-)^2 \sin^2\theta \cos^{-2}\theta$$
$$\times (n^+ \cos\theta_t)^{-3}(n^+ \cos\theta + n^- \cos\theta_t)^{-4}(n^- \cos\theta_t - n^+ \cos\theta)^{-1}$$
$$\times \{\cos^2\theta_t[n^-n^+\epsilon^- \cos^6\theta + 3(\epsilon^-)^2 \cos^5\theta \cos\theta_t$$
$$+ 2n^+n^-(\epsilon^- - \epsilon^+)\cos^4\theta \cos^2\theta_t - 2(4\epsilon^+\epsilon^- - (\epsilon^+)^2 - (\epsilon^-)^2)\cos^3\theta \cos^3\theta_t$$
$$- 2n^+n^-(\epsilon^- - \epsilon^+)\cos^2\theta \cos^4\theta_t + 3(\epsilon^+)^2 \cos\theta \cos^5\theta_t + n^-n^+\epsilon^+ \cos^6\theta_t]$$
$$+ (\cos^2\theta_t - \sin^2\theta)(n^+ \cos\theta + n^- \cos\theta_t)^2$$
$$\times [2n^+n^- \cos^2\theta \cos^2\theta_t + (\cos\theta \cos\theta_t + \sin\theta \sin\theta_t)$$
$$\times (\epsilon^+ \cos^2\theta_t + \epsilon^- \cos^2\theta + n^+n^- \cos\theta \cos\theta_t)]$$

$$
\begin{aligned}
&+ 4\epsilon^{+} \cos^3\theta \cos^3\theta_t (n^{+}\cos\theta + n^{-}\cos\theta_t)^2 \\
&+ 6\sin^2\theta(\epsilon^{+}n^{+}\cos\theta_t + \epsilon^{-}n^{-}\cos\theta)(n^{+}\cos\theta + n^{-}\cos\theta_t) \\
&\times \cos^2\theta\cos^2\theta_t + (\epsilon^{+})^{-1}\sin^4\theta[n^{+}n^{-}(\epsilon^{-})^2\cos^4\theta + 3(\epsilon^{-}\cos\theta)^3\cos\theta_t \\
&+ 4n^{+}n^{-}\epsilon^{+}\epsilon^{-}\cos^2\theta\cos^2\theta_t + 3(\epsilon^{+}\cos\theta_t)^3\cos\theta + n^{+}n^{-}(\epsilon^{+})^2\cos^4\theta_t] \\
&+ 2(\epsilon^{-} - \epsilon^{+})n^{+}\cos\theta_t(n^{+}\cos^3\theta + 3n^{-}\cos^2\theta\cos\theta_t \\
&+ 3n^{+}\cos\theta\cos^2\theta_t + n^{-}\cos^3\theta_t)\}] \,. \qquad (2.18)
\end{aligned}
$$

Here R_s and R_p are the reflectances of the film for s- and p-polarized light, which are expressed in terms of the reflectances $R_s^0(d)$ and $R_p^0(d)$ of a plane parallel film of the same material with thickness d (the average film thickness), and contributions due to surface roughness. The same is done for the transmittances T_s and T_p and the ellipsometric coefficient $r = r_p/r_s$, where r_p and r_s are the amplitudes of p- and s-polarized light (cf. eq. (I.5.23)). The losses of energy from the beams Q_s and Q_p are split up into contributions $Q_s^0(d)$ and $Q_p^0(d)$, due to absorption in the film, and contributions due to the loss of energy by scattering by the rough surfaces. The functions f, g and h are complicated functions of the angles θ and θ_t and the dielectric constants ϵ^- and ϵ^+, which will not be given here explicitly (see later). The terms with $\sigma^2(\omega/c)^2$ in eqs. (2.12)–(2.17) have been derived earlier[3,7]). It is interesting to note that the relative reflectivities $R_s/R_s^0(d)$ and $R_p/R_p^0(d)$, as well as the relative transmittivities $T_s/T_s^0(d)$ and $T_p/T_p^0(d)$, are equal, as long as these terms are only considered. Note that in expression (2.18) for the ellipsometric coefficient r the term with $\sigma^2(\omega/c)^2$ is lacking. From eqs. (2.12)–(2.18) one can see that the long correlation length limit leads to a series expansion in terms of the quantity $(c/\omega)^2 l^{-2}$. Therefore a necessary condition for this expansion to hold is $l \gg (c/\omega) = \lambda/2\pi$. In refs. 3 and 7 an alternative condition was given, namely $4\pi r_c \cos\theta_1 \gg \lambda$, where r_c denotes the radius of curvature and θ_1 is the angle between the local normal and the direction of the incident beam. For very smooth surfaces θ_1 is approximately equal to the angle of incidence, and the average magnitude of the radius of curvature is roughly l^2/σ. It follows that for small surface roughness (i.e. $\sigma \ll \lambda/2\pi$) this condition is not stringent enough, since it also allows $l \approx \lambda/2\pi$, in which case the above mentioned expansion breaks down. As we have seen above $l \gg c/\omega = \lambda/2\pi$, so that in general the terms with σ^2/l^2 are much smaller than the terms with $\sigma^2(\omega/c)^2$. Experimentally it will therefore be very difficult, if possible at all, to detect the effect of the (σ^2/l^2) terms on the reflectance, transmittance and the energy loss in the film. For that reason we have not given the explicit forms of the functions f, g and h. (For s-polarized light the σ^2/l^2 terms are much simpler, since only the coefficient γ_{tr} contributes.) In expression (2.18) for the ellipsometric coefficient, however, the term with $\sigma^2(\omega/c)^2$ vanishes, so that the relative difference $[r - r^0(d)]/r^0(d)$ of the ellipsometric coefficient of the rough and the flat film is proportional to σ^2/l^2. From this difference the σ^2/l^2 term can therefore in principle be measured experimentally.

We shall now compare our result (2.18) for the ellipsometric coefficient with results obtained previously by Ohlídal and Lukeš[4,5]). In ref. 4 these authors have used the Helmholtz–Kirchhoff integral in order to evaluate the far-away electric fields of electromagnetic waves, reflected and scattered by a rough surface, from the local electric field on this surface. This local electric field, on its turn, is calculated by supposing the validity of the so-called tangent plane approximation. In this approximation it is assumed that the local electric field on a rough surface may be calculated in every point, by approximating this surface in that point by an infinitely flat surface coinciding with the local tangent plane and using Fresnel's laws of reflection and transmission of light. The result of Ohlídal and Lukeš[4]) for identical films, their eq. (34) together with eqs. (45), (51)–(54), can be written, up to second order in the average film thickness d and the surface roughness σ, as

$$\begin{aligned} r = r^0(d)[1 - 2l^{-2}\sigma^2(\epsilon^+ - \epsilon^-)n^- \\ &\times (n^+ \cos\theta_t)^{-3}(n^+\cos\theta + n^-\cos\theta_t)^{-2}(n^+\cos\theta - n^-\cos\theta_t)^{-1} \\ &\times \{2\epsilon^+\cos\theta\cos^2\theta_t(n^+\cos\theta + n^-\cos\theta_t)(\cos^2\theta + \cos^2\theta_t) \\ &+ \sin^2\theta(n^+\epsilon^-\cos^2\theta(\cos^2\theta + \cos^2\theta_t) + 3n^-\epsilon^-\cos\theta\cos\theta_t \\ &+ n^-(\epsilon^- + \epsilon^+)\cos\theta\cos\theta_t(\cos^2\theta_t - \sin^2\theta) + 2n^+(\epsilon^- + \epsilon^+)\cos^4\theta_t)\}] \,. \end{aligned} \tag{2.19}$$

In ref. 5 Ohlídal and Lukeš also use the tangent plane approximation in order to calculate the local electromagnetic field on the rough surface, but the far-away electromagnetic field is calculated with the help of the so-called Stratton–Chu–Silver integral. Their result, eq. (14) of ref. 5, can be written for thin films on rough surfaces, up to second order in d and σ, as

$$\begin{aligned} r = r^0(d)[1 - 2l^{-2}\sigma^2(\epsilon^+ - \epsilon^-)n^-\sin^2\theta\,(n^+\cos\theta_t)^{-3}(n^+\cos\theta + n^-\cos\theta_t)^{-2} \\ &\times (n^+\cos\theta - n^-\cos\theta_t)^{-1}\{2\cos\theta(n^+\cos\theta + n^-\cos\theta_t) \\ &\times (\epsilon^-\cos^2\theta + \epsilon^+\cos^2\theta_t)\cos^2\theta_t + n^+\epsilon^-\cos^2\theta(\cos^2\theta + \cos^2\theta_t) \\ &+ 3n^-\epsilon^-\cos\theta\cos\theta_t + n^-(\epsilon^+ + \epsilon^-)\cos\theta\cos\theta_t(\cos^2\theta_t - \sin^2\theta) \\ &+ 2n^+(\epsilon^+ + \epsilon^-)\cos^4\theta_t\}] \,. \end{aligned} \tag{2.20}$$

One can immediately see that expression (2.19) cannot be correct, since in the case of normal incidence ($\theta = 0$) one would find:

$$r = -1 + 8l^{-2}\sigma^2 n^-(n^+)^{-1}, \tag{2.21}$$

instead of the correct value $r = -1$. Note that eqs. (2.18) and (2.20) do fulfil this condition. We therefore only need to compare these two equations. Since both are complicated expressions, this will be done only for special angles θ. In fact we will examine here how the so-called Brewster angle θ_B is changed by the (σ^2/l^2) terms in both expressions, eqs. (2.18) and (2.20). This Brewster angle will be defined by the equation

$$\mathrm{Re}\, r(\theta_B) = 0\,. \tag{2.22}$$

For a plane substrate, without film, this angle is given by

$$\theta_B^0 = \arccos\{n^-(\epsilon^+ + \epsilon^-)^{-1/2}\}\,. \tag{2.23}$$

If a plane parallel film with dielectric constant $\epsilon = \epsilon' + \mathrm{i}\epsilon''$ and thickness d is present on top of this substrate, the Brewster angle is changed by the amount

$$\begin{aligned}\Delta\theta_B^{(f)} = &-(\omega/c)dn^-n^+\epsilon^+(\epsilon^+ + \epsilon^-)^{-3/2}(\epsilon^+ - \epsilon^-)^{-1}\epsilon''(\epsilon^+\epsilon^-|\epsilon|^{-2} - 1)\\ &+ (\omega/c)^2 d^2 n^+ n^- \epsilon^+(\epsilon^+ + \epsilon^-)^{-2}(\epsilon^+ - \epsilon^-)^{-1}\\ &\times [\epsilon'\{\epsilon^+ + \epsilon^- - 2\epsilon^+\epsilon^-(\epsilon^+ - \epsilon^-)^{-1} - (\epsilon^+ + \epsilon^-)^2(\epsilon^+ - \epsilon^-)^{-1}\\ &+ \epsilon^+\epsilon^-(\epsilon^+ + \epsilon^-)|\epsilon|^{-2} - 2(\epsilon^+\epsilon^-)^2(\epsilon^+ - \epsilon^-)^{-1}|\epsilon|^{-2}\}\\ &- \tfrac{1}{2}(\epsilon'^2 - \epsilon''^2)\{1 - 2(\epsilon^+ + \epsilon^-)(\epsilon^+ - \epsilon^-)^{-1} + (\epsilon^+\epsilon^-)^2|\epsilon|^{-4}\}\\ &- \tfrac{1}{2}\epsilon''^2(\epsilon^+ + \epsilon^-)^{-1}(\epsilon^+ - \epsilon^-)^{-1}\epsilon^+(\epsilon^+ - 3\epsilon^-)(\epsilon^+\epsilon^-|\epsilon|^{-2} - 1)^2\\ &- \tfrac{1}{2}\{(\epsilon^+ + \epsilon^-)^2 + 2\epsilon^+\epsilon^- - 6(\epsilon^+ + \epsilon^-)\epsilon^+\epsilon^-(\epsilon^+ - \epsilon^-)^{-1}\}]\,. \end{aligned} \tag{2.24}$$

For absorbing films ($\epsilon'' \neq 0$) one may neglect in general the second order terms in this expression, so that

$$\Delta\theta_B^{(f)} \simeq -(\omega/c)dn^-n^+\epsilon^+(\epsilon^+ + \epsilon^-)^{-3/2}(\epsilon^+ - \epsilon^-)^{-1}\epsilon''(\epsilon^+\epsilon^-|\epsilon|^{-2} - 1)\,. \tag{2.25}$$

For non-absorbing films, eq. (2.24) becomes

$$\begin{aligned}\Delta\theta_B^{(f)} = &\tfrac{1}{2}(\omega/c)^2 d^2 n^+ n^- \epsilon^+(\epsilon^+ - \epsilon)(\epsilon - \epsilon^-)(\epsilon^+ + \epsilon^-)^{-2}(\epsilon^+ - \epsilon^-)^{-1}\\ &\times \{\epsilon^+\epsilon^-(\epsilon)^{-2} - (\epsilon^+ + \epsilon^-)(\epsilon)^{-1} + 4\epsilon^+\epsilon^-(\epsilon)^{-1}(\epsilon^+ - \epsilon^-)^{-1}\\ &+ 1 - 2(\epsilon^+ + \epsilon^-)(\epsilon^+ - \epsilon^-)^{-1}\}\,. \end{aligned} \tag{2.26}$$

The change of the Brewster angle by the surface roughness follows from eqs. (2.22) and (2.18):

$$\Delta\theta_B^{(c)} = -l^{-2}\sigma^2(\epsilon^+ - \epsilon^-)(\epsilon^+ + \epsilon^-)^{-1}(n^- n^+ \epsilon^+)^{-1}\{(\epsilon^+)^2 + 2(\epsilon^-)^2 + 4\epsilon^+\epsilon^-\}. \tag{2.27}$$

The total change of the Brewster angle, due to the film and roughness is, up to second order in $(\omega/c)d$ and σ/l, equal to the sum of the contributions, eqs. (2.24) and (2.27):

$$\Delta\theta_B = \Delta\theta_B^{(f)} + \Delta\theta_B^{(c)}. \tag{2.28}$$

Let us now compare with the change in Brewster angle, due to surface roughness, found from expression (2.20), of Ohlídal and Lukeš:

$$\Delta\theta_B^{(c)'} = -l^{-2}\sigma^2(\epsilon^+ + \epsilon^-)^{-2}(n^- n^+ \epsilon^+)^{-1}$$
$$\times \{(\epsilon^+)^4 + 4(\epsilon^+)^3\epsilon^- + 3(\epsilon^+\epsilon^-)^2 + 6\epsilon^+(\epsilon^-)^3 + 2(\epsilon^-)^4\}. \tag{2.29}$$

We see that this expression differs qualitatively from ours, eq. (2.27), in the sense that $\Delta\theta_B^{(c)}$ changes sign, when the light is incident from the substrate into the ambient, instead of from the ambient into the substrate, whereas the sign of $\Delta\theta_B^{(c)'}$ remains unaffected under this operation. This is also the case with the change in Brewster angle, due to the surface roughness, obtained from the incorrect expression (2.19), which angle is given by

$$\Delta\theta_B^{(c)''} = -l^{-2}\sigma^2(n^- n^+ \epsilon^+)^{-1}\{(\epsilon^+)^2 + 2(\epsilon^-)^2 + 2\epsilon^+\epsilon^-\}. \tag{2.30}$$

Now it is in fact this last formula which has been tested experimentally by Ohlídal and Lukeš[4]) on rough silicon crystals (covered with a thin SiO_2 film). Since ϵ^+ is rather large for Si and $\epsilon^- = 1$, we find that all expressions, eqs. (2.27), (2.29) and (2.30), yield approximately the same result. Their experiment is therefore not conclusive for the correctness of their theory.

In the next section we shall show that the difference in results obtained by Ohlídal and Lukeš[4,5]) and in the present paper is due to an inconsistency in the assumptions made by these authors in order to calculate the local electromagnetic field on the rough surface.

3. The electromagnetic field on the rough surface

In the previous section we have seen that our calculation of the influence of surface roughness on the optical properties leads for the ellipsometric coefficient to an equation (cf. eq. (2.27)) that differs from the results obtained by Ohlídal and Lukeš[4,5]) (cf. eqs. (2.29) and (2.30)). We shall now examine the origin of this difference. In ref. 4 Ohlídal and Lukeš calculate the ellipsometric coefficient of a rough surface, or of a system substrate-thin film with rough boundaries, using the Helmholtz–Kirchhoff integral*

$$\boldsymbol{E}_P = \iint_S \left[\boldsymbol{E} \frac{\partial \psi}{\partial n} - \psi \frac{\partial \boldsymbol{E}}{\partial n} \right] \mathrm{d}S , \tag{3.1}$$

where $\boldsymbol{E}_P$ is the electric field of the wave, reflected and diffracted by this surface, at a point P, situated inside the Fraunhofer diffraction zone. In eq. (3.1) $\boldsymbol{E}$ and $\partial \boldsymbol{E}/\partial n$ are the electric field on the surface and its derivative, normal to the surface, whereas S is the illuminated part of this surface. The function ψ is defined by

$$\psi = (4\pi R)^{-1} \exp(\mathrm{i} n_P \omega R/c) \exp(-\mathrm{i} \boldsymbol{k}_2 \cdot \boldsymbol{r}) , \tag{3.2}$$

where R is the distance from P to the origin, situated at the surface, $\boldsymbol{k}_2$ is the wave vector of the diffracted light, with length $|\boldsymbol{k}_2| = n_P\omega/c$, and n_P is the refractive index of the medium in which P is situated (i.e. for reflected light $n_P = n^-$ and for transmitted light $n_P = n^+$).

In ref. 5 the Stratton–Chu–Silver integral was used. Here the electric field at P is given by

$$\boldsymbol{E}_P = \mathrm{i} \boldsymbol{k}_2 \wedge \iint_S [\boldsymbol{n} \wedge \boldsymbol{E} - (n_P)^{-1} \boldsymbol{n}_2 \wedge (\boldsymbol{n} \wedge \boldsymbol{H})] \psi \, \mathrm{d}S , \tag{3.3}$$

with $\boldsymbol{n}_2 = \boldsymbol{k}_2/|\boldsymbol{k}_2|$ the unit vector in the direction of the diffracted light, $\boldsymbol{n}$ the local normal unit vector and $\boldsymbol{E}$ and $\boldsymbol{H}$ the electric and magnetic fields at the rough surface.

To calculate $\boldsymbol{E}_P$, using either eq. (3.1) or eq. (3.3), requires knowledge of the electromagnetic field at (in fact just above) the rough surface. The exact form of this field is, however, in general unknown, so that one has to make certain approximations. A usual assumption, made e.g. by Beckmann[7]) and by Ohlídal and Lukeš[4,5]), is that, in the case the radius of curvature of the rough surface is

* In formulae related to the work of Ohlídal and Lukeš we shall adopt their notation of capital letters for the fields. Otherwise, however, we shall use our convention of denoting fluctuating unaveraged fields by lower case letters and bulk fields by capital letters.

much larger than the wavelength of the incident light, one may use the so-called tangent plane approximation (Kirchhoff). In this approximation the electromagnetic field on the surface is expressed in terms of the local Fresnel coefficients $\hat{R}_s$ and $\hat{R}_p$ for s- and p-polarized light (see e.g. ref. 4 eqs. (6) and (7)). For the electric field $\boldsymbol{E}$ one so finds:

$$\boldsymbol{E} = [(1+\hat{R}_s)(\boldsymbol{a}\cdot\boldsymbol{t})\boldsymbol{t} + (1-\hat{R}_p)(\boldsymbol{a}\cdot\boldsymbol{d})\boldsymbol{d} + 2\hat{R}_p(\boldsymbol{a}\cdot\boldsymbol{d})(\boldsymbol{d}\cdot\boldsymbol{n})\boldsymbol{n}]E_0\,, \tag{3.4}$$

where $\boldsymbol{a}$ is the unit polarization vector of the incident light $\boldsymbol{E}_0$, $\boldsymbol{t} = (\boldsymbol{n}_1 \wedge \boldsymbol{n})[1-(\boldsymbol{n}_1\cdot\boldsymbol{n})^2]^{-1/2}$, $\boldsymbol{d} = \boldsymbol{n}_1 \wedge \boldsymbol{t}$, with $\boldsymbol{n}_1$ the unit vector in the direction of the incident wave (see ref. 5).

In our theory we have consistently taken into account all contributions up till second order in the film thickness d and the root mean square value σ of the surface roughness over the wavelength λ and the correlation length l. The long correlation length limit (see section 2) was also considered by Ohlídal and Lukeš[4,5]). We therefore expect that the tangent plane approximation is the cause of the difference between eqs. (2.29) and (2.30), obtained by Ohlídal and Lukeš, and our equation (2.27). We have the possibility to verify this, because in paper I, eq. (3.18), we have found a general expression for the electromagnetic fields $\boldsymbol{n}_e \equiv (e_x, e_y, d_z)$ and $\boldsymbol{n}_m \equiv (h_x, h_y, b_z)$, valid up to second order in d/λ and σ/λ:

$$\begin{aligned} \boldsymbol{n}_e &= \boldsymbol{N}_e^{(0)} - \boldsymbol{K}_0\cdot\boldsymbol{p}^s\delta(z) + \boldsymbol{L}_0\cdot\boldsymbol{m}^s\delta(z)\,,\\ \boldsymbol{n}_m &= \boldsymbol{N}_m^{(0)} - \boldsymbol{L}\cdot(1-\boldsymbol{\xi}_0\cdot\boldsymbol{K}_0)\cdot\boldsymbol{p}^s\delta(z) - (\boldsymbol{K}+\boldsymbol{L}\cdot\boldsymbol{\xi}_0\cdot\boldsymbol{L}_0)\cdot\boldsymbol{m}^s\delta(z)\,. \end{aligned} \tag{3.5}$$

Here $\boldsymbol{N}_e^{(0)}$ and $\boldsymbol{N}_m^{(0)}$ are the fields that would be present in space in the absence of surface roughness and film, i.e. in the case of a perfect flat substrate. The propagators $\boldsymbol{K}_0$ and $\boldsymbol{L}_0$ are given by eqs. (I.3.19) in terms of the propagators $\boldsymbol{K}$ and $\boldsymbol{L}$, which are defined by eqs. (I.3.10) and (I.3.11), together with eq. (I.3.7); $\boldsymbol{\xi}_0$ is given by eq. (I.3.15). In eq. (3.5) $\boldsymbol{K}_0\cdot\boldsymbol{p}^s\delta(z)$ is a short-hand notation for $\int \boldsymbol{K}_0(\boldsymbol{r}_\parallel, z, t|\boldsymbol{r}_\parallel', z', t')\cdot\boldsymbol{p}^s(\boldsymbol{r}_\parallel', t)\delta(z')\,\mathrm{d}\boldsymbol{r}_\parallel'\,\mathrm{d}z'\,\mathrm{d}t'$ in coordinate representation, etc. The surface polarization and magnetization densities $\boldsymbol{p}^s(\boldsymbol{r}_\parallel, t)$ and $\boldsymbol{m}^s(\boldsymbol{r}_\parallel, t)$ are given by eq. (I.2.27). Eq. (3.5) gives the correct electromagnetic fields everywhere in space, except in the region where $\min[f^-(\boldsymbol{r}_\parallel), 0] < z < f^+(\boldsymbol{r}_\parallel)$ (see fig. 1: shaded area). This is a consequence of the fact that $\boldsymbol{p}^s$ and $\boldsymbol{m}^s$ were constructed by analytic extension of the electromagnetic fields from the surfaces $f^\pm(\boldsymbol{r}_\parallel)$ to the plane $z = 0$. With eq. (3.5) we can therefore evaluate the electromagnetic field, up to second order in d/λ and σ/λ, on the upper surface of the shaded region in fig. 1, more precisely, on $z = f^-(\boldsymbol{r}_\parallel) - 0$, if $f^-(\boldsymbol{r}_\parallel) < 0$, and on $z = -0$, if $f^-(\boldsymbol{r}_\parallel) > 0$. By analytic extension (see below) we then easily find also the field on $z = f^-(\boldsymbol{r}_\parallel) - 0$, if $f^-(\boldsymbol{r}_\parallel) > 0$. Substituting the electromagnetic fields on $z =$

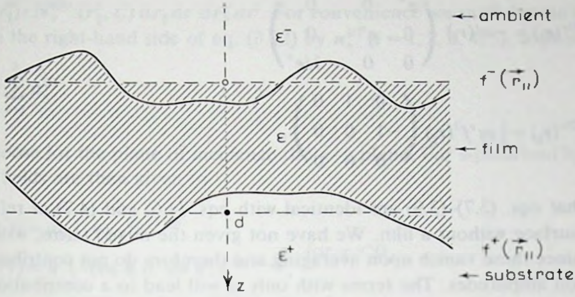


Fig. 1. Cross section of a thin film on a rough surface.

$f^-(\boldsymbol{r}_\parallel)-0$, calculated in this way, into the right-hand side of the Helmholtz–Kirchhoff integral, eq. (3.1), or the Stratton–Chu–Silver integral, eq. (3.3), one finds the correct second order expression for the electric field $\boldsymbol{E}_P$ at an arbitrary point P in the ambient. We will follow this procedure in the next section to calculate the reflected amplitude in the case of s-polarized light. It is found that the results, obtained from the Helmholtz–Kirchhoff integral and the Stratton–Chu–Silver integral, are identical to those obtained by the method of section 2. The extension to the more complicated case of p-polarized light is straightforward but, since the calculations are lengthy and the results again identical to those obtained in section 2, this will not be given here.

For the identical film the upper and lower surface are related by

$$f^-(\boldsymbol{r}_\parallel)=f^+(\boldsymbol{r}_\parallel)-d\,. \tag{3.6}$$

Using this relation in eqs. (I.2.23)–(I.2.27) the expressions for the polarization and magnetization densities simplify considerably, after application of the boundary conditions (I.2.15). Omitting terms with the average film thickness d, and replacing $f^-(\boldsymbol{r}_\parallel)$ by $f(\boldsymbol{r}_\parallel)$, one obtains

$$\begin{aligned} \boldsymbol{p}^s &= \sum_\nu \xi_b^{(1)\nu}\cdot\boldsymbol{n}_e^\nu+\sum_\nu \xi_b^{(2)\nu}\cdot c^{-1}\frac{\partial}{\partial t}\boldsymbol{n}_m^\nu\,,\\ \boldsymbol{m}^s &= \sum_\nu \xi_b^{(2)\nu}\cdot c^{-1}\frac{\partial}{\partial t}\boldsymbol{n}_e^\nu\,, \end{aligned} \tag{3.7}$$

where $\boldsymbol{n}_e^\nu$ and $\boldsymbol{n}_m^\nu$ are the fields on both sides ($\nu=+$ or $-$) of the plane $z=0$ (see paper I) and where the susceptibilities are given by

$$\xi_b^{(1)\nu}(r_\parallel) = -\nu f(r_\parallel) \begin{pmatrix} \epsilon^\nu & 0 & 0 \\ 0 & \epsilon^\nu & 0 \\ 0 & 0 & -1/\epsilon^\nu \end{pmatrix}, \tag{3.8}$$

$$\xi_b^{(2)\nu}(r_\parallel) = \tfrac{1}{2}\nu\epsilon^\nu f^2(r_\parallel) \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{3.9}$$

Note that eqs. (3.7)–(3.9) are identical with eqs. (4.6) and (4.7) of ref. 8 for a rough surface without a film. We have not given the mixed terms, with both d and f, since these vanish upon averaging and therefore do not contribute to the reflection amplitudes. The terms with only d will lead to a contribution of the average film thickness on the amplitudes. The omission of these terms is motivated by the fact that we are here only interested in the influence of surface roughness on the electromagnetic field. (Note that in section 2 there was no ambiguity in the value of $\Delta\theta_B^{(f)}$ found by Ohlídal and Lukeš[4,5]) and by us. It was the value of $\Delta\theta_B^{(c)}$ which was different in the various methods.)

