Electron-magnon scattering in elementary ferromagnets from first principles: lifetime broadening and kinks

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We study the electron-magnon scattering in bulk Fe, Co, and Ni within the framework of many-body perturbation theory implemented in the full-potential linearized augmented-plane-wave method. To this end, a $k$-dependent self-energy describing the scattering of electrons and magnons is constructed from the solution of a Bethe-Salpeter equation for the two-particle (electron-hole) Green function, in which single-particle Stoner and collective spin-wave excitations (magnons) are treated on the same footing. Partial self-consistency is achieved by the alignment of the chemical potentials. The resulting renormalized electronic band structures exhibit strong spin-dependent lifetime effects close to the Fermi energy, which are strongest in Fe. The renormalization gives rise to kinks in the electronic band dispersion at low binding energies, which we attribute to electron scattering with spatially extended spin waves. Furthermore, we find a band anomaly at larger binding energies in iron, which results from a coupling of the quasi-hole with single-particle excitations that form a peak in the Stoner continuum.

I. INTRODUCTION

The interaction of electrons and spin excitations plays a fundamental role for a wide variety of phenomena. For example, spin-polarized currents depolarize due to their interaction with magnons [11], the characteristic temperature dependence of the tunneling magnetoresistance (TMR) [2] is determined by the electron scattering by magnons, and in nanospINTRONICS the spin and charge currents that flow through nanostructures can be strongly affected by the electron-magnon interaction [4,5]. Moreover, it is speculated that the electron-magnon interaction is the origin of the superconductivity in Fe pnictides [6,10].

The elementary ferromagnets Fe, Co, and Ni are promising model systems to study the electron-magnon interaction. They form a class of immediately correlated materials in which localized $d$ states close to the Fermi level are embedded in a free-electron-like band structure. While density-functional theory (DFT), employing either the local-spin-density (LSDA) or the generalized gradient approximation (GGA) to the exchange-correlation potential, is able to capture the ground-state properties of these materials, it fails to yield accurate excited-state properties. For example, the exchange splitting of these materials is often overestimated within DFT calculations. Nickel is an extreme case in that the exchange splitting is almost a factor of two too large. In addition, recent angle-resolved photoemission spectroscopy (ARPES) experiments reveal pronounced differences in the quasiparticle dispersion compared to DFT. ARPES measurements, for example, have exhibited anomalous kinks in the band dispersion of iron surface states [11] and nickel bulk states [12]. These kinks appear at binding energies much larger than what one would expect for a phonon-mediated band renormalization. It has been speculated that these band anomalies are a footprint of electron-magnon scattering because the energetic position of the kinks corresponds to typical magnon energies in the materials. Furthermore, a strongly spin-dependent band broadening is observed in angle-resolved [13] and two-photon photoemission experiments [14], which again indicates that the electron-magnon interaction plays an important role in 3$d$ ferromagnets.

The appearance of kinks in the electronic band dispersion and the strong band broadening of the quasiparticle peaks clearly go beyond the scope of a mean-field theory such as DFT and calls for a genuine many-body description. One such method is the dynamical mean-field theory (DMFT), which maps the interacting many-body system onto an Anderson impurity model [15,16]. DMFT relies on the choice of model parameters, the effective intra-atomic interaction parameters $U$ and $J$ as well as the intra- and inter-atomic hopping parameters. In applications to real materials, the latter are usually taken from a L(S)DA mean-field solution. An early implementation [17] of this LSDA+DMFT method showed that majority hole states are strongly damped in iron. This result has been confirmed by Grechnev et al. [18] who studied Fe, Co, and Ni with the same approach. They found a strong damping of the majority quasiparticle states and, in addition, a shallow satellite feature below the bottom of the $d$ bands. A comparison of ARPES spectra to LSDA+DMFT calculations [19,21] revealed that the agreement between experiment and theory is considerably improved with respect to LSDA, but still the linewidths and the effective masses tend to be underestimated compared to experiment. So far, no evidence of band anomalies (kinks) has been found in the theoretical studies.

