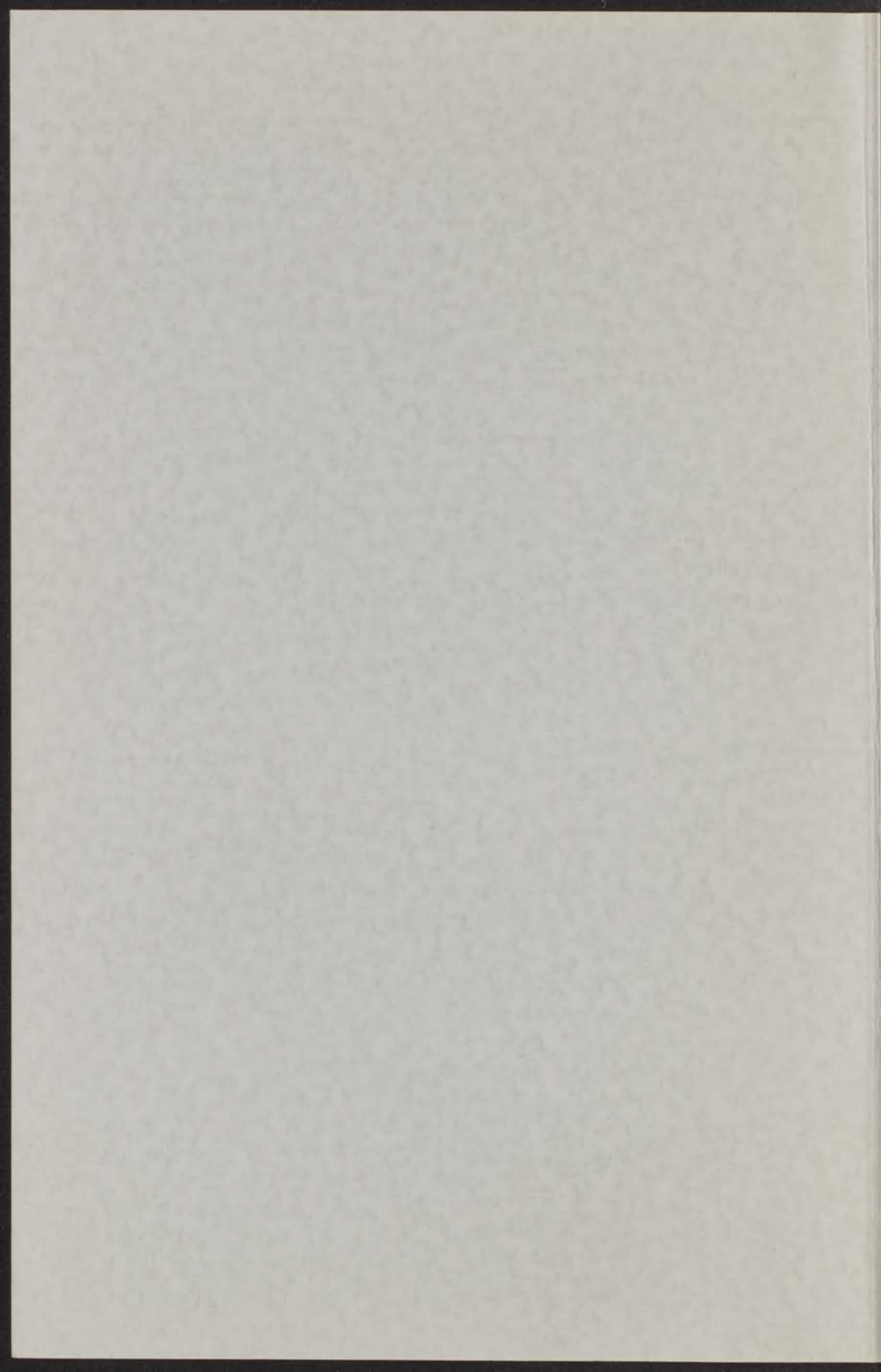


ON SOME ASPECTS OF THE  
ELECTRODYNAMIC BEHAVIOUR  
OF MATERIAL SYSTEMS

J. DE GOEDE



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## STELLINGEN

### I

Het klassieke extinctietheorema van Ewald en Oseen is reeds een gevolg van de fenomenologische Maxwell-theorie en de fenomenologische materiaalvergelijkingen.

Hoofdstuk I van dit proefschrift.

### II

Een statistische uitdrukking voor de transversale geleidingsvermogens-tensor van een materieel systeem kan met behulp van een projectieoperator-methode worden afgeleid.

Hoofdstuk II van dit proefschrift.

### III

Voor een klassiek electronengas kan een projectieoperator gevonden worden, die voor golflengten veel groter dan de Debye-afschermingslengte het longitudinale geleidingsvermogen geeft. Dit longitudinale geleidingsvermogen voldoet aan Kramers-Kronig relaties.

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## IV

Voor het probleem van de niet-stationaire diffusie door een membraan, zoals geformuleerd door Hoffmann en Unbehend, kan een exacte oplossing worden gegeven. Voor lange tijden wijkt deze aanzienlijk af van de door Hoffmann en Unbehend gegeven benadering.

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## V

Uit de theorieën van de optische activiteit van Maaskant en Oosterhoff en Terwiel en Mazur kunnen Onsager-Casimir relaties worden afgeleid voor de kruiscoëfficiënten in de uitdrukkingen voor de polarisatie en de magnetisatie.

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## VI

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## VII

De beschouwingen van Mandel over de fenomenologische theorie van de diëlectrische permittiviteit kunnen worden uitgebreid tot willekeurige media en uitwendige wisselvelden.

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## VIII

In de chemie van de vaste stof is het ionogene model nog steeds het beste uitgangspunt voor beschouwingen betreffende de kristalstructuur.

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De conclusie van Segal, dat de elektrische fluctuaties in kikkerhuiden veroorzaakt worden door het proces, dat het actieve transport van natrium regelt, is twijfelachtig.

Segal, J.R., *Biophys. J.* 12 (1972) 1371.

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BEHAVIOUR OF MATERIAL SYSTEMS

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1973

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ON SOME ASPECTS OF THE ELECTRODYNAMIC  
BEHAVIOUR OF MATERIAL SYSTEMS

# ON SOME ASPECTS OF THE ELECTRODYNAMIC BEHAVIOUR OF MATERIAL SYSTEMS

PROEFSCHRIFT

TER VERKRIJGING VAN DE GRAAD VAN DOCTOR IN  
DE WISKUNDE EN NATUURWETENSCHAPPEN AAN  
DE RIJKSUNIVERSITEIT TE LEIDEN, OP GEZAG VAN  
DE RECTOR MAGNIFICUS DR. A. E. COHEN,  
HOGLERAAR IN DE FACULTEIT DER LETTEREN,  
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JACOB DE GOEDE  
GEBOREN TE AMSTERDAM IN 1934

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PROEFSCHRIFT

Promotor: PROF. DR. P. MAZUR

DE VERKRIJGING VAN DE GRAAD VAN DOCTOR IN  
DE WISSENSCHAPPEN VAN DE NATUURWETENSCHAPPEN AAN  
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Ter nagedachtenis aan mijn vader  
Aan mijn moeder  
Aan Marijke  
Aan Anne Marie

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## INTRODUCTION

In the domain of linear, macroscopic electrodynamics the behaviour of material bodies in an electromagnetic field may be characterized by such material parameters as the generalized, wave vector and frequency dependent dielectric tensor or the generalized, wave vector and frequency dependent conductivity tensor. These parameters enable one to determine the resulting electromagnetic field at any point in space and time when the material system is subjected to an arbitrary external electromagnetic field. It is the task of statistical mechanics to provide, in terms of the microscopic properties of the system, expressions for these material constants. The conventional way to obtain such expressions is to first derive a statistical expression for the "external" susceptibility tensor characterizing the current response of the system to an external electromagnetic field and then to eliminate the external field by means of a relation between the external field and the Maxwell field of the system. One obtains in this way an expression for the conductivity tensor in terms of the external susceptibility.

In the theory of optics <sup>1)</sup>, one is interested in the behaviour of material systems, when they are perturbed by fields which are generated by external sources outside the material system ("light"). In particular one studies the propagation of normal waves in material bodies characterized by indices of refraction. In one such theory, the so-called rigorous dispersion theory, the extinction theorem of Ewald and Oseen plays an important role. <sup>3)</sup> Due to the principle of linear superposition the total electric field inside the medium consists of the sum of the incident wave and the induced wave. As the velocity of light in a medium is in

general different from the velocity of light in vacuum, the incident light has to be cancelled by the induced field. How this comes about is the context of the extinction theorem. It states that the incident electric field at any point inside the medium is cancelled ("extinguished") by the electric field generated by the induced current density at the boundary of the material substance. The method by which this theorem, together with an expression of the index of refraction, is obtained in this theory, seems to imply that the extinction of the incident wave should be understood on the basis of microscopic considerations.

In this thesis we shall on the one hand attempt to elucidate the meaning of the extinction theorem and on the other hand develop a method by means of projection operator techniques which yields directly the required statistical expressions for the material parameters of the system.

In chapter I we derive, starting from the general solution of the classical boundary-value problem for electromagnetic fields, formal extinction theorems for the incident electromagnetic field. These expressions are applicable to any medium and are nothing but general identities. Introducing material equations the true extinction theorem of the Ewald-Oseen type is derived for linear, isotropic, homogeneous media. The importance of surface effects arising from discontinuities in the material parameters is demonstrated. This analysis proves that the extinction theorem is a consequence of the macroscopic Maxwell equations together with constitutive relations. We may therefore infer that in the traditional theories this theorem should rather be interpreted as a consistency check with Maxwell's equations. In chapter II we show, in the framework of linear response theory, that by means of projection operator techniques an expression for the transverse part of the wave vector and frequency dependent conductivity tensor can be found directly in terms of the transverse part of a current current correlation function with a modified propagator. From the structure of this expression Kramers-

Kronig relations follow in a standard way. The fact that we were not able to find an analogous expression for the longitudinal conductivity is connected to the circumstance that there can be no general proof of Kramers-Kronig relations for the longitudinal conductivity as was pointed out by Martin. <sup>1)</sup> It was also stressed by Martin that the failure of a physical system to obey Kramers-Kronig relations should not be interpreted as a breakdown of causality.

To compute the conductivity tensor we must include the electromagnetic interactions between the particles. Their inclusion considerably complicates all calculations. It is therefore common practice to employ self-consistent field methods as a first approximation. <sup>4)</sup> These methods are based on the assumption that the conductivity tensor, which represents the response to the total average electromagnetic field, may be obtained by evaluating the response to an external electromagnetic field for a model system with an effective Hamiltonian in which part of the electromagnetic interaction is neglected. In the next approximation one then consider so-called local field (Lorentz field) corrections. <sup>5)</sup> It is stated in the literature <sup>6)</sup>, that local field corrections are not important in plasmas. It is not clear, whether such self-consistent field methods are reasonable in the case of e.g. a molecular fluid. One of the attractive features of the expressions for the electromagnetic transport coefficients in terms of commutator correlation functions with a modified propagator is the circumstance that they form a convenient starting point to study the validity of the above mentioned self-consistent field methods.

In chapter III we derive so-called "inverse" extinction theorems from the conventional linear response theory for material systems to an external electromagnetic field. These inverse extinction theorems express the fact that the induced current density and the total electric field at any point inside a medium is equal to a current or field generated by the external electric field at the boundary of the medium. Such inverse extinction

theorems are also obtained directly from the phenomenological Maxwell theory. Therefore, as in the case of the usual extinction theorem, these relations are a consequence of the Maxwell equations together with material equations. Together with the physical interpretation of these relations, some aspects of the linear response theory for translationally invariant systems are discussed. We also give an interpretation of the commutator correlation functions in the linear response theory, i.e. the "external" susceptibilities, in terms of the macroscopic propagator of electrodynamics and the conductivity tensor of the medium.



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## CHAPTER I

### ON THE EXTINCTION THEOREM IN ELECTRODYNAMICS

#### Synopsis

Starting from the general solution of the classical boundary-value problem for electromagnetic fields, formal extinction theorems for the incident electromagnetic field are derived which are applicable to any medium and are nothing but general identities. Introducing material equations, the true extinction theorem of the Ewald-Oseen type is then derived from these identities for linear, isotropic, homogeneous systems. The importance of surface effects arising from discontinuities in the material parameters is demonstrated. This treatment establishes the intimate connection between the classical boundary-value problem and the extinction theorem.

1. *Introduction.* The Ewald-Oseen extinction theorem was originally obtained within the frame work of the so-called rigorous dispersion theory<sup>1,2,3</sup>, starting from an analysis of the integral equation for a linear, isotropic dielectric of volume  $V$ , subjected to an external electric field†

$$P(\mathbf{r}\omega) = \rho\alpha(\omega)[\mathbf{E}^e(\mathbf{r}\omega) + \int_{s(\mathbf{r})}^{\Sigma} \text{curl}_{\mathbf{r}} \text{curl}_{\mathbf{r}} g(\mathbf{r}|\mathbf{r}'; k) P(\mathbf{r}'\omega) dv']. \quad (1.1)$$

Here  $P(\mathbf{r}\omega)$  is the polarization per unit volume at position  $\mathbf{r}$  and frequency  $\omega$ ,  $\mathbf{E}^e(\mathbf{r}\omega)$  the external (incident) electric field,  $\alpha(\omega)$  the mean molecular polarizability,  $\rho$  the density of the medium and  $g$  the scalar Green function

$$g(\mathbf{r}|\mathbf{r}'; k) = \frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{4\pi|\mathbf{r}-\mathbf{r}'|} \quad \text{with} \quad k = \omega/c. \quad (1.2)$$

In eq. (1.1) a small sphere centered around  $\mathbf{r}$  and bounded by  $s(\mathbf{r})$  is excluded from the integration domain which is bounded by the surface  $\Sigma$  of the medium. Formula (1.1) is obtained from microscopic considerations. As

† All time-dependent quantities are Fourier-transformed in time, *e.g.*,

$$\mathbf{E}^e(\mathbf{r}\omega) = \int_{-\infty}^{+\infty} \mathbf{E}^e(\mathbf{r}t) e^{i\omega t} dt.$$

We use rationalized gaussian units throughout.

shown by Rosenfeld<sup>2</sup>), effects of radiation damping and parts of density fluctuations may be included in the definition of  $\alpha$ .

It was now possible through a careful analysis of the integral equation to derive the Lorentz-Lorenz formula for the index of refraction  $n$  of the medium

$$(n^2 - 1)/(n^2 + 2) = \frac{1}{3}\rho\alpha. \quad (1.3)$$

At the same time the analysis yields the result

$$\mathbf{E}^e(\mathbf{r}\omega) + \frac{1}{k^2(n^2 - 1)} \times \text{curl}_{\mathbf{r}} \text{curl}_{\mathbf{r}} \int_{\Sigma} [\mathbf{P}(\mathbf{r}'\omega) g_n(\mathbf{r}|\mathbf{r}'; k) - g(\mathbf{r}|\mathbf{r}'; k) \mathbf{P}_n(\mathbf{r}')] d\sigma = 0. \quad (1.4)$$

The surface integral in (1.4) is performed over the boundary  $\Sigma$  of the system. The index  $n$  denotes the normal derivatives at the boundary in the outward direction. Formula (1.4) expresses the fact that the external field  $\mathbf{E}^e$  at any point in the medium is cancelled by a field due to the polarization at the surface of the medium. This "most remarkable feature of the analysis"<sup>4</sup> which became known as the Ewald-Oseen "Ausloeschungssatz"<sup>5</sup>, seemed to imply that the extinction of the incident field should be understood on the basis of the integral equation (1.1) and therefore on microscopic considerations.

It should be noted at this point that a statistical-mechanical treatment of a system of interacting polarizable point particles (atoms, molecules) does not yield the integral equation (1.1), but an equation containing additional terms due to fluctuations of the molecular dipole moments. It has been shown, however, that the analysis of the latter equation leads to a modified Lorentz-Lorenz formula and again to the extinction theorem (1.4)<sup>6</sup>. For optically active media, similar treatments have been given<sup>3, 7, 8, 9</sup>, leading to a formula for the optical rotatory power and again to an extinction theorem. Again, therefore, it would seem that the extinction theorem is obtained ultimately on the basis of a microscopic analysis.

Recently Sein<sup>10</sup>), in an elegant note, has criticized this last point of view and has shown that the extinction theorem is already contained in the Maxwell equations and the material equations. Sein assumes both the charge and current densities to be non-singular everywhere in the material medium and outside the medium. He derives with the use of the Hertz vector a general statement equivalent to a formal extinction theorem. From this statement he derives for the case of a non-magnetic dielectric the usual form of the Ewald-Oseen extinction theorem. However, the assumption of non-singular charge and current densities is certainly too restrictive. Indeed, even for a non-magnetic homogeneous, isotropic dielectric, there will be a



surface-charge density due to the discontinuity of the dielectric constant at the boundary of the medium, while for a medium which is also magnetic, there will be in addition a surface-current density due to the discontinuity of the magnetic permeability. Furthermore it would seem that a formulation in terms of a Hertz vector is not the most transparent approach to the problem, since this quantity cannot under all circumstances easily be eliminated in favour of physical charge and current densities.

In this chapter we pursue the ideas put forward by Sein and show that one can indeed derive the extinction theorem entirely on the basis of the Maxwell equations and the material equations. In doing so, we shall allow the charge and current densities to be singular at the surface of the medium and present the analysis entirely in terms of physical fields.

In section 2 we discuss the well-known classical boundary-value problem for electromagnetic fields and their sources. This problem consists in expressing the electromagnetic field at an interior point in terms of the values of the electromagnetic field over an enclosing surface and the sources inside this surface. At first, we represent the discontinuities at the boundary of the system by a small transition layer in which the physical properties change very rapidly but continuously.

The solution of the boundary-value problem is shown in section 3 to lead to formal extinction theorems (identities) for the incident electromagnetic field, thus showing the intimate connection between the classical boundary-value problem and the extinction theorems.

The formal extinction theorems are cast into various forms by means of transformation formulae collected in appendix A. These forms enable us to discuss static problems and derive later on true extinction theorems of the Ewald-Oseen type using the material equations characterizing the medium. Finally, in section 3, we go over to real discontinuities at the boundary of the system by a limiting procedure introducing in this way surface-charge and surface-current densities.

In section 4 we introduce suitable material equations which describe linear, homogeneous and isotropic media which are superconducting, conducting, dielectric, optically active and magnetic. We then derive the corresponding explicit form of the extinction theorem for the incident electric field. For the case of a simple dielectric the general extinction theorem reduces to the usual form of Ewald and Oseen (1.4). The case of a dielectric with magnetic properties is also discussed.

Some conclusions that may be drawn from the treatment presented, are discussed in section 5.

*2. Maxwell's equations and the classical boundary-value problem.* We consider a material system, bounded by a regular closed surface  $\Sigma$ . The system is acted upon by an external electromagnetic field with electric and mag-

netic field vectors  $\mathbf{E}^e(\mathbf{r}\omega)$  and  $\mathbf{B}^e(\mathbf{r}\omega)$ , where  $\mathbf{r}$  is the three-dimensional position vector and  $\omega$  the frequency. The coupling of the external electromagnetic field and the material system gives rise to an additional electromagnetic field. The total electromagnetic field at  $\mathbf{r}$ ,  $\mathbf{E}(\mathbf{r}\omega)$  and  $\mathbf{B}(\mathbf{r}\omega)$  is a solution of the Maxwell equations

$$\text{curl } \mathbf{E}(\mathbf{r}\omega) - \frac{i\omega}{c} \mathbf{B}(\mathbf{r}\omega) = 0, \quad (2.1)$$

$$\text{curl } \mathbf{B}(\mathbf{r}\omega) + \frac{i\omega}{c} \mathbf{E}(\mathbf{r}\omega) = \frac{1}{c} \mathbf{I}(\mathbf{r}\omega), \quad (2.2)$$

$$\text{div } \mathbf{E}(\mathbf{r}\omega) = \rho(\mathbf{r}\omega), \quad (2.3)$$

$$\text{div } \mathbf{B}(\mathbf{r}\omega) = 0, \quad (2.4)$$

with  $\mathbf{I}(\mathbf{r}\omega)$  and  $\rho(\mathbf{r}\omega)$  the (total) current and charge densities, respectively. We assume, as is usually done, that in a small transition layer located between the regular surfaces  $\Sigma^-$  just inside the material system and  $\Sigma^+$  just outside the material system, the source terms  $\mathbf{I}(\mathbf{r}\omega)$  and  $\rho(\mathbf{r}\omega)$  change very rapidly but continuously from their values at  $\Sigma^-$  to zero at  $\Sigma^+$ .

