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The theory of indirect resonant inelastic X-ray scattering on magnons

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Abstract – Recent experiments reveal indirect resonant inelastic X-ray scattering (RIXS) to be a new probe of spin dynamics. Here I derive the cross-section for magnetic RIXS and determine the momentum-dependent four-spin correlation function that it measures. These results show that this technique offers information on spin dynamics that is complementary to, *e.g.*, neutron scattering. As an example the RIXS spectrum of Heisenberg antiferromagnets is calculated by considering a half filled single band Hubbard model at strong coupling and zero temperature. It turns out that only scattering processes that involve at least two magnons are allowed. Other selection rules imply that the scattering intensity vanishes for specific transferred momenta $\mathbf{q}$, in particular for $\mathbf{q} = 0$. All results agree very well with recent experimental data.

Introduction. – Resonant inelastic X-ray scattering (RIXS) is a technique that is rapidly developing due to the recent increase in brilliance of the new generation synchrotron X-ray sources [1]. High flux photon beams with energies that are tunable to various resonant edges are now becoming available. RIXS has two important advantages. First, it is sensitive to excitations that are difficult to observe by other techniques. An example are direct *d*-*d* excitations in cuprates or manganites [2–7]. Second, unlike optical methods it can probe directly the momentum dependence of such excitations.

Experiments are performed at the K-edges of transition metal ions, where the frequency of the incident hard X-rays is tuned to match the energy of an atomic $1s$-$4p$ transition (5–10 keV). From the theoretical side it is clear that in this case the scattering intensity is related to the *charge dynamics* of the system under study [8–11]. Actually it was recently established that the RIXS intensity is proportional to the dielectric loss function of the *d*-electrons [12–15].

A recent breakthrough was achieved by J. P. Hill *et al.* [16], who observed that RIXS on the high-temperature superconductors $La_{2-x}Sr_xCuO_4$ and $Nd_{2-x}Ce_xCuO_4$ picks up *spin dynamics*. This triggers the question what kind of spin correlation function actually is measured in magnetic RIXS and how this is different from other probes of spin dynamics —in particular inelastic neutron scattering. The aim of this letter is to answer that question.

To this end the scattering amplitude for magnetic RIXS is expressed in terms of an intrinsic dynamic *four-spin correlation function* of the system that is probed. Moreover I derive selection rules that are related to the symmetry of the underlying spin Hamiltonian. It turns out that the first allowed magnetic scattering process is a two-magnon scattering one. As an example the formalism is used to calculate the indirect RIXS spectrum of Heisenberg antiferromagnets as function of transferred momentum $\mathbf{q}$. The scattering intensity vanishes at $(0,0)$ and —for the ordered Heisenberg antiferromagnet— also at the antiferromagnetic lattice vector $\mathbf{q} = (\pi, \pi)$. The scattering intensity around this particular point in the Brioullin zone is actually very sensitive to longer range magnetic couplings. These results are in agreement with the experimental observations of J. P. Hill and coworkers [16]. This illustrates the point that the correlation functions and selection rules of magnetic RIXS are very different from neutron scattering —a technique that in essence measures a *two-spin* correlation function.

These results in this paper I have first presented in the preprint ref. [17] and were later used and expanded upon by Nagao and Igarashi [18] to include magnon interaction

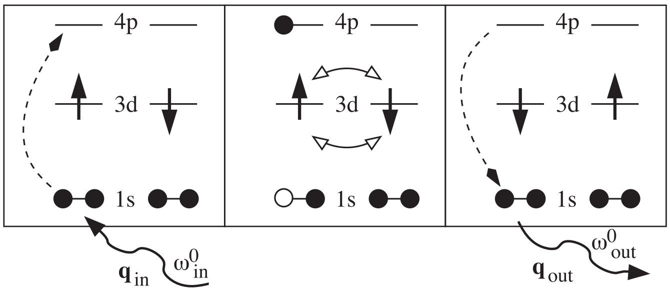

Fig. 1: Schematic representation of the magnetic RIXS scattering process at a transition metal K-edge. Left: the incoming photon (energy $\omega^0_{\rm in}$, momentum $\mathbf{q}_{\rm in}$) induces an electronic transition from a $1s$ to a $4p$ level. Middle: exchange interaction between $3d$ electrons in the presence of the core-hole. Right: de-excitation and outgoing photon ($\omega^0_{\rm out}$, $\mathbf{q}_{\rm out}$).

effects and by Forte *et al.* [19] to include ring-exchange processes, finite temperatures and higher-order scattering.

