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Correlation functions measured by indirect resonant inelastic X-ray scattering

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Abstract. – We express the cross-section for indirect resonant inelastic X-ray scattering in terms of an intrinsic dynamic correlation function of the system that is studied with this technique. We find that the cross-section is a linear combination of the charge response function and the dynamic longitudinal spin density correlation function of the electrons that couple to the core-hole. This result is asymptotically exact for both strong and weak local core-hole potentials. The relative charge and spin contribution to the inelastic spectral weight can be changed by varying the incident photon energy.

Introduction. – 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. Such resonant X-ray scattering experiments [1] are performed on, *e.g.*, the K-edges of transition metal ions, where the frequency of the X-rays is tuned to match the energy of an atomic $1s$-$4p$ transition, which is around 5–10 keV [2–7]. At this resonant energy 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 excitations of electrons near the Fermi level. The strong Coulomb potential of the $1s$ core-hole causes, *e.g.*, very low-energy electron-hole excitations in the valence/conduction band: the core-hole potential is screened by the valence electrons. When the excited $4p$-electron recombines with the $1s$ core-hole and the outgoing photon is emitted, the system can therefore be left behind in an excited final state. Experimentally, the momentum $\boldsymbol{q}$ and energy ω of the elementary excitation are determined from the difference in energy and momentum between incoming and outgoing photons. Since the excitations are caused by the core-hole, we refer to this scattering mechanism as *indirect* resonant inelastic X-ray scattering (RIXS).

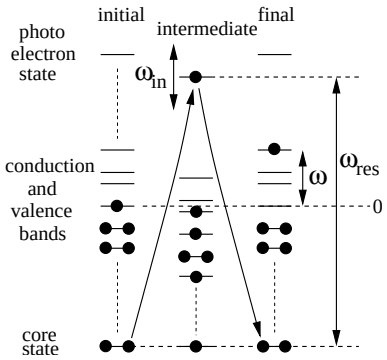

Fig. 1 – Schematic representation of the indirect resonant inelastic X-ray scattering (RIXS) process.

RIXS is rapidly developing due to the recent increase in brilliance of the new-generation synchrotron X-ray sources. High flux photon beams with energies that are tunable to resonant edges are now becoming available. At present an energy resolution in the order of 100 meV can be reached. In the near feature it seems experimentally feasible for RIXS to become sensitive to the low-energy excitations of the solid, where excitation energies are of the order of room temperature. Such low-lying electronic excitations can, for example, be collective features such as plasmons, magnons, orbitons, excitons and single-particle–like continua related to the band structure. RIXS provides a new tool to study these elementary excitations.

For the interpretation of spectroscopic data, it is very important to express the scattering cross-section for a technique in terms of physical correlation functions. It is well known, for example, that in standard optical experiments the charge density response function (or dielectric function) at $\boldsymbol{q} \simeq 0$ is measured. In electron energy loss experiments this linear response function is measured at finite transferred momentum.

In this paper, we determine the dynamical correlation function that is measured in indirect resonant inelastic X-ray scattering. For a local core-hole potential, the dynamical correlation function turns out to be a linear combination of the charge density and longitudinal spin density response function. We show that the actual linear combination that is measured depends on the energy of the incoming photons and we determine the precise energy dependence of its coefficients. The expressions for the scattering cross-section that we derive are asymptotically exact in the two limits of either a very strong or a very weak local core-hole potential. Moreover, our approach permits a controlled expansion away from these two limits.

Series expansion of the scattering cross section. – The Kramers-Heisenberg formula [8–10] for the resonant X-ray scattering cross-section is [11]

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

where A_{fi} is the scattering amplitude from the initial (i) to a final (f) state of the system. The sum is over all final states. The momentum and energy of the incoming/outgoing photons is $\boldsymbol{q}_{\mathrm{in/out}}$ and $\omega^0_{\mathrm{in/out}}$ and the loss energy $\omega = \omega^0_{\mathrm{out}} - \omega^0_{\mathrm{in}}$ is equal to the energy difference between the final and initial state $\omega_{fi} = E_f - E_i$. In the following, we will take the energy of the initial state as reference energy: $E_i \equiv 0$. The resonant energy is ω_{res}, n denotes the intermediate states and $\hat{D}$ the (dimensionless) dipole operator that describes the excitation from initial to intermediate state and the de-excitation from intermediate to final state. 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: $\omega_{\rm in} = \omega_{\rm in}^0 - \omega_{\rm res}$ and at resonance when the incoming photon energy is tuned precisely to the K-edge $\omega_{\rm in} = 0$). E_n is the energy of the intermediate state $|n\rangle$ with respect to the resonance energy.