While allowing for an efficient treatment of many-body effects, the DMFT method suffers from some badly controlled approximations. Above all, the usage of the impurity model essentially amounts to neglecting the momentum dependence of the many-body scattering processes. Furthermore, the choice of the model parameters...
introduces an element of arbitrariness. Often they are fitted to experiment, which limits the predictive power of the method. Finally, since LSDA already contains the electron-electron interaction in an approximate way, it is necessary to apply a double-counting correction, for which no unique definition exists.

In this work, we employ an alternative description that avoids the usage of a model. The single-particle wave functions and propagators are allowed to extend over the whole (infinite) crystal. In this way, the momentum dependence is retained, and there is no need for model parameters nor a double-counting correction. Many-body perturbation theory (MBPT) is employed to construct an approximation to the electronic self-energy \( \Sigma(r, r'; \omega) \). This effective scattering potential describes many-body exchange and correlation scattering processes that an electron or hole experiences as it propagates through a many-electron system. In other words, the self-energy connects the non-interacting mean-field system to the real interacting system. The solution of the Dyson equation then yields the single-particle Green function of the interacting system, and the imaginary part of which is directly related to the photoemission spectra \[22\].

The most popular approximation to the electronic self-energy is the \( GW \) approximation, which has been shown to yield accurate band structures for a wide range of materials. For example, it is known to have a strong effect on the band gaps of semiconductors and insulators \[23\], which are corrected from their (usually underestimated) DFT values towards experiment. The \( GW \) method has also been applied to the elementary ferromagnets, where it partly cures the shortcomings of LSDA. While the 3d band width is typically overestimated in LSDA, the \( GW \) approximation applied to iron \[24\] and nickel \[25\, 26\] yields results in better agreement to the experimental values \[27\, 34\]. The explicit electron-electron scattering of the \( GW \) self-energy gives rise to a lifetime broadening of the bands, which is, however, too small to explain the broadening seen in photoemission experiments, indicating that the \( GW \) self-energy misses some important scattering processes.

In this work, we derive from the Hedin equations \[35\] a first-principles self-energy approximation that describes the scattering of electrons and magnons. The Hedin equations are a set of integro-differential equations that, if solved self-consistently, would yield, in principle, the exact self-energy for a many-electron system. A full self-consistent solution is not possible in practice, but the Hedin equations can be used to derive approximations to the self-energy. For example, the \( GW \) approximation results from a single cycle through the equations starting from \( \Sigma = 0 \). By iteration of the Hedin equations, one systematically generates more and more higher-order self-energy diagrams. In this way, we identify and select those scattering diagrams that are relevant for the coupling of electrons to many-body spin excitations. Then, summing these ladder diagrams to all orders in the interaction yields the \( GT \) approximation, where \( T \) stands for the magnon propagator, which describes the correlated motion of an electron-hole pair with opposite spins. It is shown that the lowest-order diagram of the \( T \) matrix is of third order in \( W \), which renders double-counting corrections with \( GW \) unnecessary. The \( GT \) approximation has a similar mathematical form as the \( GW \) self-energy in that it is given by a product of the single-particle Green function \( G \) and the effective magnon propagator \( T \). The implementation is, however, complicated by the fact that this \( T \) matrix is a four-point quantity: It depends on four points in space. A numerical implementation has been realized by employing a basis set of Wannier functions, which allow for an efficient truncation of the four-point quantity in real space.

The theoretical foundation of the \( GT \) self-energy is sketched in Sec. \[II\]. Its implementation within the SPES code \[26\] is described in Sec. \[III\]. We analyze the many-body renormalization of the band structure of the bulk elementary ferromagnets iron, cobalt, and nickel in Sec. \[IV\].

The coupling of electrons to spin excitations leads to a pronounced spin-dependent lifetime broadening of the quasiparticle states. The lifetime broadening, which is particularly strong in iron, can lead to a complete loss of the quasiparticle character in certain energy regions of the electronic spectrum. The electron-magnon scattering can give rise to anomalous quasiparticle dispersion behaviors, which acquire the form of a kink in the vicinity of the Fermi level. Furthermore, we find a band anomaly at higher binding energy in iron, in agreement with a very recent ARPES study. Section \[V\] gives a summary.