Substituting eq. (3.7) into eq. (3.5) for the n_e field, one finds

$$n_e = N_e^{(0)} - \sum_\nu K_0 \cdot \xi_b^{(1)\nu} \cdot n_e^\nu - \sum_\nu K_0 \cdot \xi_b^{(2)\nu} \cdot c^{-1}\frac{\partial}{\partial t} n_m^\nu + \sum_\nu L_0 \cdot \xi_b^{(2)\nu} \cdot c^{-1}\frac{\partial}{\partial t} n_e^\nu, \tag{3.10}$$

where $K_0 \cdot \xi_b^{(1)\nu} \cdot n_e^\nu$ is a short-hand notation for $\int K_0(r_\parallel, z, t | r_\parallel', \; z' = 0, t') \cdot \xi_b^{(1)\nu}(r_\parallel') \cdot n_e^\nu(r_\parallel', t')\, dr_\parallel'\, dt'$, etc. Up to second order in f one may now replace n_m^ν and n_e^ν by $N_m^{(0)\nu}$ and $N_e^{(0)\nu}$, respectively, in the third and fourth term at the right-hand side of eq. (3.10), because $\xi_b^{(2)\nu}$ is already of second order (cf. eq. (3.9)). Up to first order in f one obtains from eq. (3.10):

$$n_e = N_e^{(0)} - \sum_\nu K_0 \cdot \xi_b^{(1)\nu} \cdot N_e^{(0)\nu}, \tag{3.11}$$

which may be used to replace n_e^ν in the second term at the right-hand side of eq. (3.10). In this way one obtains, correct up to second order in f, for eq. (3.10):

$$\begin{aligned} n_e = N_e^{(0)} &- K_0 \cdot \sum_\nu \xi_b^{(1)\nu} \cdot N_e^{(0)\nu} + K_0 \cdot \sum_{\nu'} \xi_b^{(1)\nu'} \cdot K_0 \cdot \sum_\nu \xi_b^{(1)\nu} \cdot N_e^{(0)\nu} \\ &- K_0 \cdot \sum_\nu \xi_b^{(2)\nu} \cdot c^{-1}\frac{\partial}{\partial t} N_m^{(0)\nu} + L_0 \cdot \sum_\nu \xi_b^{(2)\nu} \cdot c^{-1}\frac{\partial}{\partial t} N_e^{(0)\nu}, \end{aligned} \tag{3.12}$$

where $K_0 \cdot \Sigma_{\nu'} \xi_b^{(1)\nu'} \cdot K_0 \cdot \Sigma_\nu \xi_b^{(1)\nu} \cdot N_e^{(0)\nu}$ is a short-hand notation for $\int K_0(r_\parallel, z, t | r_\parallel', z' = 0, \; t') \cdot \Sigma_{\nu'} \xi_b^{(1)\nu'}(r_\parallel') \cdot K_0(r_\parallel', \; z' = \nu' 0, \; t' | r_\parallel'', \; z'' = 0, \; t'') \cdot$

$\Sigma_\nu \xi_b^{(1)\nu}(\boldsymbol{r}_\|') \cdot \boldsymbol{N}_e^{(0)\nu}(\boldsymbol{r}_\|'', t'')\,\mathrm{d}\boldsymbol{r}_\|'\,\mathrm{d}t'\,\mathrm{d}\boldsymbol{r}_\|''\,\mathrm{d}t''$. For convenience we shall denote the five terms on the right-hand side of eq. (3.12) by $\boldsymbol{n}_e^{(i)}$ ($i = 1, 2, 3, 4, 5$), so that

$$\boldsymbol{n}_e = \sum_{i=1}^{5} \boldsymbol{n}_e^{(i)} . \tag{3.13}$$

We choose for the plane of incidence the x–z plane. For s-polarized light the incident fields are then given by

$$\begin{aligned} \boldsymbol{E}_i(\boldsymbol{r}, t) &= (0, 1, 0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i\cdot\boldsymbol{r} - \omega t)} , \\ \boldsymbol{B}_i(\boldsymbol{r}, t) &= n^-(-\cos\theta, 0, \sin\theta)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i\cdot\boldsymbol{r} - \omega t)} , \end{aligned} \qquad \text{for } z < 0 , \tag{3.14}$$

where $\boldsymbol{k}_i = n^-(\omega/c)(\sin\theta, 0, \cos\theta)$. The fields reflected and transmitted by the flat interface are given by

$$\begin{aligned} \boldsymbol{E}_r(\boldsymbol{r}, t) &= r_s^0(0, 1, 0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_r\cdot\boldsymbol{r} - \omega t)} , \\ \boldsymbol{B}_r(\boldsymbol{r}, t) &= n^- r_s^0(\cos\theta, 0, \sin\theta)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_r\cdot\boldsymbol{r} - \omega t)} , \end{aligned} \qquad \text{for } z < 0 , \tag{3.15}$$

and

$$\begin{aligned} \boldsymbol{E}_t(\boldsymbol{r}, t) &= t_s^0(0, 1, 0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_t\cdot\boldsymbol{r} - \omega t)} , \\ \boldsymbol{B}_t(\boldsymbol{r}, t) &= n^+ t_s^0(-\cos\theta_t, 0, \sin\theta_t)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_t\cdot\boldsymbol{r} - \omega t)} , \end{aligned} \qquad \text{for } z > 0 , \tag{3.16}$$

where $\boldsymbol{k}_r = n^-(\omega/c)(\sin\theta, 0, -\cos\theta)$, $\boldsymbol{k}_t = n^+(\omega/c)(\sin\theta_t, 0, \cos\theta_t)$ and where r_s^0 and t_s^0 are the Fresnel amplitudes

$$\begin{aligned} r_s^0 &\equiv (n^-\cos\theta - n^+\cos\theta_t)(n^-\cos\theta + n^+\cos\theta_t)^{-1} , \\ t_s^0 &\equiv 2n^-\cos\theta(n^-\cos\theta + n^+\cos\theta_t)^{-1} . \end{aligned} \tag{3.17}$$

Therefore the first term at the right-hand side of eq. (3.12) is given by

$$\boldsymbol{n}_e^{(1)} \equiv \boldsymbol{N}_e^{(0)} = (0, 1, 0)\{\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i\cdot\boldsymbol{r} - \omega t)} + r_s^0\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_r\cdot\boldsymbol{r} - \omega t)}\} . \tag{3.18}$$

This expression is, of course, also valid for $z = f(\boldsymbol{r}_\|)$.

In the other terms at the right-hand side of eq. (3.12) the fields at $z = \pm 0$ are appearing. Using eqs. (3.14)–(3.17) we find

$$\begin{aligned} \boldsymbol{N}_e^{(0)-} &= \boldsymbol{N}_e^{(0)+} = t_s^0(0, 1, 0)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_\|\cdot\boldsymbol{r}_\| - \omega t)} , \\ \boldsymbol{N}_m^{(0)-} &= \boldsymbol{N}_m^{(0)+} = n^+ t_s^0(-\cos\theta_t, 0, \sin\theta_t)\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_\|\cdot\boldsymbol{r}_\| - \omega t)} , \end{aligned} \tag{3.19}$$

where $\boldsymbol{k}_{\parallel} = n^-(\omega/c)(\sin\theta, 0, 0)$ and where we have also applied Snell's law

$$n^- \sin\theta = n^+ \sin\theta_t . \tag{3.20}$$

Using eqs. (3.8) and (3.19), the second term at the right-hand side of eq. (3.12) can be written explicitly as

$$\boldsymbol{n}_e^{(2)} = (\epsilon^+ - \epsilon^-)t_s^0 \int \boldsymbol{K}_0(\boldsymbol{r}_\parallel, z, t|\boldsymbol{r}'_\parallel, z' = 0, t') \cdot \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} f(\boldsymbol{r}'_\parallel)\, e^{i(\boldsymbol{k}_\parallel \cdot \boldsymbol{r}'_\parallel - \omega t')} d\boldsymbol{r}'_\parallel\, dt' . \tag{3.21}$$

We first consider the case that $f(\boldsymbol{r}_\parallel) < 0$, so that $n_e^{(2)}(\boldsymbol{r}_\parallel, z, t)$ for $z = f(\boldsymbol{r}_\parallel) - 0$ is simply obtained by substitution of $z = f(\boldsymbol{r}_\parallel)$ into the right-hand side of eq. (3.21). In order to be able to compare our results with those obtained by means of the tangent plane approximation, we also assume that the rough surface is very smooth, so that we can expand $f(\boldsymbol{r}'_\parallel)$ in eq. (3.21) into a Taylor series around $\boldsymbol{r}_\parallel$:

$$f(\boldsymbol{r}'_\parallel) = f(\boldsymbol{r}_\parallel) + (\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel) \cdot \frac{\partial}{\partial \boldsymbol{r}_\parallel} f(\boldsymbol{r}_\parallel) + \frac{1}{2}(\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel)(\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel) : \frac{\partial^2}{\partial \boldsymbol{r}_\parallel \partial \boldsymbol{r}_\parallel} f(\boldsymbol{r}_\parallel) . \tag{3.22}$$

We do not consider higher order terms in the expansion since these cannot contribute to terms proportional to $\sigma^2(\omega/c)^2$ or σ^2/l^2 in r_s. As we shall see below, the tangent plane approximation is obtained by omitting the last term in the expansion (3.22). Substituting eq. (3.22) into eq. (3.21) and using eq. (I.4.19), we find, after partial Fourier transformation,

$$\begin{aligned}
\boldsymbol{n}_e^{(2)}(\boldsymbol{k}_\parallel, z, \omega') &= \int \boldsymbol{n}_e^{(2)}(\boldsymbol{r}_\parallel, z, t)\, e^{-i(\boldsymbol{k}_\parallel \cdot \boldsymbol{r}_\parallel - \omega' t)} d\boldsymbol{r}_\parallel\, dt \\
&= (\epsilon^+ - \epsilon^-)t_s^0 \int \boldsymbol{K}_0(\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel, z, t - t'|z' = 0) \cdot \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \\
&\quad \times \left\{ f(\boldsymbol{r}_\parallel) + (\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel) \cdot \frac{\partial}{\partial \boldsymbol{r}_\parallel} f(\boldsymbol{r}_\parallel) + \frac{1}{2}(\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel)(\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel) : \frac{\partial^2}{\partial \boldsymbol{r}_\parallel \partial \boldsymbol{r}_\parallel} f(\boldsymbol{r}_\parallel) \right\} \\
&\quad \times e^{i(\boldsymbol{k}_\parallel \cdot \boldsymbol{r}'_\parallel - \omega t')} e^{-i(\boldsymbol{k}_\parallel \cdot \boldsymbol{r}_\parallel - \omega' t)} d\boldsymbol{r}_\parallel\, dt\, d\boldsymbol{r}'_\parallel\, dt'
\end{aligned}$$

$$
= (\epsilon^+ - \epsilon^-) t_s^0 \int \boldsymbol{K}_0(\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel, z, t - t' | z' = 0) \cdot \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}
$$

$$
\times \left\{ f(\boldsymbol{r}_\parallel) + (\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel) \cdot \frac{\partial}{\partial \boldsymbol{r}_\parallel} f(\boldsymbol{r}_\parallel) + \frac{1}{2} (\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel)(\boldsymbol{r}'_\parallel - \boldsymbol{r}_\parallel) : \frac{\partial^2}{\partial \boldsymbol{r}_\parallel \partial \boldsymbol{r}_\parallel} f(\boldsymbol{r}_\parallel) \right\}
$$

$$
\times \mathrm{e}^{-\mathrm{i}\{\boldsymbol{k}_\parallel \cdot (\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel) - \omega'(t - t')\}} \, \mathrm{e}^{\mathrm{i}\{(\boldsymbol{k}'_\parallel - \boldsymbol{k}_\parallel) \cdot \boldsymbol{r}'_\parallel - (\omega - \omega')t'\}} \, \mathrm{d}(\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel) \, \mathrm{d}(t - t') \, \mathrm{d}\boldsymbol{r}'_\parallel \, \mathrm{d}t'
$$

$$
= (\epsilon^+ - \epsilon^-) t_s^0 \left\{ f(\boldsymbol{r}_\parallel) - \mathrm{i} \frac{\partial f(\boldsymbol{r}_\parallel)}{\partial \boldsymbol{r}_\parallel} \cdot \frac{\partial}{\partial \boldsymbol{k}_\parallel} - \frac{1}{2} \frac{\partial^2 f(\boldsymbol{r}_\parallel)}{\partial \boldsymbol{r}_\parallel \partial \boldsymbol{r}_\parallel} : \frac{\partial^2}{\partial \boldsymbol{k}_\parallel \partial \boldsymbol{k}_\parallel} \right\}
$$

$$
\times \boldsymbol{K}_0(\boldsymbol{k}_\parallel, z, \omega' | z' = 0) \cdot \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \delta(\boldsymbol{k}'_\parallel - \boldsymbol{k}_\parallel) \delta(\omega - \omega')
$$

$$
= (\epsilon^+ - \epsilon^-) t_s^0 \left[\left\{ f(\boldsymbol{r}_\parallel) - \mathrm{i} \frac{\partial f(\boldsymbol{r}_\parallel)}{\partial \boldsymbol{r}_\parallel} \cdot \frac{\partial}{\partial \boldsymbol{k}_\parallel} - \frac{1}{2} \frac{\partial^2 f(\boldsymbol{r}_\parallel)}{\partial \boldsymbol{r}_\parallel \partial \boldsymbol{r}_\parallel} : \frac{\partial^2}{\partial \boldsymbol{k}_\parallel \partial \boldsymbol{k}_\parallel} \right\} \right.
$$

$$
\left. \times \boldsymbol{K}_0(\boldsymbol{k}_\parallel, z, \omega | z' = 0) \right]_{\boldsymbol{k}_\parallel = \boldsymbol{k}_\parallel} \cdot \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \delta(\boldsymbol{k}'_\parallel - \boldsymbol{k}_\parallel) \delta(\omega - \omega') \,. \tag{3.23}
$$

Fourier transforming back to the original variables $\boldsymbol{r}_\parallel$, z and t one obtains

$$
\boldsymbol{n}_e^{(2)}(\boldsymbol{r}, t) = (\epsilon^+ - \epsilon^-) t_s^0 \left[\left\{ f(\boldsymbol{r}_\parallel) - \mathrm{i} \frac{\partial f(\boldsymbol{r}_\parallel)}{\partial \boldsymbol{r}_\parallel} \cdot \frac{\partial}{\partial \boldsymbol{k}_\parallel} - \frac{1}{2} \frac{\partial^2 f(\boldsymbol{r}_\parallel)}{\partial \boldsymbol{r}_\parallel \partial \boldsymbol{r}_\parallel} : \frac{\partial^2}{\partial \boldsymbol{k}_\parallel \partial \boldsymbol{k}_\parallel} \right\} \right.
$$

$$
\left. \times \boldsymbol{K}_0(\boldsymbol{k}_\parallel, z, \omega | z' = 0) \right]_{\boldsymbol{k}_\parallel = \boldsymbol{k}_\parallel} \cdot \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \mathrm{e}^{\mathrm{i}(\boldsymbol{k}_\parallel \cdot \boldsymbol{r}_\parallel - \omega t)} \,. \tag{3.24}
$$

The explicit form of the propagator $\boldsymbol{K}_0(\boldsymbol{k}_\parallel, z, \omega | z' = 0)$ for $z < 0$ is found from eq. (B.17), together with eqs. (B.15), (B.16), (B.18) and (B.19)–(B.28) of ref. 9:

$$
\boldsymbol{K}_0(\boldsymbol{k}_\parallel, z, \omega | z' = 0) = \mathrm{i} \, \mathrm{e}^{-\mathrm{i} k_\perp^- z} \begin{pmatrix} A_{11} & A_{12} & A_{13} \\ A_{21} & A_{22} & A_{23} \\ A_{31} & A_{32} & A_{33} \end{pmatrix} ,
$$

$$
A_{11} = -k_\perp^+ k_\perp^- (\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1} k_x^2 k_\parallel^{-2} - (\omega/c)^2 (k_\perp^+ + k_\perp^-)^{-1} k_y^2 k_\parallel^{-2} ,
$$

$$
A_{12} = -\{ k_\perp^+ k_\perp^- (\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1} - (\omega/c)^2 (k_\perp^+ + k_\perp^-)^{-1} \} k_x k_y k_\parallel^{-2} ,
$$

$$A_{13} = -\epsilon^+ k_\perp^- k_x(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1},$$

$$A_{21} = -\{k_\perp^+ k_\perp^-(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1} - (\omega/c)^2(k_\perp^+ + k_\perp^-)^{-1}\}k_x k_y k_\parallel^{-2},$$

$$A_{22} = -k_\perp^+ k_\perp^-(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1} k_y^2 k_\parallel^{-2} - (\omega/c)^2(k_\perp^+ + k_\perp^-)^{-1} k_x^2 k_\parallel^{-2}, \tag{3.25}$$

$$A_{23} = -\epsilon^+ k_\perp^- k_y(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1},$$

$$A_{31} = -\epsilon^- k_\perp^+ k_x(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1},$$

$$A_{32} = -\epsilon^- k_\perp^+ k_y(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1},$$

$$A_{33} = -\epsilon^+ \epsilon^- k_\parallel^2(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1},$$

where

$$k_\perp^+ = (\epsilon^+ \omega^2/c^2 - k_\parallel^2)^{1/2}, \quad k_\perp^- = (\epsilon^- \omega^2/c^2 - k_\parallel^2)^{1/2}. \tag{3.26}$$

Substitution of eq. (3.25) into eq. (3.24) gives

$$n_e^{(2)}(r, t) = \mathrm{i}(\epsilon^+ - \epsilon^-)t_s^0 \left[\left\{f(r_\parallel) - \mathrm{i}\frac{\partial f(r_\parallel)}{\partial r_\parallel}\cdot\frac{\partial}{\partial k_\parallel} - \frac{1}{2}\frac{\partial^2 f(r_\parallel)}{\partial r_\parallel \partial r_\parallel}:\frac{\partial^2}{\partial k_\parallel \partial k_\parallel}\right\} \times \mathrm{e}^{-\mathrm{i}k_\perp^- z}\begin{pmatrix} A_{12} \\ A_{22} \\ A_{32}\end{pmatrix}\right]_{k_\parallel = k_\parallel^i} \mathrm{e}^{\mathrm{i}(k_\parallel^i \cdot r_\parallel - \omega t)}. \tag{3.27}$$

Carrying out the differentiations with respect to $k_\parallel$, we find after substitution of $k_\parallel^i = n^-(\omega/c)(\sin\theta, 0, 0)$, with eq. (3.26),

$$n_e^{(2)}(r, t) = (\epsilon^+ - \epsilon^-)t_s^0 \exp\{\mathrm{i}(\omega/c)(n^- x \sin\theta - n^- z\cos\theta - ct)\}\begin{pmatrix} A_x \\ A_y \\ A_z\end{pmatrix}, \tag{3.28}$$

where

$$A_x = \sin\theta(n^+)^{-1}(n^+\cos\theta + n^-\cos\theta_t)\frac{\partial f}{\partial y}, \tag{3.29}$$

$$A_y = -\mathrm{i}(\omega/c)(n^-\cos\theta + n^+\cos\theta_t)^{-1} f(r_\parallel)$$
$$- \sin\theta(1 + \mathrm{i}(\omega/c)zn^+\cos\theta_t)(n^-\cos\theta + n^+\cos\theta_t)^{-1}$$
$$\times (n^+\cos\theta\cos\theta_t)^{-1}\frac{\partial f}{\partial x}$$

$$-\tfrac{1}{2}\mathrm{i}(c/\omega)(n^+n^-)^{-1}\{2(n^+\cos\theta + n^-\cos\theta_t)^{-1}$$

$$-(1+\mathrm{i}(\omega/c)zn^+\cos\theta_t)(n^-\cos\theta + n^+\cos\theta_t)^{-1}(\cos\theta\cos\theta_t)^{-1}\}\frac{\partial^2 f}{\partial y^2}$$

$$-\tfrac{1}{2}(n^-\cos\theta + n^+\cos\theta_t)^{-1}[-\mathrm{i}(c/\omega)\{\epsilon^+\cos^2\theta\cos^2\theta_t$$

$$+\sin^2\theta(\epsilon^+\cos^2\theta_t + \epsilon^-\cos^2\theta + n^+n^-\cos\theta\cos\theta_t)\}$$

$$\times(n^-)^{-1}(n^+\cos\theta\cos\theta_t)^{-3} + z(n^+\cos\theta_t + 2n^-\cos\theta\sin^2\theta$$

$$+\mathrm{i}(\omega/c)zn^+n^-\cos\theta\cos\theta_t\sin^2\theta)(n^+n^-\cos^3\theta\cos\theta_t)^{-1}]\frac{\partial^2 f}{\partial x^2}. \tag{3.30}$$

$$A_z = -n^-\cos\theta_t(n^+\cos\theta + n^-\cos\theta_t)^{-1}\frac{\partial f}{\partial y}. \tag{3.31}$$

In the above expressions we have omitted terms proportional to $\partial^2 f/\partial x\partial y$, since such terms will not contribute to the reflection amplitude r_s, as we shall see below.

The value of $\boldsymbol{n}_e^{(2)}(\boldsymbol{r},t)$ for $z = f(\boldsymbol{r}_\parallel) - 0$, if $f(\boldsymbol{r}_\parallel) < 0$, is obtained from eq. (3.28), together with eqs. (3.29)–(3.31), by substitution of $z = f(\boldsymbol{r}_\parallel)$. It will, however, immediately be clear that, since the functions appearing in these equations are analytic in z, the above value for $f(\boldsymbol{r}_\parallel) > 0$ is found in the same way. The value of $\boldsymbol{n}_e^{(2)}(\boldsymbol{r},t)$ on the upper surface of the (identical) film in the ambient (see fig. 1) is therefore found by substitution of $z = f(\boldsymbol{r}_\parallel)$ into eqs. (3.28)–(3.31).

In an analogous way one can calculate the other terms in the right-hand side of eq. (3.12). For the third term we find with eq. (3.13)

$$\boldsymbol{n}_e^{(3)}(\boldsymbol{r},t) = (\epsilon^+ - \epsilon^-)^2 t_s^0(n^-\cos\theta + n^+\cos\theta_t)^{-1}f(\boldsymbol{r}_\parallel)$$

$$\times\exp\{\mathrm{i}(\omega/c)(n^-x\sin\theta - n^-z\cos\theta - ct)\}$$

$$\times\Big[-(\omega/c)^2(n^-\cos\theta + n^+\cos\theta_t)^{-1}f(\boldsymbol{r}_\parallel)$$

$$-\tfrac{1}{2}(n^+n^-)^{-1}\{2(n^+\cos\theta + n^-\cos\theta_t)^{-1}$$

$$-(n^-\cos\theta + n^+\cos\theta_t)^{-1}(\cos\theta\cos\theta_t)^{-1}\}\frac{\partial^2 f}{\partial y^2}$$

$$+\tfrac{1}{2}\{\epsilon^+\cos^2\theta\cos^2\theta_t + \sin^2\theta(\epsilon^+\cos^2\theta_t + \epsilon^-\cos^2\theta$$

$$+n^+n^-\cos\theta\cos\theta_t)\}(n^-\cos\theta + n^+\cos\theta_t)^{-1}$$

$$\times(n^-)^{-1}(n^+\cos\theta\cos\theta_t)^{-3}\frac{\partial^2 f}{\partial x^2}\Big]\begin{pmatrix}0\\1\\0\end{pmatrix}, \tag{3.32}$$

where we have neglected all derivatives of second order terms, since they will not contribute to r_s. In the same way one has

$$n_e^{(4)}(r, t) = \tfrac{1}{2}(\omega/c)^2(\epsilon^+ - \epsilon^-)t_s^0 n^+ \cos\theta_t(n^- \cos\theta + n^+ \cos\theta_t)^{-1}$$

$$\times f^2(r_\parallel)\exp\{i(\omega/c)(n^- x\sin\theta - n^- z\cos\theta - ct)\}\begin{pmatrix}0\\1\\0\end{pmatrix}. \tag{3.33}$$

For the fifth term at the right-hand side of eq. (3.12) we need the explicit form of the propagator L_0, eq. (I.3.19). In appendix A we find, for $z < 0$,

$$L_0(k_\parallel, z, \omega|z' = 0) = i(\omega/c)\,e^{-ik_\perp^- z}\begin{pmatrix}B_{11} & B_{12} & B_{13}\\ B_{21} & B_{22} & B_{23}\\ B_{31} & B_{32} & B_{33}\end{pmatrix},$$

$$\begin{aligned}
B_{11} &= -\{k_\perp^+(k_\perp^+ + k_\perp^-)^{-1} - \epsilon^+ k_\perp^-(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1}\}k_x k_y k_\parallel^{-2},\\
B_{12} &= -k_\perp^+(k_\perp^+ + k_\perp^-)^{-1}k_y^2 k_\parallel^{-2} - \epsilon^+ k_\perp^-(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1}k_x^2 k_\parallel^{-2},\\
B_{13} &= -k_y(k_\perp^+ + k_\perp^-)^{-1},\\
B_{21} &= k_\perp^+(k_\perp^+ + k_\perp^-)^{-1}k_x^2 k_\parallel^{-2} + \epsilon^+ k_\perp^-(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1}k_y^2 k_\parallel^{-2},\\
B_{22} &= \{k_\perp^+(k_\perp^+ + k_\perp^-)^{-1} - \epsilon^+ k_\perp^-(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1}\}k_x k_y k_\parallel^{-2},\\
B_{23} &= k_x(k_\perp^+ + k_\perp^-)^{-1},\\
B_{31} &= \epsilon^+\epsilon^- k_y(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1},\\
B_{32} &= -\epsilon^+\epsilon^- k_x(\epsilon^+ k_\perp^- + \epsilon^- k_\perp^+)^{-1},\\
B_{33} &= 0.
\end{aligned} \tag{3.34}$$

With this expression we find

$$n_e^{(5)}(r, t) = \tfrac{1}{2}(\omega/c)^2(\epsilon^+ - \epsilon^-)t_s^0 n^+ \cos\theta_t(n^- \cos\theta + n^+ \cos\theta_t)^{-1}$$

$$\times f^2(r_\parallel)\exp\{i(\omega/c)(n^- x\sin\theta - n^- z\cos\theta - ct)\}\begin{pmatrix}0\\1\\0\end{pmatrix}. \tag{3.35}$$

From eqs. (3.13), (3.18), (3.28)–(3.31), (3.32), (3.33) and (3.35) we finally obtain the following second order expression for the electric field at the ambient side of the rough surface of an identical film:

$$\epsilon^-(r,t) = \sum_{i=1}^{s} \begin{pmatrix} n_{e,x}^{(i)}(r,t) \\ n_{e,y}^{(i)}(r,t) \\ (\epsilon^-)^{-1} n_{e,z}^{(i)}(r,t) \end{pmatrix}$$

$$= \exp\{i(\omega/c)(n^- x \sin\theta - n^- z \cos\theta - ct)\} \begin{pmatrix} A'_x \\ A'_y \\ A'_z \end{pmatrix}, \tag{3.36}$$

with

$$A'_x = (\epsilon^+ - \epsilon^-) t_s^0 \sin\theta (n^+)^{-1} (n^+ \cos\theta + n^- \cos\theta_t)^{-1} \frac{\partial f}{\partial y}, \tag{3.37}$$

$$\begin{aligned}
A'_y = {} & \exp\{2i(\omega/c) n^- z \cos\theta\} + r_s^0\{1 + 2i(\omega/c) n^- \cos\theta f(r_\parallel) \\
& - 2(\omega/c)^2 \epsilon^- \cos^2\theta\, f^2(r_\parallel)\} - 2(\epsilon^+ - \epsilon^-) n^- \sin\theta (n^+ \cos\theta_t)^{-1} \\
& \times (n^- \cos\theta + n^+ \cos\theta_t)^{-2} (1 + i(\omega/c) z n^+ \cos\theta_t) \frac{\partial f}{\partial x} \\
& - i(c/\omega)(\epsilon^+ - \epsilon^-) \cos\theta (n^+)^{-1} (n^- \cos\theta + n^+ \cos\theta_t)^{-1} \\
& \times \{2(n^+ \cos\theta + n^- \cos\theta_t)^{-1} - (1 + i(\omega/c) z n^+ \cos\theta_t) \\
& \times (n^- \cos\theta + n^+ \cos\theta_t)^{-1} (\cos\theta \cos\theta_t)^{-1}\} \frac{\partial^2 f}{\partial y^2} \\
& - (\epsilon^+ - \epsilon^-)(n^- \cos\theta + n^+ \cos\theta_t)^{-2} (n^+ \cos^2\theta \cos^3\theta_t)^{-1} \\
& \times [-i(c/\omega)(\epsilon^+)^{-1}\{\epsilon^+ \cos^2\theta \cos^2\theta_t + \sin^2\theta(\epsilon^+ \cos^2\theta_t \\
& + \epsilon^- \cos^2\theta + n^+ n^- \cos\theta \cos\theta_t)\} + z \cos^2\theta_t (n^+ \cos\theta_t \\
& + 2n^- \cos\theta \sin^2\theta + i(\omega/c) z n^+ n^- \cos\theta \cos\theta_t \sin^2\theta)] \frac{\partial^2 f}{\partial x^2} \\
& - (\epsilon^+ - \epsilon^-)^2 \cos\theta (n^+)^{-1} (n^- \cos\theta + n^+ \cos\theta_t)^{-2} \\
& \times \{2(n^+ \cos\theta + n^- \cos\theta_t)^{-1} - (n^- \cos\theta + n^+ \cos\theta_t)^{-1} \\
& \times (\cos\theta \cos\theta_t)^{-1}\} f(r_\parallel) \frac{\partial^2 f}{\partial y^2} \\
& + (\epsilon^+ - \epsilon^-)^2 \{\epsilon^+ \cos^2\theta \cos^2\theta_t + \sin^2\theta(\epsilon^+ \cos^2\theta_t + \epsilon^- \cos^2\theta \\
& + n^+ n^- \cos\theta \cos\theta_t)\} (n^- \cos\theta + n^+ \cos\theta_t)^{-3} (\cos\theta)^{-2} \\
& \times (n^+ \cos\theta_t)^{-3} f(r_\parallel) \frac{\partial^2 f}{\partial x^2},
\end{aligned} \tag{3.38}$$

$$A'_z = -(\epsilon^+ - \epsilon^-) t_s^0 \cos\theta_t (n^-)^{-1} (n^+ \cos\theta + n^- \cos\theta_t)^{-1} \frac{\partial f}{\partial y}, \tag{3.39}$$

where $z = f(r_\parallel)$. The above expression for the electric field will be used in the next section for the evaluation of the Helmholtz–Kirchhoff and Stratton–Chu–Silver integrals.