In the intermediate state a core-hole and a photo-excited electron are present. When we take the Coulomb interaction between the intermediate-state core-hole and the valence band electrons into account, we obtain a finite inelastic scattering amplitude. In that case there is a non-zero probability that the system ends up in a final state with an electron-hole excitation in the valence/conduction band, see fig. 1.

The intermediate state, however, 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. The Heisenberg time-energy uncertainty relationship then implies that the core-hole energy has an appreciable uncertainty. This uncertainty appears in the formalism above as the core-hole energy broadening Γ which is proportional to the inverse core-hole lifetime. Note that the lifetime broadening only appears in the intermediate states and not in the final or initial states as these both have very long lifetimes. This implies that the core-hole broadening does not present an intrinsic limit to the experimental resolution of RIXS: the loss energy ω is completely determined by kinematics.

In a resonant scattering process, the measured system is generally strongly perturbed. Formally this is clear from the Kramers-Heisenberg formula (1), in which both the energy and the wave function of the intermediate state —where a strongly perturbing core-hole is present— appear. In canonical optical/electron energy loss experiments the probing photon/electron is a weak perturbation and a measurement is fully described within linear response theory. The response to a core-hole in RIXS, however, is highly non-linear. So it might seem that the only possibility for determining RIXS amplitudes that one is left with is to evaluate the Kramers-Heisenberg expression numerically. For this all initial-, intermediate- and final-state energies and wave functions need to be known exactly, so that in practice a direct evaluation is only possible for systems that, for example, consist of a small cluster of atoms [12].

In this letter, however, we present a method to calculate the RIXS response functions without immediately resorting to numerics. We show that under the appropriate conditions one can integrate out the intermediate states from the Kramers-Heisenberg expression. After doing so, we can directly relate RIXS amplitudes to linear charge and spin response functions of the unperturbed system. For non-resonant scattering, one is familiar with the situation that the scattering intensity is proportional to a linear response function. Such is also expected for resonant scattering if one assumes the perturbing core-hole potential to be weak [13–15], but it is a quite unexpected result for the physical situation in which the core-hole potential is (very) strong.

Let us proceed by formally expanding the scattering amplitude in a power series

$$A_{fi} = \frac{\omega_{\rm res}}{\omega_{\rm in} - i\Gamma} \sum_{l=0}^{\infty} M_l, \qquad \text{with } M_l = \sum_n \left(\frac{E_n}{\omega_{\rm in} - i\Gamma} \right)^l \langle f|\hat{D}|n\rangle\langle n|\hat{D}|i\rangle, \tag{2}$$

where we introduced the scattering matrix elements M_l. We denote the denominator of the expansion parameter as $\Delta = \omega_{\rm in} - i\Gamma$, which is a complex number. Its minimum $|\Delta| = \Gamma$ is reached when the incoming photon energy is tuned precisely to the K-edge resonance. Expanding the scattering matrix elements we find that

$$M_l = \frac{1}{\Delta^l} \sum_n \langle f|\hat{D}|n\rangle (E_n)^l \langle n|\hat{D}|i\rangle = \frac{1}{\Delta^l} \langle f|\hat{D}(H_{\rm int})^l \hat{D}|i\rangle, \tag{3}$$

where $H_{\rm int}$ is the Hamiltonian in the intermediate state. We thus obtain the following series expansion for the resonant cross-section:

$$\left.\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega}\right|_{\rm res} = \mathrm{const}\sum_f \left|\frac{\omega_{\rm res}}{\Delta}\sum_{l=0}^{\infty} M_l\right|^2 \delta(\omega-\omega_{fi}). \tag{4}$$

The formal radius of convergence of this power series is given by $E_n^2/(\omega_{\rm in}^2+\Gamma^2)$, so that the series is obviously convergent when the incoming X-ray energy is far enough below the resonance, *i.e.* when $|\omega_{\rm in}| \gg 0$. But also at resonance, when $\omega_{\rm in}=0$ the series is convergent for intermediate energies that are smaller than the core-hole broadening Γ. Formal convergence conditions, however, need not distract us as in the following we will be performing complete re-summations of this series.