Series expansion of the scattering cross-section. – The probability for X-rays to be scattered from a solid-state system can be enhanced by orders of magnitude when the energy of the incoming photons is in the vicinity of an electronic eigenmode of the system —*i.e.* in the vicinity of a resonance. At a transition metal K-edge a $1s$ electron from the inner atomic core is excited into an empty $4p$ state, see fig. 1. In transition metal systems the empty $4p$ states are far (10–20 eV) above the Fermi level, so that the X-rays do not cause direct transitions of the $1s$ electron into the lowest $3d$-like conduction bands of the system. Still this technique is sensitive to low-energy excitations of the d-electrons because the Coulomb potential of $1s$ core-hole can couple to, *e.g.*, very low energy electron-hole excitations when the system is metallic. Since the charge excitations are caused by the core-hole, this scattering mechanism is sometimes referred to as *indirect* RIXS.

In this letter, however, I will consider insulating systems —in particular Mott insulators, where the only remaining low-energy degrees of freedom are the spin ones. So in order to determine the RIXS scattering amplitude one needs to establish how in this case the core-hole couples to *magnetic* excitations. The starting point is the Kramers-Heisenberg formula for the resonant scattering cross-section [20][1]

$$\left.\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega}\right|_{\rm res} \propto \sum_f |A_{fi}|^2\ \delta(\omega-\omega_{fi}),$$
$$\text{with}\quad A_{fi}=\omega_{\rm res}\sum_n \frac{\langle f|\hat{D}|n\rangle\langle n|\hat{D}|i\rangle}{\omega_{\rm in}-E_n-i\Gamma}, \qquad (1)$$

and f and i denote the final and initial state of the system, respectively. The sum over f is over all final states. The momentum and energy of the incoming/outgoing photons is $\mathbf{q}_{\rm in/out}$ and $\omega^0_{\rm in/out}$ and the loss energy $\omega=\omega^0_{\rm out}-\omega^0_{\rm in}$ is equal to the energy difference between the final and initial state $\omega_{fi}=E_f-E_i$. In the following the ground-state energy of the system will be taken as reference energy: $E_i=0$. In the scattering amplitude A_{fi} the resonant energy is $\omega_{\rm res}$, n denotes the intermediate states and $\hat{D}$ is the dipole operator that describes the excitation from initial to intermediate state and the de-excitation from intermediate to final state. The dipole operator is given in more detail in for instance ref. [12]. The energy of the incoming X-rays with respect to the resonant energy is $\omega_{\rm in}$ (this energy can thus either be negative or positive: $\omega_{\rm in}=\omega^0_{\rm in}-\omega_{\rm res}$) and E_n is the energy of intermediate state $|n\rangle$ with respect to the resonance energy. The last important detail is that the intermediate state is not a steady state. The highly energetic $1s$ core-hole quickly decays, *e.g.*, via Auger processes and the core-hole lifetime is very short. This leads to a core-hole energy broadening Γ which is proportional to the inverse core-hole lifetime.

To calculate RIXS amplitudes, I proceed by formally expanding the scattering amplitude in a power series [12]

$$A_{fi}=\frac{\omega_{\rm res}}{\Delta}\sum_{l=1}^{\infty}\frac{1}{\Delta^l}\langle f|\hat{D}(H_{\rm int})^l\hat{D}|i\rangle, \qquad (2)$$

where $\Delta=\omega_{\rm in}-i\Gamma$ and the Hamiltonian in the intermediate state $H_{\rm int}$ were introduced. For a further expansion of this scattering amplitude it is essential to split up the intermediate state Hamiltonian into two parts: $H_{\rm int}=H_0+H_1$, where H_0 is the Hamiltonian of the system without core-hole and H_1 the part of the Hamiltonian that is active in the presence of a core-hole.