In order to compare the electric field obtained above with that found by means of the tangent plane approximation we substitute $z = f(r_\parallel)$ into eqs. (3.36)–(3.39). One then obtains

$$e^-(r_\parallel, f(r_\parallel), t) = t_s^0 \exp\{i(\omega/c)(n^- x \sin\theta + n^- f \cos\theta - ct)\} \begin{pmatrix} A''_x \\ A''_y \\ A''_z \end{pmatrix}, \tag{3.40}$$

with

$$A''_x = (\epsilon^+ - \epsilon^-) \sin\theta (n^+)^{-1} (n^+ \cos\theta + n^- \cos\theta_t)^{-1} \frac{\partial f}{\partial y}, \tag{3.41}$$

$$\begin{aligned} A''_y = {} & 1 - (\epsilon^+ - \epsilon^-) \sin\theta (n^- \cos\theta + n^+ \cos\theta_t)^{-1} (n^+ \cos\theta \cos\theta_t)^{-1} \frac{\partial f}{\partial x} \\ & + (\epsilon^+ - \epsilon^-) \cos\theta_t (n^-)^{-1} (n^+ \cos\theta + n^- \cos\theta_t)^{-1} \left(\frac{\partial f}{\partial y}\right)^2 \\ & + \tfrac{1}{2} (\epsilon^+ - \epsilon^-)^2 \sin^2\theta (n^+ \cos\theta \cos\theta_t)^{-2} (n^- \cos\theta + n^+ \cos\theta_t)^{-2} \left(\frac{\partial f}{\partial x}\right)^2, \end{aligned} \tag{3.42}$$

$$A''_z = -(\epsilon^+ - \epsilon^-) \cos\theta_t (n^-)^{-1} (n^+ \cos\theta + n^- \cos\theta_t)^{-1} \frac{\partial f}{\partial y}. \tag{3.43}$$

Note that in eq. (3.40) $\exp\{i(\omega/c) n^- f \cos\theta\}$ appears, instead of $\exp\{-i(\omega/c) n^- f \cos\theta\}$ which would follow from direct substitution of $z = f(r_\parallel)$ into eq. (3.36). We have furthermore replaced $f(\partial^2 f/\partial x^2)$ by $-(\partial f/\partial x)^2$ and $f(\partial^2 f/\partial y^2)$ by $-(\partial f/\partial y)^2$ in the derivation of the above equations, omitting the differences $\partial(f \partial f/\partial x)/\partial x$ and $\partial(f \partial f/\partial y)/\partial y$ which again do not contribute to r_s (see below).

In order to calculate the electric field, as given by the tangent plane approximation, we must make the following substitutions in eq. (3.4) (see ref. 10):

$$\boldsymbol{a} = (0, 1, 0)\,,$$

$$\boldsymbol{n} = \left\{1 + \left(\frac{\partial f}{\partial x}\right)^2 + \left(\frac{\partial f}{\partial y}\right)^2\right\}^{-1}\left(-\frac{\partial f}{\partial x}, -\frac{\partial f}{\partial y}, 1\right)$$

$$= \left(-\frac{\partial f}{\partial x}, -\frac{\partial f}{\partial y}, 1 - \tfrac{1}{2}\left(\frac{\partial f}{\partial x}\right)^2 - \tfrac{1}{2}\left(\frac{\partial f}{\partial y}\right)^2\right),$$

$$\boldsymbol{n}_1 = (\sin\theta, 0, \cos\theta)\,,$$

$$\boldsymbol{t} = \left(\cos\theta(\sin\theta)^{-1}\frac{\partial f}{\partial y}, -1 + \tfrac{1}{2}(\sin\theta)^{-2}\left(\frac{\partial f}{\partial y}\right)^2, -\frac{\partial f}{\partial y}\right), \tag{3.44}$$

$$\boldsymbol{d} = \left(\cos\theta - \tfrac{1}{2}\cos\theta(\sin\theta)^{-2}\left(\frac{\partial f}{\partial y}\right)^2, (\sin\theta)^{-1}\frac{\partial f}{\partial y}, -\sin\theta + \tfrac{1}{2}(\sin\theta)^{-1}\left(\frac{\partial f}{\partial y}\right)^2\right),$$

$$\hat{R}_s = (n^- \cos\varphi - n^+ \cos\varphi_t)(n^- \cos\varphi + n^+ \cos\varphi_t)^{-1}\,,$$

$$\hat{R}_p = (n^+ \cos\varphi - n^- \cos\varphi_t)(n^+ \cos\varphi + n^- \cos\varphi_t)^{-1}\,,$$

$$\cos\varphi = \boldsymbol{n}\cdot\boldsymbol{n}_1 = \cos\theta - \sin\theta\,\frac{\partial f}{\partial x} - \tfrac{1}{2}\cos\theta\left(\frac{\partial f}{\partial x}\right)^2 - \tfrac{1}{2}\cos\theta\left(\frac{\partial f}{\partial y}\right)^2,$$

$$\cos\varphi_t = (n^+)^{-1}(\epsilon^+ - \epsilon^- + \epsilon^- \cos^2\varphi)^{1/2}\,,$$

$$\boldsymbol{E}_0 = \exp\{\mathrm{i}(\omega/c)(n^- x \sin\theta + n^- f\cos\theta - ct)\}\boldsymbol{a}\,.$$

Substituting these second order expressions into the right-hand side of eq. (3.4), we find

$$\boldsymbol{E}^-(\boldsymbol{r}_\parallel, f(\boldsymbol{r}_\parallel), t) = t_s^0 \exp\{\mathrm{i}(\omega/c)(n^- x\sin\theta + n^- f\cos\theta - ct)\}\begin{pmatrix} A''_x \\ A'''_y \\ A''_z \end{pmatrix}, \tag{3.45}$$

where A''_x and A''_z are given respectively by eqs. (3.41) and (3.43), whereas

$$A'''_y = 1 - (\epsilon^+ - \epsilon^-)\sin\theta(n^- \cos\theta + n^+\cos\theta_t)^{-1}(n^+\cos\theta\cos\theta_t)^{-1}\frac{\partial f}{\partial x}$$

$$+ \tfrac{1}{2}(\epsilon^+ - \epsilon^-)\{2\cos\theta_t(n^-\cos\theta + n^+\cos\theta_t)(n^+\cos\theta + n^-\cos\theta_t)^{-1}$$

$$- n^-(n^-\cos\theta + n^+\cos\theta_t)^{-1}\}(n^+ n^-\cos\theta_t)^{-1}\left(\frac{\partial f}{\partial y}\right)^2$$

$$- \tfrac{1}{2}(\epsilon^+ - \epsilon^-)(n^+\cos\theta + 2n^-\cos\theta_t\sin^2\theta)$$

$$\times (n^-\cos\theta + n^+\cos\theta_t)^{-1}(\epsilon^+\cos\theta\cos^3\theta_t)^{-1}\left(\frac{\partial f}{\partial x}\right)^2. \tag{3.46}$$

As we see the y-component of the electric field in the tangent plane approximation is in second order different from our expression for the y-component of that field. As we have already mentioned (after eq. (3.22)), the tangent plane approximation can be obtained by using, instead of eq. (3.22), the expansion

$$f(\boldsymbol{r}_\parallel') = f(\boldsymbol{r}_\parallel) + (\boldsymbol{r}_\parallel' - \boldsymbol{r}_\parallel)\cdot\frac{\partial}{\partial \boldsymbol{r}_\parallel} f(\boldsymbol{r}_\parallel)\,. \tag{3.47}$$

In appendix B we demonstrate that, if we use this expansion and consistently neglect all terms with second and higher derivatives of f, we find an expression for the electric field identical to eq. (3.45). Comparing eqs. (3.45) and (3.40), or rather eqs. (3.46) and (3.42), one sees that the electric field on the rough surface, as given by the tangent plane approximation, is equal to the rigorous second order electric field except for the terms proportional to $(\partial f/\partial x)^2$ and $(\partial f/\partial y)^2$. From the previous arguments it will be clear that this difference is a direct consequence of the neglect of surface curvature. One can easily demonstrate that the neglected terms do contribute to the reflection amplitude r_s by examining $\langle \boldsymbol{e}(\boldsymbol{r}, t)\exp\{-\mathrm{i}(\omega/c)n^-(x\sin\theta - f\cos\theta)\}\rangle$, which is proporational to the first term in the Helmholtz–Kirchhoff integral, eq. (3.1). Using eqs. (3.36)–(3.39) we find

$$\begin{aligned}\langle \boldsymbol{e}^- \exp\{\mathrm{i}(\omega/c)n^- f\cos\theta\}\rangle &= \Big\{1 - 2(\omega/c)^2\epsilon^-\cos^2\theta\langle f^2\rangle + \tfrac{1}{2}(\epsilon^+ - \epsilon^-)\\ &\times [n^+n^-\cos\theta - (\epsilon^- - 2\epsilon^+)\cos\theta_t](n^-\cos\theta + n^+\cos\theta_t)^{-1}\\ &\times (n^+n^-\cos\theta_t)^{-1}(n^+\cos\theta + n^-\cos\theta_t)^{-1}\Big\langle\Big(\frac{\partial f}{\partial y}\Big)^2\Big\rangle + (\epsilon^+ - \epsilon^-)\\ &\times (2\cos^2\theta_t\sin^2\theta + \cos^2\theta)(n^-\cos\theta + n^+\cos\theta_t)^{-1}\\ &\times (n^+\cos^2\theta\cos^3\theta_t)^{-1}\Big\langle\Big(\frac{\partial f}{\partial x}\Big)^2\Big\rangle\Big\} t_s^0 \exp\{\mathrm{i}(\omega/c)n^- x\sin\theta\}\,\mathrm{e}^{-\mathrm{i}\omega t}\begin{pmatrix}0\\1\\0\end{pmatrix}, \end{aligned}\tag{3.48}$$

whereas with eqs. (3.45), (3.46), (3.41) and (3.43) we find

$$\begin{aligned}\langle \boldsymbol{E}^- \exp\{\mathrm{i}(\omega/c)n^- f\cos\theta\}\rangle &= \Big\{1 - 2(\omega/c)^2\epsilon^-\cos^2\theta\langle f^2\rangle + \tfrac{1}{2}(\epsilon^+ - \epsilon^-)\\ &\times [\cos\theta_t(2\epsilon^+\cos^2\theta_t + 2\epsilon^-\cos^2\theta - \epsilon^-) + \cos\theta\, n^+n^-(4\cos^2\theta_t - 1)]\\ &\times (n^+n^-\cos\theta_t)^{-1}(n^-\cos\theta + n^+\cos\theta_t)^{-1}(n^+\cos\theta + n^-\cos\theta_t)^{-1}\Big\langle\Big(\frac{\partial f}{\partial y}\Big)^2\Big\rangle\end{aligned}$$

$$-\tfrac{1}{2}(\epsilon^+ - \epsilon^-)(n^+ \cos\theta + 2n^- \cos\theta_t \sin^2\theta)(n^- \cos\theta + n^+ \cos\theta_t)^{-1}$$

$$\times (\epsilon^+ \cos\theta \cos^3\theta_t)^{-1} \left\langle \left(\frac{\partial f}{\partial x}\right)^2 \right\rangle \Big\} t_s^0 \exp\{i(\omega/c)n^- x \sin\theta\}\, e^{-i\omega t} \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}. \tag{3.49}$$

In the above derivations use has been made of the following relations, which follow by differentiation of $\langle f \rangle = 0$ and $\langle f^2 \rangle = \text{const.}$ with respect to x and y and from isotropy in the x–y plane:

$$\left\langle \frac{\partial f}{\partial x} \right\rangle = \left\langle \frac{\partial f}{\partial y} \right\rangle = 0, \quad \left\langle f \frac{\partial f}{\partial x} \right\rangle = \left\langle f \frac{\partial f}{\partial y} \right\rangle = 0,$$

$$\left\langle \frac{\partial^2 f}{\partial x \partial y} \right\rangle = 0, \quad \left\langle f \frac{\partial^2 f}{\partial x \partial y} \right\rangle = -\left\langle \left(\frac{\partial f}{\partial x}\right)\left(\frac{\partial f}{\partial y}\right) \right\rangle = 0, \tag{3.50}$$

$$\left\langle f \frac{\partial^2 f}{\partial x^2} \right\rangle = -\left\langle \left(\frac{\partial f}{\partial x}\right)^2 \right\rangle, \quad \left\langle f \frac{\partial^2 f}{\partial y^2} \right\rangle = -\left\langle \left(\frac{\partial f}{\partial y}\right)^2 \right\rangle.$$

It is evident from eq. (3.50) why it was allowed to omit terms proportional to $\partial^2 f/\partial x \partial y$ in the above calculations of the fields, and to replace $f(\partial^2 f/\partial x^2)$ and $f(\partial^2 f/\partial y^2)$ by $-(\partial f/\partial x)^2$ and $-(\partial f/\partial y)^2$, respectively.

The terms with $\langle(\partial f/\partial x)^2\rangle$ and $\langle(\partial f/\partial y)^2\rangle$ in eqs. (3.48) and (3.49) are clearly different. It follows that, even for very smooth surfaces, the curvature may not be neglected in a second order calculation of the electromagnetic field. Therefore it is not surprising that, using the incomplete field on the surface, as given by the tangent plane approximation, one finds results with the Helmholtz–Kirchhoff and the Stratton–Chu–Silver integral that are different from ours.

In the next section we shall prove that, using the correct second order expressions (3.36)–(3.39) in the Helmholtz–Kirchhoff integral, eq. (3.1), and in the Stratton–Chu–Silver integral, eq. (3.3), one finds in both cases the same values for r_s and $R_s = |r_s|^2$ as obtained in section 2 (eq. (2.12)).

4. The far field

In this section we will calculate the reflection amplitude r_s using the Helmholtz–Kirchhoff and Stratton–Chu–Silver integrals. In fact, now that we have derived the explicit expression for the electric field on the rough surface, eqs.

(3.36)–(3.39), the evaluation of these integrals is straightforward. Therefore we will not give these calculations in every detail. First we consider the Helmholtz–Kirchhoff integral, eq. (3.1). Using eqs. (3.2), (3.36) and (3.44) one finds in the direction of specular reflection:

$$E_p = -(4\pi R)^{-1}\, e^{in^-(\omega/c)R}\, e^{-i\omega t} \iint_S \left[\left\{ 2i(\omega/c)n^-\left(\cos\theta + \frac{\partial f}{\partial x}\sin\theta\right) - \begin{pmatrix} -\partial f/\partial x \\ -\partial f/\partial y \\ 1 \end{pmatrix} \cdot \frac{\partial}{\partial r} \right\} \begin{pmatrix} A_x' \\ A_y' \\ A_z' \end{pmatrix}\right]_{z=f(\eta)} dx\, dy\,, \tag{4.1}$$

where we have also used that the surface elements are given by

$$\begin{aligned} dS &= dx\left\{1+\left(\frac{\partial f}{\partial x}\right)^2\right\}^{1/2} dy\left\{1+\left(\frac{\partial f}{\partial y}\right)^2\right\}^{1/2} \\ &= dx\, dy\left\{1+\tfrac{1}{2}\left(\frac{\partial f}{\partial x}\right)^2+\tfrac{1}{2}\left(\frac{\partial f}{\partial y}\right)^2\right\}. \end{aligned} \tag{4.2}$$

In order to calculate r_s one must average the field E_p. One then obtains

$$\langle E_p\rangle = -(4\pi R)^{-1}\, e^{in^-(\omega/c)R}\, e^{-i\omega t} L^2 \left\langle \left[\left\{ 2i(\omega/c)n^-\left(\cos\theta + \frac{\partial f}{\partial x}\sin\theta\right) - \begin{pmatrix} -\partial f/\partial x \\ -\partial f/\partial y \\ 1 \end{pmatrix} \cdot \frac{\partial}{\partial r} \right\} \begin{pmatrix} A_x' \\ A_y' \\ A_z' \end{pmatrix}\right]_{z=f(\eta)} \right\rangle, \tag{4.3}$$

where L is the linear dimension of the illuminated area.

Let us first consider the x- and z-components of the far field $\langle E_p\rangle$. From eqs. (3.37) and (3.39) one sees that both A_x' and A_z' are proportional to $\partial f/\partial y$. The only non-vanishing contributions are therefore of the form $(\partial f/\partial y)^2$, as follows from the relations (3.50). One easily verifies, however, that no such terms will appear in eq. (4.3). It follows that the x- and z-component of $\langle E_p\rangle$ are equal to zero.

To find the y-component of $\langle E_p\rangle$ we substitute A_y', eq. (3.38), into eq. (4.3). After some algebra one finds

$$\langle E_{p,y}\rangle = -i(\omega/c)n^- \cos\theta L^2 (2\pi R)^{-1} r_s^0\, e^{in^-(\omega/c)R}\, e^{-i\omega t}\Big[1 - 2(\omega/c)^2\epsilon^- \cos^2\theta\langle f^2\rangle$$
$$-(\epsilon^+ - \epsilon^-)\cos\theta(n^+)^{-1}\{2(n^+\cos\theta + n^-\cos\theta_t)^{-1}$$

$$
- (n^- \cos\theta + n^+ \cos\theta_t)^{-1}(\cos\theta \cos\theta_t)^{-1}\} \left\langle \left(\frac{\partial f}{\partial y}\right)^2 \right\rangle
$$

$$
+ (\epsilon^+ - \epsilon^-)\{\epsilon^+ \cos^2\theta \cos^2\theta_t + \sin^2\theta(\epsilon^+ \cos^2\theta_t
$$

$$
+ \epsilon^- \cos^2\theta + n^+ n^- \cos\theta \cos\theta_t)\}(n^- \cos\theta + n^+ \cos\theta_t)^{-1}
$$

$$
\times (n^+ \cos\theta_t)^{-3}(\cos\theta)^{-2} \left\langle \left(\frac{\partial f}{\partial x}\right)^2 \right\rangle \Big] . \tag{4.4}
$$

We now substitute $\langle f^2 \rangle \equiv \sigma^2$ and $\langle (\partial f/\partial x)^2 \rangle = \langle (\partial f/\partial y)^2 \rangle = 2\sigma^2/l^2$, which follows by evaluating $-\partial^2 S(r)/\partial x^2|_{x=0}$, where the correlation function is given by eqs. (2.2) and (2.4). We finally obtain

$$
\langle \boldsymbol{E}_P(\boldsymbol{r}) \rangle = -\,\mathrm{i}(\omega/c) n^- \cos\theta\, L^2 (2\pi R)^{-1} r_s^0\, \mathrm{e}^{\mathrm{i}(\omega/c)(n^- x \sin\theta - n^- z \cos\theta - ct)} \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}
$$

$$
\times [1 - 2\epsilon^-(\omega/c)^2 \sigma^2 \cos^2\theta + 2(\epsilon^+ - \epsilon^-) l^{-2} \sigma^2 \sin^2\theta
$$

$$
\times \cos^{-2}\theta (n^+ \cos\theta_t)^{-3} \{2 n^+ n^- \cos^2\theta \cos^2\theta_t
$$

$$
\times (n^+ \cos\theta + n^- \cos\theta_t)^{-1} + (\epsilon^+ \cos^2\theta_t + \epsilon^- \cos^2\theta
$$

$$
+ n^+ n^- \cos\theta \cos\theta_t)(n^- \cos\theta + n^+ \cos\theta_t)^{-1}\}] , \tag{4.5}
$$

where we have also used the fact that the point P is situated in the direction of specular reflection. This expression for the field is in agreement with eq. (I.5.2), except for the factor $-\mathrm{i}(\omega/c) n^- \cos\theta L^2 (2\pi R)^{-1}$ in eq. (4.5). The usual way to overcome this difficulty is to compare the obtained field with that reflected by the flat surface, see e.g. ref. 7. One so finds

$$
r_s = r_s^0 [1 - 2\epsilon^-(\omega/c)^2 \sigma^2 \cos^2\theta + 2(\epsilon^+ - \epsilon^-) l^{-2} \sigma^2 \sin^2\theta
$$

$$
\times \cos^{-2}\theta (n^+ \cos\theta_t)^{-3} \{2 n^+ n^- \cos^2\theta \cos^2\theta_t (n^+ \cos\theta + n^- \cos\theta_t)^{-1}
$$

$$
+ (\epsilon^+ \cos^2\theta_t + \epsilon^- \cos^2\theta + n^+ n^- \cos\theta \cos\theta_t)(n^- \cos\theta + n^+ \cos\theta_t)^{-1}\}] . \tag{4.6}
$$

To calculate the far field with the Stratton–Chu–Silver integral, eq. (3.3), one requires the explicit form of the magnetic field $\boldsymbol{H}$ at the rough surface. This field is most easily obtained from the electric field $\boldsymbol{E}$, eqs. (3.36)–(3.39), by applying Maxwell's equation $(\partial/\partial \boldsymbol{r}) \wedge \boldsymbol{E} = \mathrm{i}(\omega/c)\boldsymbol{B}$. Having thus found the magnetic field, the evaluation of the Stratton–Chu–Silver integral is mathematically very similar to that of the Helmholtz–Kirchhoff integral; for that reason we do not give the explicit calculation here. The final expression for the far away field $\langle \boldsymbol{E}_P \rangle$ one obtains is identical to eq. (4.5). Also the corresponding expression for the amplitude r_s is therefore identical to eq. (4.6). We want to stress that it is by no means obvious that these two integrals should lead to the same result, since the terms that appear in the calculation of these integrals are quite different. Whereas in the Helmholtz–Kirchhoff integral $\langle E_{P,y} \rangle$ is determined solely by A'_y, one finds that in the Stratton–Chu–Silver integral also terms originating from A'_x and A'_z contribute to $\langle E_{P,y} \rangle$.

We will now apply the method of section 2 to the calculation of the reflection amplitude r_s, in order to be able to compare the results. According to eq. (I.5.19) we have

$$r_s = r_s^0\{1 + 2i(\omega/c)\gamma_{tt}^{(c)} n^-(\epsilon^- - \epsilon^+)^{-1} \cos\theta + 4(\omega/c)^2 \tau^{(c)} n^- n^+ (\epsilon^- - \epsilon^+)^{-1} \cos\theta \cos\theta_t\}, \tag{4.7}$$

since we consider here only the influence of surface roughness. Substituting the expressions found in section 2 for the coefficients $\gamma_{tt}^{(c)}$ and $\tau^{(c)}$, eqs. (2.8) and (2.11), one finds

$$\begin{aligned} r_s = r_s^0[1 &- 2\epsilon^-(\omega/c)^2\sigma^2 \cos^2\theta + 2(\epsilon^+ - \epsilon^-)l^{-2}\sigma^2 \sin^2\theta \\ &\times \cos^{-2}\theta (n^+ \cos\theta_t)^{-3}\{2n^+ n^- \cos^2\theta \cos^2\theta_t (n^+ \cos\theta + n^- \cos\theta_t)^{-1} \\ &+ (\epsilon^+ \cos^2\theta_t + \epsilon^- \cos^2\theta + n^+ n^- \cos\theta \cos\theta_t)(n^- \cos\theta + n^+ \cos\theta_t)^{-1}\}]. \end{aligned} \tag{4.8}$$

which is identical to eq. (4.6). Thus we have demonstrated the equivalency of the different methods. On the other hand, when we consider the length and intricacy of the calculations, the advantage of using our method of the constitutive coefficients instead of the Helmholtz–Kirchhoff or Stratton–Chu–Silver integral is obvious.

5. Discussion

A major conclusion to be drawn from this paper is that the tangent plane approximation, though intuitively appealing, cannot lead to correct results for the reflection and transmission amplitudes. Our objection against the tangent plane approximation is based on the simple equations $\langle (\partial f/\partial x)^2 \rangle = -\langle f \partial^2 f/\partial x^2 \rangle$ and $\langle (\partial f/\partial y)^2 \rangle = -\langle f \partial^2 f/\partial y^2 \rangle$, which hold for illuminated surfaces of which the linear dimension is much larger than the correlation length. These relations demonstrate that the curvature terms with $\partial^2 f/\partial x^2$ and $\partial^2 f/\partial y^2$ are just as important in the local field as the terms with $(\partial f/\partial x)^2$ and $(\partial f/\partial y)^2$. Therefore even for rough surfaces where the radius of curvature is extremely large one may not neglect the curvature terms. Since in the tangent plane approximation these terms are neglected one can expect to obtain correctly only the $\langle f^2\rangle/\lambda^2$ terms in the reflection and transmission amplitudes, which are almost trivial. The above argument was given for the case of small surface roughness ($\sigma \ll \lambda/2\pi$). It is easily extended to the case of moderately rough surfaces ($\sigma \lesssim \lambda/2\pi$) by using the relation

$$2i(\omega/c)n^- \cos\theta\langle(\partial f/\partial x)^2 \exp\{2i(\omega/c)n^- \cos\theta\ f\}\rangle$$
$$= -\langle\partial^2 f/\partial x^2 \exp\{2i(\omega/c)n^- \cos\theta\ f\}\rangle\,.$$

An interesting point to observe is that in the analysis of Ohlídal and Lukeš[4,5]) the Helmholtz–Kirchhoff integral and the Stratton–Chu–Silver integral lead to different results, whereas we find that the integrals lead to identical results. In principle both integrals give exact results when the field is exactly known over the whole surface. Since this is in general not the case one can obtain an approximate solution by calculating the field in the directly illuminated area, neglecting the secondary field on the remaining part of the surface. In this approximation the Stratton–Chu–Silver integral will lead to better results than the Helmholtz–Kirchhoff integral, since it compensates the discontinuity in the field by charge densities on the contour of the irradiated area[11]). The Stratton–Chu–Silver integral can be expressed in the Helmholtz–Kirchhoff integral with additional contour integrals, see eq. (8.15.30) of ref. 11. Now for illuminated surfaces of which the linear dimension is much larger than the wavelength of light the contribution of these contour integrals to the reflection and transmission amplitudes is negligible compared with the surface integral. One should therefore expect that the two integrals give the same result. The remark of Ohlídal and Lukeš[5]), when discussing the difference in their results, that the expression for the ellipsometric coefficient obtained with the Stratton–Chu–Silver integral is more general than that obtained with the Helmholtz–Kirchhoff integral seems therefore out of place. The difference in results indicates an inconsistency in their approach. In this connection we also want to point once more at the wrong result, obtained by Ohlídal and Lukeš[4]) with the Helmholtz–Kirchhoff integral, that the ellipsometric coefficient for normally incident light is not equal to -1 (see also eq. (2.21) of the present paper).