Spinless fermions. – We first calculate the resonant X-ray cross-section in the case where the valence and conduction electrons are effectively described by a single band of spinless fermions: spin, and orbital degrees of freedom of the valence electron system are suppressed. Physically, this situation can be realized in a fully saturated ferromagnet. The final and initial states of the system are then determined by a Hamiltonian H_0 that describes the electrons around the Fermi level. The generic form of the full many-body Hamiltonian is

$$H_0 = \sum_{i,j} t_{ij}(c_i^\dagger c_j + c_j^\dagger c_i) + c_i^\dagger c_i V_{ij} c_j^\dagger c_j, \tag{5}$$

where i and j denote lattice sites with lattice vectors $\boldsymbol{R}_i$ and $\boldsymbol{R}_j$ and i,j range from 1 to N, where N is the number of sites in the system. The hopping amplitudes of the valence electrons are denoted by t_{ij} and the $c/c^\dagger$-operators annihilate/create such electrons. The Coulomb interaction between valence electrons is $V_{ij}=V_{|\boldsymbol{R}_i-\boldsymbol{R}_j|}$, as the Coulomb interaction only depends on the relative distance between two particles.

The intermediate states are eigenstates of the Hamiltonian $H_{\rm int}=H_0+H_{\rm core}$, where $H_{\rm core}$ accounts for the Coulomb coupling between the intermediate state core-hole and the valence electrons:

$$H_{\rm core} = \sum_{i,j} s_i s_i^\dagger V_{ij}^c c_j^\dagger c_j, \tag{6}$$

where s_i creates a core-hole on site i. We assume that the core-hole is fully localized and has no dispersion. We will see shortly that this leads to major simplifications in the theoretical treatment of indirect RIXS. The core-hole – valence electron interaction is attractive: $V^c<0$. The dipole operators are given by

$$\hat{D} = \sum_i e^{-i\boldsymbol{q}_{\rm in}\cdot\boldsymbol{R}_i} s_i p_i^\dagger + e^{i\boldsymbol{q}_{\rm out}\cdot\boldsymbol{R}_i} s_i^\dagger p_i + \mathrm{h.c.}, \tag{7}$$

where $p^\dagger$ creates a photo-excited electron in a $4p$ state and h.c. denotes the Hermitian conjugate of both terms.

In order to calculate the cross-section, we need to evaluate the operator $(H_{\rm int})^l$ in eq. (3). A direct evaluation of this operator is complicated by the fact that the initial- and intermediate-state Hamiltonians do not commute. We therefore proceed by expanding $(H_{\rm int})^l$ in a series that contains the leading terms to the scattering cross-section for both strong and weak core-hole potentials. After that we will do a full re-summation of that series. We expand

$$(H_{\rm int})^l\hat{D}|i\rangle = (H_0+H_{\rm core})^l\hat{D}|i\rangle \simeq \sum_{m=0}^{l}(H_0)^m(H_{\rm core})^{l-m}\hat{D}|i\rangle, \tag{8}$$

where we used $[H_0, \hat{D}] = 0$ and $H_0|i\rangle \equiv 0$. For strong core-hole potentials the expansion is dominated by the $m = 0$ term $(H_{\rm core})^l$. Also for weak core-hole potentials we obtain the correct leading terms. It is straightforward to show that for a strong core-hole potential the first correction to the expansion above is smaller by a factor t/Γ and for a weak core-hole potential by a factor V^c/Γ. Thus, the expansion is well behaved as in the appropriate limits the leading-order corrections are vanishing. We can estimate these parameters for, *e.g.*, the high-T_c superconductors. At the copper K-edge $\Gamma_{\rm Cu} \simeq 1.5\,{\rm eV}$ and the local core-hole potential for the $3d$ electrons is $V_d^c \simeq 7.5\,{\rm eV}$ with an effective hopping of $t_{dd} \simeq 0.5\,{\rm eV}$, which clearly brings this system in the limit of a strong core-hole potential [4].