II. THEORY

Our goal is to construct a self-energy that describes the many-body renormalization due to the scattering of electrons and magnons. Here, we should understand the term \textit{magnon} to comprise not only the collective spin-wave excitations but also the single-particle Stoner excitations, which provide a decoherence channel, through which the spin waves acquire a finite lifetime. In our formulation, the two types of spin excitations are treated on the same footing. They are intimately coupled to each other (two sides of the same coin) and cannot, as a matter of principle, be treated separately. In this sense, the self-energy will describe the dressing of a propagating particle (electron or hole) through the creation and annihilation of collective spin waves and single-particle Stoner excitations alike. We note that the spin-orbit coupling is neglected.

Figure \[I\] shows the self-energy expansion in Feynman diagrams up to third order in the screened interaction \( W \) as derived from the Hedin equations. Cutting the expansion at the first order yields the \( GW \) self-energy, an approximation commonly used in computational condensed-matter physics. Despite its success; however, it lacks a number of many-body scattering effects, for example, particle-particle (electron-electron and hole-hole) and electron-hole scattering as well as
higher-order exchange processes. To account for these, the $GW$ method has been combined \cite{36}--\cite{39} with the $T$-matrix theory, in which the self-energy is expanded in terms of infinitely many ladder diagrams, describing the correlated motion of two particles (electrons or holes). This $GW + T$ approach is motivated phenomenologically: While $GW$ describes the correlation of intinerant $s$ and $p$ states, the ladder diagrams account for correlation effects taking place in localized states. However, as the $T$-matrix self-energy contains the Hartree “tadpole” diagram and the direct term of the second Born approximation, which is also contained in the $GW$ self-energy \cite{58}, a double-counting correction would be required in this approach \cite{39}.

Instead, we come back to the expansion shown in Fig. 1. Apart from the $GW$ self-energy, this expansion consists of the second- and third-order screened exchange diagrams and two direct diagrams (the ones of third order in the first row) that also appear in the $T$-matrix approach. These two diagrams have the characteristic form of ladder diagrams where the rungs of the ladder correspond to the screened interaction and the rails correspond to single-particle Green functions. They describe the correlated motion of an electron-hole pair and a particle pair (electron-electron or hole-hole), respectively. If the electron and hole are of opposite spins, the corresponding propagator can be seen as part of a solution of the Bethe-Salpeter equation (BSE) for the transverse magnetic response function \cite{59}--\cite{62}. The full BSE solution would comprise ladder diagrams of all orders, including the first and second as well as the infinitely many higher-order diagrams. For example, the collective spin-wave excitations arise from a resumma-

Figure 1: Feynman diagrams of the expansion of the Hedin self-energy up to the third order in the screened interaction $W$ denoted by an arrow. The Green function is denoted by a wiggly line. The $GW$ self-energy thus describes the emission and re-absorption of magnons in much the same way as $GW$ describes the corresponding processes for plasmons. Note that, after emission of the magnon, the spin of the propagating particle has flipped, since the magnon carries a total spin of unity.

\begin{align}
\Sigma^\sigma(13) &= -i \int d^4 d^4 T^\sigma\sigma' (12, 34) G^\sigma' (42) \tag{1}
\end{align}
This is a technical document discussing the electron-magnon self-energy in the ladder approximation. It introduces the concept of the Goldstone mode and its contribution to the self-energy. The document explains how to deal with spin-wave delta peaks and the importance of including non-local effects in the interaction.

**Figure 3:** Feynman diagram of the electron-magnon self-energy in the ladder approximation. Notation as in Fig. 1. It is assumed that the Green functions that are solely connected by the screened interaction have opposite spins.

with the spin indices $\sigma$ and $\sigma' = -\sigma$ is written in terms of the interacting single-particle Green function $G^\sigma$ and an electron-hole $T$ matrix. We have used the short-hand notation $1 = (r_1, t_1)$, and atomic units are used throughout.

In the ladder approximation, the multiple-scattering $T$ matrix is the solution of the BSE

$$T^{\sigma\sigma'}(12, 34) = \int d\varepsilon d\varepsilon' W(12)K^{\sigma\sigma'}(12, 56)$$

with the screened interaction $W(12)$ in the random-phase approximation (RPA) and the free electron-hole pair propagator

$$K^{\sigma\sigma'}(12, 34) = iG^{\sigma}(13)G^{\sigma'}(12).$$

Equation (2) defines the $T$ matrix to contain the ladder diagrams to all orders starting from the third order in $W$.