We have demonstrated in section 4 that the two integrals both lead to the same result as obtained with our method of the constitutive coefficients in section 2. The advantage of using the latter method is clear, when one considers its simplicity in contrast to the long and complicated calculation of the local electromagnetic field on the rough surface, necessary for the evaluation of the integrals. This basic simplicity allows one to extend the analysis to more complicated systems without difficulty. One can study films where the four correlation functions of the upper and lower surface are no longer equal and where the substrate is absorbing. One can also, in principle, evaluate the next term in the expression for the coefficients, proportional to $\sigma^2(c/\omega)^2 l^{-4}$.

An advantage of the approach with the tangent plane approximation is that it allows one to study surfaces which are moderately rough ($\sigma \lesssim \lambda/2\pi$), whereas our theory is restricted to small surface roughness ($\sigma \ll \lambda/2\pi$). Since the two conditions $\sigma \ll \lambda/2\pi$ and $\lambda/2\pi \ll l$ in our theory will be limiting to the practical applications of the theory (how stringent these conditions are in practice will also depend on the different dielectric constants), one would prefer to extend our theory to moderate surface roughness. This would, however, require a reformulation of the theory, since the surface polarization and magnetization densities $\boldsymbol{p}^s$ and $\boldsymbol{m}^s$ can no longer be constructed by analytic extension of the electromagnetic fields to the plane $z = 0$. One possibility would be to define these densities with respect to local planes in surface areas large compared with the wavelength and small compared with the correlation length.

Appendix A

The construction of the propagator $\boldsymbol{L}_0$

In this appendix we shall derive the explicit expression for the propagator $\boldsymbol{L}_0$. To this end it is most convenient to express $\boldsymbol{L}_0$ in terms of the propagator $\boldsymbol{K}_0$, which was explicitly constructed in appendix B of ref. 9. We start with the definitions of these two propagators, eq. (I.3.19),

$$\boldsymbol{K}_0 = (1 + \boldsymbol{K} \cdot \xi_0)^{-1} \cdot \boldsymbol{K}, \qquad \boldsymbol{L}_0 = (1 + \boldsymbol{K} \cdot \xi_0)^{-1} \cdot \boldsymbol{L}, \tag{A.1}$$

from which one immediately obtains the relation

$$\boldsymbol{L}_0 = \boldsymbol{K}_0 \cdot \boldsymbol{K}^{-1} \cdot \boldsymbol{L}. \tag{A.2}$$

Using the expressions for the propagators $\boldsymbol{K}$ and $\boldsymbol{L}$ in $\boldsymbol{k}, \omega$ representation, eqs.

(I.3.10), (I.3.11) and (I.3.7), we obtain after some calculation

$$L_0k, \omega) = (c/\omega)k_\parallel^{-2}\boldsymbol{K}_0(k, \omega)\cdot \begin{pmatrix} -k_xk_yk_z & -k_y^2k_z & k_\parallel^2k_y \\ k_x^2k_z & k_xk_yk_z & -k_\parallel^2k_x \\ -(\omega/c)^2k_y & (\omega/c)^2k_x & 0 \end{pmatrix} . \tag{A.3}$$

As already indicated in appendix B of ref. 9 it is more convenient to have these propagators in the $(k_\parallel, z, \omega)$ representation. After a one-dimensional Fourier transformation we find

$$\boldsymbol{L}_0(k_\parallel, z, \omega|z') = (c/\omega)k_\parallel^{-2}\int \mathrm{d}z''\boldsymbol{K}_0(k_\parallel, z, \omega|z'')\cdot\{\delta(z''-z')\boldsymbol{A} - \mathrm{i}\delta'(z''-z')\boldsymbol{B}\}, \tag{A.4}$$

where we have introduced the matrices

$$\boldsymbol{A} \equiv \begin{pmatrix} 0 & 0 & k_\parallel^2k_y \\ 0 & 0 & -k_\parallel^2k_x \\ -(\omega/c)^2k_y & (\omega/c)^2k_x & 0 \end{pmatrix} . \tag{A.5}$$

and

$$\boldsymbol{B} \equiv \begin{pmatrix} -k_xk_y & -k_y^2 & 0 \\ k_x^2 & k_xk_y & 0 \\ 0 & 0 & 0 \end{pmatrix} . \tag{A.6}$$

We can now substitute the expression for the propagator $\boldsymbol{K}_0(k_\parallel, z, \omega|z')$, which is given by eq. (B.17) of ref. 9, but where the coefficients $\gamma^\pm$ and $\beta^\pm$ in the reflection and transmission tensors $\boldsymbol{R}^\pm$ and $\boldsymbol{T}^\pm$ must be omitted. Thus we find the general expression for $\boldsymbol{L}_0$:

$$\begin{aligned} \boldsymbol{L}_0(k_\parallel, z, \omega|z') = {} & (c/\omega)k_\parallel^{-2}\sum_{\nu,\nu'} \eta^\nu(z)\eta^{\nu'}(z') \\ & \times[\{\boldsymbol{K}_{e^\nu}(k_\parallel, z-z', \omega)\cdot[\boldsymbol{A} + k_\perp^\nu\,\mathrm{sign}(z-z')\boldsymbol{B}] \\ & + \boldsymbol{K}_{e^\nu}(k_\parallel, z+z', \omega)\cdot\boldsymbol{R}^\nu\cdot[\boldsymbol{A} - k_\perp^\nu\,\mathrm{sign}(z+z')\boldsymbol{B}]\}\delta(\nu-\nu') \\ & + \boldsymbol{K}_{e^\nu}(k_\parallel, z - k_\perp^{\nu'}z'/k_\perp^\nu, \omega)\cdot\boldsymbol{T}^{\nu'}\cdot[\boldsymbol{A} + k_\perp^{\nu'}\,\mathrm{sign}(z-z')\boldsymbol{B}]\delta(\nu+\nu')] , \end{aligned} \tag{A.7}$$

where the various quantities in this formula are given by eqs. (B.3), (B.15)–(B.28) of ref. 9. Eq. (3.34) follows now straightforwardly from this expression by taking the limit $z'\to 0$ (the results for the limits $z'\to -0$ and $z'\to +0$ are identical, as a consequence of the continuity of the n_m-field), and carrying out the matrix multiplications.

Appendix B

The tangent plane approximation

We will prove in this appendix that the electric field

$$e^-(r_\parallel, f(r_\parallel, t), t) = t_s^0 \exp\{i(\omega/c)(n^- x \sin\theta + f\cos\theta - ct)\}\begin{pmatrix} a_x \\ a_y \\ a_z \end{pmatrix} \tag{B.1}$$

one obtains using expansion (3.47) is identical to that found with the tangent plane approximation, eq. (3.45). As we have seen in section 3 (see text after eq. (3.47)) we therefore have to demonstrate only that the terms with $(\partial f/\partial x)^2$ and $(\partial f/\partial y)^2$ in a_y are equal to those in A''_y, eq. (3.46). Let us denote by $a_y^{(i)}$ ($i = 1, 2, 3, 4, 5$) the contributions of the fields $n_e^{(i)}$, eq. (3.13), to a_y, and furthermore by $C_y^{(i)}$ the terms with $(\partial f/\partial x)^2$ and $(\partial f/\partial y)^2$ in $a_y^{(i)}$. We then obtain, in an analogous way as in section 3, from eq. (3.12) in the approximation eq. (3.47)

$$C_y^{(1)} = C_y^{(2)} = 0\,, \tag{B.2}$$

$$\begin{aligned} C_y^{(3)} = {} & [(\epsilon^+ - \epsilon^-)^2 \sin^2\theta(n^-\cos\theta + n^+\cos\theta_t)^{-2}(n^+\cos\theta\cos\theta_t)^{-2} \\ & + (\epsilon^+ - \epsilon^-)^2\{\epsilon^+\cos^2\theta\cos^2\theta_t + \sin^2\theta(\epsilon^+\cos^2\theta_t + \epsilon^-\cos^2\theta \\ & + n^+n^-\cos\theta\cos\theta_t)\}(n^-\cos\theta + n^+\cos\theta_t)^{-2}(n^+\cos\theta\cos\theta_t)^{-3} \\ & \times (n^-)^{-1}]\left(\frac{\partial f}{\partial x}\right)^2 + [(\epsilon^+ - \epsilon^-)^2\sin^2\theta(n^+\cos\theta + n^-\cos\theta_t)^{-2}(\epsilon^+)^{-1} \\ & + (\epsilon^+ - \epsilon^-)\cos\theta(n^-\cos\theta + n^+\cos\theta_t)(n^+\cos\theta + n^-\cos\theta_t)^{-2} \\ & \times (n^-)^{-1} - (\epsilon^+ - \epsilon^-)^2\{2(n^+\cos\theta + n^-\cos\theta_t)^{-1} \\ & - (n^-\cos\theta + n^+\cos\theta_t)^{-1}(\cos\theta\cos\theta_t)^{-1}\}(n^+n^-)^{-1} \\ & \times (n^-\cos\theta + n^+\cos\theta_t)^{-1}]\left(\frac{\partial f}{\partial y}\right)^2, \end{aligned} \tag{B.3}$$

$$\begin{aligned} C_y^{(4)} = {} & -\tfrac{1}{2}(\epsilon^+ - \epsilon^-)\{\epsilon^+\cos^2\theta\cos^2\theta_t + \sin^2\theta(\epsilon^+\cos^2\theta_t \\ & + \epsilon^-\cos^2\theta + n^+n^-\cos\theta\cos\theta_t)\}(n^-\cos\theta + n^+\cos\theta_t)^{-1} \end{aligned}$$

$$\times (n^- \epsilon^+ \cos^3\theta \cos^2\theta_t)^{-1} \left(\frac{\partial f}{\partial x}\right)^2$$

$$+ \tfrac{1}{2}(\epsilon^+ - \epsilon^-) \cos\theta_t (n^-)^{-1} \{2(n^+ \cos\theta + n^- \cos\theta_t)^{-1}$$

$$- (n^- \cos\theta + n^+ \cos\theta_t)^{-1} (\cos\theta \cos\theta_t)^{-1}\} \left(\frac{\partial f}{\partial y}\right)^2 , \tag{B.4}$$

$$C_y^{(5)} = -\tfrac{1}{2}(\epsilon^+ - \epsilon^-)^2 \{\epsilon^+ \cos^2\theta \cos^2\theta_t + \sin^2\theta (\epsilon^+ \cos^2\theta_t$$

$$+ \epsilon^- \cos^2\theta + 2n^+ n^- \cos\theta \cos\theta_t)\} (n^- \cos\theta + n^+ \cos\theta_t)^{-2}$$

$$\times (n^+ \cos\theta \cos\theta_t)^{-3} (n^-)^{-1} \left(\frac{\partial f}{\partial x}\right)^2$$

$$+ \tfrac{1}{2}(\epsilon^+ - \epsilon^-)^2 (n^- \cos\theta + n^+ \cos\theta_t)^{-1} (n^+ n^-)^{-1}$$

$$\times \{2(n^+ \cos\theta + n^- \cos\theta_t)^{-1} - (n^- \cos\theta + n^+ \cos\theta_t)^{-1}$$

$$\times (\cos\theta \cos\theta_t)^{-1}\} \left(\frac{\partial f}{\partial y}\right)^2 . \tag{B.5}$$

Taking the sum of these contributions one finds the terms with $(\partial f/\partial x)^2$ and $(\partial f/\partial y)^2$ in a_y, which we will denote by C_y:

$$C_y = -\tfrac{1}{2}(\epsilon^+ - \epsilon^-)(n^+ \cos\theta + 2n^- \cos\theta_t \sin^2\theta)$$

$$\times (n^- \cos\theta + n^+ \cos\theta_t)^{-1} (\epsilon^+ \cos\theta \cos^3\theta_t)^{-1} \left(\frac{\partial f}{\partial x}\right)^2$$

$$+ \tfrac{1}{2}(\epsilon^+ - \epsilon^-) \{2 \cos\theta_t (n^- \cos\theta + n^+ \cos\theta_t)$$

$$\times (n^+ \cos\theta + n^- \cos\theta_t)^{-1} - n^- (n^- \cos\theta + n^+ \cos\theta_t)^{-1}\}$$

$$\times (n^+ n^- \cos\theta_t)^{-1} \left(\frac{\partial f}{\partial y}\right)^2 . \tag{B.6}$$

Clearly these terms are identical to those in eq. (3.46).

References

1) M.M. Wind and J. Vlieger, Physica **125A** (1984) 75.
2) A.M. Albano, D. Bedeaux and J. Vlieger, Physica **102A** (1980) 105.
3) I. Ohlídal, K. Navrátil and F. Lukeš, J. Opt. Soc. Am. **61** (1971) 1630.
4) I. Ohlídal and F. Lukeš, Opt. Acta **19** (1972) 817.

5) I. Ohlídal and F. Lukeš, Opt. Commun. **7** (1973) 76.
6) B.J.A. Zielińska and D. Bedeaux, Physica **112A** (1982) 265.
7) P. Beckmann and A. Spizzichino, The Scattering of Electromagnetic Waves from Rough Surfaces (Pergamon, Oxford, 1963) pp. 93, 29, 20, 22.
8) B.J.A. Zielińska, D. Bedeaux and J. Vlieger, Physica **107A** (1981) 91.
9) J. Vlieger and D. Bedeaux, Physica **82A** (1976) 221.
10) A.K. Fung, Appl. Sci. Res. **18** (1967) 50.
11) J.A. Stratton, Electromagnetic Theory (McGraw-Hill, New York 1941), pp. 468, 469.

THE POLARIZABILITY OF A TRUNCATED SPHERE ON A SUBSTRATE I

1. INTRODUCTION

In a previous paper [1)] a statistical theory was developed for the surface dielectric susceptibilities of thin island films. Local field effects, due to the interaction of the particles, were taken into account. The theory was applied [2)] to discontinuous gold films on a glass substrate. Using the electron micrographs of the films studied experimentally by Norrman, Andersson, Granqvist and Hunderi [3)], we determined the average polarizability of the islands, assuming that they were to a good approximation prolate spheroids with their long axes parallel to the surface of the substrate. The effects due to the mirror images in the substrate were taken into account in dipole approximation. Furthermore the polarizability density auto correlation functions were determined for the films, from which the optical thickness could be evaluated, using the theory developed in the first paper. Then the transmittance curves were calculated in the optical and infra red region and compared with the experimental curves [3)]. The agreement for all films was good, if the average of the dielectric constants of the substrate and the ambient was used as the inter-island dielectric constant.

In the present paper we shall study the optical properties of particles of different shapes, namely truncated spheres on a substrate. As in reference 2, the particles are assumed to be much smaller than the wavelength of light, so that they can be considered as point dipoles for the calculation of the far field, but, in contrast with ref. 2, mirror image effects due to the substrate will be taken into account rigorously to arbitrary order in the multipole moments.

In reference 4 Berreman has introduced a method to calculate the influence of hemispherical bumps or pits in a paper on the optical behaviour of a crystal surface. In section 2 we extend this method to the more general case of a truncated sphere small compared to the wavelength on a substrate with a dielectric constant that may differ from those of the ambient and the particle. An infinite set of inhomogeneous linear equations for the multipole coefficients is derived, where the matrix elements are given in the form of integrals. Approximate solutions can be obtained by truncating this set. Expressions for the resulting polarizabilities of the particle, parallel and normal to the surface, and the optical properties for a distribution of truncated spheres on the surface are given.

In sections 3 to 5 we shall examine a number of special cases, where the integrals can easily be evaluated analytically. In section 3 we treat the case of a sphere on (or above) a substrate. This system has also been considered by Ruppin [5]. Since he uses bispherical coordinates he finds a set of linear equations which differs from ours, but gives rise to identical numerical results. In section 4 we consider a hemisphere on a substrate. For a particle with dielectric constant equal to that of the substrate this is identical to the system considered by Berreman [4]. The more general case of a hemispherical particle on a substrate was also treated by Chauvaux and Meessen [6]. Finally in section 5 the case of a very thin spherical cap on a substrate is considered. We find that in lowest order the polarizabilities parallel and normal to the surface are independent of the dielectric constant of the substrate.

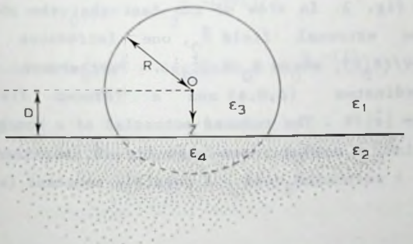
In the general case of a truncated sphere on a substrate the evaluation of the above mentioned matrix elements is more complicated. In a future paper we shall develop a systematic method to perform this calculation analytically.

2. THE POLARIZABILITY OF A TRUNCATED SPHERE

We consider a truncated sphere of radius R with a frequency dependent dielectric constant $\varepsilon_3(\omega)$ on a substrate with a dielectric constant $\varepsilon_2(\omega)$ and surrounded by an ambient with a dielectric constant $\varepsilon_1(\omega)$. The surface of the substrate is planar. See also figures 1a and 1b for a cross section of the system normal to the substrate and through the centre of the sphere. The centre of the sphere is chosen as the origin of our coordinate system. The positive z axis is chosen normal to the surface in the direction of the substrate. The surface of the substrate is given by z=D where $|D|<R$.

In fig. 1a we have drawn a situation where the centre of the sphere lies above the substrate (D>0) and in fig. 1b the case that the centre lies below the substrate (D<0). In the analysis it is convenient to replace the substrate in region 4 by a different material with a dielectric constant $\varepsilon_4(\omega)$. It is then clear that the two situations drawn in fig. 1a and 1b are in fact identical if one interchanges ε_1 and ε_2 (ambient and substrate) and also ε_3 and ε_4 (particle and substrate). In view of the above we may choose $0 < D < R$, as we will further proceed to do.

a



b

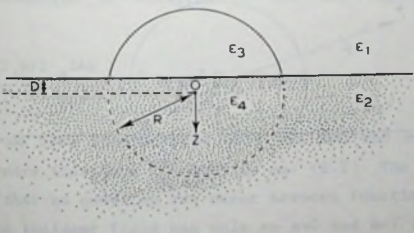


Fig. 1 Cross section of a truncated sphere on a substrate, (a) with D>0 and (b) with D<0.

For particles small compared to the minimum wavelength of the incident radiation in the media involved retardation effects become unimportant in and around the particle. We may therefore consider the incident field to be homogeneous and use the Laplace equation for the potential

$$\Delta\phi(\vec{r}) = 0 \tag{2.1}$$

in the particle, the ambient and the substrate (Δ is the Laplace operator). At the surface of the particle and the substrate the potential and the dielectric constant times the normal derivative of the potential must be continuous.

Since the centre of the sphere coincides with the ambient-substrate interface only for the special case of a hemispherical particle, we have to extend Berreman's method [4] of constructing the potentials. For this purpose we use image dipoles and multipoles situated in (0,0,2D) to describe the field due to the surface charge distribution on the substrate, see fig. 2. In view of the fact that the potential will be linear in the external field $\vec{E}_0$, one introduces [4] a reduced potential $\phi \equiv -V/(E_0R)$, where $E_0 \equiv |\vec{E}_0|$. Furthermore one introduces spherical coordinates (ρ,Θ,ϕ) and a reduced distance to the origin $r \equiv \rho/R = |\vec{r}|/R$. The reduced potential of a homogeneous external field $\vec{E}_0 = E_0(\sin\Theta_0, 0, \cos\Theta_0)$, see also fig. 2, may then be written as $\phi = r\cos\Theta\cos\Theta_0 + r\sin\Theta\sin\Theta_0\cos\phi$. A possible constant (and E

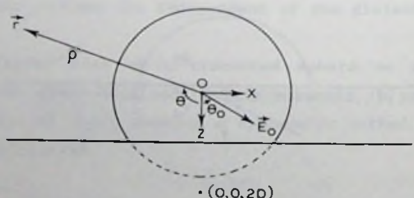


Fig. 2 Choice of the coordinate system for a truncated sphere on a substrate.

independent) contribution to this incident potential has been chosen zero. The total reduced potential in region 1 can now be written as

$$\begin{aligned}\Phi_1 = {} & r\cos\Theta\cos\Theta_0 + r\sin\Theta\sin\Theta_0\cos\Phi \\ & + \sum_{j=1}^{\infty} r^{-j-1}\{A_{1j}P_j^0(\cos\Theta) + B_{1j}P_j^1(\cos\Theta)\cos\Phi\} \\ & + \sum_{j=1}^{\infty} \{A'_{1j}V_j^0(r,\cos\Theta) + B'_{1j}V_j^1(r,\cos\Theta)\cos\Phi\} \quad . \qquad (2.2)\end{aligned}$$

The terms with the multipole coefficients A_{1j} and B_{1j} give the contributions to the reduced potential in region 1 due to the multipole of order j induced in the sphere by the external field. Similarly the terms with A'_{1j} and B'_{1j} give the contributions to the reduced potential in region 1 due to the image multipoles of the j^{th} order situated in (0,0,2D), which are used to describe the influence of the substrate. The functions $V_j^0(r,\cos\Theta)$ and $V_j^1(r,\cos\Theta)$ are defined by

$$\begin{aligned}V_j^m(r,\cos\Theta) \equiv {} & (r^2 - 4rr_0\cos\Theta + 4r_0^2)^{-(j+1)/2} \\ & \times P_j^m\left((r\cos\Theta - 2r_0)(r^2 - 4rr_0\cos\Theta + 4r_0^2)^{-1/2}\right) , \quad (m = 0,1) , \qquad (2.3)\end{aligned}$$

where we have introduced the truncation parameter

$$r_0 \equiv D/R \quad . \qquad (2.4)$$

We use the convention of Stratton [7] for the associated Legendre functions:

$$P_\ell^m(\eta) \equiv \frac{(1-\eta^2)^{m/2}}{2^\ell \ell!} \frac{d^{\ell+m}}{d\eta^{\ell+m}} (\eta^2 - 1)^\ell \qquad (m\geq 0) \quad . \qquad (2.5)$$

Note that we have incorporated only the m=0 and m=1 terms of the associated Legendre functions $P_j^m(\cos\Theta)$ in eq. (2.2). The symmetry of the system is such that no coupling can occur between functions of different m values and the incident field has only an m=0 and m=1 term.

For the potentials in the other three regions we obtain similar expressions:

$$\psi_2 = \psi_2' + \alpha r\cos\Theta\cos\Theta_0 + \beta r\sin\Theta\sin\Theta_0\cos\phi$$

$$+ \sum_{j=1}^{\infty} r^{-j-1}\{A_{2j}P_j^0(\cos\Theta) + B_{2j}P_j^1(\cos\Theta)\cos\phi\} \quad , \tag{2.6}$$

$$\psi_3 = \psi_3' + \sum_{j=1}^{\infty} r^j\{A_{3j}P_j^0(\cos\Theta) + B_{3j}P_j^1(\cos\Theta)\cos\phi\}$$

$$+ \sum_{j=1}^{\infty} \{A_{3j}'W_j^0(r,\cos\Theta) + B_{3j}'W_j^1(r,\cos\Theta)\cos\phi\} \quad , \tag{2.7}$$

$$\psi_4 = \psi_4' + \sum_{j=1}^{\infty} r^j\{A_{4j}P_j^0(\cos\Theta) + B_{4j}P_j^1(\cos\Theta)\cos\phi\} \quad , \tag{2.8}$$

where ψ_2' , ψ_3' and ψ_4' are constants which can not be chosen equal to zero. Furthermore α and β are constants and the functions $W_j^m(r,\cos\Theta)$ are defined by

$$W_j^m(r,\cos\Theta) = (r^2 - 4rr_0\cos\Theta + 4r_0^2)^{j/2}$$

$$\times P_j^m\left((r\cos\Theta - 2r_0)(r^2 - 4rr_0\cos\Theta + 4r_0^2)^{-1/2}\right) , \quad (m = 0,1) . \tag{2.9}$$

The boundary conditions at the surface of the substrate, z=D or $r\cos\Theta = r_0$, are easily solved as a consequence of our choice of potentials. The continuity of the potentials ψ_1 and ψ_2 along this boundary leads to the equation

$$-\psi_2' + (1 - \alpha)r_0\cos\Theta_0 + (1 - \beta)(r^2 - r_0^2)^{1/2}\sin\Theta_0\cos\phi$$

$$+ \sum_{j=1}^{\infty} \{A_{1j} + (-1)^j A_{1j}' - A_{2j}\}(-1)^j V_j^0(r,r_0/r)$$

$$+ \sum_{j=1}^{\infty} \{B_{1j} + (-1)^{j+1} B_{1j}' - B_{2j}\}(-1)^{j+1} V_j^1(r,r_0/r)\cos\phi = 0 \quad . \tag{2.10}$$

From the continuity of the normal component of the electric displacement vector one finds

$$(\varepsilon_1 - \alpha\varepsilon_2)\cos\Theta_0$$

$$+ \sum_{j=1}^{\infty} \{\varepsilon_1 A_{1j} - \varepsilon_1(-1)^j A_{1j}' - \varepsilon_2 A_{2j}\}(-1)^{j+1} V_j^{0\prime}(r,r_0/r)$$

$$+ \sum_{j=1}^{\infty} \{\varepsilon_1 B_{1j} - \varepsilon_1(-1)^{j+1}B'_{1j} - \varepsilon_2 B_{2j}\}(-1)^j V_j^{1\,\prime}(r,r_0/r)\cos\phi = 0 \ . \quad (2.11)$$

The prime in $V_j^{m\,\prime}$ denotes differentiation with respect to z. Eqs. (2.10) and (2.11) must hold for r>1. One easily verifies that the following relations satisfy both equations

$$\psi'_2 = (1 - \varepsilon_1/\varepsilon_2)r_0\cos\Theta_0 \ , \qquad \beta = 1 \ , \qquad \alpha = \varepsilon_1/\varepsilon_2 \ ,$$

$$A_{2j} = \frac{2\varepsilon_1}{\varepsilon_1 + \varepsilon_2} A_{1j} \ , \qquad A'_{1j} = \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}(-1)^j A_{1j} \ ,$$

$$B_{2j} = \frac{2\varepsilon_1}{\varepsilon_1 + \varepsilon_2} B_{1j} \ , \qquad B'_{1j} = \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}(-1)^{j+1} B_{1j} \ . \quad (2.12)$$

In a completely analogous way one finds that the continuity of the potential and of the normal component of the displacement vector at the boundary z=D between the regions 3 and 4 can be achieved by choosing the following relations

$$\psi'_4 = \psi'_3 \ , \qquad A_{4j} = \frac{2\varepsilon_3}{\varepsilon_3 + \varepsilon_4} A_{3j} \ , \qquad A'_{3j} = \frac{\varepsilon_3 - \varepsilon_4}{\varepsilon_3 + \varepsilon_4}(-1)^j A_{3j} \ ,$$

$$B_{4j} = \frac{2\varepsilon_3}{\varepsilon_3 + \varepsilon_4} B_{3j} \ , \qquad B'_{3j} = \frac{\varepsilon_3 - \varepsilon_4}{\varepsilon_3 + \varepsilon_4}(-1)^{j+1} B_{3j} \ . \quad (2.13)$$

Note that as a consequence of the rotational symmetry the coefficients A and B do not couple.