The problem of expressing the electromagnetic field  $\mathbf{E}(\mathbf{r}\omega)$ ,  $\mathbf{B}(\mathbf{r}\omega)$  at an interior point in terms of the values of  $\mathbf{E}(\mathbf{r}\omega)$  and  $\mathbf{B}(\mathbf{r}\omega)$  over an enclosing surface  $S$  and the sources inside  $S$  (within the volume  $\Omega$  bounded by  $S$ ) constitutes the classical boundary value problem<sup>11</sup>). We follow the standard procedure<sup>12</sup>) to obtain the solution. From the Maxwell equations (2.1)–(2.4) we derive

$$-\text{curl curl } \mathbf{E}(\mathbf{r}\omega) + k^2 \mathbf{E}(\mathbf{r}\omega) = -\frac{i\omega}{c^2} \mathbf{I}(\mathbf{r}\omega), \quad (2.5)$$

and

$$-\text{curl curl } \mathbf{B}(\mathbf{r}\omega) + k^2 \mathbf{B}(\mathbf{r}\omega) = -\frac{1}{c} \text{curl } \mathbf{I}(\mathbf{r}\omega), \quad (2.6)$$

where  $k = \omega/c$ .

To obtain the solution of eqs. (2.5) and (2.6), we introduce a tensor Green function  $\mathbf{G}(\mathbf{r}|\mathbf{r}_0; k)$ , which is a solution of the equation

$$-\text{curl curl } \mathbf{G}(\mathbf{r}|\mathbf{r}_0; k) + k^2 \mathbf{G}(\mathbf{r}|\mathbf{r}_0; k) = -\delta(\mathbf{r} - \mathbf{r}_0) \mathbf{U}, \quad (2.7)$$

and obeys the Silver–Müller radiation conditions<sup>13</sup>)

$$\lim_{r \rightarrow \infty} [\mathbf{r} \times (\text{curl} + ik\mathbf{r}) \mathbf{G}(\mathbf{r}|\mathbf{r}_0; k) = 0, \quad (2.8)$$

and

$$|r\mathbf{G}(\mathbf{r}|\mathbf{r}_0; k)| \quad \text{bounded as} \quad r \rightarrow \infty, \quad (2.9)$$

uniformly with respect to the direction of  $\mathbf{r}$ . In eq. (2.7)  $\mathbf{U}$  denotes the unit tensor.



The radiation conditions (2.8) and (2.9) are the mathematical formulation of the physical assumption, that at great distances from the material sources the electromagnetic field generated by these sources represents divergent travelling waves.

The Green function in eq. (2.7) is given by

$$\mathbf{G}(\mathbf{r}|\mathbf{r}_0; k) = g(\mathbf{r}|\mathbf{r}_0; k) \mathbf{U} + \frac{1}{k^2} \text{grad grad } g(\mathbf{r}|\mathbf{r}_0; k), \quad (2.10)$$

with

$$g(\mathbf{r}|\mathbf{r}_0; k) = e^{ik|\mathbf{r}-\mathbf{r}_0|/4\pi|\mathbf{r}-\mathbf{r}_0|}. \quad (2.11)$$

The electromagnetic field for a point  $\mathbf{r}_0$  inside  $S$  is now given by

$$\begin{aligned} \mathbf{E}(\mathbf{r}_0\omega) &= \frac{i\omega}{c^2} \int_{\Omega} \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \cdot \mathbf{I}(\mathbf{r}\omega) \, dv \\ &\quad - \int_S \{ [\hat{\mathbf{n}}(\mathbf{r}) \times \text{curl } \mathbf{E}(\mathbf{r}\omega)] \cdot \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \\ &\quad - [\text{curl } \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k)] \cdot [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{E}(\mathbf{r}\omega)] \} \, d\sigma, \end{aligned} \quad (2.12)$$

and

$$\begin{aligned} \mathbf{B}(\mathbf{r}_0\omega) &= \frac{1}{c} \int_{\Omega} \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \cdot \text{curl } \mathbf{I}(\mathbf{r}\omega) \, dv \\ &\quad - \int_S \{ [\hat{\mathbf{n}}(\mathbf{r}) \times \text{curl } \mathbf{B}(\mathbf{r}\omega)] \cdot \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \\ &\quad - [\text{curl } \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k)] \cdot [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{B}(\mathbf{r}\omega)] \} \, d\sigma, \end{aligned} \quad (2.13)$$

in which  $\hat{\mathbf{n}}(\mathbf{r})$  is the outward normal on the surface  $S$ . The volume integrals in eqs. (2.12) and (2.13) give the contribution to the electromagnetic field from the sources inside  $S$ ; the surface integrals over  $S$  represent the contribution from all sources located outside  $S$ . If  $S$  recedes to infinity, the volume integrals in eqs. (2.12) and (2.13) represent the electromagnetic field generated by all the sources at finite distances which are the sources of the material system, and the surface integrals in eqs. (2.12) and (2.13) represent the external electromagnetic field  $\mathbf{E}^e(\mathbf{r}_0\omega)$  and  $\mathbf{B}^e(\mathbf{r}_0\omega)$ , generated by sources at infinity. In that case we can write eqs. (2.12) and (2.13)

$$\mathbf{E}(\mathbf{r}_0\omega) = \mathbf{E}^e(\mathbf{r}_0\omega) + \frac{i\omega}{c^2} \int_{V^+} \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \cdot \mathbf{I}(\mathbf{r}\omega) \, dv, \quad (2.14)$$

and

$$\mathbf{B}(\mathbf{r}_0\omega) = \mathbf{B}^e(\mathbf{r}_0\omega) + \frac{1}{c} \int_{V^+} \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \cdot \text{curl } \mathbf{I}(\mathbf{r}\omega), \quad (2.15)$$

where the integrals are restricted to the volume  $V^+$  bounded by  $\Sigma^+$ .

3. A formal "extinction theorem". In eqs. (2.12) and (2.13)  $S$  is an arbitrary regular surface. We may choose for this surface the surface  $\Sigma^-$ . The sources outside  $\Sigma^-$  are then sources at infinity, needed to generate the external electromagnetic fields  $\mathbf{E}^e(\mathbf{r}_0\omega)$ ,  $\mathbf{B}^e(\mathbf{r}_0\omega)$  and the sources located in the transition layer between  $\Sigma^-$  and  $\Sigma^+$  which belong to the material system. We obtain from eqs. (2.12) and (2.13), using eqs. (2.14) and (2.15) the relations

$$\begin{aligned} \mathbf{E}^e(\mathbf{r}_0\omega) - \int_{\Sigma^-} \left\{ [\text{curl } \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k)] \cdot [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{E}(\mathbf{r}\omega)] \right. \\ \left. - \frac{i\omega}{c} \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \cdot [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{B}(\mathbf{r}\omega)] \right\} d\sigma \\ + \frac{i\omega}{c^2} \int_{\Sigma^-}^{\Sigma^+} \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \cdot \mathbf{I}(\mathbf{r}\omega) dv = 0, \end{aligned} \quad (3.1)$$

and

$$\begin{aligned} \mathbf{B}^e(\mathbf{r}_0\omega) - \int_{\Sigma^-} \left\{ [\text{curl } \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k)] \cdot [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{B}(\mathbf{r}\omega)] \right. \\ \left. + \frac{i\omega}{c} \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k) \cdot [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{E}(\mathbf{r}\omega)] \right\} d\sigma \\ - \frac{1}{c} \int_{\Sigma^-}^{\Sigma^+} [\text{curl } \mathbf{G}(\mathbf{r}_0|\mathbf{r}; k)] \cdot \mathbf{I}(\mathbf{r}\omega) dv = 0, \end{aligned} \quad (3.2)$$

where we have also used the Maxwell equation (2.1) and the relations (A.9) and (A.10), derived in appendix A.

The two relations (3.1) and (3.2) which are straightforward consequences of the Maxwell equations may be interpreted as formal extinction theorems: they demonstrate indeed that the external fields at a point inside the material system are equal to a field generated by the fields (the tangential components of  $\mathbf{E}$  and  $\mathbf{B}$ ) at the surface  $\Sigma^-$ , as well as by the current density in the transition layer. The reason that we have expressed in the above relations the external fields in terms of surface integrals over  $\Sigma^-$  rather than  $\Sigma^+$ , is that we can now relate the true fields, set up at the surface, to the current density at the surface by means of the constitutive equations for the medium. This will enable us to obtain the true extinction theorem in the next section†. We could not have achieved this by choosing for our

† It should be kept in mind that the relations (3.1) and (3.2) also hold, *e.g.*, if there is only a single point charge (or no charge at all) within the boundary  $\Sigma^-$ . We wish to reserve the terminology "true extinction theorem" to the case where the electromagnetic field  $\mathbf{E}(\mathbf{r}\omega)$ ,  $\mathbf{B}(\mathbf{r}\omega)$  satisfies within the medium a wave equation with a phase velocity different from the phase velocity *in vacuo*, so that it becomes meaningful to interpret relations (3.1) and (3.2) as describing the cancellation ("Auslöschung") of the incident wave.

surface the surface  $\Sigma^+$  where the charge and current densities have fallen off to zero. Our procedure will be to finally let the surfaces  $\Sigma^-$  and  $\Sigma^+$  approach each other to form the surface  $\Sigma$ , thus going over to a real discontinuity. In this limit the contribution from the boundary layer will either disappear or approach a finite value depending on whether or not surface currents result from the discontinuities in the material parameters. It will thus turn out that the transition layer may give rise to additional contributions to the classical Ewald-Oseen extinction theorem.

Before we carry out the program sketched above, we shall first transform the relations (3.1) and (3.2) by means of the Maxwell equations and vector identities and cast them into a form more suited for our purpose.

Indeed, using the definition (2.10) of  $G(\mathbf{r}_0|\mathbf{r};k)$  and Maxwell's equations (2.1)–(2.4) we obtain from (3.1) and (3.2) the alternative forms

$$\begin{aligned} E^e(\mathbf{r}_0\omega) + \int_{\Sigma^-} \{ \mathbf{E}(\mathbf{r}\omega) \hat{\mathbf{n}}(\mathbf{r}) \cdot \text{grad } g(\mathbf{r}_0|\mathbf{r};k) \\ - g(\mathbf{r}_0|\mathbf{r};k) [\hat{\mathbf{n}}(\mathbf{r}) \cdot \text{grad } \mathbf{E}(\mathbf{r}\omega) - \hat{\mathbf{n}}(\mathbf{r}) \text{div } \mathbf{E}(\mathbf{r}\omega)] \} d\sigma \\ + \frac{i\omega}{c^2} \int_{\Sigma^-}^{\Sigma^+} g(\mathbf{r}_0|\mathbf{r};k) \mathbf{I}(\mathbf{r}\omega) dv + \int_{\Sigma^-}^{\Sigma^+} [\text{grad } g(\mathbf{r}_0|\mathbf{r};k)] \rho(\mathbf{r}\omega) dv = 0, \end{aligned} \quad (3.3)$$

and

$$\begin{aligned} B^e(\mathbf{r}_0\omega) + \int_{\Sigma^-} \left\{ [\text{grad } g(\mathbf{r}_0|\mathbf{r};k)] \mathbf{B}(\mathbf{r}\omega) \cdot \hat{\mathbf{n}}(\mathbf{r}) \right. \\ \left. + [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{B}(\mathbf{r}\omega)] \times \text{grad } g(\mathbf{r}_0|\mathbf{r};k) \right. \\ \left. - \frac{i\omega}{c} g(\mathbf{r}_0|\mathbf{r};k) [\hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{E}(\mathbf{r}\omega)] \right\} d\sigma \\ - \frac{1}{c} \int_{\Sigma^-}^{\Sigma^+} [\text{grad } g(\mathbf{r}_0|\mathbf{r};k)] \times \mathbf{I}(\mathbf{r}\omega) dv = 0. \end{aligned} \quad (3.4)$$

The necessary transformations are given in appendix A. The relations (3.3) and (3.4) are particularly well suited for the discussion of static problems.

Next, since the field  $\mathbf{E}^e(\mathbf{r}\omega)$  satisfies the source-free equation

$$-\text{curl curl } \mathbf{E}^e(\mathbf{r}\omega) + k^2 \mathbf{E}^e(\mathbf{r}\omega) = 0, \quad (3.5)$$

we obtain from relation (3.3)

$$E^e(\mathbf{r}_0\omega) + \frac{1}{k^2} \text{curl}^0 \text{curl}^0 \int_{\Sigma^-} \{ \mathbf{E}(\mathbf{r}\omega) \hat{\mathbf{n}}(\mathbf{r}) \cdot \text{grad } g(\mathbf{r}_0|\mathbf{r};k)$$



$$\begin{aligned}
 & -g(\mathbf{r}_0|\mathbf{r};k)[\hat{\mathbf{n}}(\mathbf{r})\cdot\text{grad } \mathbf{E}(\mathbf{r}\omega) - \hat{\mathbf{n}}(\mathbf{r}) \text{div } \mathbf{E}(\mathbf{r}\omega)]\} d\sigma \\
 & + \frac{i}{\omega} \text{curl}^0 \text{curl}^0 \int_{\Sigma^-}^{\Sigma^+} g(\mathbf{r}_0|\mathbf{r};k) \mathbf{I}(\mathbf{r}\omega) dv = 0, \tag{3.6}
 \end{aligned}$$

where the operation curl is performed with respect to  $\mathbf{r}_0$ . This last form of our formal extinction theorem will be used for the derivation of the true extinction theorem of the Ewald-Oseen type for the incident electric field.

Finally we let  $\Sigma^-$  and  $\Sigma^+$  approach the material surface  $\Sigma$ . Then, e.g., formula (3.6) gets the form †

$$\begin{aligned}
 & \mathbf{E}^e(\mathbf{r}_0\omega) + \lim_{\Sigma^- \rightarrow \Sigma} \frac{1}{k^2} \text{curl}^0 \text{curl}^0 \int_{\Sigma^-} \{ \mathbf{E}(\mathbf{r}\omega) \hat{\mathbf{n}}(\mathbf{r}) \cdot \text{grad } g(\mathbf{r}_0|\mathbf{r};k) \\
 & - g(\mathbf{r}_0|\mathbf{r};k)[\hat{\mathbf{n}}(\mathbf{r})\cdot\text{grad } \mathbf{E}(\mathbf{r}\omega) - \hat{\mathbf{n}}(\mathbf{r}) \text{div } \mathbf{E}(\mathbf{r}\omega)]\} d\sigma \\
 & + \frac{i}{\omega} \text{curl}^0 \text{curl}^0 \int_{\Sigma} g(\mathbf{r}_0|\mathbf{r};k) \mathbf{I}_s(\mathbf{r}\omega) d\sigma = 0, \tag{3.7}
 \end{aligned}$$

where the surface current density  $\mathbf{I}_s$  is defined through<sup>14) †</sup>

$$\mathbf{I}(\mathbf{r}\omega) = U(-F) \mathbf{i}(\mathbf{r}\omega) + \mathbf{I}_s(\mathbf{r}\omega) |\text{grad } F| \delta(F). \tag{3.8}$$

† Eq. (3.7) is identical with the following equation

$$\begin{aligned}
 & \mathbf{E}^e + \text{curl}^0 \text{curl}^0 \int_{\Sigma^-} [\mathbf{Z}\mathbf{n} \cdot \text{grad } g - g\mathbf{n} \cdot \text{grad } \mathbf{Z}] d\sigma \\
 & + \frac{i}{\omega} \text{curl}^0 \text{curl}^0 \int_{\Sigma} g \mathbf{I}_s d\sigma, \tag{3.7a}
 \end{aligned}$$

where  $\mathbf{Z}$  is the Hertz vector. The transformations leading from (3.7) to the above equation are somewhat lengthy but otherwise straightforward. Eq. (3.7a) without the term involving the surface current is precisely Sein's form of the formal extinction theorem which he obtained assuming the charge and current densities to be non-singular. However, as stated before, the last term may in general not be neglected since a surface current may exist as a consequence of discontinuities in the material constants (cf. section 4). Moreover, (3.7a) is not the most convenient form for a formal extinction theorem, since for a specific medium the Hertz vector cannot easily be directly eliminated in favour of the charge and current densities at  $\Sigma^-$ .

‡ Eq. (3.8) has to be interpreted as the limiting form of the current density  $\mathbf{I}(\mathbf{r}\omega)$  when the material layer between  $\Sigma^-$  and  $\Sigma^+$  is squeezed to form a real discontinuity. Thus  $\mathbf{I}_s$  is also defined through the limiting process.

$$\mathbf{I}_s = \lim_{\Delta F \rightarrow 0} \frac{\Delta F \mathbf{I}(\Delta F)}{|\text{grad } F|},$$

where  $\Delta F$  is the difference in value of the function  $F$  on the surfaces  $\Sigma^+$  and  $\Sigma^-$ . The notation  $\mathbf{I}(\Delta F)$  indicates that the value of the current density  $\mathbf{I}$  in the boundary layer will depend on the width of this layer. As stated before the existence of a finite value for  $\mathbf{I}_s$  is related to discontinuities occurring in the material constants of the medium (cf. section 4).

Here  $F(\mathbf{r}) = 0$  defines the surface  $\Sigma$ , such that  $F(\mathbf{r}) > 0$  refers to points outside  $\Sigma$  and  $F(\mathbf{r}) < 0$  to points inside  $\Sigma$ .

$U(F)$  is the Heaviside unit function defined as

$$U(F) = \begin{cases} 0 & \text{for } F(\mathbf{r}) < 0, \\ 1 & \text{for } F(\mathbf{r}) > 0, \end{cases} \quad (3.9)$$

and

$$\delta(F) = \frac{dU(F)}{dF}. \quad (3.10)$$

The quantity  $\mathbf{i}(\mathbf{r}\omega)$  represents the (finite) volume or bulk-current density. In the following sections  $\Sigma^-$  will always denote the limit as the inner surface approaches the boundary  $\Sigma$ .

Formula (3.3) will lead in the limit to an expression containing also a surface-charge density defined in an analogous way.

4. *The extinction theorem for linear media and non-static fields.* In this section we shall derive the extinction theorem for linear, homogeneous, isotropic systems and non-static fields. To achieve this we have to relate the electromagnetic field at the surface to the current density at the surface by means of the material equations for the media. We write the induced current density  $\mathbf{I}(\mathbf{r}\omega)$  as a sum of three terms, *viz.*

$$\mathbf{I}(\mathbf{r}\omega) = \mathbf{I}_c(\mathbf{r}\omega) - i\omega\mathbf{P}(\mathbf{r}\omega) + c \operatorname{curl} \mathbf{M}(\mathbf{r}\omega). \quad (4.1)$$

Here  $\mathbf{I}_c(\mathbf{r}\omega)$  is the current density of the free charges,  $\mathbf{P}(\mathbf{r}\omega)$  the polarization of the bound charges and  $\mathbf{M}(\mathbf{r}\omega)$  the magnetization of the bound charges. The usual way of making this separation is by considering a special model of the material. If one wishes to perform this separation independent of a specific model, additional physical assumptions are required<sup>15</sup>.