As already mentioned, the $T$ matrix is closely related to the transverse magnetic response function $R^{+−}(12) = \delta m^{+}(1)/\delta B^{+}(2)$, which gives the linear response of the spin density with respect to an externally applied circularly polarized magnetic field $B$. For simplicity, we omit "$+$" and "$−$" denoting the circular polarizations in the following. We now generalize the spin density to the spin density matrix $m(12)$ and the magnetic field to a non-local field $B(34)$. Then, the four-point response function $R(12, 34) = \delta m(12)/\delta B(34)$ can be written as the solution of a BSE (see, e.g., Refs. 19, 29). In matrix notation, the solution reads $R = −2(1 − K W)^{−1}K$ with the free electron-hole pair propagator of Eq. (3), which has poles at the energies of the unrenormalized Stoner excitations. The inverse $(1 − K W)^{−1}$ renormalizes these Stoner excitations, but it also gives rise to the collective spin-wave modes. Without it, one would simply obtain $−2K$, the transverse magnetic response function of the non-interacting mean-field system.

An important special case is the limit $k → 0$. In this limit (and in the absence of spin-orbit coupling), a static $B$ field that is orthogonal to the spin polarization is able to rotate the spins collectively towards the field direction even if $B$ is infinitesimally small. This leads to a spin-wave delta peak in $R$ at $k = 0$ and $\omega = 0$. For finite $k$, the spin-wave dispersion can be shown [14] to be quadratic in $k$, which, in a more general sense, corresponds to the Goldstone mode expected to arise in the case of a spontaneously broken symmetry such as ferromagnetism. The delta peak at $\omega = 0$ results from the infinite summation of ladder diagrams and will thus also appear in the $T$ matrix, where it is likely to pose a numerical problem in the evaluation of the self-energy. Therefore, we discuss its contribution to Eq. (1) in the following.

As a first step, we have to characterize the Goldstone delta peak in a more detailed way. As already mentioned, an externally applied infinitesimal transversal $B$ field can rotate the electron spins macroscopically. In order for this rotation to be rigid, the linear transversal change of the ground-state spin density matrix $m(12)$ must be proportional to $m(12)$ itself, $\delta m \propto m$. Thus, integrating the response function $R$ with the infinitesimal $B$ field should give a finite response proportional to $m$. Conversely, the inverse of $R$ should have a vanishing eigenvalue with the eigenvector $m$, i.e., $(1 − K W)m = 0$. We now sketch a proof that this is fulfilled if the Coulomb-hole screened-exchange (COHSEX) self-energy is taken as the starting point. For brevity, we use a simplified notation. The static COHSEX self-energy $\Sigma(12) = −W(12)m^{\sigma}(12) + \Sigma_{c}(1)\delta(12)$ with the density matrix $n^{\sigma}(12)$ consists of the screened exchange (Hartree-Fock exchange potential with a statically screened Coulomb potential) and a static Coulomb-hole self-energy, which has the form of a local potential. When separating off the spin-independent part $[(\Sigma^{+} + \Sigma^{-})]/2$, the remaining spin-dependent part can formally be interpreted as arising from a nonlocal magnetic field $B(12) = −W(12)m(12)/2$, which gives rise to the spin polarization in the COHSEX mean-field solution. Now we use the simple fact that rigidly rotating this $B$ field will rotate the magnetization in the same way, implying the proportionality $\delta B \propto B$ and $\delta m \propto m$ with $\delta B/B = \delta m/m$. With the magnetic response function $−2K$ of the mean-field system, corresponding to the formula for $R$ without the inverse, we can formulate this magnetic response as $−2K\delta B = \delta m$ (in matrix notation), and, with the above relationship for $\delta B$ and $\delta m$, we thus have $−2KBM = m$. Then, inserting $B = −Wm/2$ finally yields $KWm = m$. This proves that $m$ is an eigenvector of $1 − K W$ with vanishing eigenvalue, a claim that we have previously substantiated with arguments of theoretical consistency. There is another more detailed proof, which will be presented elsewhere [12].