We now turn to the boundary conditions at the surface of the sphere, r=1. We use the fact that spherical harmonics give a complete orthonormal set of functions on the surface of the sphere. Multiplying the boundary conditions on the surface of the sphere with these spherical harmonics and integrating over the surface of the sphere gives for m=0

$$\int_{-1}^{r_0} dt \int_0^{2\pi} d\phi(\psi_1 - \psi_3)_{r=1} P_k^0(t) + \int_{r_0}^{1} dt \int_0^{2\pi} d\phi(\psi_2 - \psi_4)_{r=1} P_k^0(t) = 0 \ ,$$

$$\int_{-1}^{r_0} dt \int_0^{2\pi} d\phi(\varepsilon_1 \frac{\partial}{\partial r}\psi_1 - \varepsilon_3 \frac{\partial}{\partial r}\psi_3)_{r=1} P_k^0(t)$$

$$+\int_{r_0}^{1} dt \int_0^{2\pi} d\phi\left(\varepsilon_2 \frac{\partial}{\partial r}\psi_2 - \varepsilon_4 \frac{\partial}{\partial r}\psi_4\right)_{r=1} P_k^0(t) = 0 \quad , \tag{2.14}$$

where we have introduced $t \equiv \cos\Theta$, and for m=1

$$\int_{-1}^{r_0} dt \int_0^{2\pi} d\phi(\psi_1 - \psi_3)_{r=1} P_k^1(t)\cos\phi + \int_{-1}^{r_0} dt \int_0^{2\pi} d\phi(\psi_2 - \psi_4)_{r=1} P_k^1(t)\cos\phi = 0,$$

$$\int_{-1}^{r_0} dt \int_0^{2\pi} d\phi\left(\varepsilon_1 \frac{\partial}{\partial r}\psi_1 - \varepsilon_3 \frac{\partial}{\partial r}\psi_3\right)_{r=1} P_k^1(t)\cos\phi$$

$$+\int_{r_0}^{1} dt \int_0^{2\pi} d\phi\left(\varepsilon_2 \frac{\partial}{\partial r}\psi_2 - \varepsilon_4 \frac{\partial}{\partial r}\psi_4\right)_{r=1} P_k^1(t)\cos\phi = 0 \quad . \tag{2.15}$$

Eqs. (2.14) and (2.15) hold for k = 0,1,2,3... Upon substitution of the potentials, eqs. (2.2), (2.6)-(2.8), into eqs. (2.14) and (2.15) one finds that no coupling occurs between contributions containing zeroth order Legendre functions and contributions with first order Legendre functions as a result of the orthogonality of the corresponding spherical harmonics. For the same reason we do not need equations like (2.14) and (2.15) for m>1. Using eqs. (2.12), (2.13) and also the orthogonality relations of the associated Legendre functions on the interval (-1,1),

$$\int_{-1}^{1} dt P_j^m(t) P_k^m(t) = \frac{2(j+m)!}{(2j+1)(j-m)!}\,\delta_{jk} \quad , \tag{2.16}$$

we obtain from eq. (2.14) the following set of equations for the multipole coefficients corresponding with the the normal component of the external field

$$\sum_{j=1}^{\infty} C_{kj}A_{1j} + \sum_{j=1}^{\infty} D_{kj}A_{3j} - 2\psi_3'\delta_{k0} = E_k\cos\Theta_0 \quad ,$$

$$\sum_{j=1}^{\infty} F_{kj}A_{1j} + \sum_{j=1}^{\infty} G_{kj}A_{3j} = H_k\cos\Theta_0 \quad , \quad (k = 0,1,2,3,\ldots) \quad . \tag{2.17}$$

The matrices and vectors are defined by

$$C_{kj} \equiv \frac{4\varepsilon_1\delta_{kj}}{(\varepsilon_1+\varepsilon_2)(2k+1)} - \frac{\varepsilon_1-\varepsilon_2}{\varepsilon_1+\varepsilon_2}\int_{-1}^{r_0} dt P_k^0(t)\{P_j^0(t) - (-1)^j V_j^0(1,t)\} \ ,$$

$$D_{kj} \equiv -\frac{4\varepsilon_3\delta_{kj}}{(\varepsilon_3+\varepsilon_4)(2k+1)} + \frac{\varepsilon_3-\varepsilon_4}{\varepsilon_3+\varepsilon_4}\int_{-1}^{r_0} dt P_k^0(t)\{P_j^0(t) - (-1)^j W_j^0(1,t)\} \ ,$$

$$E_k \equiv -\frac{2\varepsilon_1\delta_{k1}}{3\varepsilon_2} - \left(1 - \frac{\varepsilon_1}{\varepsilon_2}\right) \int_{-1}^{r_0} dt P_k^0(t)(t - r_0) \quad ,$$

$$F_{kj} \equiv -\frac{4\varepsilon_1\varepsilon_2(k+1)\delta_{kj}}{(\varepsilon_1+\varepsilon_2)(2k+1)}$$

$$-\frac{\varepsilon_1(\varepsilon_1-\varepsilon_2)}{\varepsilon_1+\varepsilon_2}\int_{-1}^{r_0} dt P_k^0(t)\left\{(j+1)P_j^0(t) - (-1)^j \frac{\partial V_j^0}{\partial r}(1,t)\right\} \quad ,$$

$$G_{kj} \equiv -\frac{4\varepsilon_3\varepsilon_4 k\delta_{kj}}{(\varepsilon_3+\varepsilon_4)(2k+1)}$$

$$-\frac{\varepsilon_3(\varepsilon_3-\varepsilon_4)}{\varepsilon_3+\varepsilon_4}\int_{-1}^{r_0} dt P_k^0(t)\left\{jP_j^0(t) + (-1)^j \frac{\partial W_j^0}{\partial r}(1,t)\right\} \quad ,$$

$$H_k \equiv -\frac{2\varepsilon_1\delta_{k1}}{3} \quad . \tag{2.18}$$

The first equation of eq. (2.17) for k=0 should be used to determine the unknown quantity ψ_3' in terms of the multipole coefficients. Since we are only interested in the multipole coefficients we can simply discard that equation for k=0. It is also possible to show that for k=0 the second equation of eq. (2.17) is redundant. If free charge were present in the system eq. (2.17) would contain additional terms proportional to the monopole coefficient A_{10} and the free charge density. The equation for k=0 would then relate A_{10} to the total free charge in the system. In the present case, where we do not consider free charges and have set $A_{10} = 0$, the equation vanishes identically. *We shall therefore use eq. (2.17) for $k = 1,2,3,\ldots$ only.*

From eq. (2.15) one finds for the coefficients corresponding to the parallel component of the external field

$$\sum_{j=1}^{\infty} J_{kj}B_{1j} + \sum_{j=1}^{\infty} K_{kj}B_{3j} = L_k \sin\Theta_0 \quad ,$$

$$\sum_{j=1}^{\infty} M_{kj}B_{1j} + \sum_{j=1}^{\infty} N_{kj}B_{3j} = P_k \sin\Theta_0 \quad , \quad (k = 1,2,3,\ldots) \quad , \tag{2.19}$$

with the following definitions

$$J_{kj} \equiv \frac{4\varepsilon_1 k(k+1)\delta_{kj}}{(\varepsilon_1+\varepsilon_2)(2k+1)} - \frac{\varepsilon_1-\varepsilon_2}{\varepsilon_1+\varepsilon_2}\int_{-1}^{r_0} dt P_k^1(t)\left\{P_j^1(t) + (-1)^j V_j^1(1,t)\right\} \quad ,$$

$$K_{kj} \equiv -\frac{4\varepsilon_3 k(k+1)\delta_{kj}}{(\varepsilon_3+\varepsilon_4)(2k+1)} + \frac{\varepsilon_3-\varepsilon_4}{\varepsilon_3+\varepsilon_4}\int_{-1}^{r_0} dt P_k^1(t)\left\{P_j^1(t) + (-1)^j W_j^1(1,t)\right\},$$

$$L_k \equiv -\frac{4\delta_{kl}}{3} \quad ,$$

$$M_{kj} \equiv -\frac{4\varepsilon_1\varepsilon_2 k(k+1)^2\delta_{kj}}{(\varepsilon_1+\varepsilon_2)(2k+1)} - \frac{\varepsilon_1(\varepsilon_1-\varepsilon_2)}{\varepsilon_1+\varepsilon_2}\int_{-1}^{r_0} dtP_k^1(t)\left\{(j+1)P_j^1(t) + (-1)^j \frac{\partial V_j^1}{\partial r}(1,t)\right\} \quad ,$$

$$N_{kj} \equiv -\frac{4\varepsilon_3\varepsilon_4 k^2(k+1)\delta_{kj}}{(\varepsilon_3+\varepsilon_4)(2k+1)} - \frac{\varepsilon_3(\varepsilon_3-\varepsilon_4)}{\varepsilon_3+\varepsilon_4}\int_{-1}^{r_0} dtP_k^1(t)\left\{jP_j^1(t) - (-1)^j \frac{\partial W_j^1}{\partial r}(1,t)\right\} \quad ,$$

$$P_k \equiv -\frac{4\varepsilon_2\delta_{kl}}{3} - (\varepsilon_1-\varepsilon_2)\int_{-1}^{r_0} dtP_k^1(t)P_1^1(t) \quad . \tag{2.20}$$

Clearly, once the integrals in the matrixelements in eqs. (2.18) and (2.20) have been evaluated the dipole coefficients A_{11} and B_{11} can be obtained in principle. The usual technique is to truncate the set of equations at a sufficiently large value, say M, of k and j and to solve the finite set of inhomogeneous linear equations. This yields values for the first M multipole coefficients A_{1j} and B_{1j}, which are found to be a good approximation if M is sufficiently large compared to j. One simply increases M until the value becomes stable within the desired accuracy. For j=1 M=32 is usually sufficient, depending somewhat on the dielectric constants and r_0. In a future paper we shall derive recurrence relations for these integrals, with which the matrix elements can be evaluated analytically. In this paper, in the following three sections, we shall consider some special cases, where these integrals can be calculated more easily.

The polarizability tensor of a rotationally symmetric particle on a flat substrate is given by

$$\overset{\leftrightarrow}{\alpha}{}^e = \begin{pmatrix} \alpha_\parallel^e & 0 & 0 \\ 0 & \alpha_\parallel^e & 0 \\ 0 & 0 & \alpha_\perp^e \end{pmatrix} \quad . \tag{2.21}$$

To obtain the relationship between the parallel and normal component of this tensor and the dipole coefficients we replace the particle in figs. (1a) and (1b) by a point dipole situated in (0,0,D-δ) with δ positive and δ<<R, i.e. *in the ambient just above the substrate*. The dipole

strength is chosen such that for $r>>R$ the field of this dipole is asymptotically equal to the multipole fields, eqs. (2.2) and (2.6). The polarizability is then obtained by dividing this dipole strength by the external field *in the ambient*. For the case that the centre of the truncated sphere is in the ambient, $D>0$ (fig. 1a), we thus obtain

$$\alpha_{\parallel}^{e} = -4\pi\varepsilon_1 R^3 (\sin\Theta_0)^{-1} B_{11} \quad ,$$
$$\alpha_{\perp}^{e} = -4\pi\varepsilon_1 R^3 (\cos\Theta_0)^{-1} A_{11} \quad , \tag{2.22}$$

and if the centre is in the substrate, $D<0$ (fig. 1b), we find

$$\alpha_{\parallel}^{e} = -4\pi\varepsilon_2 R^3 (\sin\Theta_0)^{-1} B_{11} \quad ,$$
$$\alpha_{\perp}^{e} = -4\pi(\varepsilon_1^{\,2}/\varepsilon_2) R^3 (\cos\Theta_0)^{-1} A_{11} \quad . \tag{2.23}$$

Using the fact that

$$\varepsilon_1 \lim_{D\downarrow 0} B_{11}(D) = \varepsilon_2 \lim_{D\uparrow 0} B_{11}(D) \quad , \tag{2.24}$$

one finds that the polarizability $\alpha_{\parallel}^{e}$ with respect to the electric field parallel to the surface of the substrate is a continuous function of D in D=0. For the polarizability normal to the surface of the substrate one has

$$\varepsilon_1^{-1} \lim_{D\downarrow 0} A_{11}(D) = \varepsilon_2^{-1} \lim_{D\uparrow 0} A_{11}(D) \quad , \tag{2.25}$$

and as a consequence $\alpha_{\perp}^{e}$ is also a continuous function of D in D=0. One can also place the dipole *in the substrate* by choosing δ negative rather than positive. To obtain the polarizability one then has to divide the dipole strength by the electric field *in the substrate*. Using the fact that E_x and E_y are continuous on the surface of the substrate one may show that $\alpha_{\parallel}^{e}$ is a continuous function of δ for $\delta=0$. In the direction normal to the substrate D_z rather than E_z is continuous and as a consequence one may show that

$$\varepsilon_1^{-2} \lim_{\delta\downarrow 0} \alpha_{\perp}^{e} = \varepsilon_2^{-2} \lim_{\delta\uparrow 0} \alpha_{\perp}^{e} \quad . \tag{2.26}$$

In fact it is more appropriate to define the polarizability in the normal direction with respect to the displacement field D_z, see refs. 1 and 8. If one defines the polarizabilities with respect to

$$\vec{N} \equiv (E_x , E_y , D_z) , \tag{2.27}$$

then for $\delta>0$ the polarizability tensor differs from that given by eq. (2.21) by a factor $(\varepsilon_1)^{-2}$ in the normal component, so that

$$\vec{\vec{\alpha}}^n = \begin{pmatrix} \alpha_\parallel^e & 0 & 0 \\ 0 & \alpha_\parallel^e & 0 \\ 0 & 0 & \alpha_\perp^e \varepsilon_1^{-2} \end{pmatrix} = \begin{pmatrix} \alpha_\parallel^n & 0 & 0 \\ 0 & \alpha_\parallel^n & 0 \\ 0 & 0 & \alpha_\perp^n \end{pmatrix} . \tag{2.28}$$

see appendix B of reference 1. Similarly one finds for $\delta<0$

$$\vec{\vec{\alpha}}^n = \begin{pmatrix} \alpha_\parallel^e & 0 & 0 \\ 0 & \alpha_\parallel^e & 0 \\ 0 & 0 & \alpha_\perp^e \varepsilon_2^{-2} \end{pmatrix} = \begin{pmatrix} \alpha_\parallel^n & 0 & 0 \\ 0 & \alpha_\parallel^n & 0 \\ 0 & 0 & \alpha_\perp^n \end{pmatrix} . \tag{2.29}$$

$\vec{\vec{\alpha}}^n$ and in particular $\alpha_\perp^n$ is a continuous function of δ in $\delta=0$. To obtain the polarizabilities of hemispherical bumps and pits on a substrate Berreman[4)] uses point dipoles located precisely at the interface between ambient and substrate ($\delta=0$). This choice has the disadvantage that the distinction between direct and image dipoles is no longer clear. As a result Berreman does not take the influence of the image dipoles into account correctly. His expression for p_x , eq. (56), must be divided by $2\varepsilon_1/(\varepsilon_1+\varepsilon_2)$ and the expression for p_z , eq. (58), must be divided by $2\varepsilon_2/(\varepsilon_1+\varepsilon_2)$.

So far we have considered only one particle. For a low density system of identical particles one finds that the surface susceptibilities with respect to the $\vec{N}$ field are equal to the polarizabilities per unit surface area with respect to this field. For $\delta>0$ one thus finds

$$\gamma = \rho\alpha_\parallel^n = \rho\alpha_\parallel^e ,$$

$$\beta = \rho\alpha_\perp^n = \rho\alpha_\perp^e \varepsilon_1^{-2} \quad , \tag{2.30}$$

where ρ is the number of particles per unit of surface area. For $\delta<0$ one finds a similar expression. For higher particle densities local field effects have to be taken into account [1]. Such a system can be described macroscopically by the following electrical surface susceptibilities:

$$\gamma = \frac{\rho\alpha_\parallel^n}{1 - \frac{1}{3}\kappa_\parallel \rho\alpha_\parallel^n} \quad ,$$

$$\beta = \frac{\rho\alpha_\perp^n}{1 - \frac{1}{3}\kappa_\perp \rho\alpha_\perp^n} \quad . \tag{2.31}$$

The parameters $\kappa_\parallel$ and $\kappa_\perp$, with the dimensionality of an inverse length, are related to some average distance between the particles. Explicit expressions for $\kappa_\parallel$ and $\kappa_\perp$ in terms of the autocorrelation function of the surface densities of the particles were derived in reference 2. One may identify $(\varepsilon_1\kappa_\parallel)^{-1}$ with the often used optical thickness of the film, see ref. 1.

The amplitudes for the reflected and transmitted s- and p-polarized electromagnetic fields are given in terms of γ and β by

$$r_s = \{n_1\cos\Theta - n_2\cos\Theta_t + i(\omega/c)\gamma\}/\{n_1\cos\Theta + n_2\cos\Theta_t - i(\omega/c)\gamma\} \quad , \tag{2.32}$$

$$t_s = 2n_1\cos\Theta/\{n_1\cos\Theta + n_2\cos\Theta_t - i(\omega/c)\gamma\} \quad , \tag{2.33}$$

$$\begin{aligned} r_p = \{&(n_2\cos\Theta - n_1\cos\Theta_t)(1 - (\omega/2c)^2\varepsilon_1\gamma\beta\sin^2\Theta) - i(\omega/c)\gamma\cos\Theta\cos\Theta_t \\ &+ i(\omega/c)n_1n_2\varepsilon_1\beta\sin^2\Theta\}/\{(n_2\cos\Theta + n_1\cos\Theta_t)(1 - (\omega/2c)^2\varepsilon_1\gamma\beta\sin^2\Theta) \\ &- i(\omega/c)\gamma\cos\Theta\cos\Theta_t - i(\omega/c)n_1n_2\varepsilon_1\beta\sin^2\Theta\} \quad , \end{aligned} \tag{2.34}$$

$$\begin{aligned} t_p = &2n_1\cos\Theta(1 + (\omega/2c)^2\varepsilon_1\gamma\beta\sin^2\Theta)/\{(n_2\cos\Theta + n_1\cos\Theta_t) \\ &\times(1 - (\omega/2c)^2\varepsilon_1\gamma\beta\sin^2\Theta) - i(\omega/c)\gamma\cos\Theta\cos\Theta_t \end{aligned}$$

$$- i(\omega/c)n_1 n_2 \varepsilon_1 \beta \sin^2\Theta\} \quad , \tag{2.35}$$

where n_1 and n_2 are the refractive indices of ambient and substrate respectively. Θ is the angle of incidence and Θ_t the angle of transmittance. These equations were first derived in reference 9 and can be obtained from eqs. (3.11) and (3.12) of that reference by putting $\gamma^- = \gamma^+ = \gamma$ and $\beta^- = \beta^+ = \beta$. (See also ref. 1).

In ref. 4 Berreman incorrectly used the free dipole propagator to calculate the far field, which resulted in the omission of certain Fresnel factors in the amplitudes. In eq. (48) of ref. 4 the second term on the r.h.s. must be multiplied by a factor $-2n_1\cos\Theta/(n_1\cos\Theta + n_2\cos\Theta_t)$, the second term in eq. (59) must be multiplied by $-2n_1\cos\Theta_t/(n_2\cos\Theta + n_1\cos\Theta_t)$ and the last term of the same equation must be multiplied by $2n_2\cos\Theta/(n_2\cos\Theta + n_1\cos\Theta_t)$. The above mentioned corrections for the polarizabilities must also be incorporated in these equations.

3. SPHERE ON (OR ABOVE) A SUBSTRATE

In this section we shall consider the special case of a sphere on a substrate. This corresponds to setting r_0 equal to 1. The following identity

$$V_j^m(r,t) = (-1)^{j+m} \sum_{\ell=m}^{\infty} \frac{(\ell + j)!\, r^\ell P_\ell^m(t)}{(\ell + m)!\,(j - m)!\,(2r_0)^{\ell+j+1}} \quad , \tag{3.1}$$

which is valid for $r < 2r_0$, is very useful for evaluating the integrals on the right hand side of eqs. (2.18) and (2.20). We also know that for the special case of a sphere region 4 disappears, so that the multipole coefficients are independent of the value of ε_4. We can use this property to make the convenient choice $\varepsilon_4 = \varepsilon_3$. Using also eq. (2.16) we obtain for eq. (2.18):

$$C_{kj} = \frac{2}{2k+1}\left\{\delta_{kj} + \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\,\frac{(k+j)!}{k!\,j!\,2^{k+j+1}}\right\} \quad ,$$

$$D_{kj} = -\frac{2\delta_{kj}}{2k+1} \quad ,$$

$$E_k = -\frac{2\delta_{k1}}{3} \quad ,$$

$$F_{kj} = \frac{2\varepsilon_1}{2k+1}\left\{-(j+1)\delta_{kj} + \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\frac{k(k+j)!}{k!j!2^{j+k+1}}\right\} \quad ,$$

$$G_{kj} = -\frac{2\varepsilon_3 k\delta_{kj}}{2k+1} \quad ,$$

$$H_k = -\frac{2\varepsilon_1\delta_{k1}}{3} \quad . \tag{3.2}$$

Similarly we obtain for eq. (2.20):

$$J_{kj} = \frac{2k(k+1)}{(2k+1)}\left\{\delta_{kj} + \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\frac{(k+j)!}{(k+1)!(j-1)!2^{j+k+1}}\right\} \quad ,$$

$$K_{kj} = -\frac{2k(k+1)\delta_{kj}}{(2k+1)} \quad ,$$

$$L_k = -\frac{4\delta_{k1}}{3} \quad ,$$

$$M_{kj} = \frac{2\varepsilon_1 k(k+1)}{(2k+1)}\left\{-(j+1)\delta_{kj} + \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\frac{k(k+j)!}{(k+1)!(j-1)!2^{j+k+1}}\right\} \quad ,$$

$$N_{kj} = -\frac{2\varepsilon_3 k^2(k+1)\delta_{kj}}{2k+1} \quad ,$$

$$P_k = -\frac{4\varepsilon_1\delta_{k1}}{3} \quad . \tag{3.3}$$

One can now eliminate the coefficients A_{3j} and B_{3j}, describing the potential inside the sphere, by multiplying the first equation of eqs. (2.17) and (2.19) by $\varepsilon_3 k$ and substracting from this the second equation. After rearranging terms we obtain

$$\sum_{j=1}^{\infty}\left\{\delta_{jk} + \frac{(\varepsilon_2 - \varepsilon_1)k(\varepsilon_1 - \varepsilon_3)}{(\varepsilon_2 + \varepsilon_1)\left((k+1)\varepsilon_1 + k\varepsilon_3\right)}\frac{(k+j)!}{k!j!2^{k+j+1}}\right\}A_{1j} = \frac{\varepsilon_1 - \varepsilon_3}{2\varepsilon_1 + \varepsilon_3}\cos\Theta_0\delta_{k1} \quad , \tag{3.4}$$

$$\sum_{j=1}^{\infty}\left\{\delta_{kj} + \frac{(\varepsilon_2 - \varepsilon_1)k(\varepsilon_1 - \varepsilon_3)}{(\varepsilon_2 + \varepsilon_1)\left((k+1)\varepsilon_1 + k\varepsilon_3\right)}\frac{(k+j)!}{(k+1)!(j-1)!2^{j+k+1}}\right\}B_{1j} = \frac{\varepsilon_1 - \varepsilon_3}{2\varepsilon_1 + \varepsilon_3}\sin\Theta_0\delta_{k1} \quad . \tag{3.5}$$

In a recent paper on the light scattering by a sphere on a substrate[10] these equations have also been obtained as the static limit of the dynamic solution.

Since the quantity $2r_0$ in eq. (3.1) is just the distance between the origin of the sphere and its image, one can easily generalize eqs. (3.4) and (3.5) to the case of a sphere of radius R a distance D above the substrate. One then obtains

$$\sum_{j=1}^{\infty} \left\{\delta_{kj} + \frac{(\varepsilon_2 - \varepsilon_1)k(\varepsilon_1 - \varepsilon_3)}{(\varepsilon_2 + \varepsilon_1)[(k + 1)\varepsilon_1 + k\varepsilon_3]} \frac{(k + j)!}{k!j!(2r_0)^{k+j+1}}\right\} A_{1j} = \frac{\varepsilon_1 - \varepsilon_3}{2\varepsilon_1 + \varepsilon_3} \cos\Theta_0 \delta_{k1} , \tag{3.6}$$

$$\sum_{j=1}^{\infty} \left\{\delta_{kj} + \frac{(\varepsilon_2 - \varepsilon_1)k(\varepsilon_1 - \varepsilon_3)}{(\varepsilon_2 + \varepsilon_1)[(k + 1)\varepsilon_1 + k\varepsilon_3]} \frac{(k + j)!}{(k + 1)!(j - 1)!(2r_0)^{k+j+1}}\right\} B_{1j} = \frac{\varepsilon_1 - \varepsilon_3}{2\varepsilon_1 + \varepsilon_3} \sin\Theta_0 \delta_{k1} , \tag{3.7}$$

where r_0 is given by eq. (2.4) as usual, but now $r_0 > 1$. These equations are very similar to those for a system of two identical spheres in a homogeneous external field. For the multipole coefficients induced by the component of the field parallel to the line connecting the centres of the spheres one merely has to put $(\varepsilon_2-\varepsilon_1)/(\varepsilon_2+\varepsilon_1) = 1$ in eq. (3.6), and for the coefficients corresponding to the normal component one has to put $(\varepsilon_2-\varepsilon_1)/(\varepsilon_2+\varepsilon_1) = -1$ in eq. (3.7).

In the following we shall restrict ourself to a sphere touching the substrate, eqs. (3.4) and (3.5). If we truncate the sets of equations at M=1, we obtain the dipole coefficients A_{11} and B_{11} in dipole approximation

$$A_{11} = \frac{\dfrac{(\varepsilon_1 - \varepsilon_3)}{(2\varepsilon_1 + \varepsilon_3)} \cos\Theta_0}{1 + \dfrac{1}{4} \dfrac{(\varepsilon_2 - \varepsilon_1)(\varepsilon_1 - \varepsilon_3)}{(\varepsilon_2 + \varepsilon_1)(2\varepsilon_1 + \varepsilon_3)}} , \tag{3.8}$$

$$B_{11} = \frac{\dfrac{(\varepsilon_1 - \varepsilon_3)}{(2\varepsilon_1 + \varepsilon_3)} \sin\Theta_0}{1 + \dfrac{1}{8} \dfrac{(\varepsilon_2 - \varepsilon_1)(\varepsilon_1 - \varepsilon_3)}{(\varepsilon_2 + \varepsilon_1)(2\varepsilon_1 + \varepsilon_3)}} . \tag{3.9}$$

Thus, one finds for the polarizabilities in dipole approximation, using eq. (2.22),

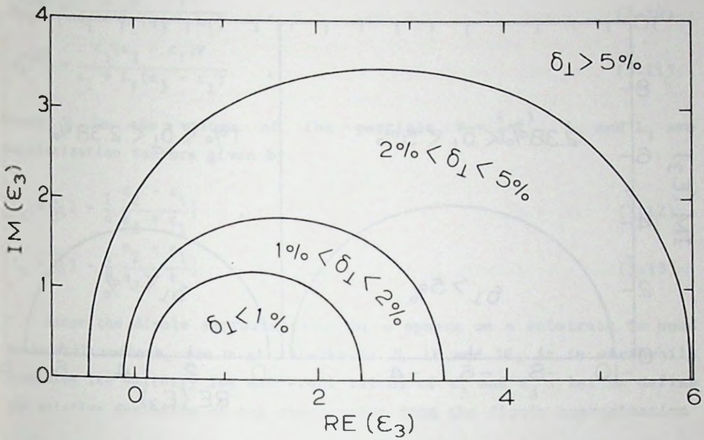


Fig. 3 Contours of constant $\delta_\perp$ as a function of ε_3 for a perfectly conducting substrate, defining the four regions $\delta_\perp$<1%, 1%<$\delta_\perp$<2%, 2%<$\delta_\perp$<5% and $\delta_\perp$>5%.

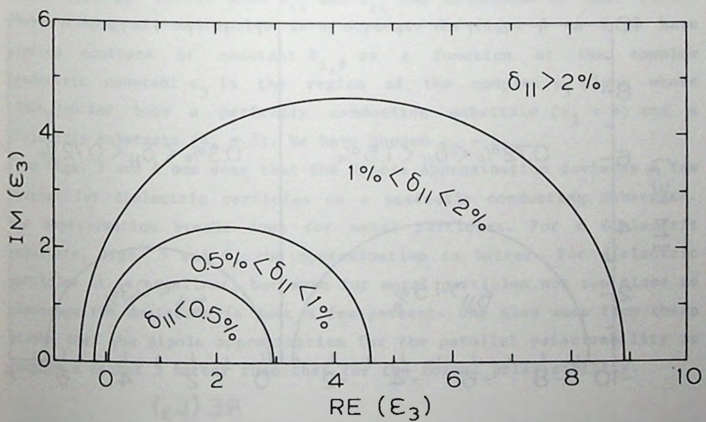


Fig. 4 Contours of constant $\delta_\parallel$ as a function of ε_3 for a perfectly conducting substrate, defining the four regions $\delta_\parallel$<0.5%, 0.5%<$\delta_\parallel$<1%, 1%<$\delta_\parallel$<2% and $\delta_\parallel$>2%.