Spin Hamiltonian with core-hole. – In the following I will calculate the magnetic resonant X-ray cross-section in a Mott-Hubbard insulator at zero temperature. The assumption is that this system is described by a single band Hubbard model at strong coupling and at half-filling. In this case the electrons are localized and the only low-energy degree of freedom is their spin. It is well known that in a Mott-Hubbard insulator the magnetic exchange integrals are determined by a virtual hopping process of electrons. Hopping amplitudes of the valence electrons are denoted by t_{ij} where i and j denote lattice sites with lattice vectors $\mathbf{R}_i$ and $\mathbf{R}_j$. The Coulomb interaction between electrons at the same site is U, so that in second-order perturbation theory one finds the exchange interaction $J_{ij}=2t_{ij}^2/U$ and the spin dynamics is governed by a Heisenberg spin Hamiltonian of the form

$$H_0=\sum_{i,j}J_{ij}\mathbf{S}_i\mathbf{S}_j=\sum_{\mathbf{k}}J_{\mathbf{k}}\ \mathbf{S}_{\mathbf{k}}\cdot\mathbf{S}_{-\mathbf{k}}, \qquad (3)$$

where $J_{\mathbf{k}}$ is the Fourier transform of J_{ij}. It is well known that for neutron scattering on such a spin system the *two-spin* correlation function $\sum_\alpha\int e^{-i\omega t}\langle S^\alpha_{\mathbf{q}}(0)S^\alpha_{-\mathbf{q}}(t)\rangle\mathrm{d}t$

[1]Note that where possible indices and constants are suppressed to avoid cluttering in the equations.

is measured, where the sum α is over the three spin components. We will see shortly that magnetic RIXS measures a very different *four-spin* correlation function.

In the intermediate state a core-hole is present. I assume the core-hole potential U_c to be local, *i.e.* as acting exclusively on those (valence) electrons that belong to the atom with the core-hole. When on site m a core-hole is present, the exchange interactions that involve the spin on site m become stronger, as the virtual intermediate state with two electrons on the site with the core-hole are lowered in energy by U_c. On the other hand, the virtual state with two holes present on site m is at $U+U_c$. Adding these two effects leads to the following exchange interaction between the spins on site m and j of

$$J^c_{mj} = 2t^2_{mj}\frac{U}{U^2-U_c^2} = (1+\eta)J_{mj} \tag{4}$$

and $\eta = \frac{U_c^2}{U^2-U_c^2}$. From this one obtains H_1, part of the intermediate state Hamiltonian that is active in the presence of a core-hole

$$H_1 = \eta\sum_{m,j} s_m s_m^\dagger J_{mj}\mathbf{S}_m\cdot\mathbf{S}_j, \tag{5}$$

where the operator s_m creates a core-hole on site m.

Spin-spin correlation function as measured in RIXS. – In order to finally obtain the magnetic cross-section, one needs to evaluate the operator $(H_{\rm int})^l$ in eq. (2). This is a non-trivial task. I first expand $(H_{\rm int})^l$ in a series that contains the leading terms to the scattering cross-section in lowest order in $\eta J/\Delta$. A conservative estimate gives, using for the copper K-edge $\Gamma\approx 1.5\,$eV and for the high-temperature superconductors $J\approx 125\,$meV and $U_c/U\approx 0.85$ [6,21], that at resonance $\eta J/\Delta\approx 0.22$. This makes it a suitable expansion parameter. Re-summing the leading-order terms in the series gives in the end the magnetic scattering amplitude

$$A_{fi} = \frac{\omega_{\rm res}}{\Delta}\frac{\eta}{\Delta-\omega}\langle f|\hat{O}_{\mathbf{q}}|i\rangle$$

$$\text{with}\ \ \hat{O}_{\mathbf{q}} = \sum_{\mathbf{k}} J_{\mathbf{k}}\ \mathbf{S}_{\mathbf{k}-\mathbf{q}}\cdot\mathbf{S}_{-\mathbf{k}}, \tag{6}$$

where the scattering operator is $\hat{O}_{\mathbf{q}}$ so that the magnetic correlation function that is measured in RIXS is proportional to the spin correlator $\int e^{-i\omega t}\langle\hat{O}_{\mathbf{q}}(0)\hat{O}_{-\mathbf{q}}(t)\rangle{\rm d}t$.