To continue, we have to define a basis in which the quantities are to be expanded. We will later use a Wannier product basis, but, for the present purpose, it is easier to consider products of single-particle wave functions $\{\phi_{k/m}(1)\phi^{\dagger}_{k/m}(2)\}$ as a basis, instead. The Goldstone condition $KWm = m$ gives $m$ as an eigenvector in this basis. With $m(12) = n^{\uparrow}(12) − n^{\downarrow}(12)$ and

$$n^{\sigma}(12) = \frac{1}{N} \sum_{k} \sum_{m} f^{\sigma}_{km} \phi_{km}(1)\phi^{\dagger}_{km}(2)$$

with the number $N$ of $k$ points and the occupation num-
hers $f^\pm_{km}$, we obtain after expansion of $\phi^\pm_{km}(1)$ in terms of $\{\phi^\sigma_{km}(1)\}$

\[
m(12) = \frac{1}{N} \sum_k \sum_{m,m'} (f^\pm_{km} - f^-_{km}) \langle \phi^\pm_{km} \phi^{\dagger\pm}_{km} \rangle \phi^\pm_{km}(1) \phi^{\dagger\pm}_{km}(2).
\]

(5)

According to this expression, we can immediately conclude (because of the factor $f^\pm_{km} - f^-_{km}$) that the nonzero eigenvector elements are those where one of the states $(km \uparrow)$ or $(km' \downarrow)$ is occupied and the other is unoccupied. If the spin-up and spin-down states are not too different from each other, we can also see that the combination $(km \uparrow)$ occupied and $(km' \downarrow)$ unoccupied gives a sizable contribution, while the other term is negligible (because of the small overlap).

These considerations are helpful in the discussion of the Goldstone-mode contribution to the GT self-energy

\[
\Sigma^\sigma(\omega) = -\frac{i}{2\pi} \int_{-\infty}^{\infty} T^{\sigma\sigma'}(\omega') G^{\sigma'}(\omega - \omega') d\omega',
\]

(6)

here written in simplified notation and Fourier space. The frequency integral can be evaluated along a closed contour from $-\infty$ to $\infty$ and back to $-\infty$ along an infinite semicircle in the upper or lower complex half-plane. The latter does not contribute because both the $T$ matrix and the Green function behave asymptotically as $\omega^{-1}$. When writing $G$ and $T$ as sums over poles [e.g., see Eq. (1)], the contribution of any pair of poles can be evaluated with the residue theorem, and it vanishes unless the poles are both below or both above the real frequency axis.

Let us consider the spin-down case first. The $T^{\uparrow\dagger}$ matrix has the Goldstone delta peak infinitesimally below the real frequency axis, so its contribution is proportional to $1/(\omega - \epsilon_M + i\eta)$ with $\epsilon_M = 0$. Then, in the frequency convolution of Eq. (6), only the poles of the Green function that are also below the real axis contribute, which, according to Eq. (11), are those of the unoccupied states $(km' \uparrow)$, resulting in self-energy poles at $\epsilon_{km'} + \epsilon_M = \epsilon_{km'}$. According to Eq. (8) and the discussion thereafter, the unoccupied states $(km' \uparrow)$ must be combined with occupied states $(km \downarrow)$ to form the eigenvector of the $T$ matrix that is related to the Goldstone peak. The corresponding eigenvector elements are exactly those that have been discussed above to be very small. Furthermore, because of the combination of states, only the self-energy matrix elements that couple occupied states can get a non-zero contribution from this eigenvector, for example, the diagonal element $\Sigma^\downarrow_{km}(\omega) = \langle \phi^\downarrow_{km} | \Sigma^\downarrow(\omega) | \phi^\downarrow_{km} \rangle$. The contribution would consist of poles located at frequencies larger than the Fermi energy, thus in an energy region that is less relevant for the renormalization of the occupied states $(km \downarrow)$. Analogous arguments apply to the spin-up case: The $T^{\uparrow\dagger}$ matrix has its Goldstone delta peak infinitesimally above the real frequency axis. So, it is the Green-function poles related to the occupied states $(km \downarrow)$ that now give rise to self-energy poles at $\epsilon_{km}$, the other poles do not contribute. These states have to be combined with the unoccupied states $(km' \uparrow)$—the same combination as before—to yield a contribution to the diagonal element $\Sigma^\uparrow_{km}(\omega) = \langle \phi^\uparrow_{km} | \Sigma^\uparrow(\omega) | \phi^\dagger_{km} \rangle$ with poles at frequencies below the Fermi energy. Again, these poles do not play an important role for the renormalization of the unoccupied states $(km' \uparrow)$.

