Indirect resonant inelastic X-ray scattering on magnons

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Recent experiments show that indirect resonant inelastic X-ray scattering (RIXS) is 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. The RIXS spectrum of Heisenberg antiferromagnets is calculated. 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 \( q \), in particular for \( q = 0 \). The calculated spectra agree very well with the 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 \[1\]. As a scattering technique RIXS has two important advantages. First, it is sensitive to excitations that are difficult to observe by otherwise, for example direct \( d-d \) excitations in cuprates or manganites \[2, 3, 4, 5, 6, 7\]. Second, it probes directly both the energy and momentum dependence of such excitations \[8, 9, 10, 11\]. The scattering intensity vanishes at \( (0,0) \) vanishes, as well as the longitudinal spin one – which is in fact a higher order charge response function. This implies that we need take the ultra-short lifetime expansion \[12, 13\] one step further. By doing so I find the scattering amplitude for magnetic RIXS, expressed in terms of an intrinsic dynamic four-spin correlation function of the system that is probed. Moreover I derive selection rules which 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 \( q \). The scattering intensity vanishes at \((0,0)\) and at the antiferromagnetic wavevector \( q = (\pi, \pi) \). The computed spectra all agree with the experimental data.

**FIG. 1:** Schematic representation of the magnetic RIXS scattering process at a transition metal K-edge. Left: the incoming photon (energy \( \omega^\text{in} \), momentum \( q^\text{in} \)) 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 (\( \omega^\text{out}, q^\text{out} \)).
empty 4\textit{p} state, see Fig. 11. In transition metal systems the empty 4\textit{p} states are far (10-20 eV) above the Fermi-level, so that the X-rays do not cause direct transitions of the 1\textit{s} electron into the lowest 3d-like conduction bands of the system. Still this technique is sensitive to low energy excitations of the \textit{d}-electrons because the Coulomb potential of 1\textit{s} 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, we 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 we need to establish how in this case the core-hole couples to magnetic excitations. Our starting point is the Kramers-Heisenberg formula for the resonant scattering cross section [12,10]

\[
\frac{d^2\sigma}{d\Omega d\omega_{\text{res}}} \propto \sum_{f} |A_{fi}|^2 \delta(\omega - \omega_{fi}), \quad \text{with}
\]

\[
A_{fi} = \omega_{\text{res}} \sum_{n} \frac{\langle f|\hat{D}|n\rangle\langle n|\hat{D}|i\rangle}{\omega_{in} - E_{n} - i\Gamma}. \tag{1}
\]

and \textit{f} and \textit{i} denote the final and initial state of the system, respectively. The sum is over \textit{f} is over all final states. The momentum and energy of the incoming/outgoing photons is \(\omega_{\text{in/out}}\) and the energy \(\omega_{in} - \omega_{out}\) is equal to the energy difference between the final and initial state \(E_{f} - E_{i}\). In the following we will take the groundstate energy of our system as reference energy: \(E_{i} = 0\). In the scattering amplitude \(A_{fi}\) the resonant energy is \(\omega_{\text{res}}\), \textit{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 \(\omega_{in}\) (this energy can thus either be negative or positive: \(\omega_{in} = \omega_{in}^0 - \omega_{\text{res}}\)) and \(E_{in}\) 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 1\textit{s} core-hole quickly decays e.g. via Auger processes and the core-hole life-time is very short. This leads to a core-hole energy broadening \(\Gamma\) which is proportional to the inverse core-hole life-time.

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

\[
A_{fi} = \frac{\omega_{\text{res}}}{\Delta} \sum_{l=1}^{\infty} \frac{1}{\Delta^l} \langle f|\hat{D}(\hat{H}_{\text{int}})^l|\hat{D}|i\rangle \tag{2}
\]

where we introduced \(\Delta = \omega_{in} - i\Gamma\) and the Hamiltonian \(\hat{H}_{\text{int}}\) in the intermediate state. For a further expansion of this scattering amplitude it is essential that we split up the intermediate state Hamiltonian into two parts: \(H_{\text{int}} = H_{0} + H_{1}\), 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.** We will calculate the resonant X-ray cross section in a Mott-Hubbard insulator at zero temperature. We assume 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. We denote the hopping amplitudes of the valence electrons 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 we have the exchange interaction \(J_{ij} = 2t_{ij}^2/U\) and the spin dynamics is governed by a Heisenberg spin Hamiltonian of the form

\[
H_{0} = \sum_{i,j} J_{ij} S_{i} S_{j} = \sum_{k} J_{k} S_{k} \cdot S_{-k}, \tag{3}
\]

where \(J_{k}\) 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 \(\sum_{\alpha} \int e^{-i\omega t} \langle S_{\alpha}(0)S_{\alpha}^{\alpha}(t)\rangle dt\) is measured, where the sum \(\alpha\) 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. We 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 \(m\) becomes 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

\[
J_{m,j}^c = 2t_{m,j}^2 U_{c} U_{2}^{1/2} = (1 + \eta)J_{mj} \tag{4}
\]

and \(\eta = \frac{U^2}{U_{c} U_{2}}\). From this we obtain \(H_{1}\), part of the intermediate state Hamiltonian that is active in the presence of a core-hole

\[
H_{1} = \eta \sum_{m,j} s_{m} s_{m}^{\dagger} J_{m,j} S_{m} \cdot S_{j}, \tag{5}
\]

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, we need to evaluate the operator \((\hat{H}_{\text{int}})^l\) in equation (2).