This expression is surprisingly simple and elegant. It shows that indeed momentum resolved indirect RIXS probes a momentum-dependent four-spin correlation function. From expression (6) it is immediately clear why experimentally the magnetic RIXS intensity vanishes at zero transferred momentum, *i.e.* at $\mathbf{q}=0$. In that case the correlation function is nothing but the steady-state Hamiltonian of the system ($\hat{O}_{\mathbf{q}=0}\propto H_0$). Thus $|i\rangle$ and $|f\rangle$ are eigenstates of this magnetic scattering operator, which makes inelastic scattering impossible[2]. This is in stark contrast with conventional two-magnon Raman scattering in the optical or UV range. That technique is also sensitive to a four-spin correlation function [22,23], but a quite different one. This is obvious considering the fact conventional Raman scattering is restricted to $\mathbf{q}=0$ —precisely the momentum transfer where RIXS vanishes. Therefore also these two techniques offer complementary information on spin dynamics.

Two-magnon scattering in antiferromagnets. – From eq. (6) immediately another selection rule follows. The projection of the total spin on the z-axis, $S^z_{tot}=\sum_i S^z_i$ commutes with both H_0 and $\hat{O}_{\mathbf{q}}$. Therefore S^z_{tot} is conserved during the scattering process, which implies that creation of a *single* magnon by the core-hole is not possible. But the creation of two magnons (with opposite z-projections) is allowed and this is therefore the lowest-order transversal spin scattering process that contributes to indirect RIXS. Also four-magnon scattering is in principle allowed, but it is of higher order and hence smaller, therefore it is not taken into account in the linear spinwave analysis that follows. Note that on the grounds of symmetry it is possible, in principle, to have magnetic scattering *without* creating any additional magnons in the scattering process. Physically this situation can only arise at finite temperature, when a magnon with momentum $\mathbf{k}$ that is present in the ground state is scattered to $\mathbf{k}+\mathbf{q}$ by the core-hole. This implies that magnetic RIXS has an interesting temperature dependence —which is, however, beyond the present scope [19].

I now apply the theory above to two-dimensional bipartite $S=1/2$ antiferromagnets and determine the two-magnon RIXS spectrum as a function of transferred momentum, at zero temperature. To this end, the Hamiltonian H_0 and correlation function $\hat{O}_{\mathbf{q}}$ are bosonized within linear spinwave theory, where $S^+_i\to a^\dagger_i$, $S^-_i\to a_i$ and $S^z_i\to\frac{1}{2}-n_i$, with boson creation/annihilation operators $a_i/a^\dagger_i$ and number operator $n_i=a^\dagger_i a_i$. After a Boguliobov transformation into the boson operators $\alpha_i/\alpha^\dagger_i$ one finds $\alpha^\dagger_{\mathbf{k}}=u_{\mathbf{k}}a^\dagger_{\mathbf{k}}+v_{\mathbf{k}}a_{-\mathbf{k}}$ with

$$u_{\mathbf{k}} = \sqrt{\frac{J_{\mathbf{k}=0}}{\epsilon_{\mathbf{k}}}+\frac{1}{2}},\qquad v_{\mathbf{k}} = {\rm sign}[{\rm J}_{\mathbf{k}}]\sqrt{\frac{J_{\mathbf{k}=0}}{\epsilon_{\mathbf{k}}}-\frac{1}{2}} \tag{7}$$

and $\epsilon_{\mathbf{k}}=2\sqrt{J^2_{\mathbf{k}=0}-J^2_{\mathbf{k}}}$, then the Hamiltonian reduces to $H_0^{LSW}=\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}\alpha^\dagger_{\mathbf{k}}\alpha_{\mathbf{k}}$. It is straightforward to show now that within linear spinwave theory the two magnon part of the magnetic scattering operator is