Let us for the moment consider the strong core-hole potential limit and keep in the expansion only the term $m = 0$. Due to the fact that in the intermediate state only one core-hole is present and that this core-hole has no dispersion, we have

$$M_l(V^c \gg t) = \frac{1}{\Delta^l}\langle f| \sum_i e^{i\boldsymbol{q}\cdot\boldsymbol{R}_i} \left(\sum_j V_{ij}^c c_j^\dagger c_j \right)^l |i\rangle, \tag{9}$$

where the transfered momentum $\boldsymbol{q} \equiv \boldsymbol{q}_{\rm out} - \boldsymbol{q}_{\rm in}$.

The first important observation is that the term $l = 0$ does not contribute to the inelastic X-ray scattering intensity because $M_0 = \langle f| \sum_i e^{i\boldsymbol{q}\cdot\boldsymbol{R}_i}|i\rangle = N\delta_{\boldsymbol{q},0}\delta_{f,i}$, which only contributes to the elastic scattering intensity at $\boldsymbol{q} = \boldsymbol{0}$ and other multiples of the reciprocal lattice vectors. From inspection of eq. (2) we see immediately that the $l = 0$ term actually vanishes irrespective of the strength of the core-hole potential. This is of relevance when we consider the scattering cross-section in the so-called "fast-collision approximation" [16]. This approximation corresponds to the limit where the core-hole lifetime broadening is the largest energy scale in system ($\Gamma \to \infty$ so that $|\Delta| \to \infty$). In this limit only the $l = 0$ term contributes to the indirect RIXS amplitude and thus the resonant inelastic signal vanishes. In any theoretical treatment of indirect resonant scattering one therefore needs to go beyond the fast-collision approximation [17].

The second observation is that M_l is a l-particle correlation function. If one measures far away from resonance, where $|\Delta|$ is large (because of a large $|\omega_{\rm in}|$) the scattering cross-section is dominated by the $l = 1$, one-particle density response function. When the incoming photon energy approaches the resonance, gradually the two, three, four etc. particle response functions add more and more spectral weight to the inelastic scattering amplitude. Generally, these multi-particle response functions interfere. The main point of this paper is that these multi-particle correlation functions collapse onto a single dynamic single particle (charge) and two-particle (spin) correlation function when the local core-hole potential is local.

Now we determine the general charge/spin response functions that are measured in RIXS. Such only is possible because it is justified in hard X-ray electron spectroscopies to consider the core-hole potential to be local. This corresponds to the widely used Anderson impurity approximation in the theoretical analysis of, *e.g.*, X-ray absorption and photo-emission, introduced in refs. [18–20]. This approximation is reasonable as the Coulomb potential is certainly largest on the atom where the core-hole is located and a deep core-hole has no appreciable dispersion. By inserting the local core-hole potential $V_{ij}^c = U_c\delta_{ij}$ in eq. (8) we find after performing the sum over m the scattering matrix elements

$$M_l^{sf} = \frac{1}{\Delta^l}\frac{U_c^l - E_f^l}{1 - E_f/U_c}\langle f| \sum_i e^{i\boldsymbol{q}\cdot\boldsymbol{R}_i} c_i^\dagger c_i |i\rangle. \tag{10}$$

Using that $\sum_i e^{i\boldsymbol{q}\cdot\boldsymbol{R}_i} c_i^\dagger c_i = \sum_{\boldsymbol{k}} c_{\boldsymbol{k}+\boldsymbol{q}}^\dagger c_{\boldsymbol{k}} \equiv \rho_{\boldsymbol{q}}$ is the density operator, we now perform the sum

over l in eq. (2) and obtain

$$A_{fi}^{sf} = P_1(\omega, U_c)\langle f|\rho_{\boldsymbol{q}}|i\rangle, \quad \text{with } P_1(\omega, U_c) \equiv \frac{U_c\ \omega_{\rm res}}{(\Delta - U_c)(\Delta - \omega)}, \tag{11}$$