We assume that the following constitutive relations hold:

$$\mathbf{I}_c(\mathbf{r}\omega) = [-\Lambda(\omega)/i\omega + \sigma(\omega)] U(-F) \mathbf{E}^-(\mathbf{r}\omega), \quad (4.2)$$

$$\mathbf{P}(\mathbf{r}\omega) = [\varepsilon(\omega) - 1] U(-F) \mathbf{E}^-(\mathbf{r}\omega) + iv_1(\omega) U(-F) \mathbf{B}^-(\mathbf{r}\omega), \quad (4.3)$$

and

$$\mathbf{M}(\mathbf{r}\omega) = -iv_2(\omega) U(-F) \mathbf{E}^-(\mathbf{r}\omega) + [1 - \mu(\omega)^{-1}] U(-F) \mathbf{B}^-(\mathbf{r}\omega), \quad (4.4)$$

where  $\Lambda$  is the parameter for superconductivity,  $\sigma$  is the conductivity,  $\varepsilon$  the dielectric constant,  $v_1$  and  $v_2$  are parameters for optical activity and  $\mu$  is the magnetic permeability. The function  $U(-F)$  has been introduced into eqs. (4.2)–(4.4) as a formal way to indicate that the material constants have a discontinuity at the boundary of the system. In eqs. (4.2)–(4.4) the

functions  $\mathbf{E}^-(\mathbf{r}\omega)$  and  $\mathbf{B}^-(\mathbf{r}\omega)$  are such that

$$\left. \begin{aligned} \mathbf{E}(\mathbf{r}\omega) &= \mathbf{E}^-(\mathbf{r}\omega) \\ \mathbf{B}(\mathbf{r}\omega) &= \mathbf{B}^-(\mathbf{r}\omega) \end{aligned} \right\} \quad \text{when} \quad F(\mathbf{r}) < 0. \quad (4.5)$$

By means of the eqs. (4.2)–(4.4)  $\mathbf{i}(\mathbf{r}\omega)$  and  $\mathbf{I}_s(\mathbf{r}\omega)$  in eq. (3.8) become

$$\begin{aligned} \mathbf{i}(\mathbf{r}\omega) &= -i\omega[\varepsilon_{\text{eff}}(\omega) - 1] \mathbf{E}^-(\mathbf{r}\omega) \\ &\quad + \omega[v_1(\omega) + v_2(\omega)] \mathbf{B}^-(\mathbf{r}\omega) + c[1 - \mu(\omega)^{-1}] \text{curl } \mathbf{B}^-(\mathbf{r}\omega), \end{aligned} \quad (4.6)$$

and

$$\begin{aligned} \mathbf{I}_s(\mathbf{r}\omega) &= icv_2(\omega) \frac{\text{grad } F(\mathbf{r})}{|\text{grad } F(\mathbf{r})|} \times \mathbf{E}^-(\mathbf{r}\omega) \\ &\quad - c[1 - \mu(\omega)^{-1}] \frac{\text{grad } F(\mathbf{r})}{|\text{grad } F(\mathbf{r})|} \times \mathbf{B}^-(\mathbf{r}\omega). \end{aligned} \quad (4.7)$$

In eq. (4.6) we have used the abbreviation

$$\varepsilon_{\text{eff}}(\omega) = -\frac{A(\omega)}{\omega^2} + \frac{i}{\omega} \sigma(\omega) + \varepsilon(\omega). \quad (4.8)$$

The normal  $\hat{\mathbf{n}}(\mathbf{r})$ , pointing from the region  $F < 0$  into the region  $F > 0$ , is given by

$$\hat{\mathbf{n}}(\mathbf{r}) = \text{grad } F / |\text{grad } F|. \quad (4.9)$$

Upon substitution into eq. (4.7) we obtain for the surface-current density†

$$\mathbf{I}_s(\mathbf{r}\omega) = icv_2(\omega) \hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{E}^-(\mathbf{r}\omega) - c[1 - \mu(\omega)^{-1}] \hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{B}^-(\mathbf{r}\omega). \quad (4.10)$$

In accordance with the statement made in section 3, the surface-current density results from discontinuities in the material parameters, *i.e.* formally from the occurrence of the function  $U(-F)$  in eqs. (4.2)–(4.4).

Note that only the material parameters of the magnetization  $\mathbf{M}(\mathbf{r}\omega)$  appear in the formula for the surface-current density‡. Eq. (4.6) may be considered as a volume-material equation and eq. (4.10) as a surface-material equation. Using eqs. (2.1) and (2.5) to eliminate the last term on the right-hand side of eq. (4.6), the latter can be put into the form

$$\mathbf{i}(\mathbf{r}\omega) = -i\omega\alpha(\omega) \mathbf{E}^-(\mathbf{r}\omega) - \frac{ic^2}{\omega} \beta(\omega) \text{curl } \mathbf{E}^-(\mathbf{r}\omega), \quad (4.11)$$

† Usually the surface-current density is defined by  $(1/c)\mathbf{I}_s = \hat{\mathbf{n}} \times [\mathbf{B}^+ - \mathbf{B}^-]$  where  $\mathbf{B}^+$  is the magnetic field just outside the medium. Using the relation  $\mathbf{B} = \mathbf{H} + \mathbf{M}$  and the condition that the tangential component of  $\mathbf{H}$  is continuous, one then arrives also at a surface-current density as given by eq. (4.10).

‡ The material parameters occurring in eqs. (4.2) and (4.3) for the current  $\mathbf{I}_e$  and the polarization  $\mathbf{P}$  give rise to a surface-charge density.



in which

$$\alpha(\omega) = \epsilon_{\text{eff}}(\omega) \mu(\omega) - 1, \quad (4.12)$$

and

$$\beta(\omega) = k[v_1(\omega) + v_2(\omega)] \mu(\omega). \quad (4.13)$$

The electromagnetic properties inside the medium may also be characterized by two indices of refraction  $n_+(\omega)$  and  $n_-(\omega)$ , determining the phase velocity for right-handed and left-handed circularly polarized waves respectively. We show in appendix B that  $n_+(\omega)$  and  $n_-(\omega)$  are related to the parameters  $\alpha(\omega)$  and  $\beta(\omega)$  in the following way

$$n_+ n_- - 1 = \alpha, \quad (4.14)$$

and

$$\Delta n = n_+ - n_- = \beta/k. \quad (4.15)$$

Eq. (4.11) may now be inverted (*cf.* appendix B), using Maxwell's equations and also the relations (4.14) and (4.15) to yield

$$\mathbf{E}^-(\mathbf{r}\omega) = \frac{i}{\omega} f_1(n_+, n_-) \mathbf{i}(\mathbf{r}\omega) - \frac{ic}{\omega^2} f_2(n_+, n_-) \text{curl } \mathbf{i}(\mathbf{r}\omega), \quad (4.16)$$

in which the functions  $f_1$  and  $f_2$  are given by

$$f_1(n_+, n_-) = \frac{(n_+ n_- - 1) + \Delta n^2}{(n_+ n_- - 1)^2 - \Delta n^2}, \quad (4.17)$$

and

$$f_2(n_+, n_-) = \frac{\Delta n}{(n_+ n_- - 1)^2 - \Delta n^2}. \quad (4.18)$$

The extinction theorem of the Ewald-Oseen type is now obtained, *e.g.* for the electric field, by substituting eq. (4.16) into the formal extinction theorem (3.7). We then obtain

$$\begin{aligned} \mathbf{E}^e + \frac{1}{k^2} \text{curl}^0 \text{curl}^0 \int_{\Sigma^-} \left\{ \left[ \frac{i}{\omega} f_1 \mathbf{i} - i \frac{c}{\omega^2} f_2 \text{curl } \mathbf{i} \right] \hat{\mathbf{n}} \cdot \text{grad } g \right. \\ \left. - g \hat{\mathbf{n}} \cdot \text{grad} \left[ \frac{i}{\omega} f_1 \mathbf{i} - i \frac{c}{\omega^2} f_2 \text{curl } \mathbf{i} \right] - \frac{i}{\omega} g \hat{\mathbf{n}} \text{div } \mathbf{i} \right\} d\sigma \\ + \frac{i}{\omega} \text{curl}^0 \text{curl}^0 \int_{\Sigma} g \mathbf{I}_s d\sigma = 0, \end{aligned} \quad (4.19)$$

where we have also used the continuity equation (inside the medium)

$$-i\omega\rho(\mathbf{r}\omega) + \text{div } \mathbf{i}(\mathbf{r}\omega) = 0. \quad (4.20)$$

At this point we should like to make the following remarks. Firstly, one may not disregard *a priori* the possibility that an external (transverse) electromagnetic field even in the case of a homogeneous (but finite) system, can give rise to an electric field inside the medium, which has also a longitudinal component. According to the continuity equation (4.20) and eq. (4.11) this can only happen if

$$\alpha(\omega) = -1. \quad (4.21)$$

Excluding this possibility, the term with  $\text{div } \mathbf{i}$  in the integrand of eq. (4.19) vanishes. Secondly, we note that the extinction theorem can be expressed entirely in terms of the induced volume-current density at  $\Sigma^-$ , the indices of refraction and the material parameters appearing in the magnetization if the surface-current density is eliminated by means of eq. (4.10) and subsequently the fields  $\mathbf{E}^-$  and  $\mathbf{B}^- = c(i\omega)^{-1} \text{curl } \mathbf{E}^-$  using eq. (4.16). We illustrate this for a non-gyrotropic ( $v_1 = v_2 = 0$ ) system for which also  $\mathcal{A} = 0$ ,  $\sigma = 0$ . In this case eq. (4.10) reduces to

$$\mathbf{I}_s(\mathbf{r}\omega) = -c[1 - \mu(\omega)^{-1}] \hat{\mathbf{n}}(\mathbf{r}) \times \mathbf{B}^-(\mathbf{r}\omega), \quad (4.22)$$

and eq. (4.16) to

$$\mathbf{E}^-(\mathbf{r}\omega) = \frac{\mathbf{i}}{\omega} \frac{1}{n^2 - 1} \mathbf{i}(\mathbf{r}\omega), \quad (4.23)$$

so that, using also eq. (2.1)

$$\mathbf{B}^-(\mathbf{r}\omega) = \frac{c}{\omega^2} \frac{1}{(n^2 - 1)} \text{curl } \mathbf{i}(\mathbf{r}\omega). \quad (4.24)$$

Applying eqs. (4.22) and (4.24) to the extinction theorem (4.19) we obtain

$$\begin{aligned} E^e + \frac{\mathbf{i}}{\omega} \frac{1}{k^2(n^2 - 1)} \text{curl}^0 \text{curl}^0 \int_{\Sigma^-} [\mathbf{i} \hat{\mathbf{n}} \cdot \text{grad } g \\ - g \hat{\mathbf{n}} \cdot \text{grad } \mathbf{i} - g(1 - \mu^{-1}) \hat{\mathbf{n}} \times \text{curl } \mathbf{i}] d\sigma = 0. \end{aligned} \quad (4.25)$$

The last contribution to the surface integral arises entirely as a consequence of the singular current density at the surface.

If we now specialize to a non-magnetic ( $\mu = 1$ ) system, eq. (4.25) reduces to the usual statement of the Ewald-Oseen extinction theorem

$$\begin{aligned} E^e(\mathbf{r}_0\omega) + \frac{1}{k^2(n^2 - 1)} \text{curl}^0 \text{curl}^0 \int_{\Sigma^-} [\mathbf{P}(\mathbf{r}\omega) \hat{\mathbf{n}}(\mathbf{r}) \cdot \text{grad } g(\mathbf{r}_0|\mathbf{r}; k) \\ - g(\mathbf{r}_0|\mathbf{r}; k) \hat{\mathbf{n}}(\mathbf{r}) \cdot \text{grad } \mathbf{P}(\mathbf{r}\omega)] d\sigma = 0, \end{aligned} \quad (4.26)$$

since then  $\mathbf{I}_s = 0$ ,  $\mathbf{i} = \mathbf{I} = -i\omega\mathbf{P}$  in view of eqs. (4.1) and (4.22).

5. *Conclusions.* The main conclusion to be drawn from the present analysis is the following. The extinction theorem was derived originally within the framework of a theory for the index of refraction. The emphasis in such a treatment was on the calculation of the refractive index in terms of molecular quantities. The extinction theorem was obtained as a by-product. Since, as we have shown in agreement with the ideas put forward by Sein, the extinction theorem is already contained in Maxwell's equations and material equations for a specific medium, we may infer that in the traditional theories this theorem should rather be interpreted as a consistency check with Maxwell's equations. This also implies that theories of the index of refraction, in which the extinction theorem is not manifestly derived, but in which Maxwell's equations are satisfied, are equally satisfactory, since they will contain the extinction theorem implicitly†. A theory of the latter type is the linear response theory in statistical mechanics which yields susceptibilities containing true absorption in contradiction with theories based on (1.1).

Note furthermore the importance of contributions from surface currents to the extinction theorem. The existence of such terms demonstrates that the separation of the total current into a polarization current and a magnetization current is not completely arbitrary any more. While in bulk *e.g.* the magnetic properties may be completely accounted for by redefining the dielectric constant [*e.g.* eqs. (4.11) and (4.12)], this is not true for the surface current which contains the magnetic permeability  $\mu$  separately [*e.g.* eq. (4.10)]. This is the reason why in the extinction theorem (4.25), there occurs not only the index of refraction of the system, but also again the permeability  $\mu$ . Thus again, while for optically active systems the bulk current (and the rotatory power) contains only the sum  $v_1 + v_2$ , the surface current (and the extinction theorem) contains  $v_2$  characterizing the magnetization separately<sup>17</sup>). Finally, it may be mentioned that the formal extinction theorems, (3.3) and (3.4), can also be used to discuss static problems.

In particular one may think of a perfect diamagnet, characterized by  $\mu = 0$  and therefore  $\mathbf{B} = 0$  inside the medium. Eq. (3.4) then yields the simple result, using also the fact that  $\mathbf{I}_s = c\hat{\mathbf{n}} \times \{\mathbf{B}^+ - \mathbf{B}^-\} = c\hat{\mathbf{n}} \times \mathbf{H}^+$  is finite and non-vanishing,

$$\mathbf{B}^e(\mathbf{r}_0, 0) - \frac{1}{c} \int_{\Sigma} [\text{grad } g(\mathbf{r}_0 | \mathbf{r}; 0)] \times \mathbf{I}_s(\mathbf{r}, 0) d\sigma = 0, \quad (5.1)$$

which illustrates the "extinction" of the applied magnetic field by the surface current in the Meissner effect. Alternatively, we could have arrived

† This point does not seem to be appreciated in general<sup>10</sup>).



at a formula with the same physical content, using London's equation for a superconductor [cf. eq. (4.2)].

$$\mathbf{I}(\mathbf{r}\omega) = -\frac{\Lambda(\omega)}{i\omega} U(-F) \mathbf{E}^-(\mathbf{r}\omega), \quad (5.2)$$

which leads to

$$\text{curl } \mathbf{i}(\mathbf{r}\omega) = -\frac{\Lambda(\omega)}{c} \mathbf{B}^-(\mathbf{r}\omega), \quad \mathbf{I}_s = 0. \quad (5.3)$$

Eliminating  $\mathbf{B}$  from eq. (3.4) by means of eq. (5.3) gives

$$\mathbf{B}^e - \frac{c}{\Lambda} \int_{\Sigma^-} [(\text{curl } \mathbf{i}) \cdot \hat{\mathbf{n}} \text{ grad } g + (\hat{\mathbf{n}} \times \text{curl } \mathbf{i}) \times \text{grad } g] d\sigma = 0, \quad (5.4)$$

expressing the fact that from this point of view  $\mathbf{B}^e$  is cancelled by the finite current density at the surface.

The simple static problem considered here clearly demonstrates again the connection between the boundary-value problem and the "extinction" theorem.

#### APPENDIX A

In this appendix we give the transformation formulae we need to obtain the alternative forms of the formal extinction theorems (3.3) and (3.4) from the identities (3.1) and (3.2). To this end we use a version of Green's theorem<sup>12</sup> applied to  $\mathbf{E}(\mathbf{r}\omega)$  and  $g(\mathbf{r}_0|\mathbf{r}; k)$ , viz.

$$\begin{aligned} \int_{V^-} (\mathbf{E}\nabla^2 g - g\nabla^2 \mathbf{E}) dv &= \int_{\Sigma^-} [(\text{grad } g) \mathbf{E} \cdot \hat{\mathbf{n}} - g \hat{\mathbf{n}} \text{ div } \mathbf{E}] d\sigma \\ &\quad - \int_{\Sigma^-} [(\text{grad } g) \times (\hat{\mathbf{n}} \times \mathbf{E}) - g \hat{\mathbf{n}} \times \text{curl } \mathbf{E}] d\sigma, \end{aligned} \quad (\text{A.1})$$

where  $V^-$  is the volume bounded by  $\Sigma^-$ .

Using the fact that

$$\text{curl } g(\mathbf{r}_0|\mathbf{r}; k) \mathbf{U} = \text{curl } G(\mathbf{r}_0|\mathbf{r}; k), \quad (\text{A.2})$$

and writing

$$\int_{V^-} (\mathbf{E}\nabla^2 g - g\nabla^2 \mathbf{E}) dv = \int_{\Sigma^-} [\mathbf{E} \hat{\mathbf{n}} \cdot \text{grad } g - g \hat{\mathbf{n}} \cdot \text{grad } \mathbf{E}] d\sigma, \quad (\text{A.3})$$

according to the usual form of Green's theorem, we obtain from eq. (A.1) the relation

$$\begin{aligned} & \int_{\Sigma^-} [(\text{curl } \mathbf{G}) \cdot (\hat{\mathbf{n}} \times \mathbf{E}) - \mathbf{G} \cdot (\hat{\mathbf{n}} \times \text{curl } \mathbf{E})] d\sigma \\ &= - \int_{\Sigma^-} [\mathbf{E} \hat{\mathbf{n}} \cdot \text{grad } g - g \hat{\mathbf{n}} \cdot \text{grad } \mathbf{E} \\ & \quad - (\text{grad } g) \mathbf{E} \cdot \hat{\mathbf{n}} + g \hat{\mathbf{n}} \text{div } \mathbf{E} + L \cdot (\hat{\mathbf{n}} \times \text{curl } \mathbf{E})] d\sigma, \end{aligned} \quad (\text{A.4})$$

in which

$$L = (1/k^2) \text{grad grad } g. \quad (\text{A.5})$$

The volume integral in eq. (3.1) can be written, using the continuity equation

$$-i\omega\rho + \text{div } \mathbf{I} = 0, \quad (\text{A.6})$$

and Gauss's theorem as

$$\begin{aligned} & \frac{i\omega}{c^2} \int_{\Sigma^-}^{\Sigma^+} \mathbf{G} \cdot \mathbf{I} dv \\ &= \frac{i\omega}{c^2} \int_{\Sigma^-}^{\Sigma^+} g \mathbf{I} dv - \text{grad}^0 \int_{\Sigma^-}^{\Sigma^+} g \rho dv + \frac{i}{\omega} \text{grad}^0 \int_{\Sigma^-} g \mathbf{I} \cdot \hat{\mathbf{n}} d\sigma. \end{aligned} \quad (\text{A.7})$$

Making use of eq. (2.5), Stokes's theorem and the fact that  $\Sigma^-$  is a closed surface, it is easily shown that

$$\begin{aligned} & \frac{i}{\omega} \text{grad}^0 \int_{\Sigma^-} g \mathbf{I} \cdot \hat{\mathbf{n}} d\sigma + \text{grad}^0 \int_{\Sigma^-} g \mathbf{E} \cdot \hat{\mathbf{n}} d\sigma \\ & \quad + \frac{1}{k^2} \int_{\Sigma^-} (\hat{\mathbf{n}} \times \text{curl } \mathbf{E}) \cdot \text{grad grad } g = 0. \end{aligned} \quad (\text{A.8})$$

Eqs. (A.4), (A.7) and (A.8), together with eq. (2.1) have been applied in the derivation of eq. (3.3).

