Ultrashort lifetime expansion for indirect resonant inelastic x-ray scattering

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In indirect resonant inelastic x-ray scattering (RIXS) an intermediate state is created with a core-hole that has an ultrashort lifetime. The core-hole potential therefore acts as a femtosecond pulse on the valence electrons. We show that this fact can be exploited to integrate out the intermediate states from the expression for the scattering cross section. By doing so we obtain an effective scattering cross section that only contains the initial and final scattering states. This effective cross section turns out to be a linear combination of the charge response function $S(\mathbf{q},\omega)$ and the dynamic longitudinal spin density correlation function, both with a resonant prefactor. This result is asymptotically exact for both strong and weak local core-hole potentials and ultrashort lifetimes. The resonant scattering prefactor is shown to be weakly temperature dependent. We also derive a sum rule for the total scattering intensity and generalize the results to multiband systems. One of the remarkable outcomes is that one can change the relative charge and spin contribution to the inelastic spectral weight by varying the incident photon energy.

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I. INTRODUCTION

Resonant inelastic x-ray scattering (RIXS) is a technique that develops rapidly due to the recent increase in brilliance of the new generation synchrotron x-ray sources, where high flux photon beams with energies that are tunable to resonant edges are now becoming widely available.[1] 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—a resonant edge—of the system. RIXS experiments 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.[2–15] At this resonant energy a $1s$ electron from the inner atomic core is excited into an empty $4p$ state, see Fig. 1.

It is a well-known fact that the $1s$ core-hole that is created in this process has an ultrashort lifetime, of the order of femtoseconds. The reason is that the core-hole has a very high energy and is prone to decay via all sorts of radiative and nonradiative processes, severely cutting down the efficiency of RIXS. In the canonical theoretical treatments of RIXS this lifetime effect is normally introduced as a core-hole broadening and disregarded from that point on.

In a previous study,[16] however, we have shown that from the theory perspective there is a great advantage to the extremely short lifetime of the core-hole. The ultrashort lifetime implies that for the electrons in the solid—particularly for the slow ones that are close to the Fermi energy—the core-hole potential is almost an instantaneous delta function in time. Although the core-hole potential by itself can be large and therefore a strong perturbation to the electrons, the very short duration of this perturbing potential allows for a systematic expansion of the scattering cross section in terms of the core-hole lifetime. Here we present a detailed derivation and various generalizations of this result. We shall see that the most important consequence of the ultrashort core-hole lifetime is that for indirect RIXS the effective scattering cross section is proportional to the charge structure factor $S(\mathbf{q},\omega)$ and the longitudinal spin structure factor that is associated with it.

The indirect RIXS process is shown schematically in Fig. 1. In transition metal systems the photoelectron is promoted from a $1s$ core-orbital to empty $4p$ states that are far (10–20 eV) above the Fermi level, so the x rays do not cause direct transitions of the $1s$ electron into the lowest $3d$-like conduction bands of the system. Still RIXS is sensitive to excitations of electrons near the Fermi level. The Coulomb potential of $1s$ 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.

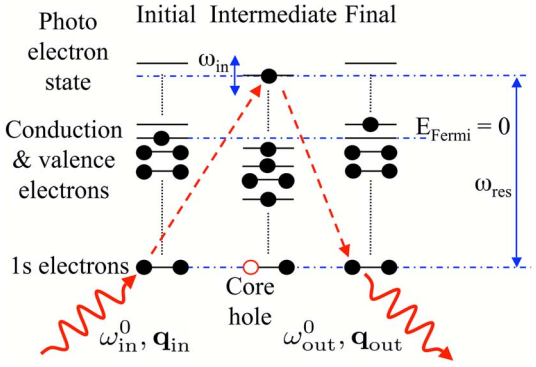

FIG. 1. (Color online) Schematic representation of the indirect resonant inelastic x-ray scattering (RIXS) process. The energy and momentum of the incoming photon are ω^0_{in} and $\mathbf{q}_{in}$, respectively, and of the outgoing photon ω^0_{out} and $\mathbf{q}_{out}$. The potential of the core-hole in the intermediate state scatters the valence and conduction electrons. The detuning energy ω_{in} quantifies how far the incoming photon energy is tuned away from the resonant edge at ω_{res}.

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 $\mathbf{q}$ and energy ω of the elementary excitation is 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).

At present energy resolutions of about 100 meV can be reached. In the near future 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. Recently it has been shown that also magnetic excitations, magnons, can be measured in RIXS.[17,18] Other interesting low-lying electronic excitations that potentially can be probed by RIXS are, for example, collective features such as plasmons, orbitons, and excitons, but also single-particlelike continua related to the band structure. RIXS provides a new tool to study these elementary excitations.

In this paper, we derive in detail the dynamical correlation function that is measured in indirect resonant inelastic x-ray scattering. We aim to give a full and self-contained derivation of the results that were presented in a previous Letter[16] and we elaborate on several generalizations. In particular we will show that for local core-hole potentials and ultrashort lifetimes, the dynamical correlation function is a linear combination of the charge density and longitudinal spin density response function. For a single band system 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. A sum rule is derived and we generalize these results to the case of finite temperature and multiband systems.