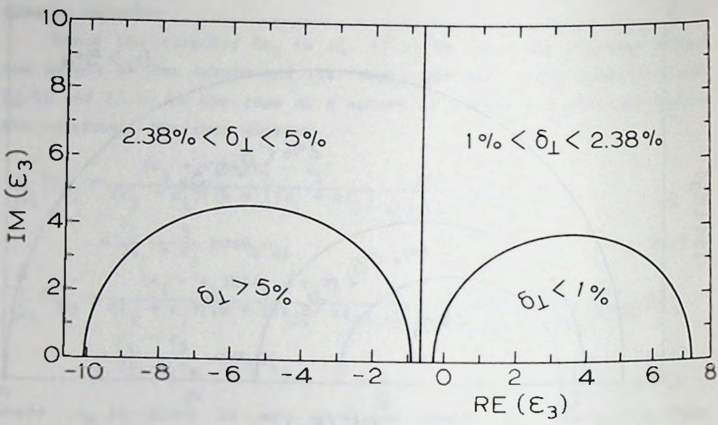


Fig. 5 Contours of constant $\delta_\perp$ as a function of ε_3 for a dielectric substrate ($\varepsilon_2 = 3$), defining the four regions $\delta_\perp<1\%$, $1\%<\delta_\perp<2.38\%$, $2.38\%<\delta_\perp<5\%$ and $\delta_\perp>5\%$.

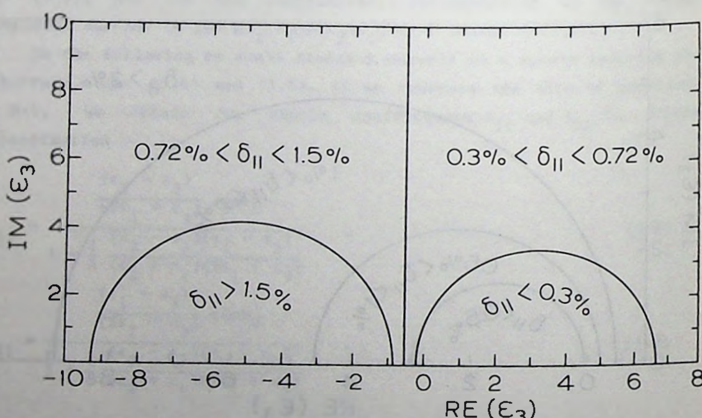


Fig. 6 Contours of constant $\delta_{\parallel}$ as a function of ε_3 for a dielectric substrate ($\varepsilon_2 = 3$), defining the four regions $\delta_{\parallel}<0.3\%$, $0.3\%<\delta_{\parallel}<0.72\%$, $0.72\%<\delta_{\parallel}<1.5\%$ and $\delta_{\parallel}>1.5\%$.

$$\alpha_{\perp}^{e(d)} = \frac{\varepsilon_1(\varepsilon_3 - \varepsilon_1)V}{\varepsilon_1 + L_{\perp}(\varepsilon_3 - \varepsilon_1)} , \tag{3.10}$$

$$\alpha_{\parallel}^{e(d)} = \frac{\varepsilon_1(\varepsilon_3 - \varepsilon_1)V}{\varepsilon_1 + L_{\parallel}(\varepsilon_3 - \varepsilon_1)} , \tag{3.11}$$

where V is the volume of the particle, $V = \frac{4}{3}\pi R^3$. $L_{\perp}$ and $L_{\parallel}$ are depolarization factors given by

$$L_{\perp} = \frac{1}{3}\left[1 - \frac{1}{4}\frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + \varepsilon_1}\right] , \tag{3.12}$$

$$L_{\parallel} = \frac{1}{3}\left[1 - \frac{1}{8}\frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + \varepsilon_1}\right] . \tag{3.13}$$

Since the dipole approximation for a sphere on a substrate is used by several authors, see e.g. references 2, 11 and 12, it is worthwhile examining its validity for different values of ε_2 and ε_3. Let us define the relative deviation of the exact value from the dipole approximation

$$\delta_{\perp,\parallel} \equiv |(\alpha^{e}_{\perp,\parallel} - \alpha^{e(d)}_{\perp,\parallel})/\alpha^{e}_{\perp,\parallel}| , \tag{3.14}$$

where $\alpha_{\perp}^{e(d)}$ and $\alpha_{\parallel}^{e(d)}$ are given by eqs. (3.10) and (3.11), and $\alpha^{e}_{\perp,\parallel}$ follow from eq. (2.22) with A_{11} and B_{11} the solutions of eqs. (3.4), (3.5), taking all multipoles into account. In figs. 3 to 6 we have plotted contours of constant $\delta_{\perp,\parallel}$ as a function of the complex dielectric constant ε_3 in the region of the complex ε_3 plane where $\mathrm{Im}(\varepsilon_3)>0$ for both a perfectly conducting substrate ($\varepsilon_2 \to \infty$) and a dielectric substrate ($\varepsilon_2 = 3$). We have chosen $\varepsilon_1 = 1$.
From figs. 3 and 4 one sees that the dipole approximation deviates a few percent for dielectric particles on a perfectly conducting substrate. The approximation breaks down for metal particles. For a dielectric substrate, figs. 5 and 6, the approximation is better. For dielectric particles it is excellent, but even for metal particles not too close to resonance the deviation is just a few percent. One also sees from these graphs that the dipole approximation for the parallel polarizability is roughly a factor 3 better than that for the normal polarizability.

4. HEMISPHERE ON SUBSTRATE

In this section we shall consider the case of a hemispherical particle on a substrate. This case can be obtained by putting $r_0 = 0$ and $\varepsilon_4 = \varepsilon_2$ in the general formulae eqs. (2.17) - (2.20). Setting first $r_0 = 0$ in eqs. (2.3) and (2.9) one finds that the functions $V_j^m(r,\cos\Theta)$ and $W_j^m(r,\cos\Theta)$ reduce to $r^{-j-1}P_j^m(\cos\Theta)$ and $r^jP_j^m(\cos\Theta)$, respectively. We will follow Berreman's procedure [4] and introduce the matrix

$$R_{kj} \equiv \int_{-1}^{0} dt P_k^0(t) P_j^0(t) \quad , \tag{4.1}$$

so that the coefficients in eq. (2.18) can be written as

$$\begin{aligned}
C_{kj} &= \frac{4\varepsilon_1\delta_{kj}}{(\varepsilon_1 + \varepsilon_2)(2k+1)} - \left(1 - (-1)^j\right) \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2} R_{kj} \quad , \\
D_{kj} &= -\frac{4\varepsilon_3\delta_{kj}}{(\varepsilon_3 + \varepsilon_2)(2k+1)} + \left(1 - (-1)^j\right) \frac{\varepsilon_3 - \varepsilon_2}{\varepsilon_3 + \varepsilon_2} R_{kj} \quad , \\
E_k &= -\frac{2\varepsilon_1\delta_{k1}}{3\varepsilon_2} - \left(1 - \frac{\varepsilon_1}{\varepsilon_2}\right) R_{k1} \quad , \\
F_{kj} &= -\frac{4\varepsilon_1\varepsilon_2(k+1)\delta_{kj}}{(\varepsilon_1 + \varepsilon_2)(2k+1)} - \left(1 + (-1)^j\right) \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \varepsilon_1 (j+1) R_{kj} \quad , \\
G_{kj} &= -\frac{4\varepsilon_3\varepsilon_2 k\delta_{kj}}{(\varepsilon_3 + \varepsilon_2)(2k+1)} - \left(1 + (-1)^j\right) \frac{\varepsilon_3 - \varepsilon_2}{\varepsilon_3 + \varepsilon_2} \varepsilon_3 j R_{kj} \quad , \\
H_k &= -\frac{2\varepsilon_1\delta_{k1}}{3} \quad .
\end{aligned} \tag{4.2}$$

Using the properties of the functions R_{kj} one can eliminate the multipole coefficients A_{3j} and obtain the following equations

$$\begin{aligned}
&\frac{(k+1)\varepsilon_1(\varepsilon_2 + \varepsilon_3) + k\varepsilon_3(\varepsilon_1 + \varepsilon_2)}{(2k+1)(\varepsilon_1 - \varepsilon_3)} A_{1k} + \varepsilon_1 \sum_{j=2}^{\infty}{}^{(E)} (j+1) R_{kj} A_{1j} \\
&\quad = \frac{1}{6}(\varepsilon_1 + \varepsilon_2)\delta_{k1}\cos\Theta_0 \quad , \qquad k = \text{odd} \quad , \\
&\frac{(k+1)(\varepsilon_1 + \varepsilon_2) + k(\varepsilon_2 + \varepsilon_3)}{k(2k+1)(\varepsilon_1 - \varepsilon_3)} \varepsilon_1 A_{1k} - \varepsilon_2 \sum_{j=1}^{\infty}{}^{(O)} R_{kj} A_{1j} \\
&\quad = \frac{1}{2}(\varepsilon_1 + \varepsilon_2) R_{k1}\cos\Theta_0 \quad , \qquad k = \text{even} \quad .
\end{aligned} \tag{4.3}$$

Here O refers to odd values of j and k and E to even values. The matrix elements of R_{kj} in eq. (4.3) are most easily derived from the relation[13]

$$R_{kj} = \frac{P_k^0(0)\,\frac{dP_j^0}{dt}(0) - P_j^0(0)\,\frac{dP_k^0}{dt}(0)}{k(k+1) - j(j+1)} \quad , \quad k \neq j \quad , \tag{4.4}$$

and are found to be

$$R_{kj} = \frac{(-1)^{\frac{1}{2}(k+j-1)}\,k!\;j!}{2^{k+j-1}(k-j)(k+j+1)\left\{\left(\frac{k}{2}\right)!\;\left(\frac{j-1}{2}\right)!\right\}^2} \;,\; k = \text{even},\; j = \text{odd}. \tag{4.5}$$

Similarly we introduce the matrix

$$U_{kj} \equiv \int_{-1}^{0} dt P_k^1(t) P_j^1(t) \quad , \tag{4.6}$$

and we find for eq. (2.20):

$$J_{kj} = \frac{4\varepsilon_1 k(k+1)\delta_{kj}}{(\varepsilon_1+\varepsilon_2)(2k+1)} - \left(1 + (-1)^j\right)\frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2} U_{kj} \quad ,$$

$$K_{kj} = -\frac{4\varepsilon_3 k(k+1)\delta_{kj}}{(\varepsilon_3+\varepsilon_2)(2k+1)} + \left(1 + (-1)^j\right)\frac{\varepsilon_3 - \varepsilon_2}{\varepsilon_3 + \varepsilon_2} U_{kj} \quad ,$$

$$L_k = -\frac{4\delta_{k1}}{3} \quad ,$$

$$M_{kj} = -\frac{4\varepsilon_1\varepsilon_2 k(k+1)^2\delta_{kj}}{(\varepsilon_1+\varepsilon_2)(2k+1)} - \left(1 - (-1)^j\right)\frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\varepsilon_1(j+1)U_{kj} \quad ,$$

$$N_{kj} = -\frac{4\varepsilon_3\varepsilon_2 k^2(k+1)\delta_{kj}}{(\varepsilon_3+\varepsilon_2)(2k+1)} - \left(1 - (-1)^j\right)\frac{\varepsilon_3 - \varepsilon_2}{\varepsilon_3 + \varepsilon_2}\varepsilon_3 j U_{kj} \quad ,$$

$$P_k = -\frac{4\varepsilon_2\delta_{k1}}{3} - (\varepsilon_1 - \varepsilon_2)U_{k1} \quad . \tag{4.7}$$

After eliminating the coefficients B_{3j} we obtain the following set of equations for the coefficients B_{1j}

$$\frac{(k+1)(\varepsilon_1+\varepsilon_2) + k(\varepsilon_2+\varepsilon_3)}{(2k+1)(\varepsilon_1-\varepsilon_3)}(k+1)\varepsilon_1 B_{1k} - \varepsilon_2 \sum_{j=2}^{\infty}{}^{(E)} U_{kj}B_{1j}$$

$$= \frac{1}{3}(\varepsilon_1+\varepsilon_2)\delta_{k1}\sin\Theta_0 \quad , \quad k = \text{odd} \quad ,$$

$$\frac{(k+1)\varepsilon_1(\varepsilon_2+\varepsilon_3) + k\varepsilon_3(\varepsilon_1+\varepsilon_2)}{(2k+1)(\varepsilon_1-\varepsilon_3)}k(k+1)B_{1k} + \varepsilon_1 \sum_{j=1}^{\infty}{}^{(O)}(j+1)U_{kj}B_{1j}$$

$$= \frac{1}{2}(\varepsilon_1 + \varepsilon_2)U_{k1}\sin\Theta_0 \quad , \qquad k = \text{even} \quad . \tag{4.8}$$

where the elements of U_{kj} are given by

$$U_{kj} = \frac{(-1)^{\frac{1}{2}(k+j-1)}(k+1)!\; j!}{2^{k+j-3}k(k-j)(k+j+1)\{(\frac{k-2}{2})!\;(\frac{j-1}{2})!\}^2} \;,\; k = \text{even},\; j = \text{odd} \;, \tag{4.9}$$

which result can be obtained from the relation[13]

$$U_{kj} = \frac{P_k^1(0)\,\frac{dP_j^1}{dt}(0) - P_j^1(0)\,\frac{dP_k^1}{dt}(0)}{k(k+1) - j(j+1)} \quad , \qquad k \neq j \quad . \tag{4.10}$$

For a discussion of the properties of the solutions of eqs. (4.3) and (4.8) we refer to Chauvaux and Meessen [6] and Berreman [4].

5. THIN SPHERICAL CAP ON SUBSTRATE

The third application we shall consider in this article is that of a very thin cap on a substrate. This geometry can be obtained from the general case of section 2 by setting $\varepsilon_3 = \varepsilon_1$ and taking $1 - r_0 \ll 1$. Since the dielectric constants of ambient, substrate and particle are now equal to ε_2, ε_1 and ε_4, respectively, it is convenient to make the substitutions $\varepsilon_2 \to \varepsilon_1$, $\varepsilon_1 \to \varepsilon_2$ and $\varepsilon_4 \to \varepsilon_3$, so that the dielectric constant of the ambient is, as usual, ε_1, etc. It is useful to introduce the (reduced) height of the particle

$$h \equiv 1 - r_0 \quad , \tag{5.1}$$

where $h \ll 1$, so that one can expand the multipole coefficients in this parameter. To transform the integrals in eqs. (2.18) and (2.20), we use eq. (3.1) and also the identities

$$W_j^m(r,t) = \sum_{\ell=m}^{j} \frac{(-1)^{j+\ell}(j+m)!(2r_0)^{j-\ell} r^\ell P_\ell^m(t)}{(\ell+m)!(j-\ell)!} \quad , \qquad (m = 0,1) \tag{5.2}$$

Together with the orthogonality relations eq. (2.16) we can now write eq. (2.18) in the form

$$C_{kj} = \frac{2\delta_{kj}}{2k+1} + \frac{2(\varepsilon_2 - \varepsilon_1)(j+k)!}{(\varepsilon_1 + \varepsilon_2)(2k+1)j!k!(2r_0)^{k+j+1}}$$

$$+ \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_1 + \varepsilon_2} \int_{1-h}^{1} dt P_k^0(t)\{P_j^0(t) - (-1)^j V_j^0(1,t)\} \quad ,$$

$$D_{kj} = - \frac{2\delta_{kj}}{2k+1} - \frac{2(\varepsilon_2 - \varepsilon_3)(-1)^k j!(2r_0)^{j-k}}{(\varepsilon_2 + \varepsilon_3)(2k+1)k!(j-k)!}$$

$$- \frac{\varepsilon_2 - \varepsilon_3}{\varepsilon_2 + \varepsilon_3} \int_{1-h}^{1} dt P_k^0(t)\{P_j^0(t) - (-1)^j W_j^0(t)\} \quad ,$$

$$E_k = - \frac{2\delta_{k1}}{3} + (1 - \frac{\varepsilon_2}{\varepsilon_1}) \int_{1-h}^{1} dt P_k^0(t)(t - 1 + h) \quad ,$$

$$F_{kj} = - \frac{2(k+1)\delta_{kj}}{2k+1} + \frac{2(\varepsilon_2 - \varepsilon_1)k(j+k)!}{(\varepsilon_1 + \varepsilon_2)(2k+1)j!k!(2r_0)^{k+j+1}}$$

$$+ \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_1 + \varepsilon_2} \int_{1-h}^{1} dt P_k^0(t)\{(j+1)P_j^0(t) - (-1)^j \frac{\partial V_j^0}{\partial r}(1,t)\} \quad ,$$

$$G_{kj} = - \frac{2k\delta_{kj}}{2k+1} - \frac{2(\varepsilon_2 - \varepsilon_3)(-1)^k k j!(2r_0)^{j-k}}{(\varepsilon_2 + \varepsilon_3)(2k+1)k!(j-k)!}$$

$$+ \frac{\varepsilon_2 - \varepsilon_3}{\varepsilon_2 + \varepsilon_3} \int_{1-h}^{1} dt P_k^0(t)\{jP_j^0(t) + (-1)^j \frac{\partial W_j^0}{\partial r}(1,t)\} \quad .$$

$$H_k = - \frac{2\delta_{k1}}{3} \quad , \tag{5.3}$$

where we have divided the second equation of eq. (2.17) by ε_2. We have used the convention that $1/(j-k)! = 0$ for $k > j$. Similarly we find for eq. (2.20)

$$J_{kj} = \frac{2k(k+1)\delta_{kj}}{2k+1} + \frac{2(\varepsilon_2 - \varepsilon_1)k(k+1)(j+k)!}{(\varepsilon_1 + \varepsilon_2)(2k+1)(k+1)!(j-1)!(2r_0)^{k+j+1}}$$

$$+ \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_1 + \varepsilon_2} \int_{1-h}^{1} dt P_k^1(t)\{P_j^1(t) + (-1)^j V_j^1(1,t)\} \quad ,$$

$$K_{kj} = -\frac{2k(k+1)\delta_{kj}}{2k+1} + \frac{2(\varepsilon_2-\varepsilon_3)(-1)^k k(k+1)(j+1)!(2r_0)^{j-k}}{(\varepsilon_2+\varepsilon_3)(2k+1)(k+1)!(j-k)!}$$

$$-\frac{\varepsilon_2-\varepsilon_3}{\varepsilon_2+\varepsilon_3}\int_{1-h}^{1} dtP_k^1(t)\{P_j^1(t) + (-1)^j W_j^1(1,t)\} \quad ,$$

$$L_k = -\frac{4\delta_{k1}}{3} \quad ,$$

$$M_{kj} = -\frac{2k(k+1)^2\delta_{kj}}{2k+1} + \frac{2(\varepsilon_2-\varepsilon_1)k^2(k+1)(j+k)!}{(\varepsilon_1+\varepsilon_2)(2k+1)(k+1)!(j-1)!(2r_0)^{k+j+1}}$$

$$+\frac{\varepsilon_2-\varepsilon_1}{\varepsilon_1+\varepsilon_2}\int_{1-h}^{1} dtP_k^1(t)\{(j+1)P_j^1(t) + (-1)^j \frac{\partial V_j^1}{\partial r}(1,t)\} \quad ,$$

$$N_{kj} = \frac{2k^2(k+1)\delta_{kj}}{2k+1} + \frac{2(\varepsilon_2-\varepsilon_3)(-1)^k k^2(k+1)(j+1)!(2r_0)^{j-k}}{(\varepsilon_2+\varepsilon_3)(2k+1)(k+1)!(j-1)!}$$

$$+\frac{\varepsilon_2-\varepsilon_3}{\varepsilon_2+\varepsilon_3}\int_{1-h}^{1} dtP_k^1(t)\{jP_j^1(t) - (-1)^j \frac{\partial W_j^1}{\partial r}(1,t)\} \quad ,$$

$$P_k = -\frac{4\delta_{k1}}{3} + (1-\frac{\varepsilon_1}{\varepsilon_2})\int_{1-h}^{1} dtP_k^1(t)P_1^1(t) \quad . \tag{5.4}$$

Again, the second equation of eq. (2.19) has been divided by ε_2.

In the limit $h \to 0$ the particle disappears and the only non-zero multipole coefficients are A_{31} and B_{31}, corresponding to the external field:

$$A_{31} = \frac{\varepsilon_2+\varepsilon_3}{2\varepsilon_3}\cos\Theta_0 \quad , \tag{5.5}$$

$$B_{31} = \frac{\varepsilon_2+\varepsilon_3}{2\varepsilon_2}\sin\Theta_0 \quad . \tag{5.6}$$

Therefore it is convenient to introduce new coefficients, defined by

$$A'_{3j} \equiv A_{3j} - \frac{\varepsilon_2+\varepsilon_3}{2\varepsilon_3}\delta_{j1}\cos\Theta_0 \quad , \tag{5.7}$$

$$B'_{3j} \equiv B_{3j} - \frac{\varepsilon_2 + \varepsilon_3}{2\varepsilon_2}\,\delta_{j1}\sin\Theta_0 \quad . \tag{5.8}$$

In terms of these coefficients eqs. (2.17) and (2.19) can be written as

$$\sum_{j=1}^{\infty} C_{kj}A_{1j} + \sum_{j=1}^{\infty} D_{kj}A'_{3j} = E'_k\cos\Theta_0 \quad ,$$
$$\sum_{j=1}^{\infty} F_{kj}A_{1j} + \sum_{j=1}^{\infty} G_{kj}A'_{3j} = H'_k\cos\Theta_0 \quad , \quad (k = 1,2,3,....) \quad , \tag{5.9}$$

and

$$\sum_{j=1}^{\infty} J_{kj}B_{1j} + \sum_{j=1}^{\infty} K_{kj}B'_{3j} = L'_k\sin\Theta_0 \quad ,$$
$$\sum_{j=1}^{\infty} M_{kj}B_{1j} + \sum_{j=1}^{\infty} N_{kj}B'_{3j} = P'_k\sin\Theta_0 \quad , \quad (k = 1,2,3,....) \quad . \tag{5.10}$$

From eqs. (2.18), (2.20), (5.3), (5.4) and (5.7)-(5.10) it follows that E'_k, H'_k, L'_k and P'_k are given by

$$E'_k = \frac{\varepsilon_2(\varepsilon_1 - \varepsilon_3)}{\varepsilon_1\varepsilon_3}\int_{1-h}^{1} dt P_k^0(t)(t - 1 + h) \quad ,$$

$$H'_k = 0 \quad ,$$

$$L'_k = 0 \quad ,$$

$$P'_k = \frac{\varepsilon_3 - \varepsilon_1}{\varepsilon_2}\int_{1-h}^{1} dt P_k^1(t)P_1^1(t) \quad . \tag{5.11}$$

Expanding E'_k and P'_k into powers of h one finds that they are of second order, and therefore all multipole coefficients are of second order in h. The integrals on the r.h.s. of eqs. (5.3) and (5.4) are at least linear in h. Neglecting the contributions of these integrals to the multipole coefficients we can eliminate the coefficients A'_{3j} and B'_{3j} from eqs. (5.9) and (5.10). We then obtain, correct up to second order in h,

$$A_{1k} = \frac{\varepsilon_2(\varepsilon_1 - \varepsilon_3)k}{2\varepsilon_1\varepsilon_3}\cos\Theta_0\int_{1-h}^{1} dt P_k^0(t)(t - 1 + h) \quad , \tag{5.12}$$

$$B_{1k} = \frac{(\varepsilon_1 - \varepsilon_3)}{2\varepsilon_2 k(k + 1)}\sin\Theta_0\int_{1-h}^{1} dt P_k^1(t)P_1^1(t) \quad . \tag{5.13}$$

For the dipole coefficients we thus find

$$A_{11} = \frac{\varepsilon_2(\varepsilon_1 - \varepsilon_3)h^2(1 - \frac{h}{3})}{4\varepsilon_1\varepsilon_3} \cos\Theta_0 \quad , \tag{5.14}$$

$$B_{11} = \frac{(\varepsilon_1 - \varepsilon_3)h^2(1 - \frac{h}{3})}{4\varepsilon_2} \sin\Theta_0 \quad , \tag{5.15}$$

and the corresponding polarizabilities, eq. (2.23), are

$$\alpha_\perp^e = \frac{\varepsilon_1(\varepsilon_3 - \varepsilon_1)}{\varepsilon_3} V \quad , \tag{5.16}$$

$$\alpha_\parallel^e = (\varepsilon_3 - \varepsilon_1)V \quad , \tag{5.17}$$

where V, the volume of the particle, is given by

$$V = \pi R^3 h^2 (1 - \frac{h}{3}) \quad . \tag{5.18}$$

A system of these particles in the low density limit, eq. (2.30), is characterized by the surface susceptibilities

$$\beta = \frac{\varepsilon_3 - \varepsilon_1}{\varepsilon_3\varepsilon_1} \langle f \rangle \quad , \tag{5.19}$$

$$\gamma = (\varepsilon_3 - \varepsilon_1) \langle f \rangle \quad , \tag{5.20}$$

where $\langle f \rangle$ is the average film thickness, given by

$$\langle f \rangle = \rho V \quad . \tag{5.21}$$

It is interesting to note that these surface susceptibilities are independent of the dielectric constant of the substrate, ε_2. In fact they are equal to the surface susceptibilities which describe the presence of a thin homogeneous film of thickness $\langle f \rangle$ and dielectric constant ε_3 on top of a substrate. In ref. 14, an article on the optical properties of thin films on rough surfaces, we derived expressions for the surface susceptibilities in terms of the average film thickness and height-height correlation functions. If we apply that model to the present system we obtain in lowest order the same result for the coefficients γ and β.

REFERENCES

1. J. Vlieger and D. Bedeaux, Thin Solid Films, 69 (1980) 107
2. D. Bedeaux and J. Vlieger, Thin Solid Films, 102 (1983) 265
3. S. Norrman, T. Andersson, C.G. Granqvist and O. Hunderi, Phys. Rev. B, 18 (1978) 674
4. D.W. Berreman, Phys. Rev., 163 (1967) 855
5. R. Ruppin, Surface Science, 127 (1983) 108
6. R. Chauvaux and A. Meessen, Thin Solid Films, 62 (1979) 125
7. J.A. Stratton, Electromagnetic Theory (McGraw-Hill, New York, 1941), p. 401
8. J. Vlieger and D. Bedeaux, Physica, 82A (1976) 221
9. D. Bedeaux and J. Vlieger, Physica, 67 (1973) 55
10. P.A. Bobbert and J. Vlieger, Physica A (1986)
11. T. Yamaguchi, S. Yoshida and A. Kinbara, Thin Solid Films, 21 (1974) 173
12. V.V. Truong and G.D. Scott, J. Opt. Soc. Am. 67 (1977) 502
13. L. Pauling and E.B. Wilson, Introduction to Quantum Mechanics (McGraw-Hill, New York, 1935), p. 448
14. M.M. Wind and J. Vlieger, Physica, 125A (1984) 75

THE POLARIZABILITY OF A TRUNCATED SPHERE

ON A SUBSTRATE II

1. INTRODUCTION

In a previous paper [1], hereafter referred to as I, we developed a general method for the calculation of the polarizability of a truncated spherical particle on a substrate, assuming that this particle is much smaller than the wavelength of the incident light. By choosing the method of image multipoles we incorporated all effects of the particle-substrate interaction. The boundary conditions for the potential and the normal component of the displacement vector at the surface of the sphere then yielded an infinite set of inhomogeneous linear equations for the multipole coefficients. The matrixelements in these equations contained integrals involving Legendre functions. In paper I we demonstrated that for the special cases of a sphere, hemisphere and thin cap these integrals could be evaluated easily.