The Maxwell equation (2.2) yields the relation

$$\begin{aligned} & \int_{\Sigma^-} (\hat{\mathbf{n}} \times \text{curl } \mathbf{B}) \cdot \mathbf{G} d\sigma \\ &= \int_{\Sigma^-} \left[ -\frac{i\omega}{c} (\hat{\mathbf{n}} \times \mathbf{E}) \cdot \mathbf{G} + \frac{1}{c} (\hat{\mathbf{n}} \times \mathbf{I}) \cdot \mathbf{G} \right] d\sigma. \end{aligned} \quad (\text{A.9})$$

In the derivation of eq. (3.2) the relation (A.9) and the identity

$$\begin{aligned} & \frac{1}{c} \int_{\Sigma^-}^{\Sigma^+} \mathbf{G} \cdot \text{curl } \mathbf{I} \, dv \\ &= -\frac{1}{c} \int_{\Sigma^-} \mathbf{G} \times \mathbf{I} \cdot \hat{\mathbf{n}} \, d\sigma - \frac{1}{c} \int_{\Sigma^+} (\text{curl } \mathbf{G}) \cdot \mathbf{I} \, d\sigma. \end{aligned} \quad (\text{A.10})$$

have been used. Again, using Stokes's theorem and the fact that  $\Sigma^-$  is a closed surface, one finds after straight-forward calculations

$$\frac{i\omega}{c} \int_{\Sigma^-} (\hat{\mathbf{n}} \times \mathbf{E}) \cdot \mathbf{I} \, d\sigma = \text{grad}^0 \int_{\Sigma^-} g \mathbf{B} \cdot \hat{\mathbf{n}} \, d\sigma. \quad (\text{A.11})$$

Upon substitution of relation (A.11) into eq. (3.2) we obtain, noting also that, e.g.,

$$(\text{curl } \mathbf{G}) \cdot \mathbf{I} = (\text{grad } g) \times \mathbf{I}, \quad (\text{A.12})$$

the formal extinction theorem (3.4).

#### APPENDIX B

For the sake of completeness we derive in this appendix the relations (4.14) and (4.15). To this end we consider the differential equation for  $\mathbf{E}^-(\mathbf{r}\omega)$

$$\begin{aligned} & -\text{curl curl } \mathbf{E}^-(\mathbf{r}\omega) + k^2[1 + \alpha(\omega)] \mathbf{E}^-(\mathbf{r}\omega) \\ & + \beta(\omega) \text{curl } \mathbf{E}^-(\mathbf{r}\omega) = 0, \end{aligned} \quad (\text{B.1})$$

which follows from eq. (2.5) when we eliminate  $\mathbf{I}(\mathbf{r}\omega)$  by means of eqs. (3.8) and (4.11). We look for solutions of  $\mathbf{E}^-(\mathbf{r}\omega)$  representing transverse plane waves propagated in the  $z$  direction, *viz.*

$$\begin{aligned} E_x^-(\mathbf{r}\omega) &= E_x^-(\omega) e^{iknz}, \\ E_y^-(\mathbf{r}\omega) &= E_y^-(\omega) e^{iknz}, \\ E_z^-(\mathbf{r}\omega) &= 0. \end{aligned} \quad (\text{B.2})$$

Substitution into (B.1) yields two equations for  $E_x^-(\omega)$  and  $E_y^-(\omega)$ . Setting the coefficient determinant equal to zero gives the dispersion equation

$$(n^2 - 1 - \alpha)^2 = (\beta^2/k^2) n^2. \quad (\text{B.3})$$

The roots of eq. (B.3) determine two values of the refractive index:  $n_+$  and  $n_-^\dagger$ .

† From two values  $\pm(n^2)^\dagger$  we choose that one which corresponds to a wave propagated in the positive  $z$  direction. The choice of the  $n$  value with the other sign simply corresponds to a change in sign of the direction of propagation.



The solutions for the transverse plane waves are

$$E_x^-(\omega) + iE_y^-(\omega) = 0, \quad (\text{B.4})$$

corresponding to a right-handed polarized wave with refractive index  $n_+$  and

$$E_x^-(\omega) - iE_y^-(\omega) = 0, \quad (\text{B.5})$$

corresponding to a left-handed circularly polarized wave with refractive index  $n_-$ . Therefore  $n_+$  and  $n_-$  satisfy the equations

$$n_+^2 - 1 - \alpha = (\beta/k) n_+, \quad (\text{B.6})$$

and

$$n_-^2 - 1 - \alpha = -(\beta/k) n_-, \quad (\text{B.7})$$

from which follow the eqs. (4.14) and (4.15) by addition and subtraction. When the medium is stable, the waves must be damped in the direction of propagation. From this it follows that  $\text{Im } n^2 \geq 0$ . In that case the inequality

$$\text{Im } \beta^2(\omega) \geq -2k^2 \text{Im } \alpha(\omega), \quad (\text{B.8})$$

has to be satisfied by the material parameters  $\alpha$  and  $\beta$ .

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## PROJECTION OPERATORS AND THE TRANSVERSE ELECTRICAL CONDUCTIVITY TENSOR OF A MATERIAL SYSTEM

Synopsis

Starting from a Hamiltonian describing non relativistic charged particles with spin, the conventional linear response theory to obtain the conductivity tensor of a many body system is reviewed. A projection operator is then defined which yields the correct expression for the transverse part of the conductivity tensor in terms of the transverse part of a current current commutator correlation function with a modified propagator. Furthermore one can show that a second projection operator may be defined which yields the transverse part of the impedance for transverse normal waves. The difficulties are discussed which arise if one attempts to derive an analogous expression for the longitudinal conductivity.

1. Introduction. The conventional way to obtain a statistical mechanical expression for the electrical, wave vector and frequency dependent conductivity tensor of a many body system is to first derive a commutator correlation function ("external" susceptibility tensor) expression for the current response of the system to an external electromagnetic field. Eliminating then the external field by means of a relation between the external field and the average Maxwell field in the system, one obtains an expression for the conductivity tensor in terms of the external susceptibility tensor. This is the procedure followed e.g. by Kadanoff and



Martin<sup>1)</sup> and elegantly reviewed in a subsequent paper by Martin.<sup>2)</sup> On the other hand expressions for wave vector and frequency dependent transport coefficient in fluids, such as the viscosity, the thermal conductivity etc. have been derived by means of projection operator techniques.<sup>3)</sup> In this chapter we show that a projection operator can be found which yields the correct expression for the transverse part of the wave vector and frequency dependent conductivity tensor. The transverse part of the conductivity tensor is then directly defined as the transverse part of a current current commutator correlation function with a modified propagator. It follows then simply that the real part and the imaginary part of the transverse conductivity tensor satisfy the Kramers-Kronig relations. We were not able to express in an analogous way the longitudinal part of the conductivity tensor. We shall discuss this point in more detail in the last section of this chapter.

In section 2 we define the Hamiltonian of a system of non-relativistic point charges with spin in interaction with the radiation field and list the relevant commutation relations. In section 3 we briefly review the linear response theory for this system when it is subjected to an external electromagnetic field. For consistency we show that the average fields and sources (charge and current densities) in linear response satisfy Maxwell's equations as should be expected. In section 4 we establish the connection between the conductivity tensor and the response function of the current density to an external electric field along conventional lines. In section 5 we then define a projection operator which enables us to express the transverse part of the conductivity tensor as a modified current current commutator correlation function. Finally in section 6 we define an alternative projection operator which allows to obtain an expression for the impedance for transverse normal waves only, i.e. when the wave vector  $\vec{k}$  and the frequency  $\omega$  satisfy the transverse dispersion relation in the medium.

## 2. The Hamiltonian of the unperturbed system.

We consider a system of  $N$  point particles (electrons and nuclei) labelled  $j = 1, 2, \dots, N$  with charges  $e_j$ , masses  $m_j$ , spins  $\vec{s}_j$  and positions  $\vec{r}_j$ . The particles are confined to a volume  $V$  and interact with the electromagnetic field generated by all the charges and their motions. The electromagnetic field is supposed to extend over all space.

The Hamiltonian of the non-relativistic particles and the electromagnetic field is given by <sup>4,5)</sup> \*

$$H_0 = H_P + H_C + H_S + H_R, \quad (2.1)$$

where

$$H_P = \sum_{j=1}^N \frac{\{\vec{p}_j - \frac{e_j}{c} \vec{a}(\vec{r}_j)\}^2}{2m_j} \quad (2.2)$$

is the kinetic energy of the particles,

$$H_C = \frac{1}{2} \sum_{i \neq j}^N \frac{e_i e_j}{4\pi |\vec{r}_i - \vec{r}_j|} \quad (2.3)$$

is the static coulomb energy of the particles,

$$H_S = - \sum_{j=1}^N \frac{e_j}{2m_j c} \vec{g}_j \cdot \vec{s}_j \cdot \text{curl } \vec{a}(\vec{r}_j) \quad (2.4)$$

is the spin energy in the non-relativistic approximation and

$$H_R = \frac{1}{2} \int dv \left[ \vec{e}^{\text{tr}}(\vec{r})^2 + \{\text{curl } \vec{a}(\vec{r})\}^2 \right] \quad (2.5)$$

is the energy of the radiation field.

In eq. (2.5) the integration is performed over infinite space. In relation (2.2)  $\vec{p}_j$  is the canonical conjugate momentum to  $\vec{r}_j$ ,  $\vec{e}^{\text{tr}}(\vec{r})$  the transverse

\* Rationalized gaussian units are used throughout.

component of the electric field,  $\vec{a}(\vec{r})$  the vector potential of the electromagnetic field in the radiation gauge i.e.

$$\text{div } \vec{a}(\vec{r}) = 0. \quad (2.6)$$

Finally,  $\frac{e_i}{2m_i c} g_i$  is the gyromagnetic ratio of particle  $i$  and  $c$  the velocity of light in vacuo.

In the quantum mechanical case, the fundamental dynamical variables satisfy the commutation relations (4,5)

$$[r_i^\alpha, r_j^\beta] = [p_i^\alpha, p_j^\beta] = 0, \quad (2.7)$$

$$[r_i^\alpha, p_j^\beta] = i\hbar \delta_{ij} \delta^{\alpha\beta}, \quad (2.8)$$

$$[a^\alpha(\vec{r}), a^\beta(\vec{r}')] = [e^{\text{tra}}(\vec{r}), e^{\text{tr}\beta}(\vec{r}')] = 0, \quad (2.9)$$

$$[e^{\text{tra}}(\vec{r}), a^\beta(\vec{r}')] = i\hbar c \delta^{\text{tra}\beta}(\vec{r}-\vec{r}'), \quad (2.10)$$

and

$$[s_i^\alpha, s_j^\beta] = i\hbar \delta_{ij} \delta^{\alpha\beta\gamma} s_i^\gamma, \quad (2.11)$$

with the convention, that the occurrence of identical greek indices implies a summation over these indices. In relations (2.8) and (2.11)  $\delta_{ij}, \delta^{\alpha\beta}$  denote Kronecker deltas. In relation (2.10)  $\delta^{\text{tra}\beta}(\vec{r}-\vec{r}')$  denotes the transverse Dirac delta function and is given by

$$\delta^{\text{tra}\beta}(\vec{r}-\vec{r}') \equiv \delta^{\alpha\beta} \delta(\vec{r}-\vec{r}') + \frac{\partial}{\partial r^\alpha} \frac{\partial}{\partial r^\beta} \frac{1}{4\pi |\vec{r}-\vec{r}'|}. \quad (2.12)$$

The Levi-Civita tensor  $\delta^{\alpha\beta\gamma}$  in formula (2.11) is defined as

$$\delta^{\alpha\beta\gamma} = \begin{cases} 1 & \text{if } \alpha, \beta, \gamma = 1, 2, 3, \text{ cycl.} \\ -1 & \text{if } \alpha, \beta, \gamma = 2, 1, 3, \text{ cycl.} \\ 0 & \text{otherwise} \end{cases} \quad (2.13)$$

The operator for the total electric field is

$$\vec{e}(\vec{r}) = \vec{e}^{tr}(\vec{r}) + \vec{e}^l(\vec{r}), \quad (2.14)$$

with

$$\vec{e}^l(\vec{r}) = -\text{grad} \int \frac{\rho_m(\vec{r}')}{4\pi|\vec{r}-\vec{r}'|} dv'. \quad (2.15)$$

The charge density operator  $\rho_m(\vec{r})$  is defined by

$$\rho_m(\vec{r}) = \sum_{j=1}^N e_j \delta(\vec{r}-\vec{r}_j). \quad (2.16)$$

The magnetic field operator is given by

$$\vec{b}(\vec{r}) = \text{curl} \vec{a}(\vec{r}) \quad (2.17)$$

and the current density operator by

$$\begin{aligned} \vec{j}(\vec{r}) = & \frac{1}{2} \sum_{j=1}^N e_j \{ \vec{r}_j \delta(\vec{r}-\vec{r}_j) + \delta(\vec{r}-\vec{r}_j) \vec{r}_j \} + \\ & + \sum_{j=1}^N \frac{e_j}{2m_j} g_j \text{curl} \vec{s}_j \delta(\vec{r}-\vec{r}_j), \end{aligned} \quad (2.18)$$

where

$$\vec{r}_j = \frac{\vec{p}_j - \frac{e_j}{c} \vec{a}(\vec{r}_j)}{m_j} \quad (2.19)$$

is the velocity operator of particle  $j$ .

The Hamiltonian (2.1) yields for the electromagnetic fields, as canonical equations of motion in the Heisenberg picture, the Maxwell-Lorentz equations in operator form:

$$\text{curl} \vec{e}(\vec{r}, t) = -\frac{1}{c} \dot{\vec{b}}(\vec{r}, t), \quad (2.20)$$

$$\text{curl} \vec{b}(\vec{r}, t) = \frac{1}{c} \dot{\vec{e}}(\vec{r}, t) + \frac{1}{c} \vec{j}(\vec{r}, t), \quad (2.21)$$

with

$$\text{div} \vec{e}(\vec{r}, t) = \rho_m(\vec{r}, t) \quad (2.22)$$



and

$$\operatorname{div} \vec{b}(\vec{r}, t) = 0, \quad (2.23)$$

with

$$\vec{e}(\vec{r}, t) = e^{\frac{i}{\hbar} H_0 t} \vec{e}(\vec{r}) e^{-\frac{i}{\hbar} H_0 t} \equiv e^{i L_0 t} \vec{e}(\vec{r}), \quad (2.24)$$

where  $L_0$  is the Liouville operator, and similarly for all other time dependent operators occurring in the equations (2.20) - (2.23).

### 3. Linear response to an external electromagnetic field; the macroscopic Maxwell equations.

We now assume that the system described in section 2 interacts with an external \* (classical) electromagnetic field characterized by a vector potential  $\vec{A}_e(\vec{r}, t)$  and a scalar potential  $\phi_e(\vec{r}, t)$ , with

$$\lim_{t \rightarrow -\infty} \vec{A}_e(\vec{r}, t) = 0 \quad (3.1)$$

and

$$\lim_{t \rightarrow -\infty} \phi_e(\vec{r}, t) = 0. \quad (3.2)$$

The total Hamiltonian of the system is then

$$H = H_0 + H_e(t) \quad (3.3)$$

where  $H_0$  is defined by (2.1) and

$$H_e(t) = -\frac{1}{c} \int d\vec{v} \vec{j}(\vec{r}) \cdot \vec{A}_e(\vec{r}, t) + \int d\vec{v} \rho_m(\vec{r}) \phi_e(\vec{r}, t). \quad (3.4)$$

Here the term quadratic in the external vector potential  $\vec{A}_e(\vec{r}, t)$  has been neglected since we shall be interested only in the linear response of the system described by  $H_0$  to the external field. We furthermore assume that

\* It is not necessarily implied, that the "external sources", generating the external electromagnetic field, are located outside the material system, but only that they are externally controlled.

the system is in thermodynamic equilibrium before the external fields are switched on and is described by a density operator  $\rho^{eq}$  which commutes with  $H_0$

$$[H_0, \rho^{eq}] = 0. \quad (3.5)$$

According to standard procedure<sup>2,6)</sup> we then find for the average electromagnetic field  $\vec{E}(\vec{r}, t)$ ,  $\vec{B}(\vec{r}, t)$  in linear response to the external field

$$\vec{E}(\vec{r}, t) = \vec{E}_e(\vec{r}, t) + \langle \vec{e}(\vec{r}) \rangle + \frac{1}{i\hbar} \int_{-\infty}^t d\tau \langle [\vec{e}(\vec{r}, t-\tau), H_e(\tau)] \rangle \quad (3.6)$$

and

$$\vec{B}(\vec{r}, t) = \vec{B}_e(\vec{r}, t) + \langle \vec{b}(\vec{r}) \rangle + \frac{1}{i\hbar} \int_{-\infty}^t d\tau \langle [\vec{b}(\vec{r}, t-\tau), H_e(\tau)] \rangle. \quad (3.7)$$

Here brackets denote ensemble averaging e.g.

$$\langle \vec{e}(\vec{r}, t) \rangle = \text{Tr } \rho^{eq} \cdot \vec{e}(\vec{r}, t). \quad (3.8)$$

In the relations (3.6) and (3.7) we have allowed for persistent electric and magnetic fields at  $t \rightarrow -\infty$ .

The external fields  $\vec{E}_e(\vec{r}, t)$  and  $\vec{B}_e(\vec{r}, t)$  are found from the external potentials according to

$$\vec{E}_e(\vec{r}, t) = -\frac{1}{c} \dot{\vec{A}}_e(\vec{r}, t) - \text{grad } \phi_e(\vec{r}, t) \quad (3.9)$$

and

$$\vec{B}_e(\vec{r}, t) = \text{curl } \vec{A}_e(\vec{r}, t). \quad (3.10)$$

Let us also consider the average charge and current densities  $\rho(\vec{r}, t)$  and  $\vec{J}(\vec{r}, t)$ . In linear response we find

$$\rho(\vec{r}, t) = \rho_e(\vec{r}, t) + \langle \rho_m(\vec{r}) \rangle + \frac{1}{i\hbar} \int_{-\infty}^t \langle [\rho_m(\vec{r}, t-\tau), H_e(\tau)] \rangle \quad (3.11)$$

and

$$\vec{J}(\vec{r}, t) = \vec{J}_e(\vec{r}, t) + \langle \vec{J}(\vec{r}) \rangle - \frac{\omega_p^2(\vec{r})}{c} \vec{A}_e(\vec{r}, t) + \frac{1}{i\hbar} \int_{-\infty}^t d\tau \langle [\vec{J}(\vec{r}, t-\tau), H_e(\tau)] \rangle, \quad (3.12)$$

where

$$\omega_p^2(\vec{r}) = \left\langle \sum_{j=1}^N \frac{e_j^2}{m_j} \delta(\vec{r} - \vec{r}_j) \right\rangle, \quad (3.13)$$

with  $\omega_p(\vec{r})$  the plasma frequency. The term with the plasma frequency (3.13) arises from the fact that the current density operator in the perturbed system given by

$$\vec{J}(\vec{r}) = \sum_{j=1}^N \frac{e_j^2}{m_j c} \delta(\vec{r} - \vec{r}_j) \vec{A}_e(\vec{r}, t), \quad (3.14)$$

where  $\vec{J}(\vec{r})$  is the current density operator of the unperturbed system (equation 2.18), contains already a term linear in the external vector potential.

In the relations (3.11) and (3.12)  $\rho_e(\vec{r}, t)$  and  $\vec{J}_e(\vec{r}, t)$  are the external charge and current densities which are the sources of the external fields according to the Maxwell equations

$$\text{curl } \vec{E}_e(\vec{r}, t) = -\frac{1}{c} \dot{\vec{B}}_e(\vec{r}, t), \quad (3.15)$$

$$\text{curl } \vec{B}_e(\vec{r}, t) = \frac{1}{c} \dot{\vec{E}}_e(\vec{r}, t) + \frac{1}{c} \vec{J}_e(\vec{r}, t), \quad (3.16)$$

$$\text{div } \vec{E}_e(\vec{r}, t) = \rho_e(\vec{r}, t) \quad (3.17)$$

and

$$\text{div } \vec{B}_e(\vec{r}, t) = 0. \quad (3.18)$$

In agreement with equations (3.6) and (3.7) we have allowed for persistent sources which generates the electric and magnetic field at  $t \rightarrow -\infty$ .