$$\begin{aligned}\hat{O}^{LSW}_{\mathbf{q}} \ =\ & \sum_{\mathbf{k}>0}\big[(J_{\mathbf{k}-\mathbf{q}/2}+J_{\mathbf{k}+\mathbf{q}/2})(u_{\mathbf{k}-\mathbf{q}/2}u_{\mathbf{k}+\mathbf{q}/2}\\ &+v_{\mathbf{k}-\mathbf{q}/2}v_{\mathbf{k}+\mathbf{q}/2})-(J_0+J_{\mathbf{q}})(u_{\mathbf{k}-\mathbf{q}/2}v_{\mathbf{k}+\mathbf{q}/2}\\ &+v_{\mathbf{k}-\mathbf{q}/2}u_{\mathbf{k}+\mathbf{q}/2})\big]\left(\alpha^\dagger_{\mathbf{k}-\mathbf{q}/2}\alpha^\dagger_{-\mathbf{k}-\mathbf{q}/2}+{\rm h.c.}\right),\end{aligned} \tag{8}$$

[2]This holds for the expansion in lowest order of $\eta J/\Delta$; higher-order terms might in principle give rise to some residual spin-related scattering intensity at $\mathbf{q}=0$.

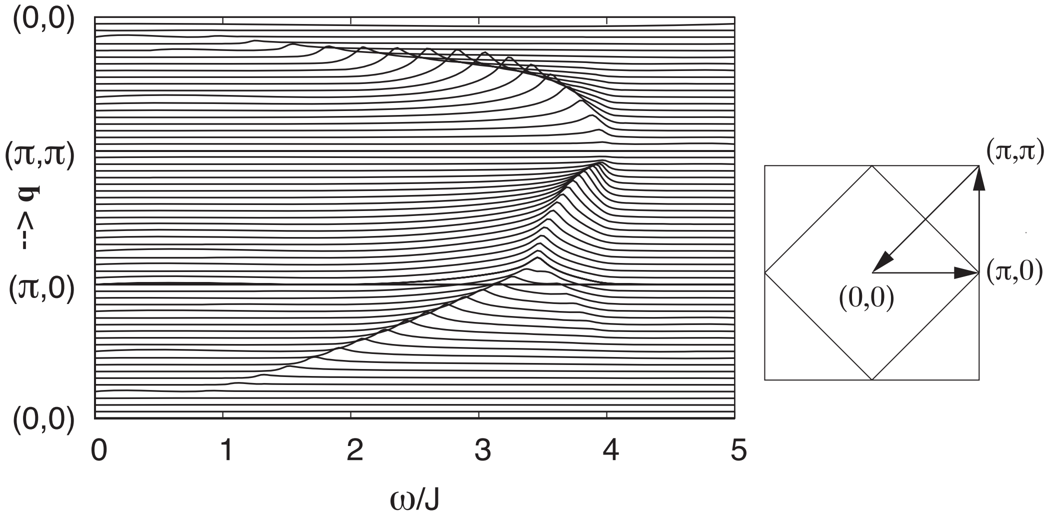

Fig. 2: Left: RIXS spectrum for a nearest-neighbor Heisenberg antiferromagnet with exchange interaction J as a function of transferred momentum $\mathbf{q}$ for a cut through the Brioullin zone that is shown on the right.

where $\mathbf{q}$ is the total momentum of the two-magnon excitation. The resulting RIXS spectrum is shown in fig. 2, for a cut through the Brioullin zone indicated by the right-hand side of the figure. There are several remarkable features in the spectrum.

First of all the spectral weight vanishes not only at $\mathbf{q} = (0, 0)$, but also at $\mathbf{q} = (\pi, \pi)$. This in agreement with the experimental observations [16]. It is straightforward to see that this selection rule is due to the antiferromagnetic ordering that occurs in the nearest-neighbor Heisenberg Hamiltonian. In the scattering operator (8), the terms $J_{\mathbf{k}-\mathbf{q}/2} + J_{\mathbf{k}+\mathbf{q}/2}$ and $J_0 + J_{\mathbf{q}}$ are strictly zero at $\mathbf{q} = (\pi, \pi)$. It is actually easy to show that in any bipartite magnetic system the RIXS intensity always vanishes at (π, π) if this scattering vector is a reciprocal lattice vector. This holds for instance also for a Heisenberg Hamiltonian with weak second and third neighbor exchange intereactions (J' and J'', respectively). This is illustrated by the calculation RIXS spectrum for an extended Heisenberg antiferromagnet with $J' = J'' = J/20$, shown in fig. 3. The longer-range couplings transfer spectral weight to scattering vectors around (π, π), but do not induce weight at precisely that wave vector.