In the present paper we shall consider the general case of a truncated sphere on a substrate. In section 2 we show that the above mentioned integrals are polynomials in the truncation parameter (the distance of the centre of the truncated sphere to the substrate divided by its radius). We derive a complete set of recurrence relations for these integrals, by means of which all matrixelements can be evaluated explicitly. At the end of section 2 we give a summary of the evaluation of the coefficients in the polynomials for those who want to skip the details of the derivation. In section 3 we apply the method to the calculation of the parallel and normal components of the polarizability of a truncated spherical gold particle on a sapphire substrate in the optical region and discuss the results. We then calculate the transmittance for perpendicularly incident light in the optical region of a system consisting of such gold particles in a two-dimensional square lattice on a sapphire substrate. The results are compared with values obtained experimentally by Niklasson and Craighead [2,3] for the transmittance of square arrays of small gold particles on sapphire. The agreement between the theoretical and experimental curves is very good, though a discrepancy between the observed and fitted particle size remains to be explained.

2. RECURRENCE RELATIONS

In paper I we derived a linear set of equations for the multipole coefficients describing the scalar electric potential in and around a truncated sphere on a substrate in a homogeneous external electric field. The matrixelements in these equations contained rather complicated integrals involving Legendre functions. In this section we shall derive a complete set of recurrence relations by means of which these integrals can be evaluated explicitly. Let us first define the integrals involved:

$$Q^m_{kj}(r_0) \equiv \int_{-1}^{r_0} dt P^m_k(t) P^m_j(t) \quad , \tag{2.1}$$

$$S^m_{kj}(r,r_0) \equiv \int_{-1}^{r_0} dt P^m_k(t) V^m_j(r,t) \quad , \tag{2.2}$$

$$T^m_{kj}(r,r_0) \equiv \int_{-1}^{r_0} dt P^m_k(t) W^m_j(r,t) \quad , \tag{2.3}$$

where r_0 is the truncation parameter, defined as the distance of the centre of the truncated sphere to the substrate divided by the radius; k and j are integers with $k \geq m$, $j \geq m$ and m is 0, corresponding to an external electric field perpendicular to the substrate, or 1, the parallel case. The functions $V^m_j(r,t)$ and $W^m_j(r,t)$ were defined in eqs. (I.2.3), (I.2.9) as

$$V^m_j(r,t) \equiv (r^2 - 4rr_0t + 4r_0^2)^{-(j+1)/2}$$

$$\times P_j^m((rt - 2r_0)(r^2 - 4rr_0t + 4r_0^2)^{-1/2}) , \quad (m = 0,1) , \tag{2.4}$$

$$W_j^m(r,t) \equiv (r^2 - 4rr_0t + 4r_0^2)^{j/2}$$

$$\times P_j^m((rt - 2r_0)(r^2 - 4rr_0t + 4r_0^2)^{-1/2}) , \quad (m = 0,1) . \tag{2.5}$$

The functions $S_{kj}^m(r,r_0)$ appear in the matrixelements, eqs. (I.2.18) and (I.2.20), only as $S_{kj}^m(1,r_0)$ and $[\frac{\partial}{\partial r}S_{kj}^m(r,r_0)]_{r=1}$. The same holds for the functions $T_{kj}^m(r,r_0)$.

We shall now proceed to derive recurrence relations for these functions. The remarkable fact that all these functions are polynomials in r_0 , a result that we shall prove below, will then allow one to determine the functions explicitly. First we derive the relationship between $S_{kj}^m(1,r_0)$ and $[\frac{\partial}{\partial r}S_{kj}^m(r,r_0)]_{r=1}$ and also between $T_{kj}^m(1,r_0)$ and $[\frac{\partial}{\partial r}T_{kj}^m(r,r_0)]_{r=1}$. If one introduces the variable

$$x \equiv r_0/r , \tag{2.6}$$

eqs. (2.2), (2.3) can be written alternatively as

$$S_{kj}^m(r,xr) = r^{-j-1} \int_{-1}^{xr} dt P_k^m(t)(1 - 4xt + 4x^2)^{-(j+1)/2}$$

$$\times P_j^m((t - 2x)(1 - 4xt + 4x^2)^{-1/2}) , \tag{2.7}$$

$$T_{kj}^m(r,xr) = r^j \int_{-1}^{xr} dt P_k^m(t)(1 - 4xt + 4x^2)^{j/2}$$

$$\times P_j^m((t - 2x)(1 - 4xt + 4x^2)^{-1/2}) , \tag{2.8}$$

where we have also used eqs. (2.4), (2.5). If one now differentiates these expressions with respect to r and then sets r equal to 1, one obtains the recurrence relations

$$[\frac{\partial}{\partial r}S_{kj}^m(r,r_0)]_{r=1} = -(j+1)S_{kj}^m(1,r_0) - r_0 \frac{\partial}{\partial r_0}S_{kj}^m(1,r_0)$$

$$+ r_0(-1)^{j+m}\frac{d}{dr_0}Q_{kj}^m(r_0) , \tag{2.9}$$

$$[\frac{\partial}{\partial r}T_{kj}^m(r,r_0)]_{r=1} = jT_{kj}^m(1,r_0) - r_0 \frac{\partial}{\partial r_0}T_{kj}^m(1,r_0)$$

$$+ r_0(-1)^{j+m}\frac{d}{dr_0}Q^m_{kj}(r_0) \quad . \tag{2.10}$$

With these equations all functions $[\frac{\partial}{\partial r}S^m_{kj}(r,r_0)]_{r=1}$ and $[\frac{\partial}{\partial r}T^m_{kj}(r,r_0)]_{r=1}$ can be determined once $S^m_{kj}(1,r_0)$, $T^m_{kj}(1,r_0)$ and $Q^m_{kj}(r_0)$ are known.

Since we can now focus our attention on $S^m_{kj}(1,r_0)$ and $T^m_{kj}(1,r_0)$ it is convenient to denote these quantities by $S^m_{kj}(r_0)$ and $T^m_{kj}(r_0)$. For the following step we shall use the identities

$$V^m_j(1,t) = (-1)^{j+m}\sum_{\ell=m}^{\infty}\frac{(\ell+j)!P^m_\ell(t)}{(\ell+m)!(j-m)!(2r_0)^{\ell+j+1}} \ , \quad r_0 > \tfrac{1}{2} \ , \tag{2.11}$$

$$V^m_j(1,t) = \sum_{\ell=j}^{\infty}\frac{(\ell-m)!(2r_0)^{\ell-j}P^m_\ell(t)}{(\ell-j)!(j-m)!} \ , \quad r_0 < \tfrac{1}{2} \ , \tag{2.12}$$

$$W^m_j(1,t) = \sum_{\ell=m}^{j}\frac{(-1)^{j+\ell}(j+m)!(2r_0)^{j-\ell}P^m_\ell(t)}{(\ell+m)!(j-\ell)!} \quad . \tag{2.13}$$

Two of these expansions where also used in I, cf. eqs. (I.3.1), (I.5.2). The validity of these three relations can easily be verified by multiplying both sides of the equations by q^j , with $|q| < 1$, summing over j and using the generating function of the Legendre polynomials. Assuming for the moment that $r_0 > \frac{1}{2}$, we find for the derivative of $S^m_{kj}(r_0)$ with respect to r_0

$$\frac{d}{dr_0}S^m_{kj}(r_0) = P^m_k(r_0)P^m_j(-r_0) - 2(-1)^{j+m}\sum_{\ell=m}^{\infty}\frac{(\ell+j+1)!Q^m_{k\ell}(r_0)}{(\ell+m)!(j-m)!(2r_0)^{\ell+j+2}} \tag{2.14}$$

where we have used eqs. (2.1), (2.2) and (2.11). Using the same equations we can simplify the second term on the r.h.s. of eq. (2.14) and after rearranging terms we find

$$2(j-m+1)S^m_{k,j+1}(r_0) = \frac{d}{dr_0}S^m_{kj}(r_0) - (-1)^{j+m}\frac{d}{dr_0}Q^m_{kj}(r_0) \quad . \tag{2.15}$$

One easily verifies that the case $r_0 < \frac{1}{2}$, where one has to use eq. (2.12) instead of eq. (2.11), yields the same recurrence relation, which is therefore valid for all values of r_0 . Note that we occasionally insert punctuation marks between indices to avoid confusion. One similarly obtains from eqs. (2.3) and (2.13) the following recurrence relation for $T^m_{kj}(r_0)$:

$$\frac{d}{dr_0}T^m_{kj}(r_0) = -2(j+m)T^m_{k,j-1}(r_0) + (-1)^{j+m}\frac{d}{dr_0}Q^m_{kj}(r_0) \quad . \tag{2.16}$$

Eqs. (2.15) and (2.16) can be used to express the functions $S^m_{kj}(r_0)$ and $T^m_{kj}(r_0)$ in terms of $S^m_{k,j-1}(r_0)$ and $T^m_{k,j-1}(r_0)$ respectively, assuming the functions $Q^m_{kj}(r_0)$ to be known. For the case of the $T^m_{kj}(r_0)$ functions one has to be careful. Since one there has to integrate, a possible constant term cannot be determined this way. Using eqs. (2.1) and (2.3) one easily verifies that this constant term should be chosen such that

$$T^m_{kj}(0) = Q^m_{kj}(0) = \int_{-1}^{0} dt P^m_k(t)P^m_j(t) \quad . \tag{2.17}$$

In paper I we encountered this expression already in the discussion of the hemisphere on a substrate. Explicit expressions for $Q^m_{kj}(0)$ are given for the case k=even, j=odd by eq. (I.4.5) for m=0 and eq. (I.4.9) for m=1. Combining these results into a single equation one finds

$$Q^m_{kj}(0) = \frac{(-1)^{\frac{1}{2}(k+j-1)}(k+1)!\ j!}{2^{k+j-2m-1}(k-m+1)(k-j)(k+j+1)\{(\frac{k-2m}{2})!\ (\frac{j-1}{2})!\}^2} \ ,$$

$$k = \text{even},\ j = \text{odd} \ . \tag{2.18}$$

Note that the functions $Q^m_{kj}(r_0)$, eqs. (2.1), are symmetric in the indices k and j. For k+j=even one easily finds

$$Q^m_{kj}(0) = \frac{(k+m)!}{(2k+1)(k-m)!}\delta_{kj} \quad . \tag{2.19}$$

We now have to evaluate the functions $S^m_{kj}(r_0)$ and $T^m_{kj}(r_0)$ for the lowest value of the index j, i.e. for j=m, since all functions of higher index j can then be evaluated by means of eqs. (2.15) - (2.19). For $T^m_{km}(r_0)$ this evaluation is straightforward, and one finds, using eqs. (2.3) and (2.13), the simple result

$$T^{m}_{km}(r_0) = Q^{m}_{km}(r_0) \quad . \tag{2.20}$$

For the more complicated $S^{m}_{km}(r_0)$ functions we shall consider the two cases $m=0$ and $m=1$ separately. Let us first examine $m=0$. From eqs. (2.2) and (2.4) one finds

$$S^{0}_{k0}(r_0) = \int_{-1}^{r_0} dt P^{0}_{k}(t)(1 - 4r_0 t + 4r_0^{2})^{-1/2} \quad . \tag{2.21}$$

We first differentiate eq. (2.21) with respect to r_0:

$$\frac{d}{dr_0} S^{0}_{k0}(r_0) = P^{0}_{k}(r_0) + 2 \int_{-1}^{r_0} dt P^{0}_{k}(t)(t - 2r_0)(1 - 4r_0 t + 4r_0^{2})^{-3/2} \quad . \tag{2.22}$$

Integration by parts and multiplication of both sides of this equation by r_0 yields

$$r_0 \frac{d}{dr_0} S^{0}_{k0}(r_0) = P^{0}_{k}(-1) - \int_{-1}^{r_0} dt (1 - 4r_0 t + 4r_0^{2})^{-1/2} \times \left\{(t - 2r_0)\frac{d}{dt} P^{0}_{k}(t) + P^{0}_{k}(t)\right\} \quad . \tag{2.23}$$

Substracting from this equation the same equation with k-2 instead of k we obtain after some rearrangement of terms

$$r_0 \frac{d}{dr_0} S^{0}_{k0}(r_0) + (k + 1) S^{0}_{k0}(r_0) = (4k - 2) r_0 S^{0}_{k-1,0}(r_0) + r_0 \frac{d}{dr_0} S^{0}_{k-2,0}(r_0) - (k - 2) S^{0}_{k-2,0}(r_0) + \delta_{k0} - \delta_{k1} \quad . \tag{2.24}$$

In deriving eq. (2.24) we have also used the following well known recurrence relations for the Legendre functions (see e.g. reference 4)

$$\frac{d}{dt} P^{m}_{k+1}(t) - \frac{d}{dt} P^{m}_{k-1}(t) = (2k + 1) P^{m}_{k}(t) \quad , \tag{2.25}$$

and

$$(2k + 1) t P^{m}_{k}(t) = (k - m + 1) P^{m}_{k+1}(t) + (k + m) P^{m}_{k-1}(t) \quad . \tag{2.26}$$

Using eq. (2.24) it follows by induction that $S^{0}_{k0}(r_0)$ is a polynomial of degree k. Note the fact that additional terms in $S^0_{k0}(r_0)$ of the form Cr_0^{-k-1} (C a constant), which do not contribute to the left hand side of eq. (2.24), can be excluded because of their singular behaviour in $r_0 = 0$. From eq. (2.15) we now verify that $S^0_{kj}(r_0)$ is a polynomial of degree $k + j - 1 + \delta_{j0}$.

For m=1 we proceed along the same lines. The functions $S^1_{k1}(r_0)$ are given by

$$S^1_{k1}(r_0) = \int_{-1}^{r_0} dt P^1_k(t)(1 - t^2)^{1/2}(1 - 4r_0 t + 4r_0^2)^{-3/2} \quad . \tag{2.27}$$

Differentiating with respect to r_0, integrating by parts and multiplying with r_0 we obtain

$$r_0 \frac{d}{dr_0} S^1_{k1}(r_0) + S^1_{k1}(r_0) = - \int_{-1}^{r_0} dt(1 - 4r_0 t + 4r_0^2)^{-3/2}(t - 2r_0)$$
$$\times \{(1 - t^2)\frac{d}{dt}P^1_k(t) - tP^1_k(t)\}(1 - t^2)^{-1/2} \quad . \tag{2.28}$$

At this point we need another two recurrence relations for the Legendre functions [4]

$$(1 - t^2)\frac{d}{dt}P^m_k(t) = (k + 1)tP^m_k(t) - (k - m + 1)P^m_{k+1}(t) \quad , \tag{2.29}$$

and

$$(1 - t^2)\frac{d}{dt}P^m_k(t) = - ktP^m_k(t) + (k + m)P^m_{k-1}(t) \quad . \tag{2.30}$$

We apply eq. (2.30) to eq. (2.28) and eq. (2.29) to eq. (2.28) with k-2 instead of k. We now choose a suitable linear combination of the equation for k and that for k-2 so that we can use eq. (2.26). The equation then simplifies considerably and we finally obtain

$$\frac{k - 1}{k + 1}\{r_0 \frac{d}{dr_0} S^1_{k1}(r_0) + (k + 2)S^1_{k1}(r_0)\} = (4k - 2)r_0 S^1_{k-1,1}(r_0)$$
$$+ \frac{k}{k - 2}\{r_0 \frac{d}{dr_0} S^1_{k-2,1}(r_0) - (k - 3)S^1_{k-2,1}(r_0)\} \quad . \tag{2.31}$$

The results eqs. (2.24) and (2.31) can be combined into a single equation:

$$\frac{k-m}{k+m}\left\{r_0 \frac{d}{dr_0} S^m_{km}(r_0) + (k+m+1) S^m_{km}(r_0)\right\} = (4k-2) r_0 S^m_{k-1,m}(r_0)$$

$$+ \frac{k+m-1}{k-m-1}\left\{r_0 \frac{d}{dr_0} S^m_{k-2,m}(r_0) - (k-m-2) S^m_{k-2,m}(r_0)\right\}$$

$$+ (\delta_{k0} - \delta_{k1})\delta_{m0} \quad . \quad (m = 0,1) \tag{2.32}$$

By the same argument as used after eq. (2.26) we can prove that $S^1_{k1}(r_0)$ is a polynomial in r_0. Since eq. (2.31) yields no results for k=1 and k=2 we evaluate $S^1_{k1}(r_0)$ for these two values of k by simply performing the integrations in eq. (2.27). This yields

$$S^1_{11}(r_0) = \frac{2}{3} - \frac{1}{2} r_0 \quad ,$$

$$S^1_{21}(r_0) = -\frac{3}{4} + \frac{12}{5} r_0 - \frac{3}{2} r_0^{\,2} \quad . \tag{2.33}$$

Consequently $S^1_{k1}(r_0)$ is a polynomial of degree k. From eq. (2.15) it then follows that $S^1_{kj}(r_0)$ is a polynomial of degree k+j-1. It is interesting to point out that the functions in eq. (2.33) are unequal to zero for $r_0 = -1$, contradictory to what follows from eq. (2.2). The reason for this is that the first derivative of the functions $S^m_{kj}(r_0)$ is discontinuous in $r_0 = -\frac{1}{2}$, and therefore the results above are not valid for $r_0 < -\frac{1}{2}$. Because we have chosen $1 > r_0 > 0$ in paper I these discontinuities cause no inconveniences.

We now turn to the functions $Q^m_{kj}(r_0)$. It is obvious that these are polynomials of degree k+j+1. Applying eq. (2.25) twice to eq. (2.1) we obtain the recurrence relation

$$\frac{j-m+1}{2j+1} Q^m_{k,j+1}(r_0) = \frac{k-m+1}{2k+1} Q^m_{k+1,j}(r_0) + \frac{k+m}{2k+1} Q^m_{k-1,j}(r_0)$$

$$- \frac{j+m}{2j+1} Q^m_{k,j-1}(r_0) \quad . \tag{2.34}$$

Finally we have to derive recurrence relations for these functions for the case j=m. For m=0 one easily finds, using eq. (2.25), the relation

$$\frac{d}{dr_0} Q^0_{k+1,0}(r_0) = (2k+1) Q^0_{k0}(r_0) + \frac{d}{dr_0} Q^0_{k-1,0}(r_0) + \delta_{k,-1} - \delta_{k0} \quad . \tag{2.35}$$

For the slightly more complicated m=1 case one obtains

$$\frac{k}{k+2}\frac{d}{dr_0}Q^1_{k+1,1}(r_0) = (2k+1)Q^1_{k1}(r_0) + \frac{k+1}{k-1}\frac{d}{dr_0}Q^1_{k-1,1}(r_0) \quad . \tag{2.36}$$

Combination of eqs. (2.35) and (2.36) yields

$$\frac{k-m+1}{k+m+1}\frac{d}{dr_0}Q^m_{k+1,m}(r_0) = (2k+1)Q^m_{km}(r_0) + \frac{k+m}{k-m}\frac{d}{dr_0}Q^m_{k-1,m}(r_0)$$

$$+ (\delta_{k,-1} - \delta_{k0})\delta_{m0} \quad . \tag{2.37}$$

For m=1 we give the functions $Q^m_{km}(r_0)$ for the values k=1 and k=2, since these cannot be obtained from eq. (2.36):

$$Q^1_{11}(r_0) = \frac{2}{3} + r_0 - \frac{1}{3}r_0{}^3 \quad ,$$

$$Q^1_{21}(r_0) = -\frac{3}{4} + \frac{3}{2}r_0{}^2 - \frac{3}{4}r_0{}^4 \quad . \tag{2.38}$$

Since one has to integrate in eq. (2.37) to obtain $Q^m_{k+1,m}(r_0)$, the constant term will be undetermined. This term is equal to $Q^m_{k+1,m}(0)$, which is just a special case of eqs. (2.18) and (2.19).

In paper I we gave a physical argument to demonstrate that the second equation of eq. (I.2.17) for k=0 is redundant. Using the results derived in this section it is possible to verify this mathematically. One can prove that the matrixelements F_{0j} and G_{0j} in this equation are identical to zero, and thus the equation is trivially satisfied.

We shall now briefly summarize those results of this section required to evaluate the integrals in the matrixelements explicitly. First one should evaluate the functions $Q^m_{kj}(r_0)$. This can be done by using eqs. (2.18) and (2.19) to find $Q^m_{km}(0)$ and next eqs. (2.37) and (2.38) to obtain the $Q^m_{km}(r_0)$ for $r_0 \neq 0$. The functions $Q^m_{kj}(r_0)$ for j>m then follow from eq. (2.34). To obtain the polynomials $S^m_{kj}(r_0)$ one first evaluates $S^m_{km}(r_0)$ with the help of eqs. (2.32) and (2.33); the $S^m_{kj}(r_0)$ for j>m are then found using eq. (2.15). The functions $T^m_{kj}(r_0)$ are first evaluated for j=m by means of eq. (2.20) and next for j>m with eqs. (2.17), (2.18) and (2.19) for $r_0 = 0$ and with eq. (2.16) for $r_0 \neq 0$. Finally the functions $[\frac{\partial}{\partial r}S^m_{kj}(r,r_0)]_{r=1}$ and $[\frac{\partial}{\partial r}T^m_{kj}(r,r_0)]_{r=1}$ are found using eqs. (2.9) and (2.10).

3. APPLICATION: GOLD PARTICLES ON SAPPHIRE

Using the recurrence relations derived in the previous section it is relatively easy to evaluate numerically the parallel and normal polarizabilities of a truncated sphere on a substrate. In this section we shall consider an application of our model. We calculate the parallel and normal polarizability of a truncated spherical gold particle on a sapphire substrate in the region of optical wavelengths (400 – 750 nm). Using the values for the polarizability we then calculate the transmittance for perpendicularly incident light of a system of such particles in a square lattice and compare the results with values obtained experimentally by Niklasson and Craighead [2,3].

For the dielectric constant of gold we use the bulk values from Johnson and Christy [5]. The refractive index of sapphire shows a little dispersion in the optical region, but for simplicity we shall use the value $n_2 = 1.77$ throughout. For the ambient we take $\varepsilon_1 = 1$. In figure 1 we have plotted the real part of the parallel component of the polarizability divided by the volume of the particle as a function of the wavelength for six values of the parameter r_0 : 1.0 (sphere), 0.8, 0.6, 0.4, 0.2 and 0.0 (hemisphere). In figures 2, 3 and 4 similar plots are made for the imaginary part of the parallel polarizability and the real and imaginary parts of the normal polarizability. The convergence as a function of M, the number of multipoles taken into account in the particle-substrate interaction (see paper I), is found to be good for all the above values of r_0 in the whole wavelength region. In fact, the absolute value of the relative deviation between the polarizability calculated with M=16 and that with M=8 is for the sphere less than 0.4% for the normal case and less than 0.1% for the parallel case. For the hemisphere the relative deviation is less than 3.5% for the normal polarizability; for the parallel polarizability the deviation reaches values as high as 13.3% near resonance, but this is mainly a result of a slight shift of the sharp resonance peak towards longer wavelengths as M is increased. If we decrease r_0 further, to negative values, the convergence becomes even worse. For this reason we restrict ourselves here to positives values of r_0. Figures 1-4 show the polarizability calculated for M=16.

For both the parallel and the normal component of the polarizability the main feature of the imaginary parts, figs. 2 and 4,

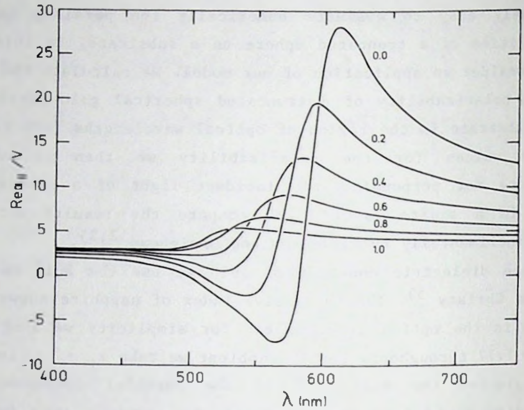


Fig. 1 Re($\alpha^e_\parallel$/V) for a gold particle on sapphire, calculated as a function of wavelength for different values of r_0 .

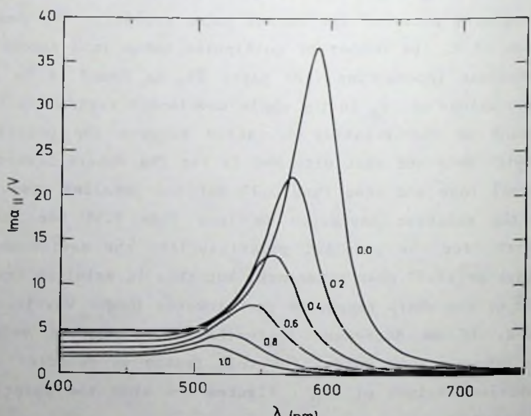


Fig. 2 Im($\alpha^e_\parallel$/V) for a gold particle on sapphire, calculated as a function of wavelength for different values of r_0 .

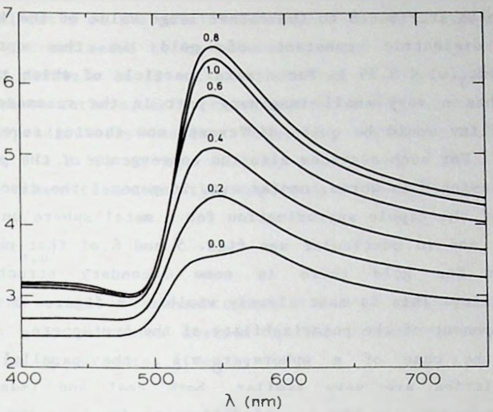


Fig. 3 $\mathrm{Re}(\alpha_{\perp}^{e}/V)$ for a gold particle on sapphire, calculated as a function of wavelength for different values of r_0 .

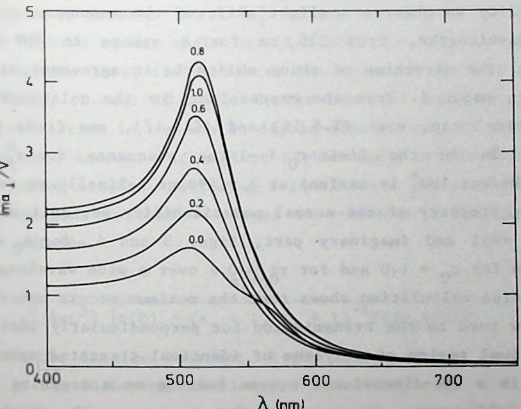


Fig. 4 $\mathrm{Im}(\alpha_{\perp}^{e}/V)$ for a gold particle on sapphire, calculated as a function of wavelength for different values of r_0 .

is the strong isolated resonance peak. The real parts, figs. 1 and 3, also show the typical behaviour associated with a single resonance. This effect can be attributed to the rather large value of the imaginary part of the dielectric constant of gold in the optical region ($1.03 < \mathrm{Im}\varepsilon_3(\omega) < 5.75$). For a metal particle of which the dielectric constant has a very small imaginary part in the resonance region the polarizability would be quite different, now showing several resonance peaks [6,7]. For such a system also the convergence of the polarizability upon increasing M is worse, confer e.g. in paper I the discussion on the validity of the dipole approximation for a metal sphere on a dielectric substrate, and in particular see figs. 5 and 6 of that paper. We note that even for gold there is some secondary structure in the polarizability. This is most clearly visible in figures 3 and 4 for the normal component of the polarizability of the hemisphere.

For the case of a sphere, $r_0 = 1$, the parallel and normal polarizabilities are very similar, both real and imaginary parts. However, decreasing r_0 has a very different effect on the parallel and normal components. For the parallel case the resonance peak shifts towards longer wavelengths, from 516 nm for $r_0 = 1$ to 589 nm for $r_0 = 0$, and also the magnitude increases drastically. For the normal polarizability we observe a slight shift of the resonance peak towards shorter wavelengths, from 515 nm for a sphere to 509 nm for a hemisphere. The direction of these shifts is in agreement with results obtained in paper I. From the expressions for the polarizability of a thin spherical cap, eqs. (I.5.16) and (I.5.17), one finds that for a gold particle in the limit $r_0 \downarrow -1$ the resonance for $\alpha_\parallel^e$ occurs at $\lambda \to \infty$, whereas $\mathrm{Im}\alpha_\perp^e$ is maximal at $\lambda = 490$ nm . Finally we note that an interesting property of the normal polarizability per unit of volume is that both real and imaginary part, figs. 3 and 4, for $r_0 = 0.8$ are larger than for $r_0 = 1.0$ and for $r_0 = 0.6$ over a wide wavelength region. A more precise calculation shows that the maximum occurs around 0.94.