where we defined the resonant enhancement factor $P_1(\omega, U_c)$ and $\omega = E_f$. Thus, for spinless fermions with a local core potential, the indirect RIXS cross-section thus turns out to be the density response function with a resonant prefactor $P_1(\omega)$ that depends on the loss energy ω, on the distance from resonance $\omega_{\rm in}(= {\rm Re}[\Delta])$, on the core-hole potential U_c and on the core-hole lifetime broadening $\Gamma(= -{\rm Im}[\Delta])$. The density response function is related to the dielectric function $\epsilon(\boldsymbol{q}, \omega)$ and the dynamic structure factor $S(\boldsymbol{q}, \omega)$ [21], so that for fixed U_c we obtain

$$\left.\frac{{\rm d}^2\sigma}{{\rm d}\Omega{\rm d}\omega}\right|_{\rm res}^{sf} \propto -|P_1(\omega)|^2\ {\rm Im}\ \left[\frac{1}{V_{\boldsymbol{q}}\epsilon(\boldsymbol{q}, \omega)}\right] \propto |P_1(\omega)|^2\ S(\boldsymbol{q}, \omega). \tag{12}$$

It is important to note that $S(\boldsymbol{q}, \omega)$ is the dynamic structure factor *of the electrons that couple to the core-hole*. So for RIXS at a transition metal K-edge, $S(\boldsymbol{q}, \omega)$ is related to the charge density response function of the $3d$ electrons only [22]. The observation that the excitation spectra depend on the reduced transferred momentum $\boldsymbol{q}$ and *not* on the total momentum $\boldsymbol{q} + \boldsymbol{G}$ ($\boldsymbol{G}$ being a reciprocal lattice vector) is a strong experimental evidence for this [23]. The ability to probe the response of a certain electronic subsystem is unique for RIXS.

Fermions with spin. – We generalize the calculation above to the situation where the electrons have an additional spin degree of freedom. In the Hamiltonians (5), (6) we now include a spin index σ (with $\sigma =\uparrow$ or $\downarrow$) to the annihilation and creation operators: $c_i \to c_{i\sigma}$ and $c_j \to c_{j\sigma'}$ and sum over these indices, taking into account that the hopping part of the Hamiltonian is diagonal in the spin variables. In order to re-sum the series in eq. (2) we now need to evaluate expansions of the number operators of the kind $(n_\uparrow + n_\downarrow)^l$. Using that for $l \geq 1$

$$(n_\uparrow + n_\uparrow)^l = n_\uparrow + n_\downarrow + (2^l - 2)n_\uparrow n_\downarrow, \tag{13}$$

we obtain

$$A_{fi} = [P_1(\omega) - P_2(\omega)]\langle f|\boldsymbol{S}_{\boldsymbol{q}}^2|i\rangle + P_2(\omega)\langle f|\rho_{\boldsymbol{q}}|i\rangle, \tag{14}$$

with $P_2(\omega, U_c) = P_1(2\omega, 2U_c)/2$ and $\rho_{\boldsymbol{q}} = \rho_{\boldsymbol{q}}^\uparrow + \rho_{\boldsymbol{q}}^\downarrow$. Here we also introduced the longitudinal spin density correlation function $\boldsymbol{S}_{\boldsymbol{q}}^2 \equiv \frac{1}{S(S+1)}\sum_{\boldsymbol{k}} \boldsymbol{S}_{\boldsymbol{k}+\boldsymbol{q}} \cdot \boldsymbol{S}_{-\boldsymbol{k}}$. Transversal spin fluctuations will be considered elsewhere [24]. Clearly, the contributions to the scattering rate from the dynamic longitudinal spin correlation function and the density correlation function need to be treated on equal footing as they interfere [25]. Moreover, the spin and charge correlation functions have different resonant enhancements. For instance, when ${\rm Re}[\Delta] = U_c$, the scattering amplitude is dominated by $P_1(\omega)$ and hence by the longitudinal spin response function. At incident energies where ${\rm Re}[\Delta] = 2U_c$, on the other hand, $P_2(\omega)$ is resonating so that the contributions to the inelastic scattering amplitude of charge and spin are approximately equal.

Conclusions. – We presented a series expansion of the indirect resonant inelastic X-ray scattering amplitude, which is asymptotically exact for both small and large local core-hole potentials. By re-summing the series, we find the dynamical charge and spin correlation functions that are measured in RIXS. It is shown that, specifically, for resonant scattering at a transition metal K-edge the charge and spin response functions of the $3d$ electrons is measured.

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