The average quantities given in equations (3.6), (3.7), (3.11) and (3.12) satisfy the macroscopic Maxwell equations

$$\text{curl } \vec{E}(\vec{r}, t) = -\frac{1}{c} \dot{\vec{B}}(\vec{r}, t), \quad (3.19)$$

$$\text{curl } \vec{B}(\vec{r}, t) = \frac{1}{c} \dot{\vec{E}}(\vec{r}, t) + \vec{J}(\vec{r}, t), \quad (3.20)$$

$$\text{div } \vec{E}(\vec{r}, t) = \rho(\vec{r}, t), \quad (3.21)$$

and

$$\text{div } \vec{B}(\vec{r}, t) = 0. \quad (3.22)$$

This is directly verified by noting that the external fields and sources obey the Maxwell equations (3.15) - (3.18), using also the equations (2.20) - (2.23), the commutation relations (2.7) - (2.11) and the identity

$$\langle [\vec{e}(\vec{r}, t), \vec{j}(\vec{r}', t)] \rangle = -i\hbar \omega_p^2(\vec{r}) \delta(\vec{r} - \vec{r}') \vec{U}, \quad (3.23)$$

which follows from the basic commutation relations (2.7) - (2.11).

In relation (3.23)  $\vec{U}$  denotes the unit tensor.

From now on we shall restrict ourselves to the case that in equilibrium there are no persistent charges or currents in the material system.

Let us next consider in more detail the induced current density  $\vec{J}_{\text{ind}}(\vec{r}, t)$  (c.f. also ref. 2) and the electric field  $\vec{E}(\vec{r}, t)$  as linear functionals of the external electric field  $\vec{E}_e(\vec{r}, t)$ . As shown by Kadanoff and Martin<sup>1,2)</sup> expression (3.12) for the induced current density is gauge invariant. One can show that this is also the case for the expression (3.6) for the electric field. Therefore, without loss of generality we may choose a gauge in which  $\phi_e(\vec{r}, t) \equiv 0$ . For the induced current density

$\vec{J}_{\text{ind}}(\vec{r}, t)$  we then obtain from equation (3.12)

$$J_{\text{ind}}^\alpha(\vec{r}, t) = -\frac{\omega_p^2(\vec{r})}{c} A_e^\alpha(\vec{r}, t) -$$



$$-\frac{1}{\hbar c} \int_{-\infty}^t d\tau \int dv' \langle [j^\alpha(\vec{r}, t), j^\beta(\vec{r}', \tau)] \rangle A_e^\beta(\vec{r}', \tau). \quad (3.24)$$

We introduce the commutator correlation function

$$\vec{\chi}_{\vec{j}\vec{j}}(\vec{r}, \vec{r}'; \tau) = \frac{i}{\hbar} \langle [\vec{j}(\vec{r}, \tau), \vec{j}(\vec{r}', 0)] \rangle \quad (3.25)$$

into equation (3.24) and taking Fourier transforms with respect to time of both members of this equation we obtain, using also equation (3.9) with  $\phi_e(\vec{r}, t) \equiv 0$ ,

$$\vec{j}_{\text{ind}}(\vec{r}, \omega) = -i\omega \int_V dv' \vec{\chi}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}', \omega) \quad (3.26)$$

where

$$\vec{j}_{\text{ind}}(\vec{r}, \omega) = \int_{-\infty}^{+\infty} dt e^{i\omega t} \vec{j}_{\text{ind}}(\vec{r}, t), \quad (3.27)$$

and

$$\vec{E}_e(\vec{r}, \omega) = \int_{-\infty}^{+\infty} dt e^{i\omega t} \vec{E}_e(\vec{r}, t). \quad (3.28)$$

In equation (3.26) we have introduced the generalized "external" susceptibility

$$\vec{\chi}(\vec{r}, \vec{r}'; \omega) = \frac{1}{\omega^2} \left[ \vec{\chi}_{\vec{j}\vec{j}}(\vec{r}, \vec{r}'; \omega) - \omega_p^2(\vec{r}) \delta(\vec{r} - \vec{r}') \vec{U} \right] \quad (3.29)$$

in which

$$\vec{\chi}_{\vec{j}\vec{j}}(\vec{r}, \vec{r}'; \omega) = \int_0^{\infty} dt e^{i\omega t} \vec{\chi}_{\vec{j}\vec{j}}(\vec{r}, \vec{r}'; t) \quad (3.30)$$

and have restricted the integration to the volume  $V$  of the material system, since  $\vec{\chi}(\vec{r}, \vec{r}'; \omega)$  vanishes for points  $\vec{r}'$  outside  $V$ .

For the electric field  $\vec{E}(\vec{r}, t)$  we obtain from equation (3.6)

$$E^{\alpha}(\vec{r}, t) = E_e^{\alpha}(\vec{r}, t) - \frac{1}{i\hbar c} \int_{-\infty}^t d\tau \int dv' \langle [e^{\alpha}(\vec{r}, t), j^{\beta}(\vec{r}', \tau)] \rangle A_e^{\beta}(\vec{r}', \tau). \quad (3.31)$$

We again introduce a commutator correlation function

$$\chi_{e_j}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \tau) = \frac{i}{\hbar} \langle [e^{\vec{r}}(\vec{r}, \tau), j^{\vec{r}'}(\vec{r}', 0)] \rangle \quad (3.32)$$

and upon substitution of relation (3.32) into equation (3.31) and Fourier transformation in time, we obtain similarly to the induced current density for the total electric field

$$\vec{E}(\vec{r}, \omega) = \vec{E}_e(\vec{r}, \omega) - \frac{i}{\omega} \int_V dv' \chi_{e_j}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}', \omega), \quad (3.33)$$

where

$$\chi_{e_j}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) = \int_0^{\infty} dt e^{i\omega t} \chi_{e_j}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; t). \quad (3.34)$$

The integration in equation (3.33) may again be restricted to the volume  $V$  since  $\chi_{e_j}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega)$  vanishes for all  $\vec{r}'$  outside  $V$ .

#### 4. The generalized conductivity.

In the linear Maxwell theory the generalized conductivity  $\vec{\sigma}(\vec{r}, \vec{r}'; \omega)$  is defined as the coefficient relating the induced current density  $\vec{J}_{\text{ind}}(\vec{r}, \omega)$  and the total electric field  $\vec{E}(\vec{r}, \omega)$  in the following way

$$\vec{J}_{\text{ind}}(\vec{r}, \omega) = \int_V dv' \vec{\sigma}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}(\vec{r}', \omega). \quad (4.1)$$

Knowledge of  $\chi(\vec{r}, \vec{r}'; \omega)$ , however, is sufficient to calculate  $\vec{\sigma}(\vec{r}, \vec{r}'; \omega)$ .

For this purpose we use equations (3.26), (3.33) and (4.1) to obtain the relation

$$\int_V dV'' \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \cdot \{ \delta(\vec{r}'' - \vec{r}') \vec{U} - \frac{i}{\omega} \chi_{\vec{e}\vec{j}}(\vec{r}'', \vec{r}'; \omega) \} =$$

$$= -i\omega \chi(\vec{r}, \vec{r}'; \omega). \quad (4.2)$$

Next we note that the relation

$$\langle [\text{curl curl } \vec{e}(\vec{r}, t), \vec{j}(\vec{r}')] \rangle + \langle \left[ \frac{1}{c^2} \ddot{\vec{e}}(\vec{r}, t), \vec{j}(\vec{r}') \right] \rangle =$$

$$= \langle \left[ -\frac{1}{c^2} \dot{\vec{j}}(\vec{r}, t), \vec{j}(\vec{r}') \right] \rangle \quad (4.3)$$

is valid due to the Maxwell-Lorentz equations (2.20) and (2.21).

Using the commutation relation

$$\left[ \vec{e}(\vec{r}), \vec{j}(\vec{r}') \right] = c \left[ \text{curl curl } \vec{a}(\vec{r}), \vec{j}(\vec{r}') \right] -$$

$$- \left[ \vec{j}(\vec{r}), \vec{j}(\vec{r}') \right] = 0, \quad (4.4)$$

and the identity (3.23) we obtain for equation (4.3)

$$- \text{curl curl } \chi_{\vec{e}\vec{j}}(\vec{r}, \vec{r}'; \omega) + \frac{\omega^2}{c^2} \chi_{\vec{e}\vec{j}}(\vec{r}, \vec{r}'; \omega) =$$

$$= -\frac{i\omega^3}{c^2} \chi(\vec{r}, \vec{r}'; \omega). \quad (4.5)$$

The solution of equation (4.5) is

$$\chi_{\vec{e}\vec{j}}(\vec{r}, \vec{r}'; \omega) = \frac{i\omega^3}{c^2} \int_V dV'' \vec{G}(\vec{r}|\vec{r}'') \cdot \vec{\chi}(\vec{r}'', \vec{r}'; \omega), \quad (4.6)$$

where we have restricted the integration to the volume  $V$  of the material system, since  $\vec{\chi}(\vec{r}, \vec{r}'; \omega)$  vanishes for points  $\vec{r}$  outside  $V$ .

The tensor Green function  $\vec{G}(\vec{r}|\vec{r}'; \omega)$  in the solution (4.6) is given by

$$\vec{G}(\vec{r}|\vec{r}'; \omega) = \left\{ \vec{U} + \frac{c^2}{\omega^2} \text{grad grad} \right\} \frac{e^{-i\omega|\vec{r}-\vec{r}'|}}{4\pi|\vec{r}-\vec{r}'|}. \quad (4.7)$$

Upon substitution of equation (4.6) into equation (4.2) we find

$$\int_V d\mathbf{v}'' \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \cdot \{ \delta(\vec{r}'' - \vec{r}') \vec{U} + \frac{\omega^2}{c^2} \int_V d\mathbf{v}''' \vec{G}(\vec{r}'' | \vec{r}'''; \omega) \cdot \vec{\chi}(\vec{r}''', \vec{r}'; \omega) \} \quad (4.8)$$

Equation (4.8) provides us with the generalized conductivity of an arbitrary material system in terms of the response function  $\vec{\chi}(\vec{r}, \vec{r}'; \omega)$ . We remark that the conductivity  $\vec{\sigma}(\vec{r}, \vec{r}'; \omega)$  satisfies the (generalized) Onsager relation

$$\sigma^{\alpha\beta}(\vec{r}, \vec{r}'; \omega) = \sigma^{\beta\alpha}(\vec{r}', \vec{r}; \omega). \quad (4.9)$$

This follows from the Onsager relation satisfied by the external susceptibility  $\vec{\chi}(\vec{r}, \vec{r}'; \omega)$  namely

$$\chi^{\alpha\beta}(\vec{r}, \vec{r}'; \omega) = \chi^{\beta\alpha}(\vec{r}', \vec{r}; \omega),$$

implied by the time reversal invariance of the Hamiltonian  $H_0$  and the density operator  $\rho^{eq}$ , and the symmetry properties of the tensor Green function  $\vec{G}(\vec{r} | \vec{r}'; \omega)$ .

In an isotropic, non-gyrotropic, translationally invariant, with respect to space, system we may write after Fourier transforming with respect to space \*

$$\vec{\chi}(\vec{k}, \omega) = \chi^{tr}(k, \omega) \left\{ \vec{U} - \frac{\vec{k}\vec{k}}{k^2} \right\} + \chi^{\ell}(k, \omega) \frac{\vec{k}\vec{k}}{k} \quad (4.10)$$

and

$$\vec{\sigma}(\vec{k}, \omega) = \sigma^{tr}(k, \omega) \left\{ \vec{U} - \frac{\vec{k}\vec{k}}{k^2} \right\} + \sigma^{\ell}(k, \omega) \frac{\vec{k}\vec{k}}{k^2}. \quad (4.11)$$

In that case we obtain, from equation (4.8),

\* Fourier transforms with respect to space are defined as e.g.

$$\vec{\sigma}(\vec{k}, \omega) = \int d\mathbf{v} \vec{\sigma}(\vec{r}, \omega) e^{-i\vec{k} \cdot \vec{r}}$$

$$\frac{i}{\omega} \sigma^{\ell}(k, \omega) = \chi^{\ell}(k, \omega) \{1 - \chi^{\ell}(k, \omega)\}^{-1} \quad (4.12)$$

and

$$\frac{i}{\omega} \sigma^{\text{tr}}(k, \omega) = \chi^{\text{tr}}(k, \omega) \left\{1 - \frac{\omega^2 \chi^{\text{tr}}(k, \omega)}{\omega^2 - c^2 k^2}\right\}^{-1}, \quad (4.13)$$

where  $\sigma^{\text{tr}}$  and  $\sigma^{\ell}$  are respectively the transverse and longitudinal conductivity. Relations (4.10) - (4.13) are those given by Martin. <sup>2)</sup>

In the following section we shall derive an alternative expression for the transverse conductivity directly in terms of a modified commutator correlation function by means of a projection operator formalism. This alternative expression suggests new approximation schemes for calculating the transverse conductivity.

### 5. The transverse conductivity.

We define a conductivity tensor  $\vec{g}(\vec{r}, \vec{r}'; \omega)$  which couples the induced current density  $\vec{J}_{\text{ind}}(\vec{r}, \omega)$  to the total transverse electric field  $\vec{E}^{\text{tr}}(\vec{r}, \omega)$  induced by a transverse external electric field through the relation

$$\vec{J}_{\text{ind}}(\vec{r}, \omega) = \int \vec{g}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}^{\text{tr}}(\vec{r}', \omega) d\vec{v}' \quad (5.1)$$

with the constraint \*

$$\text{div } \vec{E}_e(\vec{r}, \omega) = 0. \quad (5.2)$$

In the relation (5.1) the integration is over all space.

In an isotropic, non-gyrotropic translationally invariant system, the transverse external electric field can only induce a transverse Maxwell

\* The condition (5.2) allows to relate the longitudinal induced field to the transverse induced field and therefore to express the induced current density solely in terms of the transverse Maxwell field equation (5.1).



field, which in turn can only be coupled to a transverse induced current density, so that the tensor  $\vec{g}(\vec{r}, \vec{r}'; \omega)$  then reduces to the scalar transverse conductivity defined in equation (4.11). We shall prove that the tensor  $\vec{g}(\vec{r}, \vec{r}'; \omega)$  has the form

$$\vec{g}(\vec{r}, \vec{r}'; \omega) = -\frac{i}{\omega} \left\{ \int_{\vec{J}\vec{J}} d\vec{v}'' \vec{g}_{\vec{J}\vec{J}}(\vec{r}, \vec{r}''; \omega) \delta^{\vec{r}}(\vec{r}'' - \vec{r}) - \omega^2 \vec{r} \delta^{\vec{r}}(\vec{r} - \vec{r}') \vec{J} \right\} \quad (5.3)$$

where the integration is over all space.

In relation (5.3) the modified commutator correlation function

$$\vec{g}_{\vec{J}\vec{J}}(\vec{r}, \vec{r}'; \omega) \text{ is given by } \vec{g}_{\vec{J}\vec{J}}(\vec{r}, \vec{r}'; \omega) = \frac{i}{\hbar} \int_0^{\infty} \left\langle \left[ \vec{J}(\vec{r}), e^{-i(1-P)L_0\tau} \vec{J}(\vec{r}') \right] \right\rangle e^{i\omega\tau} d\tau, \quad (5.4)$$

where the projection operator  $P$  is such that  $P$  acting on an arbitrary operator  $O$  is defined by

$$PO = \frac{i}{\hbar c} \left( \vec{e}(\vec{r}) \cdot \left\langle \left[ \vec{a}(\vec{r}), O \right] \right\rangle \right) d\vec{v}. \quad (5.5)$$

In the relation (5.5) the integration is over all space. One verifies with equation (2.10) that  $P$  is idempotent:  $P^2 = P$ . \*

In order to prove equation (5.3) we introduce besides the usual commutator correlation function  $\chi_{AB}^{\vec{r}}(\vec{r}, \vec{r}'; \tau)$  for two operators  $A(\vec{r})$  and  $B(\vec{r})$  given by

\* Using the Kubo transform <sup>6)</sup> the operator (5.5) can be written in a form analogous to the projection operators introduced by Mori. <sup>3)</sup> It then follows automatically that  $P$  is also hermitian with respect to a properly defined scalar product.

$$\vec{\chi}_{AB}(\vec{r}, \vec{r}'; \tau) = \frac{i}{\hbar} \langle [A(\vec{r}), e^{-iL_0 \tau} B(\vec{r}')] \rangle, \quad (5.6)$$

the modified commutator correlation function  $\vec{g}_{AB}(\vec{r}, \vec{r}'; \omega)$  defined by

$$\vec{g}_{AB}(\vec{r}, \vec{r}'; \tau) = \frac{i}{\hbar} \langle [A(\vec{r}), e^{-i(1-P)L_0 \tau} B(\vec{r}')] \rangle \quad (5.7)$$

with the one-sided Fourier transforms of equations (5.5) and (5.6)

$$\vec{\chi}_{AB}(\vec{r}, \vec{r}'; \omega) = \int_0^{\infty} \vec{\chi}_{AB}(\vec{r}, \vec{r}'; \tau) e^{i\omega \tau} d\tau \quad (5.8)$$

and

$$\vec{g}_{AB}(\vec{r}, \vec{r}'; \omega) = \int_0^{\infty} \vec{g}_{AB}(\vec{r}, \vec{r}'; \tau) e^{i\omega \tau} d\tau. \quad (5.9)$$

Using the operator identity

$$e^{-i(1-P)L_0 t} = e^{-iL_0 t} + \int_0^t d\tau e^{-i(1-P)L_0 \tau} iPL_0 e^{-iL_0(t-\tau)}, \quad (5.10)$$

we can then write

$$\begin{aligned} \vec{g}_{JJ}(\vec{r}_0, \vec{r}'; t) &= \vec{\chi}_{JJ}(\vec{r}_0, \vec{r}'; t) + \frac{1}{c} \int_0^t d\tau \int_{je} \vec{g}_{JJ}(\vec{r}_0, \vec{r}; \tau) \cdot \\ &\cdot \frac{\partial}{\partial \tau} \vec{\chi}_{aJ}(\vec{r}, \vec{r}'; t-\tau) \end{aligned} \quad (5.11)$$

or taking one-sided Fourier transforms

$$\vec{g}_{JJ}(\vec{r}_0, \vec{r}'; \omega) = \vec{\chi}_{JJ}(\vec{r}_0, \vec{r}'; \omega) + \frac{i\omega}{c} \int_{je} \vec{g}_{JJ}(\vec{r}_0, \vec{r}; \omega) \cdot \vec{\chi}_{aJ}(\vec{r}, \vec{r}'; \omega), \quad (5.12)$$

where we have also used the fact that

$$[\vec{a}(\vec{r}), \vec{j}(\vec{r}')] = 0. \quad (5.13)$$

Upon substitution of equation (5.12) into the expression (3.26) for the induced current density  $\vec{J}_{ind}(\vec{r}, \omega)$  and using the relation



$$\frac{i\omega}{c} \vec{\chi}_{\vec{a}\vec{j}}^{\vec{t}}(\vec{r}, \vec{r}'; \omega) = \vec{\chi}_{\vec{e}\vec{tr}\vec{j}}^{\vec{t}}(\vec{r}, \vec{r}'; \omega), \quad (5.14)$$

we obtain

$$\begin{aligned} \vec{J}_{\text{ind}}(\vec{r}_0, \omega) = & -\frac{i}{\omega} \int d\vec{v}' \left\{ \vec{g}_{\vec{j}\vec{j}}^{\vec{t}}(\vec{r}_0, \vec{r}'; \omega) - \omega_p^2(\vec{r}_0) \delta(\vec{r}_0 - \vec{r}') \vec{U} \right\} \cdot \vec{E}_e(\vec{r}', \omega) + \\ & + \frac{i}{\omega} \int d\vec{v}' \int d\vec{v} \vec{g}_{\vec{j}\vec{e}}^{\vec{t}}(\vec{r}_0, \vec{r}; \omega) \cdot \vec{\chi}_{\vec{e}\vec{tr}\vec{j}}^{\vec{t}}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}'; \omega). \end{aligned} \quad (5.15)$$