We now turn to the transmission for perpendicularly incident light in the optical region of a system of identical truncated spherical gold particles in a two-dimensional square lattice on a sapphire substrate. Since we shall compare the results with experimentally obtained values by Niklasson and Craighead [2,3] we mention some properties of their system. The lattice constant is 50 nm. When the system is observed from above by electron microscopy the gold particles appear to have rotational symmetry and a diameter of 32 nm with an uncertainty of 2.5 nm. The axial ratio of the particles, defined as the diameter divided by the height, is estimated to be between 1.3 and 1.7. From the observed shape of the particle it is not clear whether an oblate spheroid model, as used by these authors, or a truncated sphere model is more realistic.

For perpendicularly incident light the transmission amplitude is given by eq. (I.2.33) or eq. (I.2.35) for $\theta = 0$. The relationship between the transmittance and this amplitude is in this case

$$T_{s,p} = n_2 \, |t_{s,p}|^2 \quad . \tag{3.1}$$

Therefore the relative transmittance, i.e. the transmittance of the system with gold particles divided by the transmittance of the bare substrate, is, in percents, given by

$$T(\%) = 100/|\, 1 - i(\omega/c)\gamma(1 + n_2)^{-1}|^2 \quad . \tag{3.2}$$

Note that for perpendicular incidence only the coefficient γ, related to the parallel polarizability, is induced by the light. We now derive the relationship between γ and $\alpha_\parallel^e$. Since the density of the particles is rather high we cannot apply eq. (I.2.30), the low density limit, but must incorporate local field effects due to inter-particle interactions in the theory, cf. eq. (I.2.31). By far the most important contribution to this interaction is the static dipole-dipole term. We shall take only this term into account and neglect higher order multipole interactions and retardation effects. Following a local field analysis for the square lattice analogous to that developed in reference 8 for general island films one obtains

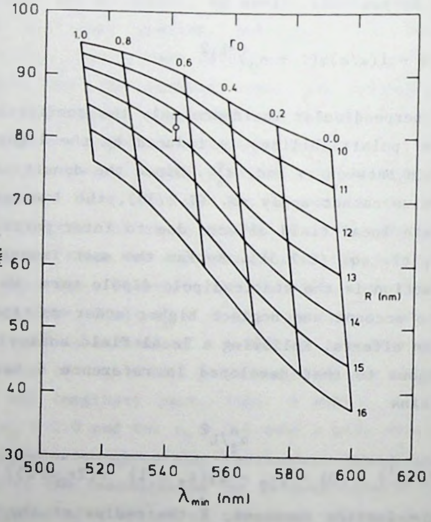
$$\gamma = \frac{\alpha_\parallel^e/L^2}{1 - (\alpha_\parallel^e/8\pi L^3)\,\{F(0) - (\varepsilon_2 - 1)(\varepsilon_2 + 1)^{-1}F(2r_0R/L)\}} \quad , \tag{3.3}$$

where L is the lattice constant, R the radius of the truncated sphere and F(x) is the following function:

$$F(x) = \sum_{m,n}{}' \{m^2 + n^2 + x^2\}^{-3/2} - 3x^2 \sum_{m,n}{}' \{m^2 + n^2 + x^2\}^{-5/2} \quad . \tag{3.4}$$

The summations are over all pairs (m,n) in the lattice except (0,0). F(x) is maximal for x=0, where it has the value 9.0336. For x>0 the function decreases monotonically and reaches half this value at x=0.565. The term with F(0) represents the interaction of the particle with the direct static dipole fields of the other particles, whereas the other term represents the interaction with the images of these dipoles. Note that the quantity $2r_0R/L$ is just the distance of the centre of the truncated sphere to its image in the substrate, divided by the lattice constant. One verifies that eq. (3.3) is indeed of the type of eq. (I.2.31), since ρ , the number of particles per unit surface area, is equal to L^{-2} , and $\kappa_\parallel$ differs not much from L^{-1} .

It is clear from eq. (3.2) that the transmittance will be mainly influenced by $\mathrm{Im}\gamma$, which in its turn is roughly related to $\mathrm{Im}\alpha_\parallel^e$. One



<u>Fig. 5</u> Position and depth of the absorption peak, calculated with bulk values for gold for different values of R and r_0 . Also shown is the minimum of the experimental transmittance.

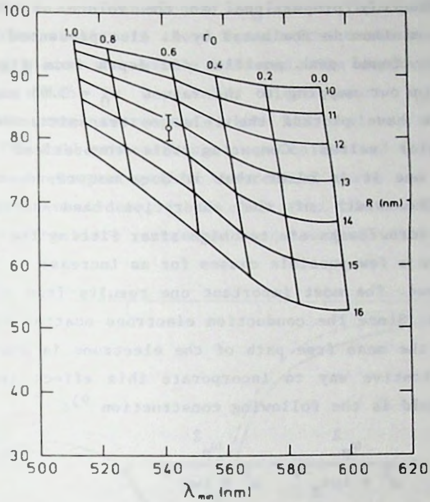


Fig 6 Position and depth of the absorption peak, calculated for different values of R and r_0 with finite size corrections incorporated. Also shown is the minimum of the experimental transmittance.

therefore expects the strong isolated resonance peak in $Im\alpha^e_\parallel$, fig. 2, to be present in the transmittance as an absorption peak. From the curve representing the experimental values for the transmittance in fig. 7, one sees that this peak is indeed present. Since the absorption peak is the main feature of the transmittance we first calculate, using the results for the polarizability and eqs. (3.2), (3.3) and (3.4), the position and depth of the minimum of this peak as a function of the truncation parameter r_0 and the radius R of the particles. The results of this calculation are represented in fig. 5. One sees that this yields a mapping between every pair λ_{min}, T_{min} on the one hand and the pair r_0, R on the other. As one would expect from fig. 2 for $Im\alpha^e_\parallel$ the position of the absorption peak is mainly determined by r_0. Since γ is approximately proportional to the volume of the particles the depth of the minimum is dominated by R. Also presented in fig. 5 is the experimentally found peak position and depth from fig. 7. This point corresponds in our mapping to the values $r_0 = 0.65$ and $R = 12.7$ nm. In fig. 7 we have plotted the relative transmittance calculated for these parameter values. Comparing this theoretical curve with the experimental one it is clear that it does not represent a good fit to the data. The width of the absorption band is too small, and consequently both flanks are too high after fitting the minimum.

There are a few possible causes for an increase of the width of the absorption band. The most important one results from the small size of the particles. Since the conduction electrons scatter at the surface of the particle the mean free path of the electrons is shorter than in the bulk. A qualitative way to incorporate this effect in the dielectric constant of gold is the following construction [9]:

$$\varepsilon(\omega) = \varepsilon_B(\omega) + \frac{\omega_P^2}{\omega^2 + i\omega\tau_B^{-1}} - \frac{\omega_P^2}{\omega^2 + i\omega\tau^{-1}} , \tag{3.5}$$

where $\varepsilon_B(\omega)$ is the bulk dielectric constant, for which we take the values of ref. 5, ω_P is the plasma frequency, τ_B is the relaxation time in the bulk metal and τ, given by [9]

$$\tau^{-1} = \tau_B^{-1} + v_F R'^{-1} , \tag{3.6}$$

is the relaxation time corrected for the finite size of the particle. v_F is the Fermi velocity and R' is some effective radius of the particle. For a truncated sphere no expression for R' is yet known. However, in reference 9 it was found that a good candidate for R' is half the short axis of the particle. We shall adopt this choice, which for a truncated sphere yields

$$R' = \frac{1}{2}(1 + r_0) R . \tag{3.7}$$

Using the values $\hbar\omega_P = 8.99$ eV, $\hbar\tau_B^{-1} = 0.027$ eV and $\hbar v_F = 0.903$ eV·nm we correct the dielectric constant of gold for the finite size of the particles with eqs. (3.5)-(3.7). Once again we calculate the position and depth of the minimum of the absorption peak in the transmittance as a function of r_0 and R. The results, drawn in figure 6, are quite similar to those found without the correction. The positions of the minima are hardly affected. The depths of the peaks have decreased, which is especially clear near $r_0 = 0.0$. The experimental data now correspond with the values $r_0 = 0.65$ and $R = 13.4$ nm. The full transmittance curve calculated for these values is drawn in fig. 7. Comparing this theoretical curve and the previous one with the experimental data, it is clear that the new curve presents a much better fit. The width corresponds quite good with the observed

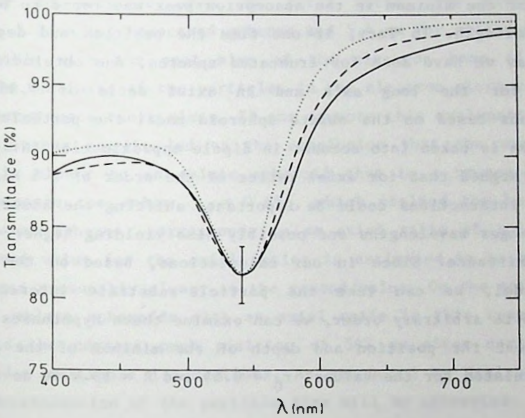


Fig. 7 Experimental relative transmittance as a function of wavelength (solid line). Theoretical curves without (dotted; R=12.7 nm, r_0=0.65) and with (dashed; R=13.4 nm, r_0=0.65) finite size correction after fitting the minimum in the absorption peak. The error bar denotes the experimental uncertainty.

value. In fact, the only discrepancies appear for the longer wavelengths. The origin of this effect is not clear to us. One notes the strong similarity between the two theoretical curves in fig. 7. Therefore using instead of eq. (3.7) a different choice for R', e.g. half the long axis of the particle, will only result in minor modifications of the curve and a slightly different fitted value for the radius.

Let us finally discuss our results, based on the truncated sphere model, and the results of Niklasson and Craighead [2,3], using the oblate spheroid model, in relation to the experimental data. Niklasson and Craighead modeled the gold particles as oblate spheroids with a long axis of 32 nm and an axial ratio of 1.5. They found that this yields a curve which is considerably lower than the experimental one. Also the position of the minimum in the absorption peak was found to be at a too short wavelength. In fact, if one fits the position and depth of the minimum, as we have done for truncated spheres, one obtains a value of 26.6 nm for the long axis and an axial ratio of 1.85. In the calculations based on the oblate spheroid model the particle-substrate interaction is taken into account in dipole approximation. Niklasson and Craighead argued that for axial ratios of the order of 1.5 higher order multipole interactions could be important, shifting the absorption peak towards longer wavelengths and possibly also yielding higher values for the transmittance. Since in our calculations, based on the truncated sphere model, we can take the particle-substrate interaction into account up to arbitrary order, we can examine these hypotheses. In table 1 we present the position and depth of the minimum of the absorption peak, calculated for the values $r_0 = 0.65$ and $R = 13.4$ nm as a function

M	λ_{min}(nm)	T_{min}(%)
1	520	83.4
2	531	81.3
4	536	82.2
8	539	81.9
16	542	81.7

Table 1. Position and depth of the absorption peak as a function of M, the number of multipoles.

of M, the number of multipoles taken into account in the interaction. One sees that for a system of truncated spheres there is indeed a considerable shift of the absorption peak in the transmittance towards longer wavelengths due to higher order multipole interactions. So it is very likely that such a shift also occurs for oblate spheroids. The other suggestion, that the relative transmittance possibly increases as a result of these higher order multipole interactions, is not supported by the results in table 1. Therefore we must consider the sizeable discrepancy between the magnitudes of the experimental and theoretical transmittances, or, equivalently, between the observed and fitted values for the particle size. One may attribute these discrepancies to the uncertainty in particle size and axial ratio. The difference in diameters, 5.2 nm for truncated spheres and 5.4 nm for oblate spheroids, is comparable with the spot size of the electron beam (5 nm). The unavoidable fuzziness of the particles in the electron micrographs leads one to overestimate their sizes. If one accepts this explanation for the size discrepancy one is led to the conclusion that the shape of the particle is closer to an oblate spheroid than to a truncated sphere. This is because the value $r_0 = 0.65$, which yielded the best fit for the truncated spheres, corresponds to an axial ratio of 1.21, whereas the observed value for the axial ratios is estimated to be between 1.3 and 1.7, and presumably closer to the second value. On the other hand, a system of oblate spheroids with an axial ratio in this range may very well yield the correct peak position of 542 nm after correcting for higher order multipole interactions. As announced by Niklasson a more precise determination of the particle size will be attempted.

REFERENCES

1. M.M. Wind, J. Vlieger and D. Bedeaux, to be published in Physica A
2. H.G. Craighead and G.A. Niklasson, Appl. Phys. Lett., 44 (1984) 1134
3. G.A. Niklasson and H.G. Craighead, Thin Solid Films, 125 (1985) 165
4. I.S. Gradshteyn and I.M. Ryzhik, Table of Integrals, Series, and Products (Academic Press, New York, 1965), p. 1005
5. P.B. Johnson and R.W. Christy, Phys. Rev. B, 6 (1972) 4370

6. R. Ruppin, Surface Science, 127 (1983) 108
7. R. Chauvaux and A. Meessen, Thin Solid Films, 62 (1979) 125
8. J. Vlieger and D. Bedeaux, Thin Solid Films, 69 (1980) 107
9. S. Norrman, T. Andersson, C.G. Granqvist and O. Hunderi, Phys. Rev. B, 18 (1978) 674

SAMENVATTING

De electromagnetische eigenschappen van oppervlakken en dunne films staan momenteel sterk in de belangstelling. Voor veel toepassingen, zoals zonnecellen, anti-reflectiecoatings, magnetische geheugensystemen en halfgeleiders, is het onderzoek naar deze eigenschappen van groot belang. Eén methode om deze eigenschappen te onderzoeken is gebruik maken van licht. Men meet dan de reflectiviteit, transmittiviteit of ellipsometrische coëfficiënt of men doet lichtverstrooiïngs-experimenten. Deze methode heeft als voordelen dat zij niet-destructief is en gevoelig voor de structuur van het oppervlak. Door de ontwikkeling van de laser is de meetnauwkeurigheid van dit soort experimenten sterk toegenomen.

Het is welbekend dat de amplituden van licht gereflecteerd of getransmitteerd door een perfect glad oppervlak gegeven worden door de Fresnel vergelijkingen. De effecten van homogene films die een dergelijk oppervlak bedekken kunnen ook eenvoudig beschreven worden. In de praktijk zijn de systemen echter vaak gecompliceerder. Oppervlakte ruwheid kan aanwezig zijn, of men is geïnteresseerd in discontinue films bestaande uit kleine metalen deeltjes. In dit proefschrift worden twee zeer verschillende theorieën ontwikkeld om de optische eigenschappen van dergelijke systemen te beschrijven. In het eerste gedeelte, bestaande uit hoofdstukken II en III, wordt een ruw oppervlak bedekt met een continue en homogene film bestudeerd. Het tweede gedeelte, hoofdstukken IV en V, beschrijft de optische eigenschappen van dunne discontinue films bestaande uit kleine afgeknotte bollen. In de eerste theorie kan de correlatielengte van de oppervlakteruwheid van de orde van grootte of langer dan de golflengte van het invallende licht zijn. Het golfkarakter van het licht is hier derhalve belangrijk en de ontwikkelde theorie is electrodynamisch. In de tweede theorie nemen we aan dat de deeltjes veel kleiner zijn dan de golflengte, zodat lokaal retardatie-effecten verwaarloosd kunnen worden en met een electrostatische theorie kan worden volstaan.

In hoofdstuk II ontwikkelen we een theorie voor de beschrijving van de optische eigenschappen van dunne, statistisch isotrope en homogene, films op ruwe oppervlakken. We nemen aan dat zowel de gemiddelde filmdikte als de amplitude van de oppervlakte ruwheid veel kleiner zijn dan de golflengte van het licht. Als uitgangspunt is gekozen voor een theorie, oorspronkelijk geformuleerd door Kröger en Kretschmann en verder ontwikkeld door Albano, Bedeaux en Vlieger. In deze methode vervangt men een grenslaag tussen twee dielectrische media door een fictief grensvlak, gesitueerd ergens in de oorspronkelijke laag. De electromagnetische velden, analytisch uitgebreid tot het nieuwe grensvlak, voldoen daar niet meer aan de gebruikelijke randcondities. De nieuwe randcondities kunnen uitgedrukt worden in termen van fictieve, fluctuerende polarisatie- en magnetisatiedichtheden. We passen deze methode toe op het systeem van een dunne film op een ruw oppervlak en leiden uitdrukkingen af voor de fluctuerende polarisatie- en magnetisatiedichtheden, correct tot op tweede orde in de gemiddelde filmdikte en de amplitude van de ruwheid. Hiermee worden de gemiddelde polarisatie- en magnetisatiedichtheden berekend, die m.b.v. een klein aantal constitutieve coëfficiënten in de gemiddelde electromagnetische velden kunnen worden uitgedrukt. Deze coëfficiënten bevatten de gemiddelde filmdikte en de hoogte-hoogtecorrelatiefuncties van de boven- en onderoppervlakken van de ruwe film. Voor parallel en loodrecht gepolariseerd licht leiden we uitdrukkingen af voor de reflectie- en transmissieamplituden in termen van deze constitutieve coëfficiënten voor willekeurige hoek van inval van het licht. De reflectiviteit, transmittiviteit en ellipsometrische coëfficiënt worden hieruit afgeleid.

In hoofdstuk III passen we de algemene theorie van het vorige hoofdstuk toe op een ruw oppervlak dat gekarakteriseerd wordt door een Gaussische correlatiefunctie met een correlatielengte veel groter dan de golflengte van licht. In deze limiet kunnen de constitutieve coëfficiënten expliciet berekend worden. De optische grootheden worden hiermee berekend en vergeleken met de resultaten verkregen met een theorie van Ohlídal, Navrátil en Lukeš, die gebaseerd is op het gebruik van diffractie-integralen. Voor de reflectiviteit en de transmittiviteit zijn de resultaten in laagste orde gelijk. Bij de ellipsometrische coëfficiënt treden echter afwijkingen op. De door ons gevonden uitdrukking verschilt zowel van het door hen met de Helmholtz-Kirchhoff-integraal gevonden resultaat als van het daarvan afwijkende door hen met de Stratton-Chu-Silver-integraal gevonden resultaat. Om de oorzaak van deze verschillen te achterhalen analyseren we het electromagnetische veld op het ruwe oppervlak in detail. We tonen aan dat de door Ohlídal, Navrátil en Lukeš gebruikte tangentiëlevlakbenadering, waarin lokaal de kromming van het ruwe oppervlak wordt verwaarloosd, inconsistent is en dat voor een correcte beschrijving de krommingstermen in het lokale electromagnetische veld moeten worden meegenomen. Substitueert men het juiste electromagnetische veld in de Helmholtz-Kirchhoff- of Stratton-Chu-Silver-integraal, dan vindt men dezelfde uitdrukkingen voor de reflectie- en transmissieamplituden als met de methode van de constitutieve coëfficiënten. Dit is bevredigend aangezien uit de Maxwell-vergelijkingen volgt dat de Helmholtz-Kirchhoff-integraal en de Stratton-Chu-Silver-integraal zonder benadering hetzelfde resultaat geven voor deze amplituden.

In hoofdstuk IV wordt een methode ontwikkeld voor de berekening van de optische eigenschappen van dunne discontinue films, bestaande uit kleine afgeknotte bolvormige deeltjes. We nemen aan dat deze deeltjes voldoende klein zijn ten opzichte van de golflengte van het licht, zodat in en rond elk deeltje het golfkarakter van de electromagnetische velden verwaarloosbaar is. In die limiet kunnen we lokaal de quasi-statische benadering gebruiken. Wanneer de oppervlaktedichtheid van de deeltjes niet al te groot is wordt de respons van een deeltje op het uitwendige veld gedomineerd door de interactie met het substraat. Derhalve richten we onze aandacht eerst op het berekenen van de quasi-statische polariseerbaarheid van één deeltje op een substraat in een homogeen uitwendig electrisch veld. We volgen een methode van Berreman en ontwikkelen de electrostatische potentiaal in en rond elk deeltje in multipolen. Om de interactie met het substraat eenvoudig te kunnen beschrijven kiezen we de methode van de beeldmultipolen. De randcondities op het boloppervlak leiden dan tot een oneindig stelsel lineaire vergelijkingen voor de multipoolcoëfficiënten, waarbij de matrixelementen tamelijk gecompliceerde integralen bevatten. Hieruit kan men benaderde oplossingen voor de polariseerbaarheid vinden door een eindig aantal multipolen in de interactie mee te nemen en dan het asymptotische gedrag als functie van dit aantal te berekenen. Voor drie speciale gevallen, een bol, een halve bol en een dun kapje, kunnen we de integralen, en derhalve de matrixelementen, zonder veel moeite berekenen. Voor het laatste geval tonen we aan dat de methode consistent is met die van hoofdstuk II. Verder laten we zien hoe, uitgaande van de polariseerbaarheid, de optische eigenschappen berekend kunnen worden van een systeem bestaande uit een groot aantal afgeknotte bolvormige deeltjes op een substraat. Evenals in de theorie van hoofdstuk II worden deze optische eigenschappen beschreven door een klein aantal constitutieve coëfficiënten.

In hoofdstuk V keren we terug tot het gecompliceerdere algemene geval van een afgeknotte bol op een substraat. We leiden een volledig stelsel recursierelaties af waarmee alle integralen in de matrixelementen systematisch berekend kunnen worden. M.b.v. deze recursierelaties is het niet moeilijk de polariseerbaarheid van een afgeknotte bol numeriek te berekenen tot op elke gewenste nauwkeurigheid. We passen de theorie toe op het geval van een goud deeltje op een saffieren substraat voor frequenties in het optische gebied. De methode blijkt in dit geval tot goede convergentie voor de polariseerbaarheid te leiden. Tenslotte berekenen we de transmittiviteit voor loodrecht invallend licht van een systeem van gouddeeltjes in een vierkant rooster op het saffieren substraat. De resultaten zijn in goede overeenstemming met door Niklasson en Craighead experimenteel gevonden waarden van deze optische grootheid.

Curriculum Vitae

Ik ben geboren op 3 september 1958 te Den Haag. In 1976 behaalde ik het diploma Gymnasium B aan de Johan de Witt scholengemeenschap te Den Haag. In september van datzelfde jaar begon ik mijn studie aan de Rijksuniversiteit Leiden. Het kandidaatsexamen N1 (hoofdvakken natuurkunde en wiskunde en bijvak sterrenkunde) werd in januari 1980 afgelegd. Het doctoraal examen natuurkunde met bijvak wiskunde behaalde ik januari 1983. Tijdens de studie werd experimenteel onderzoek verricht onder leiding van Dr. A. J. van Duyneveldt. In februari 1983 begon ik mijn werk als wetenschappelijk assistent aan het Instituut-Lorentz voor theoretische natuurkunde in Leiden, waar ik onder leiding van Dr. J. Vlieger en Prof. Dr. D. Bedeaux onderzoek heb verricht op het gebied van de optische eigenschappen van ruwe oppervlakken en dunne films. Een groot deel van de resultaten van dit onderzoek zijn weergegeven in dit proefschrift. In juni 1983 werd een conferentie over ellipsometrie in Parijs bezocht. Nauwe contacten werden onderhouden met Dr. R. Greef (Southampton) en Dr. G. A. Niklasson (Göteborg) over het experimentele werk dat zij aan dit soort systemen verrichtten. Aan de onderwijstaken werd door mij bijgedragen door het verlenen van assistentie bij het afnemen van tentamens en het geven van een werkcollege Quantumfysica II.

LIST OF PUBLICATIONS

1. J. Vlieger and M.M. Wind :
   Optical properties of thin films on rough surfaces.
   Journal de Physique, 44 (1983) C10-363

2. M.M. Wind and J. Vlieger :
   Optical properties of thin films on rough surfaces.
   Physica, 125A (1984) 75

3. M.M. Wind and J. Vlieger :
   Optical properties of thin films on rough surfaces II.
   Long correlation length limit; comparison with earlier work.
   Physica, 131A (1985) 1

4. R. Greef and M.M. Wind :
   Polar representation of ellipsometric data.
   Applied Optics, 25 (1986) 1627

5. M.M. Wind, J. Vlieger and D. Bedeaux :
   The polarizability of a truncated sphere on a substrate I.
   To be published in Physica A

6. P.A. Bobbert, M.M. Wind and J. Vlieger :
   Diffusion to a slowly growing truncated sphere on a substrate.
   To be published in Physica A

7. M.M. Wind, P.A. Bobbert, J. Vlieger and D. Bedeaux :
   The polarizability of a truncated sphere on a substrate II.
   Leiden, preprint

Apart from minor modifications, chapters II, III, IV and V of this thesis are contained in the publications 2, 3, 5 and 7 respectively.

STELLINGEN

1. Het quasi-statische concentratieveld van een puntbron temidden van een twee-dimensionale verdeling van absorberende deeltjes is zowel in het vlak van de deeltjes als loodrecht daarop gescreend, echter zwakker dan in de analoge drie-dimensionale verdeling.

2. Wanneer men over twee perfect geleidende bollen een homogeen uitwendig electrisch veld aanlegt, parallel aan het lijnstuk dat de centra van de bollen verbindt, zijn, in de limiet van rakende bollen, de hoge waarde van de polarizeerbaarheid en de slechte convergentie van deze grootheid, als functie van het aantal multipolen dat men in de interactie in rekening brengt, een gevolg van de singuliere oppervlakteladingsdichtheid die bij het contactpunt ontstaat.

3. Het door Wood gevonden resultaat voor de energie E nodig om één elementaire lading e over te brengen van een oneindig reservoir op een ongeladen bol van straal R, $E = \frac{5}{8} e^2/R$, is onjuist en dient vervangen te worden door $E = \frac{1}{2} e^2/R$, de energie van een geladen bolcondensator.

D.M. Wood, Phys. Rev. Lett. 46 (1981) 749

4. De diffusiezones, zoals die door Hills en Scharifker worden geïntroduceerd om de tijdsafhankelijke diffusiestroom te berekenen naar een verdund systeem van groeiende deeltjes op een substraat, zijn fysisch onrealistisch.

B. Scharifker en G. Hills, Electrochim. Acta 28 (1983) 879

5. Wanneer men met behulp van ellipsometrie de groei van een homogene zwak absorberende film op een substraat volgt tot deze een dikte bereikt heeft van enige malen de golflengte van het gebruikte licht, verdient het overweging de ellipsometrische hoeken ψ en Δ in een polaire representatie weer te geven.

R. Greef en M.M. Wind, Appl. Optics 25 (1986) 1627

6. De theorie, dat de vestiging van Germaanse stammen in de vijfde eeuw in Engeland en de daarop volgende onderwerping van het land gepaard ging

met het op grote schaal afslachten of verjagen van de geromaniseerde Keltische bewoners, dient bijgesteld te worden.

7. De optische eigenschappen van ruwe oppervlakken kunnen op systematische en overzichtelijke wijze beschreven worden in termen van oppervlaktesusceptibiliteiten.

Dit proefschrift, hoofdstukken II en III

8. Uitgaande van de statische dipoolcoëfficiënten van halvebolvormige onregelmatigheden op een kristaloppervlak, berekent Berreman op onjuiste wijze het electromagnetische veld dat door dit oppervlak wordt gereflecteerd.

D.W. Berreman, Phys. Rev. 163 (1967) 855

Dit proefschrift, hoofdstuk IV

9. Wanneer men licht laat invallen op een ruw oppervlak, waarvan de spreiding in de hoogteverdeling veel kleiner en de correlatielengte langs het oppervlak veel groter is dan de golflengte van dit licht, worden de laagste orde correcties van de reflectie- en transmissieamplituden op de Fresnelwaarden slechts bepaald door de hoogteverdeling van het oppervlak. Voor de berekening van de tweede orde correctietermen dient men in het lokale veld naast de termen ten gevolge van de hoogte van het oppervlak zowel die ten gevolge van de oriëntatie van het raakvlak als krommingstermen in beschouwing te nemen.

Dit proefschrift, hoofdstuk III

10. Voor de berekening van de polariseerbaarheid van een metaaldeeltje op een diëlectrisch substraat is het in het algemeen onvoldoende de wisselwerking tussen deeltje en substraat slechts in dipoolbenadering mee te nemen.

Dit proefschrift, hoofdstuk V

M.M. Wind 22 januari 1987