The other remarkable feature of the magnetic RIXS spectrum is its strong dispersion. This is apparent from fig. 2 and the upper panel of fig. 3, which shows the first moment (average peak position) of the spectrum. The maximum possible energy of a two-magnon excitation is $4J$, and the matrix elements of magnetic RIXS put much of the scattering intensity at these high energies when it is kinematically allowed. The calculations for the nearest neighbor Heisenberg antiferromagnet (fig. 3) show that the magnetic scattering disperses from about $\omega \approx 0$ around $(0, 0)$ and (π, π) to $\omega \approx 4J$ at $(\pi, 0)$ and $(\pi/2, \pi/2)$. Longer-range couplings tend to reduce the first moment of the RIXS spectrum.

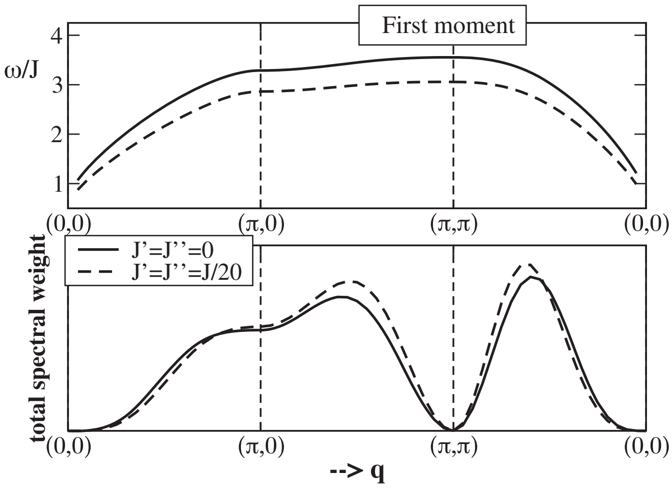

Fig. 3: Top: first moment of the RIXS spectrum and bottom: total spectral weight for a nearest-neighbor Heisenberg antiferromagnet (full line) and a Heisenberg model with in addition a second and third neighbor exchange (J' and J'', respectively) (dashed line).

The overall agreement between the theoretical results and the experimental data was already noted, where in particular the vanishing of spectral weight at particular momenta $\mathbf{q}$ and the general shape of the dispersion stand out. The data on La_2CuO_4 [16] show a maximum of the dispersion precisely where theory predicts it to be in momentum space, at around $\mathbf{q} = (\pi/2, \pi/2)$. Moreover, experimentally the peak is at an energy of around 500 meV, precisely where I find it on the basis of a nearest-neighbor Heisenberg model with $J = 135$ meV —a value that is consistent with the nearest-neighbor exchange determined by neutron scattering [24,25]. Higher-order exchange effects such as for instance the four-spin cyclic exchange, which are quite appreciable for this compound [25], will have to be taken into account in a more detailed analysis [19].

Conclusions. – In this letter I have reported on the first theoretical determination of the momentum-dependent four-spin correlation that is measured in magnetic RIXS. On the basis of this, the magnetic RIXS spectrum was calculated for Heisenberg antiferromagnets with short- and longer-range couplings. I derive selection rules that show that only scattering processes that involve at least two magnons are possible and that the scattering intensity vanishes at zero momentum transfer and at the antiferromagnetic lattice vector —which are observed in experiment. Moreover, there is quantitative agreement between theory and experiment on the two-magnon peak position at $\mathbf{q} = (\pi/2, \pi/2)$. These results show that RIXS is in principle a powerful tool to obtain new information on spin dynamics —information that is complementary to what can be obtained by other techniques such as neutron, non-resonant X-ray or conventional two-magnon Raman scattering.

* * *

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