II. SERIES EXPANSION OF THE SCATTERING CROSS SECTION

The Kramers-Heisenberg formula[19–22] for the resonant x-ray scattering cross section at finite temperature is

$$\left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{res} \propto \left\langle \sum_F |A_{\mathrm{FI}}|^2 \delta(\omega+\omega_{\mathrm{FI}}) \right\rangle_{\mathrm{T}}, \tag{1}$$

where F and I denote the final and initial state of the system, respectively. The sum is over all final states and the brackets denote the statistical average over initial states I for a temperature T. The momentum and energy of the incoming/outgoing photons is $\mathbf{q}_{\mathrm{in/out}}$ 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 $\omega_{\mathrm{FI}} = E_{\mathrm{F}} - E_{\mathrm{I}}$. In the following we will take the ground-state energy of our system as reference energy: $E_{\mathrm{gs}} \equiv 0$. The scattering amplitude A_{FI} is given by

$$A_{\mathrm{FI}} = \omega_{\mathrm{res}} \sum_n \frac{\langle F|\hat{O}|n\rangle\langle n|\hat{O}|I\rangle}{\omega_{\mathrm{in}} - E_n - i\Gamma}, \tag{2}$$

where ω_{res} is the resonant energy, n denotes the intermediate states, and $\hat{O}$ the (dimensionless) dipole operator that describes the excitation from the initial to intermediate state and the deexcitation from the intermediate to final state. The energy of the incoming x rays with respect to the resonant energy is ω_{in} (this energy can thus either be negative or positive: $\omega_{\mathrm{in}} = \omega^0_{\mathrm{in}} - \omega_{\mathrm{res}}$) and E_n is the energy of intermediate state $|n\rangle$ with respect to the resonance energy. In the intermediate state a core-hole and a photoexcited 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 nonzero probability that an electron-hole excitation is present in the final state, see Fig. 1.

Let us list, for clarity, the frequency parameters that are involved in our formulas so far. The resonant energy ω_{res} represents the energy difference in the solid between the excited $4p$ and the core-hole states (5–10 keV), while $\omega^0_{\mathrm{in}}/\omega^0_{\mathrm{out}}$ stand for the incoming/outgoing photon energy, respectively. The energy that is lost by the photon in the scattering process is $\omega = \omega^0_{\mathrm{out}} - \omega^0_{\mathrm{in}}$, which is also equal to the difference in energy of the final and initial states of the solid $\omega_{\mathrm{FI}} = E_{\mathrm{F}} - E_{\mathrm{I}}$, simply because of the energy conservation. Finally, we defined $\omega_{\mathrm{in}} = \omega^0_{\mathrm{in}} - \omega_{\mathrm{res}}$, which is the detuning energy. It is a measure of the deviation of the incoming photon energy from the resonant threshold.

Crucial to our further considerations will be the fact that the intermediate state is not a steady state. The reason is that 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, which is of the order of electron volts as the lifetime is ultrashort, of the order of femtoseconds. Note that the lifetime broadening only appears in the intermediate states and not in the final or initial states—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.

When the energy of the incoming x rays is equal to a resonant energy of the system $\omega_{in} - E_n = 0$, we see from Eqs. (1) and (2) that the resonant enhancement of the x-ray scattering cross section is $(\omega_{\mathrm{res}}/\Gamma)^2$, which is $\sim 10^6$ for a transition metal K-edge.[22]

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 potentially strongly perturbing core-hole is present—appear. This is in contrast with canonical optical/electron energy loss experiments, where the probing photon/electron presents a weak perturbation to the system that is to be measured.

To calculate RIXS amplitudes, one possibility is to numerically evaluate the Kramers-Heisenberg expression. To do so, 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.[23] In this paper, however, we show that under the appropriate conditions we

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 nonresonant scattering, one is familiar with the situation that the scattering intensity is proportional to a linear response function, but for a resonant scattering experiment this is a quite unexpected result.

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

$$A_{\mathrm{FI}} = \frac{\omega_{\mathrm{res}}}{\omega_{\mathrm{in}} - i\Gamma} \sum_{l=0}^{\infty} M_l, \tag{3}$$

where we introduced the matrix elements

$$M_l = \sum_n \left(\frac{E_n}{\omega_{\mathrm{in}} - i\Gamma} \right)^l \langle F|\hat{O}|n\rangle\langle n|\hat{O}|I\rangle. \tag{4}$$

The formal radius of convergence of this power series is given by $E_n^2/(\omega_{\mathrm{in}}^2+\Gamma^2)$, so that the series is obviously convergent when the incoming X-ray energy is, e.g., far enough below the resonance, i.e., when $|\omega_{\mathrm{in}}| \gg 0$; but also at resonance, when $\omega_{\mathrm{in}}=0$ the series is convergent for intermediate energies that are smaller than the core-hole broadening Γ. Thus this expansion is controlled for ultrashort core-hole lifetimes, which implies that Γ is large. In the following we will be performing resummations of this series.