Next we substitute into this last equation the transverse part of equation (3.33)

$$\vec{E}_e^{\text{tr}}(\vec{r}, \omega) = \vec{E}_e^{\text{tr}}(\vec{r}, \omega) - \frac{i}{\omega} \int d\vec{v}' \vec{\chi}_{\vec{e}\vec{tr}\vec{j}}^{\vec{t}}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}', \omega), \quad (5.16)$$

where  $\vec{E}_e^{\text{tr}}(\vec{r}, \omega)$  is the transverse part of the external electric field  $\vec{E}_e(\vec{r}, \omega)$ . This leads to the relation

$$\begin{aligned} \vec{J}_{\text{ind}}(\vec{r}_0, \omega) = & - \int d\vec{v} \vec{g}_{\vec{j}\vec{e}}^{\vec{t}}(\vec{r}_0, \vec{r}; \omega) \cdot \vec{E}_e^{\text{tr}}(\vec{r}, \omega) - \\ & - \frac{i}{\omega} \int d\vec{v} \left\{ \vec{g}_{\vec{j}\vec{j}}^{\vec{t}}(\vec{r}_0, \vec{r}; \omega) - \omega_p^2(\vec{r}_0) \delta(\vec{r}_0 - \vec{r}) \vec{U} \right\} \cdot \vec{E}_e(\vec{r}, \omega) + \\ & + \int d\vec{v} \vec{g}_{\vec{j}\vec{e}}^{\vec{t}}(\vec{r}_0, \vec{r}; \omega) \cdot \vec{E}_e^{\text{tr}}(\vec{r}, \omega). \end{aligned} \quad (5.17)$$

Let us now calculate the time derivative of  $\vec{g}_{\vec{j}\vec{e}}^{\vec{t}}(\vec{r}_0, \vec{r}'; t)$ . We find

$$\frac{\partial}{\partial t} \vec{g}_{\vec{j}\vec{e}}^{\vec{t}}(\vec{r}_0, \vec{r}'; t) = - \vec{g}_{\vec{j}\vec{e}}^{\vec{t}}(\vec{r}_0, \vec{r}'; t). \quad (5.18)$$

Equation (5.18) follows from equation (5.7) with  $A = \vec{j}$  and  $B = \vec{e}$  and the fact that

$$\left[ \vec{a}(\vec{r}), \vec{e}(\vec{r}') \right] = 0. \quad (5.19)$$

Similarly, we find that

$$\frac{\partial}{\partial t} \overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{b}}(\vec{r}_0, \vec{r}'; t) = 0, \quad (5.20)$$

using also equation (2.20) and the commutator (2.10). The modified commutator correlation function  $\overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{b}}(\vec{r}_0, \vec{r}'; t)$  is thus seen to be constant in time and therefore equal to its value at  $t=0$ . Due to the commutator relation (5.13), we thus have

$$\overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{b}}(\vec{r}_0, \vec{r}'; t) = 0. \quad (5.21)$$

Using equations (2.21) and (5.21) we may therefore write for equation (5.18)

$$\frac{\partial}{\partial t} \overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{e}}(\vec{r}_0, \vec{r}'; t) = \overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{j}}(\vec{r}_0, \vec{r}'; t), \quad (5.22)$$

so that the one-sided Fourier transform of equation (5.22) becomes

$$i\omega \overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{e}}(\vec{r}_0, \vec{r}'; \omega) - \omega_p^2(\vec{r}_0) \delta(\vec{r}_0 - \vec{r}') \overset{\rightarrow}{U} + \overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{j}}(\vec{r}_0, \vec{r}'; \omega) = 0. \quad (5.23)$$

We now restrict ourselves to the case that the external electric field is generated by a transverse external current density, so that condition (5.2) holds. Then equation (5.17) reduces to equation (5.1) with  $\overset{\rightarrow}{g}(\vec{r}, \vec{r}'; \omega)$  given by equation (5.3), which proves our assertion.

Thus the transverse conductivity for the homogeneous isotropic non-gyrotropic medium may indeed be found directly from the modified commutator correlation function  $\overset{\rightarrow}{g}_{\overset{\rightarrow}{j}\overset{\rightarrow}{j}}(\vec{r}, \vec{r}'; \omega)$ .

In the appendix we also explicitly show that the transverse conductivity (5.3) has in terms of the transverse response function  $\chi^{\text{tr}}(\mathbf{k}, \omega)$  precisely the form (4.13).

## 6. The impedance for transverse normal waves.

In this section we give another example of the use of the projection operator formalism developed in section 5. We define an impedance tensor  $\vec{R}(\vec{r}, \vec{r}'; \omega)$  which couples the total electric field  $\vec{E}(\vec{r}, \omega)$  inside the medium to the induced current density  $\vec{J}_{\text{ind}}(\vec{r}, \omega)$  through the relation

$$\vec{E}(\vec{r}, \omega) = \int_V \vec{R}(\vec{r}, \vec{r}'; \omega) \cdot \vec{J}_{\text{ind}}(\vec{r}', \omega) dv' \quad (6.1)$$

with the constraint that the medium is acted upon by an external electric field generated by external sources which are placed outside the material system. In the relation (6.1) the integration is over the volume  $V$  of the material system only.

We shall prove that the tensor  $\vec{R}(\vec{r}, \vec{r}'; \omega)$  has the form

$$\vec{R}(\vec{r}, \vec{r}'; \omega) = i\omega \frac{\vec{g}_{\vec{e}\vec{e}}(\vec{r}, \vec{r}'; \omega)}{\omega_p^2(\vec{r}')} \quad (6.2)$$

In relation (6.2) the modified commutator correlation function

$\vec{g}_{\vec{e}\vec{e}}(\vec{r}, \vec{r}'; \omega)$  is given by

$$\vec{g}_{\vec{e}\vec{e}}(\vec{r}, \vec{r}'; \omega) = \frac{i}{\hbar} \int_0^{\infty} \langle [\vec{e}(\vec{r}), e^{-i(1-P)L_0\tau} \vec{e}(\vec{r}')] \rangle e^{i\omega\tau} d\tau, \quad (6.3)$$

where the operator  $P$  is such that  $P$  acting on an arbitrary operator  $O$  is defined by

$$PO = -\frac{i}{\hbar} \int_V dv \frac{\vec{e}(\vec{r})}{\omega_p^2(\vec{r})} \cdot \langle [\vec{J}(\vec{r}), O] \rangle. \quad (6.4)$$

In relation (6.4) the integration is over the volume of the material system. One verifies with equation (3.23) that  $P$  is idempotent:  $P^2 = P$ . \*

\* The operator  $P$  is not hermitian with respect to the Mori scalar product. 3)

In order to prove equation (6.2) we again introduce modified commutator correlation functions and their one-sided Fourier transforms defined as in equations (5.7) and (5.9) but now with P defined by relation (6.4).

Using the operator identity (5.10) we can then write

$$\vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; t) = \vec{\chi}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; t) - \int_0^t d\tau \int_V dv \frac{\vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \tau)}{\omega_p^2(\vec{r})} \cdot \frac{\partial}{\partial \tau} \vec{\chi}_{\vec{j}\vec{j}}^{\rightarrow}(\vec{r}, \vec{r}'; t-\tau) \quad (6.5)$$

or taking one-sided Fourier transforms

$$\vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \omega) = \vec{\chi}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \omega) - i\omega \int_V dv \frac{\vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \omega)}{\omega_p^2(\vec{r})} \cdot \vec{\chi}_{\vec{j}\vec{j}}^{\rightarrow}(\vec{r}, \vec{r}'; \omega). \quad (6.6)$$

Let us now substitute equation (6.6) into formula (3.33) for the total electric field. We find

$$\vec{E}(\vec{r}_0, \omega) = \vec{E}_e(\vec{r}_0, \omega) - \frac{i}{\omega} \int_V dv' \vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}', \omega) + \int_V dv' \int_V dv \frac{\vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \omega)}{\omega_p^2(\vec{r})} \cdot \vec{\chi}_{\vec{j}\vec{j}}^{\rightarrow}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}', \omega). \quad (6.7)$$

Substituting into this last equation formula (3.26) for the induced current density we finally obtain

$$\vec{E}(\vec{r}_0, \omega) = -\frac{i}{\omega} \int_V dv' \{ i\omega \delta(\vec{r}_0 - \vec{r}') \vec{U} + \vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \omega) + i\omega \vec{g}_{\vec{e}\vec{e}}^{\rightarrow}(\vec{r}_0, \vec{r}'; \omega) \} \cdot \vec{E}_e(\vec{r}', \omega) + i\omega \int_V dv' \frac{\vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}')}{\omega_p^2(\vec{r}')} \cdot \vec{J}_{\text{ind}}(\vec{r}'). \quad (6.8)$$

We calculate now the time derivative of  $\vec{g}_{\vec{e}\vec{j}}^{\rightarrow}(\vec{r}_0, \vec{r}'; t)$ . We find



$$\frac{\partial}{\partial t} \vec{g}_{ee}(\vec{r}_0, \vec{r}'; t) = - \vec{g}_{ee}(\vec{r}_0, \vec{r}'; t). \quad (6.9)$$

This follows from equation (5.7) with  $A = B = \vec{e}$  and the fact that

$$\left[ \vec{j}(\vec{r}), \vec{e}(\vec{r}') \right] = 0. \quad (6.10)$$

Relation (6.10) is easily verified using equation (2.21) and the commutator (2.9). Similarly we obtain

$$\begin{aligned} \frac{\partial}{\partial t} \vec{g}_{eb}(\vec{r}_0, \vec{r}'; t) &= - \vec{g}_{eb}(\vec{r}_0, \vec{r}'; t) - \\ &- \int_V dv \frac{\vec{g}_{ee}(\vec{r}_0, \vec{r}; t)}{\omega^2(\vec{r})} \cdot \vec{j}_b(\vec{r}, \vec{r}'; 0). \end{aligned} \quad (6.11)$$

Using equation (2.20) and the relation (3.23) it can be shown that the right hand side of eq. (6.11) vanishes, so that

$$\frac{\partial}{\partial t} \vec{g}_{eb}(\vec{r}_0, \vec{r}'; t) = 0. \quad (6.12)$$

We thus see that  $\vec{g}_{eb}(\vec{r}_0, \vec{r}'; t)$  is constant in time and therefore equal to its value at  $t = 0$ . Due to the commutator (2.10) we thus have

$$\vec{g}_{eb}(\vec{r}_0, \vec{r}'; t) = c \text{curl}' \delta(\vec{r}_0 - \vec{r}') \vec{U}. \quad (6.13)$$

From equations (6.9), (6.13), together with the operator equation of motion (2.21) we then find

$$\frac{\partial}{\partial t} \vec{g}_{ee}(\vec{r}_0, \vec{r}'; t) = \vec{g}_{ej}(\vec{r}_0, \vec{r}'; t) + c^2 \text{curl}' \text{curl}' \delta(\vec{r}_0 - \vec{r}') \vec{U}, \quad (6.14)$$

so that the one-sided Fourier transform of equation (6.14) becomes \*

\* We restrict ourselves to frequencies  $\omega$  different from zero.

$$i\omega \vec{g}_{ee}(\vec{r}_0, \vec{r}'; \omega) = - \vec{g}_{ej}(\vec{r}_0, \vec{r}'; \omega) - \frac{i}{\omega} c^2 \text{curl}' \text{curl}' \delta(\vec{r}_0 - \vec{r}') \vec{U}. \quad (6.15)$$

This relation may be used to eliminate in eq. (6.8) the modified commutator correlation function  $\vec{g}_{ej}(\vec{r}, \vec{r}'; \omega)$  so that the expression for the total electric field becomes

$$\begin{aligned} \vec{E}(\vec{r}_0, \omega) &= \vec{E}_e(\vec{r}_0, \omega) - \frac{c^2}{\omega^2} \text{curl}^0 \text{curl}^0 \vec{E}_e(\vec{r}_0, \omega) + \\ &+ i\omega \int_V dv' \frac{\vec{g}_{ee}(\vec{r}_0, \vec{r}'; \omega)}{\omega_p^2(\vec{r}')} \cdot \vec{J}_{\text{ind}}(\vec{r}, \omega) = \\ &= - \frac{i}{\omega} \vec{J}_e(\vec{r}_0, \omega) + i\omega \int_V dv' \frac{\vec{g}_{ee}(\vec{r}_0, \vec{r}'; \omega)}{\omega_p^2(\vec{r}')} \cdot \vec{J}_{\text{ind}}(\vec{r}', \omega). \end{aligned} \quad (6.16)$$

Therefore, if we restrict ourselves to the case that the external electric field is generated by externally controlled sources which are located outside the material system, equation (6.16), for  $\vec{r}_0$  inside the material system, reduces to equation (6.1) with  $\vec{R}(\vec{r}, \vec{r}'; \omega)$  given by equation (6.2). This proves our assertion. In a translationally invariant, isotropic, non-gyrotropic system with no externally controlled sources inside the medium the transverse part of  $\vec{R}(\vec{k}, \omega)$  can thus be used to calculate the impedance for transverse normal waves and consequently also the index of refraction.

It should be noted that the transverse conductivity tensor derived in section 5 contains more information concerning the electromagnetic behaviour of the system than the transverse part of the tensor  $\vec{R}(\vec{k}, \omega)$ . In the appendix we explicitly show that the inverse transverse part of  $\vec{R}(\vec{k}, \omega)$  given by the Fourier transform of equation (6.2) is equal to the transverse conductivity (4.13) only when  $\vec{k}$  and  $\omega$  satisfy the dispersion relation in the medium.

## 7. Conclusions.

We have shown in the preceding sections by means of projection operator techniques that the transverse part of the conductivity tensor may be written as the transverse part of a current current commutator correlation function with a modified propagator, eqs. (5.3) and (5.4). From the structure of these equations Kramers-Kronig relations for the transverse conductivity follow in a standard way. We have not been able to find an analogous expression for the longitudinal part of the conductivity tensor. Indeed, had we found such an expression, we would have proved Kramers-Kronig relations also for the longitudinal conductivity. As Martin has shown, however, there can be no general proof of these relations for the longitudinal conductivity.<sup>2,7)</sup> The reason for this is connected with the fact that commutation relations for the dynamical variables of the radiation field have no analogy for the longitudinal fields. Our failure to find a projection operator which permits to write the longitudinal conductivity in a form analogous to the form obtained for the transverse conductivity is a consequence of this situation. It may be expected, however, that a projection operator can be found which yields the longitudinal conductivity  $\sigma^l(k, \omega)$  for sufficiently small values of  $\vec{k}$ , for which the Kramers-Kronig relations are satisfied.

Expressions for the conductivity in terms of commutator correlation functions with a modified time propagator should be of use for the evaluation of this quantity. Indeed one should be able to find suitable expansions for these expressions which e.g. in the case of plasmas would lead in first order to expressions for the conductivity which are found by treating the Maxwell field as an external field treated self consistently and taking a Hamiltonian for the system which in the case of the transverse conductivity contains only the coulomb interactions. We shall study this problem in more detail in a subsequent paper.

## Appendix.

In this appendix we only consider translationally invariant, isotropic non-gyrotropic material systems. For these media all Fourier transformed tensors have the form

$$\vec{T}(\vec{k}, \omega) = T^{\text{tr}}(k, \omega) \left\{ \vec{U} - \frac{\vec{k}\vec{k}}{k^2} \right\} + T^{\text{l}}(k, \omega) \frac{\vec{k}\vec{k}}{k^2}, \quad (\text{A.1})$$

where  $T^{\text{tr}}$  and  $T^{\text{l}}$  are respectively the transverse and longitudinal parts of the tensor  $\vec{T}$ .

First we will show that the transverse conductivity (5.3) in terms of the transverse response function  $\chi^{\text{tr}}$ , has the form (4.13).

For the transverse part of relation (5.12) we find

$$g_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega) = \chi_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega) + g_{\vec{j}\vec{e}}^{\text{tr}}(k, \omega) \chi_{\vec{e}\vec{j}}^{\text{tr}}(k, \omega), \quad (\text{A.2})$$

where we have used equation (5.14).

The transverse parts of equations (4.6) and (5.23) are respectively

$$\chi_{\vec{e}\vec{j}}^{\text{tr}}(k, \omega) = - \frac{i\omega}{\omega^2 - c^2 k^2} \left\{ \chi_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega) - \omega_p^2 \right\} \quad (\text{A.3})$$

and

$$i\omega g_{\vec{j}\vec{e}}^{\text{tr}}(k, \omega) = \omega_p^2 - g_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega). \quad (\text{A.4})$$

Inserting equations (A.3) and (A.4) into equation (A.2) we obtain

$$\frac{i}{\omega} g_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega) = \chi_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega) \left[ 1 - \frac{\omega_p^2 \chi_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega)}{\omega^2 - c^2 k^2} \right]^{-1}, \quad (\text{A.5})$$

where  $g_{\vec{j}\vec{j}}^{\text{tr}}$  and  $\chi_{\vec{j}\vec{j}}^{\text{tr}}$  are the transverse parts of  $\vec{g}$  and  $\vec{\chi}$  eqs. (5.3) and (3.29) respectively. It thus follows in view of equation (4.13) that

$$g_{\vec{j}\vec{j}}^{\text{tr}}(k, \omega) = \sigma^{\text{tr}}(k, \omega). \quad (\text{A.6})$$



Finally we show that the inverse transverse part of the impedance  $\vec{R}(\vec{k}, \omega)$  is equal to the transverse conductivity  $\sigma^{\text{tr}}(\vec{k}, \omega)$  when the wave vector  $\vec{k}$  and frequency  $\omega$  satisfy the dispersion relation in the medium.

For the transverse part of eq. (6.6) we find

$$g_{\vec{e}\vec{j}}^{\text{tr}}(\vec{k}, \omega) = -i\omega \frac{g_{\vec{e}\vec{e}}^{\text{tr}}(\vec{k}, \omega)}{\omega_p^2} \chi_{\vec{j}\vec{j}}^{\text{tr}}(\vec{k}, \omega) - \frac{i\omega}{\omega^2 - c^2 k^2} [\chi_{\vec{j}\vec{j}}^{\text{tr}}(\vec{k}, \omega) - \omega_p^2], \quad (\text{A.7})$$

where we have also used eq. (A.3).

The transverse part of eq. (6.15) is

$$i\omega g_{\vec{e}\vec{e}}^{\text{tr}}(\vec{k}, \omega) = -g_{\vec{e}\vec{j}}^{\text{tr}}(\vec{k}, \omega) - \frac{i}{\omega} c^2 k^2. \quad (\text{A.8})$$

Inserting equation (A.8) into eq. (A.7), also using eqs. (3.29) and (4.13), we obtain

$$-\frac{i}{\omega} \frac{\omega_p^2}{g_{\vec{e}\vec{e}}^{\text{tr}}(\vec{k}, \omega)} = -\sigma^{\text{tr}}(\vec{k}, \omega) \frac{\omega^2 - c^2 k^2}{c^2 k^2 [\omega^2 - c^2 k^2 + i\omega \sigma^{\text{tr}}(\vec{k}, \omega)] + i\omega \sigma^{\text{tr}}(\vec{k}, \omega)}. \quad (\text{A.9})$$

Therefore  $R^{\text{tr}}(\vec{k}, \omega)^{-1}$ , the inverse of the transverse part of  $\vec{R}(\vec{k}, \omega)$  which according to eq. (6.2) is equal to the left hand side of eq. (A.9), is not equal to  $\sigma^{\text{tr}}(\vec{k}, \omega)$  for all  $\vec{k}$  and  $\omega$ . Now the dispersion relation in the medium for transverse normal waves is

$$\frac{i}{\omega} \sigma^{\text{tr}}(\vec{k}, \omega) = -1 + \frac{c^2 k^2}{\omega^2}. \quad (\text{A.10})$$

Using the dispersion relation (A.10) in eq. (A.9) it thus follows that

$$R^{\text{tr}}(\vec{k}, \omega)^{-1} = -\frac{i}{\omega} \frac{\omega_p^2}{g_{\vec{e}\vec{e}}^{\text{tr}}(\vec{k}, \omega)} = \sigma^{\text{tr}}(\vec{k}, \omega) \quad (\text{A.11})$$

when  $\vec{k}$  and  $\omega$  satisfy eq. (A.10), the dispersion relation for transverse normal waves in the medium.