This is a non-trivial task. We first expand \((H_{\text{int}})^I\) 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 \(\Gamma \approx 125\) meV and \(U_c/U \approx 0.85\) \cite{6 17}, 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

\[
A_{fi} = \frac{\omega_{\text{res}}}{\Delta} \frac{\eta}{\Delta - \omega} (f|\hat{O}_q|i) \tag{6}
\]

where we find that the scattering operator \(\hat{O}_q\) to be

\[
\hat{O}_q = \sum_k J_k \vec{S}_{k-q} \cdot \vec{S}_{-k}. \tag{7}
\]

so that the magnetic correlation function that is measured in RIXS is proportional to the spin correlator \(\int e^{-i\omega t} \langle \hat{O}_q(0)\hat{O}_{-q}(t) \rangle dt\).

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 \((7)\) it is immediately clear why experimentally the magnetic RIXS intensity vanishes at zero transferred momentum, i.e. at \(q = 0\). In that case the correlation function is nothing but the steady state Hamiltonian of the system \((\hat{O}_{q=0} \propto H_0)\). Thus \(|i\rangle\) and \(|f\rangle\) are eigenstates of this magnetic scattering operator, which makes inelastic scattering impossible \cite{18}. 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 \cite{19}, but a quite different one. This is obvious considering the fact conventional Raman scattering is restricted to \(q = 0\) —precisely the momentum transfer where RIXS vanishes. Therefore also these two techniques offer complementary information on spin dynamics.

**Two-magnon scattering in antiferromagnets.** From equation \((7)\) immediately another selection rule follows. The projection of the total spin on the \(z\)-axis, \(S^z_{\text{tot}} = \sum_i S^z_i\) commutes with both \(H_0\) and \(\hat{O}_q\). Therefore \(S^z_{\text{tot}}\) 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 of higher order and therefore smaller and 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 \(k\) that is present in the groundstate is scattered to \(k + q\) by the core-hole. This implies that magnetic RIXS has an interesting temperature dependence—which is, however, beyond our present scope.

We 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 \(\hat{O}_q\) are bosonized within linear spinwave theory, where \(S^z_i \to a^+_i S^z_{\alpha} \to a_i\) and \(S^z_{\alpha} \to \frac{1}{2} - n_i\), with boson creation/annihilation operators \(a_i/a^+_i\) and number operator \(n_i = a^+_i a_i\). After a Bogoliubov transformation into the boson operators \(\alpha_i/\alpha^+_i\) we have \(\alpha^+_i = u_k a^+_k + v_k a_{-k}\) with

\[
u_k = \sqrt{\frac{J_{k=0}}{\epsilon_k}} + \frac{1}{2}, \quad u_k = \text{sign}[J_k] \sqrt{\frac{J_{k=0}}{\epsilon_k} - \frac{1}{2}} \tag{8}
\]

and \(\epsilon_k = 2\sqrt{J_{k=0} J_k}\), then the Hamiltonian reduces to

\[
H^{\text{SW}}_0 = \sum_k \epsilon_k \alpha^+_k \alpha_k. \quad \text{It is straightforward to show now that within linear spinwave theory the two magnon part of the magnetic scattering operator is}
\]

\[
\hat{O}_q^{\text{SW}} = \sum_{k>0} (J_{k-q/2} + J_{k+q/2})(u_{k-q/2} u_{k+q/2} + v_{k-q/2} v_{k+q/2})
\]

\[
+ \Big(v_{k-q/2} v_{k+q/2} - (J_0 + J_{q/2}) (u_{k-q/2} u_{k+q/2} + v_{k-q/2} v_{k+q/2})
\]

\[
+ u_{k-q/2} u_{k+q/2}) \left(\alpha_{k-q/2} \alpha_{k-q/2} + \text{h.c.}\right) \tag{9}
\]
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 Brillouin 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 at \( \mathbf{q} = (0, 0) \) and \( \mathbf{q} = (\pi, \pi) \). This is in agreement with the experimental observations [14]. From the scattering operator (9) it one sees that this selection rule is due to the antiferromagnetic ordering that occurs in the Heisenberg Hamiltonian. It is actually easy to show that the RIXS intensity always vanishes at \( (\pi, \pi) \) if this scattering vector is also a reciprocal lattice vector. This holds for instance also for a Heisenberg Hamiltonian with weak second and third neighbor exchange interactions \( (J'\text{ and } J'') \), respectively, which 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 \( (\pi, \pi) \), but do not induce weight at precisely that wavevector.

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 calculations for the nearest neighbor Heisenberg antiferromagnet (Fig. 3) show that the magnetic scattering disperses from about \( \omega \approx 0 \) around \( (0, 0) \) and \( (\pi, \pi) \) 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. The observed dispersion has a two-fold origin. It is in part due to the \( \mathbf{q} \)-dependence of the two-magnon density of states (DOS), combined with the scattering matrix elements that tend to pronounce the low energy tails of the two-magnon DOS.

The consistency at \( \mathbf{q} = (0, 0) \) and \( \mathbf{q} = (\pi, \pi) \) of the theoretical results and experimental data was already noted, but at other wave-vectors the agreement stands out even more. The data on \( \text{La}_2\text{CuO}_4 \) shows for \( \mathbf{q} = (\pi, 0) \) a peak at around 500 meV, precisely where we find it on the basis of a nearest neighbor Heisenberg model with \( J = 146 \text{ meV} \) – a value also found by the analysis of neutron scattering data [20]. Similar agreement is found at \( \mathbf{q} = (0.6\pi, 0) \) and \( \mathbf{q} = (0.6\pi, 0.6\pi) \).

**Conclusions.** We determined 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. We derive selection rules 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 theory and experiment agree very well on the two-magnon peak position and spectral weight throughout the measured Brioullin zone. 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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