We denote the denominator of the expansion parameter $\omega_{\mathrm{in}}-i\Gamma$ by the complex number Δ, so that

$$M_l = \frac{1}{\Delta^l} \sum_n \langle F|\hat{O}|n\rangle (E_n)^l \langle n|\hat{O}|I\rangle = \frac{1}{\Delta^l} \langle F|\hat{O}(H_{\mathrm{int}})^l \hat{O}|I\rangle, \tag{5}$$

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

$$\left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{res} \propto \left\langle \sum_{\mathrm{F}} \left| \frac{\omega_{\mathrm{res}}}{\Delta} \sum_{l=0}^{\infty} M_l \right|^2 \delta(\omega - \omega_{\mathrm{FI}}) \right\rangle_{\mathrm{T}}. \tag{6}$$

III. INDIRECT RIXS FOR SPINLESS FERMIONS: $T=0$

As in Ref. 16, we will first calculate the resonant x-ray cross section at zero temperature 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 determined by a Hamiltonian H_0 that describes the electrons around the Fermi level. The generic form of the full many-body Hamiltonian is

$$H_0 = \sum_{i,j} t_{ij}(c_i^\dagger c_j + c_j^\dagger c_i) + c_i^\dagger c_i V_{ij} c_j^\dagger c_j, \tag{7}$$

where i and j denote lattice sites with lattice vectors $\mathbf{R}_i$ and $\mathbf{R}_j$. Note that the sum is over each pair i,j once, with i,j ranging 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 $V_{ij}=V_{|\mathbf{R}_i-\mathbf{R}_j|}$, as the Coulomb interaction only depends on the distance between two particles.

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

$$H_c = \sum_{i,j} s_i s_i^\dagger V_{ij}^c c_j^\dagger c_j, \tag{8}$$

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

$$\hat{O} = \sum_i e^{-i\mathbf{q}_{\mathrm{in}}\cdot\mathbf{R}_i} s_i p_i^\dagger + e^{i\mathbf{q}_{\mathrm{out}}\cdot\mathbf{R}_i} s_i^\dagger p_i + \mathrm{H.c.}, \tag{9}$$

where $p^\dagger$ creates a photoexcited electron in a $4p$ state and H.c. denotes the Hermitian conjugate of both terms.

A. Short lifetime approximation: Algebraic form

In order to calculate the cross section, we need to evaluate the operator $(H_{\mathrm{int}})^l=(H_0+H_c)^l$ in Eq. (5). A direct evaluation of this operator is complicated by the fact that $[H_0,H_c]\neq 0$. We therefore proceed by approximating H_{int}^l with a series that contains the leading terms to the scattering cross section for both strong and weak core-hole potentials—as long as the core-hole lifetime is short. After that we will do a full resummation of that series. This approximation is central to the results in this paper.

Expanding $(H_0+H_c)^l$ gives a series with 2^l terms of the form

$$H_{\mathrm{int}}^l = H_c^l + \sum_{n=0}^{l-1} H_c^n H_0 H_c^{l-n-1} + \cdots + \sum_{n=0}^{l-1} H_0^n H_c H_0^{l-n-1} + H_0^l. \tag{10}$$

Using $H_0\hat{O}|I\rangle=\hat{O}H_0|I\rangle\equiv 0$, this series reduces to

$$H_{\mathrm{int}}^l \hat{O}|I\rangle = \left(H_c^l + \sum_{n=0}^{l-2} H_c^n H_0 H_c^{l-n-1} + \cdots + H_0^{l-1} H_c \right) \hat{O}|I\rangle. \tag{11}$$

Using in addition that $\langle f|\hat{O}H_0=\langle f|H_0\hat{O}=E_f\langle f|\hat{O}$, we find

$$\langle F|\hat{O}H_{\mathrm{int}}^l\hat{O}|I\rangle = \langle F|\hat{O}\Bigg(H_c^l + E_{\mathrm{F}} H_c^{l-1} + \sum_{n=1}^{l-2} H_c^n H_0 H_c^{l-n-1} + \cdots + E_{\mathrm{F}}^{l-1} H_c \Bigg) \hat{O}|I\rangle. \tag{12}$$

For strong core-hole potentials, the leading term of H_{int}^l is H_c^l. Corrections to this term contain at least one factor of H_0

and are therefore smaller by a factor of t/V^c. For weak core-hole potentials, the term H_0^l vanishes because $[H_0,\hat{O}]=0$. The leading term for this limit therefore is $E_F^{l-1}H_c$. Correction terms contain at least two factors of H_c, which make them at least a factor of V^c/t smaller.

Let us now consider the approximate expression

$$H_{\text{int}}^l\hat{O}|I\rangle \simeq \sum_{m=0}^{l} H_0^m H_c^{l-m}\hat{O}|I\rangle. \tag{13}$$

From the arguments above, it is easy to see that the leading order terms for both strong ($m=0$) and weak ($m=l-1$) core-hole potentials are included in the sum; but a large series of other terms are included as well; they can be neglected in the case that we strictly consider either limit. Including them, however, means that we consider in addition a set of higher order scattering processes. A major advantage of including these is that the terms will give rise to a smooth interpolation between the two extreme limits. Note that the $m=l$ term in Eq. (13) is 0, so that it can be removed from the sum. After performing the same manipulations as above, we obtain

$$\begin{aligned}\langle F|\hat{O}\sum_{m=0}^{l-1} H_0^m H_c^{l-m}\hat{O}|I\rangle &= \sum_{m=0}^{l-1} E_F^m\langle F|\hat{O}H_c^{l-m}\hat{O}|I\rangle \\ &= \langle F|\hat{O}[H_c^l + E_F H_c^{l-1} \\ &\quad + \cdots + E_F^{l-1}H_c]\hat{O}|I\rangle. \end{aligned}\tag{14}$$

Comparing Eqs. (12) and (14), it can be seen that the approximation (13) is exact in the limit of both strong and weak core-hole potentials.