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## Chapter III

### ON INVERSE EXTINCTION THEOREMS IN ELECTRODYNAMICS

#### Synopsis

Inverse extinction theorems in electrodynamics are derived from the conventional linear response theory for a material system to an external electromagnetic field. Such inverse extinction theorems can also be obtained directly from the phenomenological Maxwell theory.

1. Introduction. The extinction theorem in electrodynamics <sup>1,2)</sup> expresses the fact that at any point inside a finite material system the incident electric field is equal to ("cancelled" by) the field generated by the total electric field at the boundary of the system or, if a constitutive relation is introduced, equal to the field set up by the current density at the boundary. Recently <sup>3)</sup>, attempts have been made to obtain from the extinction theorem in the case of a molecular fluid a linear response relation between the current density and the external electric field. Since, as described in chapter I, linear response theory which gives rise to average fields and sources satisfying Maxwell's equations, automatically also satisfies the extinction theorem, it is interesting to investigate whether the response relations in such a theory can also be written in a form which represents an inversion of the extinction theorem.

In this chapter we show that the conventional linear response theory to an external electromagnetic field indeed yields responses of that type.



It is furthermore shown that similar relations can also be derived from the phenomenological Maxwell theory, so that inverse extinction theorems, as in the case of the usual extinction theorem, are a consequence of the Maxwell equations together with a constitutive relation.

In section 2 we derive field equations for the response functions of the induced current density and the electric field. With the use of these field equations we show in section 3 that the induced current density and the total electric field may be written in an alternative form in terms of the external electric field at the boundary of the system. These new expressions represent, so to say, an inversion of the conventional extinction theorem in electrodynamics. In section 4 finally, we derive these inverse extinction theorems from the phenomenological Maxwell theory.

2. Linear response theory for a material system to an external electromagnetic field.

In chapter II section 3 <sup>\*</sup>) we have derived the following relations for the average induced current density and the average electric field (c.f. equations II.3.26 and II.3.33)

$$\vec{j}_{\text{ind}}(\vec{r}, \omega) = -i\omega \int_V dv' \vec{\chi}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}', \omega) \quad (2.1)$$

and

$$\vec{E}(\vec{r}, \omega) = \vec{E}_e(\vec{r}, \omega) - \frac{i}{\omega} \int_V dv' \sum_{e_j} \vec{\chi}_{e_j}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}_e(\vec{r}', \omega), \quad (2.2)$$

in which  $\vec{E}_e(\vec{r}, \omega)$  was the external electric field and  $\vec{\chi}(\vec{r}, \vec{r}'; \omega)$  and  $\vec{\chi}_{e_j}(\vec{r}, \vec{r}'; \omega)$  response functions defined as (c.f. equations II.3.29, II.3.13 and II.3.34)

$$\vec{\chi}(\vec{r}, \vec{r}'; \omega) = \frac{1}{\omega^2} \left[ \sum_{jj'} \vec{\chi}_{jj'}(\vec{r}, \vec{r}'; \omega) - \omega_p^2(\vec{r}) \delta(\vec{r} - \vec{r}') \vec{U} \right] \quad (2.3)$$

with

$$\vec{\chi}_{jj'}(\vec{r}, \vec{r}'; \omega) = \frac{i}{\hbar} \int_0^{\infty} \langle [\vec{j}(\vec{r}, \tau), \vec{j}(\vec{r}', 0)] \rangle e^{i\omega\tau} d\tau, \quad (2.4)$$

$$\omega_p^2(\vec{r}) = \left\langle \sum_{j=1}^N \frac{e_j^2}{m_j} \delta(\vec{r} - \vec{r}_j) \right\rangle \quad (2.5)$$

and

$$\vec{\chi}_{e_j}(\vec{r}, \vec{r}'; \omega) = \frac{i}{\hbar} \int_0^{\infty} \langle [\vec{e}(\vec{r}, \tau), \vec{j}(\vec{r}', 0)] \rangle e^{i\omega\tau} d\tau. \quad (2.6)$$

It will now be shown that these response functions satisfy the following

\* Equations and sections of chapter II, referred to in this chapter, will be preceded by the prefix II.

field equations \*)

$$\frac{i\omega^3}{c^2} \overset{\rightarrow}{\chi}_{\rightarrow j e}(\vec{r}, \vec{r}'; \omega) = - \overset{\rightarrow}{\chi}_{\rightarrow j e}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \overleftarrow{\text{curl}}' + \frac{\omega^2}{c^2} \overset{\rightarrow}{\chi}_{\rightarrow j e}(\vec{r}, \vec{r}'; \omega) \quad (2.7)$$

and

$$\begin{aligned} \frac{i\omega}{c^2} \overset{\rightarrow}{\chi}_{\rightarrow e j}(\vec{r}, \vec{r}'; \omega) - \delta(\vec{r} - \vec{r}') \overleftarrow{\text{U}} \overleftarrow{\text{curl}}' \overleftarrow{\text{curl}}' &= \\ = - \overset{\rightarrow}{\chi}_{\rightarrow e e}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \overleftarrow{\text{curl}}' + \frac{\omega^2}{c^2} \overset{\rightarrow}{\chi}_{\rightarrow e e}(\vec{r}, \vec{r}'; \omega). \end{aligned} \quad (2.8)$$

In equation (2.7)  $\overset{\rightarrow}{\chi}_{\rightarrow j e}(\vec{r}, \vec{r}'; \omega)$  is defined as

$$\overset{\rightarrow}{\chi}_{\rightarrow j e}(\vec{r}, \vec{r}'; \omega) = \frac{i}{h} \int_0^{\infty} \langle \overset{\rightarrow}{j}(\vec{r}, \tau), \overset{\rightarrow}{e}(\vec{r}', 0) \rangle e^{i\omega\tau} d\tau \quad (2.9)$$

and in equation (2.8)  $\overset{\rightarrow}{\chi}_{\rightarrow e e}(\vec{r}, \vec{r}'; \omega)$  as

$$\overset{\rightarrow}{\chi}_{\rightarrow e e}(\vec{r}, \vec{r}'; \omega) = \frac{i}{h} \int_0^{\infty} \langle \overset{\rightarrow}{e}(\vec{r}, \tau), \overset{\rightarrow}{e}(\vec{r}', 0) \rangle e^{i\omega\tau} d\tau. \quad (2.10)$$

For the derivation of the first field equation (2.7) we observe that

$$\begin{aligned} \langle \overset{\rightarrow}{j}(\vec{r}, 0), \text{curl}' \text{curl}' \overset{\rightarrow}{e}(\vec{r}', -t) \rangle + \langle \overset{\rightarrow}{j}(\vec{r}, 0), \frac{1}{c^2} \overset{\rightarrow}{e}(\vec{r}', -t) \rangle &= \\ = \langle \overset{\rightarrow}{j}(\vec{r}, 0), -\frac{1}{c^2} \overset{\rightarrow}{j}(\vec{r}', -t) \rangle, \end{aligned} \quad (2.11)$$

where we have used the operator equation

\* For reasons of convenience we introduce the notion  $\overset{\rightarrow}{T}(\vec{r}) \overleftarrow{\text{curl}}$ , which has the following meaning:

$$\{\overset{\rightarrow}{T}(\vec{r}) \overleftarrow{\text{curl}}\}^{\alpha\beta} = \sum_{\gamma\nu} \delta^{\beta\gamma\nu} \frac{\partial}{\partial r^\gamma} T^{\alpha\nu}(\vec{r}),$$

where  $\delta^{\beta\gamma\nu}$  is the Levi-Civita tensor (c.f. II.2.13).

$$\text{curl curl } \vec{e}(\vec{r}, t) + \frac{1}{c^2} \ddot{\vec{e}}(\vec{r}, t) = -\frac{1}{c^2} \vec{j}(\vec{r}, t), \quad (2.12)$$

obtained in the usual way from the equations (II.2.20) and (II.2.21).

From equations (2.8) and (2.11) we then obtain eq. (2.7) if we also use the commutation relation

$$\begin{aligned} \left[ \vec{j}(\vec{r}, 0), \vec{e}(\vec{r}', 0) \right] &= c \left[ \vec{j}(\vec{r}, 0), \text{curl}' \text{curl}' \vec{a}(\vec{r}', 0) \right] - \\ &- \left[ \vec{j}(\vec{r}, 0), \vec{j}(\vec{r}', 0) \right] = 0, \end{aligned} \quad (2.13)$$

which follows from the basic commutation relations (II.2.7) - (II.2.11) and the identity (II.3.23).

Finally, starting from the relation

$$\begin{aligned} < \left[ \vec{e}(\vec{r}, 0), \text{curl}' \text{curl}' \vec{e}(\vec{r}', -t) \right] > + < \left[ \vec{e}(\vec{r}, 0), \frac{1}{c^2} \ddot{\vec{e}}(\vec{r}', -t) \right] > = \\ &= < \left[ \vec{e}(\vec{r}, 0), -\frac{1}{c^2} \vec{j}(\vec{r}', -t) \right] >, \end{aligned} \quad (2.14)$$

we find in a similar way equation (2.8). We note that equation (2.8)

reduces in the absence of matter ( $\chi_{\vec{j}\vec{e}}(\vec{r}, \vec{r}'; \omega) \equiv 0$ ) to the field equation of  $\chi_{\vec{j}\vec{e}}(\vec{r}, \vec{r}'; \omega)$  for the free radiation field as it should. <sup>4)</sup>

### 3. Linear response to external fields as an inverse extinction theorem.

Let us now eliminate from the Kubo formula equation (2.1)

for the induced current density the generalized external susceptibility

$\chi(\vec{r}, \vec{r}'; \omega)$  by means of the field equation (2.7). We then get

$$\begin{aligned} \vec{j}_{\text{ind}}(\vec{r}, \omega) &= \frac{c^2}{\omega^2} \int_V d\vec{v}' \left[ \left\{ \chi_{\vec{j}\vec{e}}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \overleftarrow{\text{curl}}' \right\} - \frac{\omega^2}{c^2} \chi_{\vec{j}\vec{e}}(\vec{r}, \vec{r}'; \omega) \right] \cdot \\ &\cdot \vec{E}_e(\vec{r}', \omega). \end{aligned} \quad (3.1)$$

On the other hand the external field  $\vec{E}_e(\vec{r}, \omega)$  satisfies the field equation



$$-\text{curl curl } \vec{E}_e(\vec{r}, \omega) + \frac{\omega^2}{c^2} \vec{E}_e(\vec{r}, \omega) = -\frac{i\omega}{c^2} \vec{J}_e(\vec{r}, \omega). \quad (3.2)$$

Substitution of equation (3.2) into equation (3.1) leads to

$$\begin{aligned} \vec{J}_{\text{ind}}(\vec{r}, \omega) = & \frac{c^2}{\omega^2} \int_V dv' \left[ \left\{ \vec{\chi}_{\vec{J}_e}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \overleftarrow{\text{curl}}' \right\} \cdot \vec{E}_e(\vec{r}', \omega) - \right. \\ & \left. - \vec{\chi}_{\vec{J}_e}(\vec{r}, \vec{r}'; \omega) \cdot \text{curl}' \text{curl}' \vec{E}_e(\vec{r}', \omega) \right] + \\ & + \frac{i}{\omega} \int_V dv' \vec{\chi}_{\vec{J}_e}(\vec{r}, \vec{r}'; \omega) \cdot \vec{J}_e(\vec{r}', \omega). \end{aligned} \quad (3.3)$$

Making use of Green's theorem <sup>4)</sup> for the vector  $\vec{E}_e(\vec{r}, \omega)$  and the tensor  $\vec{\chi}_{\vec{J}_e}(\vec{r}, \vec{r}'; \omega)$ , we finally obtain

$$\begin{aligned} \vec{J}_{\text{ind}}(\vec{r}, \omega) = & -\frac{c^2}{\omega^2} \int_{\Sigma} d\sigma' \left[ \left\{ \vec{\chi}_{\vec{J}_e}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \right\} \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) \} + \right. \\ & \left. + \vec{\chi}_{\vec{J}_e}(\vec{r}, \vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \right] + \\ & + \frac{i}{\omega} \int_V dv' \vec{\chi}_{\vec{J}_e}(\vec{r}, \vec{r}'; \omega) \cdot \vec{J}_e(\vec{r}', \omega) \end{aligned} \quad (3.4)$$

in which the first integral on the right-hand side is over  $\Sigma$ , the closed boundary surface of the material system and  $\hat{n}(\vec{r}')$  is the outward normal on  $\Sigma$ .

In a similar way we obtain from the Kubo relation equation (2.2), the field equations (2.8) and (3.2) and Green's theorem the following relation for the total electric field  $\vec{E}(\vec{r}, \omega)$  for a point  $\vec{r}$  inside  $V$

$$\begin{aligned} \vec{E}(\vec{r}, \omega) = & -\frac{c^2}{\omega^2} \int_{\Sigma} d\sigma' \left[ \left\{ \vec{\chi}_{\vec{e}\vec{e}}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \right\} \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) \} + \right. \\ & \left. + \vec{\chi}_{\vec{e}\vec{e}}(\vec{r}, \vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \right] + \end{aligned}$$

$$+ \frac{i}{\omega} \int_V dv' \left\{ \chi_{ee}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) - \delta(\vec{r} - \vec{r}') \vec{U} \right\} \cdot \vec{J}_e(\vec{r}', \omega) \quad (3.5)$$

and

$$\begin{aligned} \vec{E}(\vec{r}, \omega) = & \vec{E}_e(\vec{r}, \omega) + \frac{i}{\omega} \int_V dv' \chi_{ee}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \cdot \vec{J}_e(\vec{r}', \omega) - \\ & - \frac{c^2}{\omega^2} \int_{\Sigma} d\sigma' \left[ \chi_{ee}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \right] \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) + \\ & + \chi_{ee}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \} \end{aligned} \quad (3.6)$$

for a point  $\vec{r}$  outside  $V$ .

Note that equation (3.4) could also have been obtained directly from equation (3.5) using an equation for  $\chi_{ee}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega)$  analogous to equation (2.8) but involving differentiation with respect to the first argument.

Consider now the case that no "external" sources are located inside the material. Then equations (3.4) and (3.5) reduce to

$$\begin{aligned} \vec{J}_{\text{ind}}(\vec{r}, \omega) = & - \frac{c^2}{\omega^2} \int_{\Sigma} d\sigma' \left[ \chi_{je}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \right] \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) + \\ & + \chi_{je}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \} \end{aligned} \quad (3.7)$$

and

$$\begin{aligned} \vec{E}(\vec{r}, \omega) = & - \frac{c^2}{\omega^2} \int_{\Sigma} d\sigma' \left[ \chi_{ee}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \overleftarrow{\text{curl}}' \right] \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) + \\ & + \chi_{ee}^{\vec{r}, \vec{r}'}(\vec{r}, \vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \}. \end{aligned} \quad (3.8)$$

These equations have so to say the form of an "inverse" extinction theorem. In the usual extinction theorem the external electric field inside the medium is "cancelled" by a field generated by the total electromagnetic

field at the boundary of the material system, or, if constitutive relations are introduced, by the current set up by the boundary. Here we see that conversely the total electric field or the current inside the system is equal to a field or a current generated by the external field at the boundary.

Note that if the sources are removed to infinity and if we subsequently, in the case of a homogeneous medium, take the limit of an infinite material system, so that all commutator correlation functions become translationally invariant, the surface terms in equations (3.7) and (3.8) do not vanish if the medium is non absorptive or vanish if the medium is absorptive. In the latter case since the total field can only vanish if the induced electric field cancels the external field there therefore still is a coupling between the medium and the external sources at infinity, which is expressed by the equivalent equation (2.2).

It is furthermore possible to show that if all external sources are located inside the medium, the boundary terms in equations (3.4) and (3.5) vanish.

In both cases (external sources located only inside or only outside the material system) all relevant information concerning the response of the system is contained in the response function  $\vec{\chi}(\vec{r}, \vec{r}'; \omega)$ , or the commutator correlation functions  $\vec{\chi}_{ee}(\vec{r}, \vec{r}'; \omega)$ ,  $\vec{\chi}_{je}(\vec{r}, \vec{r}'; \omega)$  and  $\vec{\chi}_{ej}(\vec{r}, \vec{r}'; \omega)$ , which can be obtained from  $\vec{\chi}(\vec{r}, \vec{r}'; \omega)$  by means of field equations of the type of equations (2.7) and (2.8). This completely justifies the procedure of going to the limit of an infinite material system with external sources located at finite domains in space in order to calculate the commutator correlation functions and to derive subsequently the optical behaviour of the medium. This is the usual procedure followed e.g. by Martin.<sup>6)</sup> The idea that in order to deal with the optical properties of a material system one must necessarily consider the system to be finite, because "light (external sources at infinity) couples to

the system through its boundary", and that therefore the response of the system must be found through an inversion of the extinction theorem, is obviously based on a wrong interpretation of the content of the extinction theorem. 3) As has been shown the extinction theorem is nothing but a special form of the classical boundary value problem of electromagnetic theory, using the constitutive equations.

We have shown in this chapter that the conventional linear response theory also automatically incorporates an inversion of the extinction theorem. That these formal inverse extinction theorems do not indicate that there is a coupling to the medium through the boundary is also clear from the fact that in equations (3.4) and (3.5) the surface  $\Sigma$  may be any closed surface containing the medium and not necessarily its boundary.