B. Short lifetime approximation: Graphical representation

We can also represent the series expansion and its approximation graphically (Fig. 2). When we expand $(A+B)^l$, where A and B are noncommuting operators, each term in the series corresponds to a graph on the grid of Fig. 2(1). Each graph occurs only once and can be constructed by starting at the lower left corner of the grid and moving either to the right, representing an A, or up, representing a B. At the next vertex a new move (right or up) is made. We perform this procedure l times and in this way we can obtain 2^l distinct graphs, each corresponding to a term in the expansion of $(A+B)^l$. For example, moving l times to the right represents the term A^l and moving l times up corresponds to B^l, see Figs. 2(2) and 2(3). All other terms in the series can be constructed by moving up and right a different number of times and in different order. As we consider a fixed value of l ($l=8$ in Fig. 2), all graphs must end on the diagonal of the triangle that forms the grid. In the series for $(H_0+H_c)^l\hat{O}|I\rangle$ ($H_0=A$ and $H_c=B$) we have the simplification that terms ending with H_0 acting on the ground state give zero. These terms can thus be removed from the expansion. The graphs for this expansion now live on a reduced grid where the horizontal grid-lines at the diagonal of the triangle are absent, see Fig. 2(5): these represent all terms ending on A.

In Fig. 2 we also represent the approximate series of the right-hand side of Eq. (13). Graphically this sum corresponds

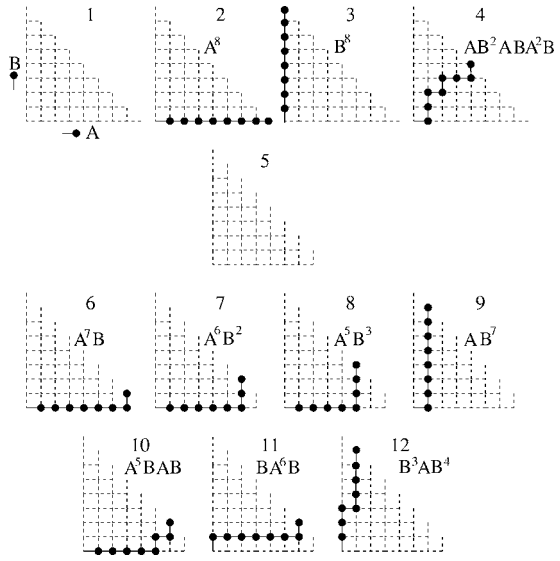

FIG. 2. Graphical representation of the expansion of $(A+B)^l$, where $A=H_0$ and $B=H_c$ are two noncommuting operators. In this example $l=8$.

to the set of graphs on the reduced grid of Fig. 2(5), with either one kink [Figs. 2(6), 2(7), 2(8), and 2(9)] or without kinks [Figs. 2(2) and 2(3)]. Thus in our approximation in Eq. (13) of the exact series for $(H_0+H_c)^l$ we neglect all graphs with two or more kinks [Figs. 2(4), 2(10), 2(11), and 2(12)]. In the limit of either very A or very large B, the graphs that we neglect correspond to subleading order corrections. When A is largest then the leading terms are, first, Fig. 2(2), which is however zero because it ends on A. The leading term is therefore of the order A^7 and shown in Fig. 2(6). Other higher order terms are shown Figs. 2(7), 2(8), 2(10) and 2(11). The last two graphs are neglected in our approximate expansion. In case B is dominating, the leading term is B^8, Fig 2(3), and next to leading is Fig 2(9), with B^7. The highest order terms that are neglected in our approximate series are of the type shown in Fig. 2(12).

C. Resummation of series for scattering cross section

In order to obtain M_l and from there the scattering amplitude A_{FI} and finally the scattering cross section, in Eq. (14) we need to evaluate expressions of the kind

$$H_c^n\hat{O}|I\rangle = H_c^{n-1}\sum_{i,l,j} s_l s_l^\dagger V_{lj}^c c_j^\dagger c_j e^{-i\mathbf{q}_{\text{in}}\cdot\mathbf{R}_i} s_i p_i^\dagger|I\rangle. \tag{15}$$

In the initial state no core-hole is present: just one core-hole is created by the dipole operator. We therefore have that $s_l s_l^\dagger s_i|I\rangle = \delta_{l,i}s_i s_i^\dagger s_i|I\rangle = \delta_{l,i}s_i|I\rangle$. Inserting this in Eq. (15), we obtain

$$H_c^n\hat{O}|I\rangle = H_c^{n-1}\sum_i e^{-i\mathbf{q}_{\text{in}}\cdot\mathbf{R}_i} s_i p_i^\dagger \sum_j V_{ij}^c c_j^\dagger c_j|I\rangle \tag{16}$$

and by recurrence

$$H_c^n\hat{O}|I\rangle = \sum_i e^{-i\mathbf{q}_{\text{in}}\cdot\mathbf{R}_i} s_i p_i^\dagger\left[\sum_j V_{ij}^c c_j^\dagger c_j\right]^n|I\rangle. \tag{17}$$

Let us for the moment consider the strong core-hole potential limit and keep in the expansion Eq. (13) only the term $m=0$. Inserting the results above in Eq. (5), we find that

$$M_l(V^c \gg t) = \frac{1}{\Delta^l}\langle F|\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}\left[\sum_j V^c_{ij}c^\dagger_j c_j\right]^l|I\rangle, \qquad (18)$$

where the transferred momentum $\mathbf{q}\equiv\mathbf{q}_{\text{out}}-\mathbf{q}_{\text{in}}$.