#### 4. Inverse extinction theorems from Maxwell theory.

We shall finally show that the inverse extinction theorems derived in section 3 from linear response theory can also be obtained, just as the conventional extinction theorem, as a consequence of the Maxwell equations together with a constitutive equation. To achieve this we start from the field equation for the total electric field which is

$$-\text{curl curl } \vec{E}(\vec{r}, \omega) + \frac{\omega^2}{c^2} \vec{E}(\vec{r}, \omega) = -\frac{i\omega}{c^2} \{ \vec{J}_{\text{ind}}(\vec{r}, \omega) + \vec{J}_e(\vec{r}, \omega) \}. \quad (4.1)$$

We introduce the constitutive equation

$$\vec{J}_{\text{ind}}(\vec{r}, \omega) = \int \vec{\sigma}(\vec{r}, \vec{r}'; \omega) \cdot \vec{E}(\vec{r}', \omega) \quad (4.2)$$

in which the conductivity tensor  $\vec{\sigma}(\vec{r}, \vec{r}'; \omega)$  vanishes for all points  $\vec{r}, \vec{r}'$  outside  $V$ , the volume of the material system. Substitution of eq. (4.2) into eq. (4.1) gives



$$\begin{aligned}
 & - \operatorname{curl} \operatorname{curl} \vec{E}(\vec{r}, \omega) + \frac{\omega^2}{c^2} \int d\vec{v}' \{ \delta(\vec{r} - \vec{r}') \vec{U} + \frac{i}{\omega} \vec{\sigma}(\vec{r}, \vec{r}'; \omega) \} \cdot \\
 & \cdot \vec{E}(\vec{r}', \omega) = - \frac{i\omega}{c^2} \vec{J}_e(\vec{r}, \omega), \quad (4.3)
 \end{aligned}$$

where the integration in the second term is over all space. We solve equation (4.3) with the help of a retarded tensor Green's function  $\vec{G}_\sigma(\vec{r}|\vec{r}'; \omega)$ , which satisfies the equation

$$\begin{aligned}
 & - \operatorname{curl} \operatorname{curl} \vec{G}_\sigma(\vec{r}|\vec{r}'; \omega) + \frac{\omega^2}{c^2} \int d\vec{v}'' \{ \delta(\vec{r} - \vec{r}'') \vec{U} + \frac{i}{\omega} \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \} \cdot \\
 & \cdot \vec{G}_\sigma(\vec{r}''|\vec{r}'; \omega) = - \delta(\vec{r} - \vec{r}') \vec{U}. \quad (4.4)
 \end{aligned}$$

The solution of eq. (5.3) can now be written as

$$\vec{E}(\vec{r}, \omega) = \frac{i\omega}{c^2} \int d\vec{v}' \vec{G}_\sigma(\vec{r}|\vec{r}'; \omega) \cdot \vec{J}_e(\vec{r}', \omega). \quad (4.5)$$

We note that the tensor Green's function  $\vec{G}_\sigma(\vec{r}|\vec{r}'; \omega)$  is the propagator in macroscopic electrodynamics. Elimination of  $\vec{J}_e(\vec{r}, \omega)$ , using eq. (3.2), gives after partial integration

$$\vec{E}(\vec{r}, \omega) = \int d\vec{v}' \vec{G}_\sigma(\vec{r}|\vec{r}'; \omega) \{ \overleftarrow{\operatorname{curl}}' \overleftarrow{\operatorname{curl}}' - \frac{\omega^2}{c^2} \vec{U} \} \cdot \vec{E}_e(\vec{r}', \omega) \quad (4.6)$$

It follows from eq. (4.4) and the Onsager relation

$$\sigma^{\alpha\beta}(\vec{r}, \vec{r}'; \omega) = \sigma^{\beta\alpha}(\vec{r}', \vec{r}; \omega) \quad (4.7)$$

that

$$\begin{aligned}
 & \vec{G}_\sigma(\vec{r}|\vec{r}'; \omega) \{ \overleftarrow{\operatorname{curl}}' \overleftarrow{\operatorname{curl}}' - \frac{\omega^2}{c^2} \vec{U} \} = \\
 & = \delta(\vec{r} - \vec{r}') \vec{U} + \frac{i\omega}{c^2} \int d\vec{v}'' \vec{G}_\sigma(\vec{r}|\vec{r}''; \omega) \cdot \vec{\sigma}(\vec{r}'', \vec{r}'; \omega), \quad (4.8)
 \end{aligned}$$

so that the integration in equation (4.6) may be restricted to the volume  $V$  of the material system for points  $\vec{r}'$  inside the medium. Equation (4.6)

can thus be written, using equation (3.2) and Green's theorem, as

$$\begin{aligned} \vec{E}(\vec{r}, \omega) = & - \int d\sigma' \left[ \vec{G}_{\sigma}(\vec{r}|\vec{r}'; \omega) \overleftarrow{\text{curl}}' \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) \} + \right. \\ & \left. + \vec{G}_{\sigma}(\vec{r}|\vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \right] + \\ & + \frac{i\omega}{c^2} \int_V \vec{G}_{\sigma}(\vec{r}|\vec{r}'; \omega) \cdot \vec{J}_e(\vec{r}', \omega). \end{aligned} \quad (4.9)$$

Using the material equation (4.2) we find for the induced current density

$$\begin{aligned} \vec{J}_{\text{ind}}(\vec{r}, \omega) = & - \int d\sigma' \left[ \int dv'' \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \cdot \vec{G}_{\sigma}(\vec{r}''|\vec{r}'; \omega) \overleftarrow{\text{curl}}' \cdot \right. \\ & \left. \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) \} + \right. \\ & \left. + \int dv'' \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \cdot \vec{G}_{\sigma}(\vec{r}''|\vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \right] + \\ & + \frac{i\omega}{c^2} \int_V dv' \int dv'' \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \cdot \vec{G}_{\sigma}(\vec{r}''|\vec{r}'; \omega) \cdot \vec{J}_e(\vec{r}', \omega). \end{aligned} \quad (4.10)$$

We now consider the case that no external sources are located inside the material. Then equations (4.9) and (4.10) reduces to

$$\begin{aligned} \vec{E}(\vec{r}, \omega) = & - \int d\sigma' \left[ \vec{G}_{\sigma}(\vec{r}|\vec{r}'; \omega) \overleftarrow{\text{curl}}' \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) \} + \right. \\ & \left. + \vec{G}_{\sigma}(\vec{r}|\vec{r}'; \omega) \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \right] \end{aligned} \quad (4.11)$$

and

$$\begin{aligned} \vec{J}_{\text{ind}}(\vec{r}, \omega) = & - \int d\sigma' \left[ \int dv'' \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \cdot \vec{G}_{\sigma}(\vec{r}''|\vec{r}'; \omega) \overleftarrow{\text{curl}}' \cdot \right. \\ & \left. \cdot \{ \hat{n}(\vec{r}') \times \vec{E}_e(\vec{r}', \omega) \} + \int dv'' \vec{\sigma}(\vec{r}, \vec{r}''; \omega) \cdot \vec{G}_{\sigma}(\vec{r}''|\vec{r}'; \omega) \cdot \right. \\ & \left. \cdot \{ \hat{n}(\vec{r}') \times \text{curl}' \vec{E}_e(\vec{r}', \omega) \} \right]. \end{aligned} \quad (4.12)$$

The equations (4.11) and (4.12) represent again inverse extinction theorems. They are here a consequence of the Maxwell equations together with a material equation. If one furthermore establishes a connection between the generalized susceptibilities, found on the basis of linear response theory, and the propagator  $\overset{\pm}{G}_\sigma$ , one can show that the inverse extinction theorems (4.11) and (4.12) are equivalent to the analogous expressions (3.8) and (3.7). This will be done in the appendix.

The solution of eq. (2.11) can now be written as

$$\vec{G}(\vec{r}, \vec{r}', \omega) = \int d\vec{r}'' \left[ \vec{G}(\vec{r}, \vec{r}'', \omega) \cdot \vec{G}(\vec{r}'', \vec{r}', \omega) + \left[ (\vec{r}, \vec{r}'', \omega) \cdot \vec{G}(\vec{r}'', \vec{r}', \omega) + (\vec{r}'', \vec{r}', \omega) \cdot \vec{G}(\vec{r}, \vec{r}'', \omega) \right] \cdot \vec{G}(\vec{r}, \vec{r}', \omega) \right]$$

We note that the tensor Green's function  $\vec{G}(\vec{r}, \vec{r}', \omega)$  is the propagator in the material system for the case that no external sources are included inside the volume  $V$ .

It is obvious that the tensor Green's function  $\vec{G}(\vec{r}, \vec{r}', \omega)$  is the propagator in the material system for the case that no external sources are included inside the volume  $V$ .

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It follows from eq. (2.11) that the tensor Green's function  $\vec{G}(\vec{r}, \vec{r}', \omega)$  is the propagator in the material system for the case that no external sources are included inside the volume  $V$ .

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## Appendix.

In this appendix we will give an interpretation of the commutator correlation functions of linear response theory in terms of the macroscopic propagator  $\overset{\rightarrow}{G}_\sigma$  and the material parameter  $\overset{\rightarrow}{\sigma}$ , proving at the same time that the inverse extinction theorems derived in section 3 are equivalent to those derived in section 4. For convenience we will use a formal matrix notation which is self explanatory. For the total electric field we may write

$$\overset{\rightarrow}{E} = \frac{i\omega}{c^2} \overset{\rightarrow}{G} \{ \overset{\rightarrow}{J}_{\text{ind}} + \overset{\rightarrow}{J}_e \} \quad (\text{A.1})$$

where

$$\overset{\rightarrow}{G}(\vec{r}|\vec{r}';\omega) = \{ \overset{\rightarrow}{U} + \frac{c^2}{\omega^2} \text{grad grad} \} \frac{e^{-i\omega|\vec{r}-\vec{r}'|}}{4\pi|\vec{r}-\vec{r}'|} \quad (\text{A.2})$$

is the vacuum propagator of electrodynamics.

On the other hand we have (c.f. equation (4.5))

$$\overset{\rightarrow}{E} = \frac{i\omega}{c^2} \overset{\rightarrow}{G}_\sigma \overset{\rightarrow}{J}_e \quad (\text{A.3})$$

and (c.f. equation (3.2))

$$\overset{\rightarrow}{E}_e = \frac{i\omega}{c^2} \overset{\rightarrow}{G} \cdot \overset{\rightarrow}{J}_e \quad (\text{A.4})$$

Using the equations (A.1), (A.3), (A.4) and the Onsager relation for  $\overset{\rightarrow}{\sigma}$  eq. (4.7) we find the relation

$$\overset{\rightarrow}{G}_\sigma = \overset{\rightarrow}{G} + \frac{i\omega}{c^2} \overset{\rightarrow}{G}_\sigma \overset{\rightarrow}{\sigma} \overset{\rightarrow}{G} \quad (\text{A.5})$$

Eliminating  $\overset{\rightarrow}{J}_e$  from (A.3) by means of (A.4) and comparing to eq. (2.2) we obtain the identification



$$\overset{\leftrightarrow}{\chi}_{\vec{e}\vec{j}} = -\frac{\omega^2}{c^2} \overset{\leftrightarrow}{G}_{\sigma} \overset{\leftrightarrow}{\sigma} \quad (\text{A.6})$$

where we have also used eq. (A.5).

In the same way it follows from equations (4.2), (A.3), (A.4) and comparison with eq. (2.1) that

$$\overset{\leftrightarrow}{\chi} = \frac{i}{\omega} \overset{\leftrightarrow}{\sigma} - \frac{1}{c^2} \overset{\leftrightarrow}{\sigma} \overset{\leftrightarrow}{G}_{\sigma} \overset{\leftrightarrow}{\sigma}. \quad (\text{A.7})$$

On the other hand we may write eq. (2.8) in the matrix form as

$$\overset{\leftrightarrow}{\chi}_{\vec{e}\vec{e}} - \overset{\leftrightarrow}{U} = -\frac{i\omega}{c^2} \overset{\leftrightarrow}{\chi}_{\vec{e}\vec{j}} \overset{\leftrightarrow}{G} + \frac{\omega^2}{c^2} \overset{\leftrightarrow}{G}. \quad (\text{A.8})$$

Upon substitution of equation (A.6) into equation (A.8) we obtain, also using equation (A.5)

$$\overset{\leftrightarrow}{\chi}_{\vec{e}\vec{e}} - \overset{\leftrightarrow}{U} = \frac{\omega^2}{c^2} \overset{\leftrightarrow}{G}_{\sigma}. \quad (\text{A.9})$$

Finally, we note, using time reversal invariance, that

$$\overset{\leftrightarrow}{\chi}_{\vec{j}\vec{e}} = \frac{\omega^2}{c^2} \overset{\leftrightarrow}{\sigma} \overset{\leftrightarrow}{G}_{\sigma}. \quad (\text{A.10})$$

It is now easily verified, using the above relations, that the inverse extinction theorems derived in section 3 are equivalent to those in section 4. Moreover we can give an interpretation of the commutator correlation function in linear response theory in terms of the macroscopic quantities  $\overset{\leftrightarrow}{G}_{\sigma}$  and  $\overset{\leftrightarrow}{\sigma}$ .

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## SAMENVATTING

In de macroscopische theorie van het electromagnetisme kan het gedrag van materiële lichamen in een electromagnetisch veld in lineaire benadering gekarakteriseerd worden door materiaalconstanten, zoals de gegeneraliseerde golfvector en frequentieafhankelijke diëlectrische tensor of de gegeneraliseerde golfvector en frequentieafhankelijke geleidingsvermogens-tensor. Deze materiaalconstanten stellen ons in staat het resulterende electromagnetische veld uit te rekenen in willekeurige ruimte-tijd-punten, wanneer het materiële systeem bloot gesteld wordt aan een willekeurig uitwendig electromagnetisch veld. Het is de taak van de statistische mechanica om uitdrukkingen voor deze materiaalparameters te leveren in termen van de microscopische eigenschappen van het systeem. Bij de conventionele methode om zulke uitdrukkingen te verkrijgen, leidt men eerst een statistische uitdrukking af voor de "uitwendige" susceptibiliteit, die de stroomresponsie van het systeem op een uitwendig electromagnetisch veld vastlegt. Vervolgens wordt het uitwendige veld geëlimineerd met behulp van een relatie tussen het uitwendige veld en het Maxwell-veld in het systeem. Men verkrijgt zodoende een uitdrukking voor het geleidingsvermogen als functie van de uitwendige susceptibiliteit.

In de theorie van de optica stelt men belang in het gedrag van materiële systemen, wanneer deze verstoord worden door licht. In het bijzonder is men geïnteresseerd in de voortplanting van normale golven in het medium, dat dan gekarakteriseerd wordt door brekingsindices. In een bepaald type optische theorie, de zogenaamde "rigorous dispersion" theorie, speelt het extinctiethorema van Ewald en Oseen een belangrijke rol. Ten gevolge van het superpositieprincipe bestaat het totale elektrische veld binnen een

medium uit de inkomende golf en de geïnduceerde golf. Daar de snelheid van het licht in een medium in het algemeen verschilt van de snelheid van het licht in vacuüm, moet de inkomende golf uitgedoofd worden door het geïnduceerde veld. Hoe dit in zijn werk gaat, is de inhoud van het extinctie-theorema. Deze stelling zegt, dat het inkomende elektrische veld in elk punt binnen het medium uitgedoofd wordt door het elektrische veld, dat voortgebracht wordt door de geïnduceerde stroomdichtheid aan het oppervlak van het materiële systeem. De manier waarop deze stelling afgeleid wordt, schijnt te impliceren, dat de uitdoving van de uitwendige lichtgolf slechts met behulp van microscopische beschouwingen begrepen kan worden.

In dit proefschrift zullen wij enerzijds trachten de betekenis van het extinctietheorema te verduidelijken, anderzijds een methode ontwikkelen met behulp van projectieoperatoren, die op directe wijze statistische uitdrukkingen geeft voor de materiaalconstanten van het systeem. Uitgaande van de algemene oplossing van het klassieke randwaardeprobleem voor electromagnetische velden, leiden we in hoofdstuk I formele extinctie-theorema's af voor het invallende electromagnetische veld. Deze uitdrukkingen zijn algemene identiteiten en kunnen gebruikt worden voor willekeurige systemen. Het ware extinctietheorema van het Ewald-Oseen type voor lineaire, isotrope, homogene media wordt daarna afgeleid door materiaalvergelijkingen in te voeren. De belangrijkheid van oppervlakte-effecten, veroorzaakt door discontinuïteiten in de materiaalconstanten, wordt aangetoond. Deze afleiding laat zien, dat het extinctietheorema reeds een gevolg is van de Maxwell-vergelijkingen en de materiaalvergelijkingen. We mogen daarom vaststellen, dat in de gebruikelijke theorieën deze stelling gezien moet worden als een uiting van het feit, dat de theorie consistent is met de Maxwell-vergelijkingen. In hoofdstuk II laten we zien hoe, met behulp van een projectieoperator-methode, in het raam van de lineaire responsietheorie een uitdrukking voor het transversale deel van de geleidingsvermogensensor gevonden kan worden in termen van het



transversale deel van een stroom-stroom-correlatiefunctie met een gemodificeerde propagator. Uit de structuur van deze uitdrukking volgen onmiddellijk Kramers-Kronig relaties. De omstandigheid, dat we niet in staat waren een analoge uitdrukking te vinden voor het longitudinale geleidingsvermogen, staat in verband met het feit, dat er geen algemeen bewijs van de Kramers-Kronig relaties gegeven kan worden voor het longitudinale geleidingsvermogen. De aanwezigheid van de electromagnetische interacties tussen de deeltjes maakt de berekening van het geleidingsvermogen buitengewoon moeilijk. Het is daarom gebruikelijk om in eerste benadering zelfconsistente veldmethoden aan te wenden. Deze methoden zijn gebaseerd op de aanname, dat de geleidingsvermogentensor verkregen kan worden door de responsie op een uitwendig electromagnetisch veld uit te rekenen voor een model-systeem met een effectieve Hamiltoniaan, waarin gedeelten van de electromagnetische wisselwerking zijn weggelaten. In een volgende benadering beschouwt men dan de zogenaamde lokale veld- (Lorentz-veld) correcties. Algemeen wordt aangenomen, dat lokale veldcorrecties niet belangrijk zijn in plasma's. Het is niet duidelijk of zo'n zelfconsistente veldmethode redelijk is in het geval van bijvoorbeeld een vloeistof bestaande uit moleculen. Het is een van de aantrekkelijke kanten van de uitdrukkingen voor de electromagnetische transportcoëfficiënten in termen van gemodificeerde commutator-correlatiefuncties, dat deze een geschikt uitgangspunt bieden om de geldigheid van de bovengenoemde zelfconsistente veldmethoden te bestuderen.

In hoofdstuk III leiden we zogenaamde "omgekeerde" extinctietheorema's af uit de conventionele lineaire responsietheorie voor materiële systemen in wisselwerking met uitwendige electromagnetische velden. Deze omgekeerde extinctietheorema's drukken uit, dat de geïnduceerde stroomdichtheid en het totale elektrische veld in elk punt binnen een medium gelijk zijn aan respectievelijk een stroom en een veld, die voortgebracht worden door het uitwendige elektrische veld aan de grens van het medium. Deze omgekeerde

extinctietheorema's kunnen ook direct verkregen worden uit de fenomenologische Maxwell-theorie. Evenals in het geval van het klassieke extinctietheorema zijn deze relaties dus een gevolg van de Maxwell-vergelijkingen tezamen met een materiaalvergelijking. Tegelijk met de fysische interpretatie van deze relaties worden enige aspecten van de lineaire responsietheorie voor translatie-invariante systemen besproken.

Tenslotte geven we een interpretatie van de commutator-correlatiefuncties der lineaire responsietheorie, d.w.z. de "uitwendige" susceptibiliteiten, in termen van de macroscopische propagator voor het elektrische veld en de geleidingsvermogentensor van het medium.

Op verzoek van de Faculteit der Wiskunde en Natuurwetenschappen volgt hier een overzicht van mijn academische studie.

In 1952 legde ik het eindexamen H.B.S.-B af aan de H.B.S., Keizersgracht te Amsterdam.

In 1957 begon ik mijn studie in de scheikunde aan de Rijksuniversiteit te Leiden en legde in februari 1961 het candidaatsexamen F af.

Onder leiding van de hoogleraren Prof.Dr. A.J. Staverman en Prof.Dr. M. Mandel (fysische chemie), Prof.Dr. L.J. Oosterhoff (theoretische organische chemie) en Prof.Dr. Van Nieuwenburg (analytische chemie), bereidde ik mij voor op het doctoraalexamen scheikunde (hoofdvak fysische chemie), dat ik in juni 1964 aflegde.

Ik was van oktober 1957 tot januari 1966 achtereenvolgens werkzaam als: studentassistent, practicumassistent, assistent en hoofdassistent op het Chemisch Laboratorium te Leiden.

Van januari 1967 tot januari 1972 was ik in dienst van de Nederlandse Organisatie voor Zuiver-Wetenschappelijk Onderzoek (Z.W.O.) als wetenschappelijk medewerker.

Van januari 1972 tot mei 1973 was ik wederom in dienst van de Rijksuniversiteit Leiden als assistent op het Chemisch Practicum op het Górlaeus Laboratorium te Leiden.

In 1966/'68 werkte ik onder leiding van Prof.Dr. P.W. Kasteleyn en Prof.Dr. L.J. Oosterhoff op het gebied van de veel-deeltjes theorie.

Sinds 1969 verrichtte ik onder leiding van Prof.Dr. P. Mazur onderzoek op het gebied van de statistische mechanica van materie in een electro-magnetisch veld. De resultaten van dit onderzoek worden in dit proefschrift beschreven.

Thans ben ik werkzaam als theoretisch fysicus op het Physiologisch Laboratorium der Rijksuniversiteit Leiden.