The first important observation is that the term $l=0$ does not contribute to the inelastic x-ray scattering intensity because $M_0=\langle F|\Sigma_i e^{i\mathbf{q}\cdot\mathbf{R}_i}|I\rangle=N\delta_{\mathbf{q},0}\delta_{\text{F,I}}$, which only contributes to the elastic scattering intensity at $\mathbf{q}=\mathbf{0}$ and other multiples of the reciprocal lattice vectors. From inspection of Eq. (4) 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."[24] This approximation corresponds to the limit where the core-hole lifetime broadening is the largest energy scale in the system ($\Gamma\to\infty$ or, equivalently, $\text{Im}[\Delta]\to-\infty$). In this limit only the $l=0$ term contributes to the indirect RIXS amplitude and the resonant inelastic signal vanishes. In any theoretical treatment of indirect resonant scattering one therefore needs to go beyond the fast-collision approximation.

Physically this vanishing of spectral weight is ultimately due to an interference effect. If we study a process in which we start from the initial state and reach a certain final state, we need to consider all different possible paths for this excitation-deexcitation process. When the core-hole broadening is very large we can reach the final state via any intermediate state and in order to obtain the scattering amplitude we thus add up coherently the contributions of all intermediate states. We then obtain $A=\Sigma_n\langle F|n\rangle\langle n|I\rangle$. When the set of intermediate states that we sum over is complete (which by definition is the case when $\Gamma\to\infty$), this leaves us with $A=\langle F|I\rangle$ which is, because of the orthogonality of eigenstates, only nonzero when the initial and final state are equal, hence only when the scattering is elastic.

The second observation is that M_l is a 2^l-particle correlation function. If we measure far away from resonance, where $|\text{Re}[\Delta]|\gg 0$, the scattering cross section is dominated by the $l=1$, two-particle, response function. When the incoming photon energy approaches the resonance, gradually the four, six, eight, etc. particle response functions add more and more spectral weight to the inelastic scattering amplitude. Generally these multiparticle response functions interfere. We will show, however, that in the local core-hole approximation the multiparticle correlation functions in expansion (13) collapse onto the dynamic two-particle (charge-charge) and four-particle (spin-spin) correlation function.

D. Local core-hole potentials

In hard x-ray electron spectroscopies one often makes the approximation that the core-hole potential is local. This corresponds to the widely used Anderson impurity approximation in the theoretical analysis of, e.g., x-ray absorption and photoemission, introduced in Refs. 25–27. This approximation is reasonable as the Coulomb potential is certainly largest on the atom where the core-hole is located.

In the present case, moreover, we can consider the potential generated by both the localized core-hole and photoexcited electron at the same time. As this exciton is a neutral object, its monopole contribution to the potential vanishes for distances larger than the exciton radius. The multipolar contributions that we are left with in this case are generally small and drop off quickly with distance.

We insert a local core-hole potential $V^c_{ij}=U\delta_{ij}$ in our equations and aim to resum the approximate series expansion in Eq. (13) for arbitrary values of the local core-hole potential. We find from Eq. (17) that

$$H^n_c\hat{O}|I\rangle = \sum_i e^{-i\mathbf{q}_{\text{in}}\cdot\mathbf{R}_i}s_{il}p^\dagger_i U^n[c^\dagger_i c_i]^n|I\rangle. \qquad (19)$$

Using that for fermions $[c^\dagger_i c_i]^n=c^\dagger_i c_i$, we obtain for our spinless fermions

$$M^{sf}_l = \frac{1}{\Delta^l}\langle F|\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}c^\dagger_i c_i|I\rangle\sum_{m=0}^{l-1}E^m_{\text{F}}U^{l-m}. \qquad (20)$$

The sum over m can easily be performed:

$$\sum_{m=0}^{l-1}E^m_{\text{F}}U^{l-m} = U^l\sum_{m=0}^{l-1}(E_{\text{F}}/U)^m = \frac{U^l-E^l_{\text{F}}}{1-E_{\text{F}}/U} \qquad (21)$$

and we obtain

$$M^{sf}_l = \frac{1}{\Delta^l}\frac{U^l-E^l_{\text{F}}}{1-E_{\text{F}}/U}\langle F|\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}c^\dagger_i c_i|I\rangle. \qquad (22)$$

Using that $\Sigma_i e^{i\mathbf{q}\cdot\mathbf{R}_i}c^\dagger_i c_i=\Sigma_{\mathbf{k}}c^\dagger_{\mathbf{k}-\mathbf{q}}c_{\mathbf{k}}\equiv\rho_{\mathbf{q}}$ is the density operator, we have to perform the sum over l in Eq. (3). The $l=0$ term is zero, as we discussed above, so that the scattering amplitude is

$$A_{\text{FI}} = \frac{\omega_{\text{res}}}{\Delta}\sum_{l=1}^{\infty}M_l. \qquad (23)$$

Using

$$\sum_{l=1}^{\infty}(U/\Delta)^l-(E_{\text{F}}/\Delta)^l = \Delta\frac{U-E_{\text{F}}}{(\Delta-U)(\Delta-E_{\text{F}})} \qquad (24)$$

we finally find that the indirect resonant inelastic scattering amplitude for spinless fermions is

$$A^{sf}_{\text{FI}} = P_1(\omega,U)\langle F|\rho_{\mathbf{q}}|I\rangle, \qquad (25)$$

where the resonant enhancement factor is $P_1(\omega,U)\equiv U\omega_{\text{res}}[(\Delta-U)(\Delta-\omega)]^{-1}$ and $\omega=E_{\text{F}}$. For spinless fermions with a local core-hole potential the scattering cross section thus turns out to be the density response function—a two-particle correlation function—with a resonant prefactor $P_1(\omega)$ that depends on the loss energy ω, the resonant energy ω_{res}, on the distance from resonance $\omega_{\text{in}}(=\text{Re}[\Delta])$, on the core-hole potential U, and on the core-hole lifetime broadening $\Gamma(=-\text{Im}[\Delta])$. We see that the resonant enhancement is largest when the energy of the incoming photons is either equal to the core-hole potential ($\omega_{\text{in}}=U$) or to the loss energy

($\omega_{in}=\omega$), which one could refer to as a "final-state resonance."

The density response function is related to the dielectric function $\epsilon(\mathbf{q},\omega)$ and the dynamic structure factor $S(\mathbf{q},\omega)$,[28] so that we obtain for the resonant scattering cross section

$$\left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{res}^{sf} \propto -|P_1(\omega)|^2 \,\mathrm{Im}\left[\frac{1}{V_{\mathbf{q}}\epsilon(\mathbf{q},\omega)}\right] \propto |P_1(\omega)|^2 S(\mathbf{q},\omega), \tag{26}$$

for a fixed value of the core-hole potential U. $V_{\mathbf{q}}$ is the Fourier transform of the Coulomb potential. For weak core-hole potentials the total scattering intensity is proportional to U^2 and for strong core-hole potentials, where $|U| \gg \Gamma$, the scattering intensity at resonance ($\omega_{in}=0$) is to first order independent of the strength of the core-hole potential. Far away from the edge, however, where $|\omega_{in}| \gg |U|$, the scattering intensity is again proportional to U^2, just as for weak core-hole potentials. Integrating $|P_1(\omega)|^2$ over all incoming photon energies, we obtain the integrated inelastic intensity at fixed loss energy ω and momentum $\mathbf{q}$,

$$\int_{-\infty}^{\infty} d\omega_{in} \left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{res}^{sf} \propto \frac{2\pi U^2 \omega_{res}^2}{\Gamma[4\Gamma^2+(U-\omega)^2]} S(\mathbf{q},\omega). \tag{27}$$

It seems that the resonant enhancement factor of the integrated intensity has a maximum when the loss energy is equal to the core-hole potential. However, the core-hole potential is attractive and therefore lower than zero, and the loss energy ω is by definition greater than zero. So the integrated intensity is maximal at energy loss $\omega=0$.

IV. INDIRECT RIXS FOR SPINLESS FERMIONS: FINITE T

In this section, we generalize the previous calculation to the case of finite temperature. The starting point is as before

$$\left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{res} \propto \frac{1}{Z}\sum_{\mathrm{I}}\sum_{\mathrm{F}} |A_{\mathrm{FI}}|^2 \delta(\omega-\omega_{\mathrm{FI}}) e^{-\beta E_{\mathrm{I}}}, \tag{28}$$

where $Z=\Sigma_{\mathrm{I}} e^{-\beta E_{\mathrm{I}}}$ is the partition function and $\beta=1/k_{\mathrm{B}}T$. Equation (28) represents the statistical average over all the initial states $|I\rangle$, where now the more general relation $H_0|I\rangle = E_{\mathrm{I}}|I\rangle$ holds.

We expand the scattering amplitude A_{FI}, using again the ultrashort lifetime of the core-hole as in Eq. (3). We are left with the evaluation of the operator $(H_{\mathrm{int}})^l$. We proceed by expanding it in the following way:

$$\begin{aligned}(H_{\mathrm{int}})^l \hat{O}|I\rangle &= (H_0+H_c)^l \hat{O}|I\rangle \\ &\simeq \sum_{n=0}^{l-1}\sum_{m=0}^{l-n-1} (H_0)^m (H_c)^{l-m-n} (H_0)^n \hat{O}|I\rangle, \end{aligned} \tag{29}$$

where we neglected the term H_0^l, as it will not contribute to the inelastic scattering cross section. This approximation reproduces the correct leading order terms, which represent the strong and weak coupling case, respectively. Moreover, it is a generalization of Eq. (13) that takes into account that the initial state is no longer the ground state so that $H_0|I\rangle = E_{\mathrm{I}}|I\rangle$. In our graphical representation, with respect to the $T=0$ case, it corresponds to retain all the additional terms, having more than one kink, that start and finish with a horizontal step. In doing this, we are neglecting again the subleading order terms $H_c^{l-1-n} H_0 H_c^n$.

After inserting expansion (29) in the expression (5) for M_l, we finally have to evaluate

$$\langle F|\hat{O}\sum_{n,m}(H_0)^m (H_c)^{l-m-n} (H_0)^n \hat{O}|I\rangle = \sum_{n,m} E_{\mathrm{F}}^m E_{\mathrm{I}}^n \langle F|\hat{O} H_c^{l-m-n} \hat{O}|I\rangle. \tag{30}$$

In the local core-hole approximation, we can resum this approximate series expansion. By using the results of Eqs. (19), we obtain for spinless fermions

$$M_l^{sf} = \frac{1}{\Delta^l}\langle F|\rho_{\mathbf{q}}|I\rangle \sum_{n,m} E_F^m E_{\mathrm{I}}^n U^{l-m-n}. \tag{31}$$

By performing the sums over n and m

$$\sum_{n,m} E_{\mathrm{F}}^m E_{\mathrm{I}}^n U^{l-m-n} = U^l \sum_{n=0}^{l-1} (E_{\mathrm{I}}/U)^n \sum_{m=0}^{l-n-1} (E_{\mathrm{F}}/U)^m, \tag{32}$$

and after summing over l, we finally obtain

$$A_{\mathrm{FI}}^{sf} = P_1(E_{\mathrm{F}},U)\frac{\Delta}{\Delta-E_{\mathrm{I}}}\langle F|\rho_{\mathbf{q}}|I\rangle. \tag{33}$$

This equation clearly shows that one of the main effects of finite temperature is to modify the resonant enhancement factor, nevertheless preserving the same structure for the scattering amplitude.

At this point we observe that at resonance $|\Delta|=\Gamma$, which is of the order of electron volts and thus several orders of magnitude larger than E_{I}, even at high temperature. This allows us to approximate the prefactor in Eq. (33) as

$$\begin{aligned} P_1(E_{\mathrm{F}},U)\frac{\Delta}{\Delta-E_{\mathrm{I}}} &\simeq P_1(\omega,U)\left(1+\frac{E_{\mathrm{I}}}{\Delta-\omega}+\cdots.\right) \\ &\times\left(1+\frac{E_{\mathrm{I}}}{\Delta}+\cdots.\right). \end{aligned} \tag{34}$$

At the lowest order in E_{I}/Γ, the prefactor is not modified by T at all, hence we conclude that the major modifications to the cross section are induced by thermal averaging of the correlation function. After integrating over all the incoming photon energies, we get the following approximate expression for the thermal average of the inelastic intensity at loss energy ω and momentum $\mathbf{q}$:

$$\left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{res,\mathrm{T}} \propto |P_1(\omega)|^2 \langle S(\mathbf{q},\omega)\rangle_{\mathrm{T}}. \tag{35}$$

In this expression the temperature dependence is entirely due to the temperature dependence of $S(\mathbf{q},\omega)$. The prefactor is in leading order temperature independent. Note that at finite temperatures also energy gain scattering occurs: the photon can gain an energy of the order of $k_B T$ from the system, which corresponds to a negative energy loss.

V. FERMIONS WITH SPIN

We generalize the calculation above to the situation where the electrons have an additional spin degree of freedom. In the Hamiltonians (7) and (8) 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 resum the series in Eq. (13) we now need to evaluate expansions of the number operators of the kind $(n_\uparrow+n_\downarrow)^l$. Using

$$(n_\uparrow + n_\downarrow)^l = n_\uparrow + n_\downarrow + n_\uparrow n_\downarrow \sum_{p=1}^{l-1} \binom{l}{p} = n_\uparrow + n_\downarrow + (2^l - 2) n_\uparrow n_\downarrow, \tag{36}$$

for $l>0$, we obtain

$$A_{\mathrm{FI}} = \langle F|P_1(\omega)[\rho_{\mathbf{q}} - 2\rho_{\mathbf{q}}^{\uparrow\downarrow}] + 2P_2(\omega)\rho_{\mathbf{q}}^{\uparrow\downarrow}|I\rangle, \tag{37}$$

with $P_2(\omega,U)=P_1(\omega,2U)/2$ and $\rho_{\mathbf{q}}^{\uparrow\downarrow} \equiv \Sigma_i e^{i\mathbf{q}\cdot\mathbf{R}_i} n_{i\uparrow} n_{i\downarrow}$. We see that in the case that each site can only be occupied by at most one valence electron, this equation immediately reduces to Eq. (25) with $\rho_{\mathbf{q}}=\rho_{\mathbf{q}}^{\uparrow}+\rho_{\mathbf{q}}^{\downarrow}$. The two terms in the scattering amplitude can also be written in terms of density and spin operators. Using $(n_{i\uparrow}-n_{i\downarrow})^2=(2S_i^z)^2=\frac{4}{3}\mathbf{S}_i^2$, we obtain $\rho_{\mathbf{q}} - 2\rho_{\mathbf{q}}^{\uparrow\downarrow}=\mathbf{S}_{\mathbf{q}}^2$, where we introduce the longitudinal spin density correlation function $\mathbf{S}_{\mathbf{q}}^2 \equiv \frac{1}{S(S+1)}\Sigma_{\mathbf{k}}\mathbf{S}_{\mathbf{k}+\mathbf{q}}\cdot\mathbf{S}_{-\mathbf{k}}$. In terms of these correlation functions the scattering amplitude for spinfull fermions is

$$A_{\mathrm{FI}} = [P_1(\omega) - P_2(\omega)]\langle F|\mathbf{S}_{\mathbf{q}}^2|I\rangle + P_2(\omega)\langle F|\rho_{\mathbf{q}}|I\rangle. \tag{38}$$

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.[29,30] Moreover, the spin and charge correlation functions have different resonant enhancements, see Fig. 3. For instance, when $\mathrm{Re}[\Delta]=U$, the scattering amplitude is dominated by $P_1(\omega)$ and hence by the longitudinal spin response function. At incident energies where $\mathrm{Re}[\Delta]=2U$, 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.

VI. MULTIBAND SYSTEMS

Let us consider systems with more than one band and take as an explicit example a transition metal with a $3d$ and a $4s$ band. The Coulomb attraction between the $1s$ core-hole and an electron in the $3d$ state (U_d) is much larger than the interaction with a $4s$ electron (U_s). Neglecting spin degrees of freedom we would naively expect that the indirect RIXS response in the two-band system is simply the sum of the responses of the two individual electronic systems, with possible interference between the two scattering channels: we expect the scattering amplitude to be equal to

$$A_{\mathrm{FI}}^{s+d} = P_1(\omega,U_d)\langle F|\rho_{\mathbf{q}}^d|I\rangle + P_1(\omega,U_s)\langle F|\rho_{\mathbf{q}}^s|I\rangle. \tag{39}$$

However, already from the calculation for the spinfull fermions we know that the situation should be more complicated,

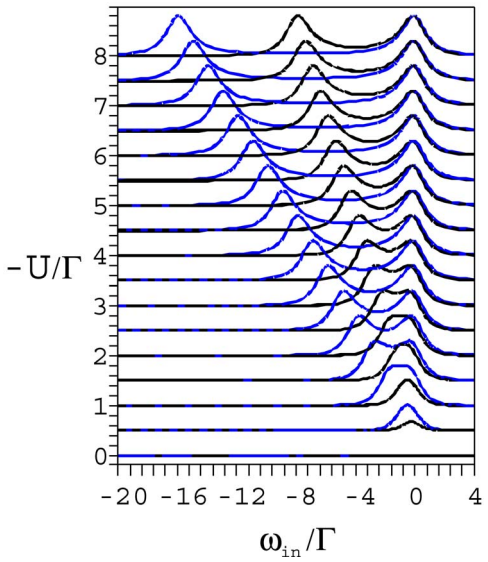

FIG. 3. (Color online) Prefactors to the scattering intensity at fixed loss energy ω as a function of incoming photon energy $\omega_{\mathrm{in}}/\Gamma$ for different values of the local core-hole potential U/Γ. The black line refers to $|P_1(U)|^2$ while the blue line (dark gray) to $|P_2(U)|^2$.

as in that case the full response function is not just the sum of the two response functions for spinless fermions. The point is that when both a $3d$ and $4s$ electron screen the core-hole, the intermediate state is at a lower energy (at $\omega_{\mathrm{in}}=U_d+U_s$) compared to the situation where only a single d/s electron screens the core-hole (with a resonance at $\omega_{\mathrm{in}}=U_d/U_s$, respectively.) In the situation that both electrons screen the core-hole, the resonance therefore appears at a different incoming photon energy.

According to Eq. (17), we now need to evaluate expressions of the sort $(U_d n^d + U_s n^s)^l$ for $l>0$. After using the binomial theorem and summing the resulting series, we obtain

$$(U_d n^d + U_s n^s)^l = U_d^l n^d + U_s^l n^s + n^d n^s [(U_d+U_s)^l - U_d^l - U_s^l], \tag{40}$$

which leads to a scattering amplitude

$$A_{\mathrm{FI}}^{sd} = A_{\mathrm{FI}}^{s+d} + [P_1(\omega, U_d+U_s) - P_1(\omega,U_d) - P_1(\omega,U_s)] \times \langle F|\rho_{\mathbf{q}}^{ds}|I\rangle, \tag{41}$$

where $\rho_{\mathbf{q}}^{ds} \equiv \Sigma_i e^{i\mathbf{q}\cdot\mathbf{R}_i} n_i^d n_i^s$. This is an interesting term, physically, as it directly measures the density correlations between the d and s electron density on a transition metal atom.

VII. CONCLUSIONS

On the basis of the ultrashort lifetime of the core-hole in the intermediate state 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. This algebraic series is also given in a graphical representation. By resumming the terms in the series, we find the dynamical charge and spin correlation functions that are measured in RIXS. The resonant prefactor is

only weakly temperature dependent. We have also derived a sum rule for the total scattering intensity and considered RIXS in both single and multiband systems. On the basis of our results, the charge and spin structure factor that is obtained from *ab initio* density functional calculations or from, e.g., Hubbard-like model Hamiltonians can directly be compared to experimental RIXS spectra. Moreover, our results open up the possibility to compare the measurements of $S(\mathbf{q},\omega)$ by RIXS and, for instance, electron energy loss spectroscopy. In this way one can actually determine experimentally the resonant scattering prefactors that we have calculated.

We should stress that four basic assumptions underly our results, which otherwise are general. First, the RIXS process that we consider is *indirect*, i.e., in the scattering process electrons are *not* directly promoted into the conduction band of the solid. Rather the inelastic scattering that we consider is due to the potential of the core-hole which is present in the intermediate state. This situation arises, for instance, at the *K*-edge of transition metal atoms, but can also occur at the *L*-edge of lanthanide ions. We assumed, furthermore, that the core-hole is localized and that its lifetime is short—very reasonable premises for the deep core-holes that are involved. The final assumption is that scattering is dominated by the coupling between the core-hole and electrons (of *d* character if we consider a transition metal *K*-edge) *on the same atom*. This is a good approximation when the *d* electrons are localized and the on-site Coulomb interaction is much larger than the one between neighboring atoms. In that sense our ultrashort lifetime expansion is expected to work very well for 3*d* systems and possibly less so for the 4*d* or 5*d* transition metal ions.

Finally, we assumed that the charge and longitudinal spin responses of the system that we consider are not vanishing, i.e., they are the leading order response function. In insulators at energies below the gap, however, these two response functions do vanish. This in principle opens a way to observe correlation functions beyond the ones that we have considered here, for instance, transversal spin or orbital response functions and thus to measure magnon dispersions or even orbiton properties with RIXS—a very exciting prospect indeed.

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