The Goldstone contribution to the GT self-energy is thus expected to be small, and numerical problems related to the Goldstone peak at $k = 0$ and $\omega = 0$ should not appear. Of course, the spin-wave peaks at finite $k$ and $\omega$ can contribute sizably to the self-energy, and we will see that these collective excitations can give rise to kinks in the electronic band dispersions.

III. IMPLEMENTATION

We have implemented the GT self-energy into the GW code SPEx [29]. The self-consistent DFT calculations are carried out with the FLEUR code [47]. Both codes rely on the all-electron full-potential linearized augmented-plane-wave method, which provides an accurate basis set for the representation of both the itinerant $s$ and $p$ states as well as the localized $d$ states. Solving the BSE [Eq. (2)] leads to an infinite series of ladder diagrams. This series appears also in the diagrammatic expansion of the transverse magnetic response function (then starting from the first order in $W$), for which we reported an implementation in the Refs. 40–43. The implementation exploits the fact that the Hamiltonian is not explicitly time dependent. In addition, the RPA screened interaction $W(r, r'; \omega)$ is approximated by its static limit $W(r, r') = W(r, r'; 0)$. As a result, the self-energy [Eq. (1)] involves only a single frequency integration. Furthermore, we restrict ourselves to calculating only the diagonal elements $\langle \phi^\sigma_{qm} | \Sigma^\sigma(\omega) | \phi^\sigma_{qm} \rangle$ of the self-energy.

\[
\Sigma^\sigma_{qm}(\omega) = -\frac{i}{2\pi} \int d^3 r_1 d^3 r_2 d^3 r_3 d^3 r_4 \phi^*_{q_{m}}(r_1) \phi^\sigma_{q_{m}}(r_3) \times \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} T^{\sigma\sigma'}(r_1, r_2; r_3, r_4; \omega'; \omega') G^{\sigma'}(r_2, r_4; \omega - \omega') d\omega' d\omega',
\]

(7)

with the momentum $q$, the band index $m$, and the spin $\sigma$ of the Bloch state $\phi^\sigma_{q_{m}}(r)$. The short-range behavior of the screened interaction $W$ in metals allows an on-site approximation to be employed [10–13]. The electron and hole are assumed to occupy the same lattice site when they interact with each other. To realize a lattice-site resolution, we formulate the theory with the help of Wannier functions

\[
w^\sigma_{Rn}(r) = \frac{1}{N} \sum_k e^{-i\mathbf{kr}} \sum_m U^\sigma_{km,n} \phi^\sigma_{km}(r),
\]

(8)

where $R$ is the lattice site, $n$ is an orbital index and $N$ is the number of $k$ points. The complex expansion coefficients $U^\sigma_{km,n}$ of the $n$th Wannier orbital are given by projection of the eigenstates onto a muffin-tin basis function.





with suitable orbital character and a subsequent Löwdin orthonormalization \([48]\). The Wannier representation of the \(T\) matrix reads

\[
T^{σσ'}(r_1, r_2; r'_3, r'_4; ω) = \sum_{R, R'} \bar{w}_{R_1}^* ω_R^{σσ'}(r_1) \bar{w}_{R_2}^{σσ}(r_2) \\
× T_{R_1R_2}^{σσ'}(R_3; R_4; ω) \bar{w}_{R_3}^{σσ}(r_3) \bar{w}_{R_4}^{σσ'}(r_4).
\]

(9)

Due to the lattice periodicity, the coefficients \(T_{R_1R_2}^{σσ'}(R_3; R_4; ω)\) only depend on the difference vector \(ΔR = R - R'\). A lattice Fourier transformation then yields

\[
T^{σσ'}_{n_1n_2,n_3n_4}(k, ω) = \sum_{ΔR} T^{σσ'}_{n_1n_2,n_3n_4;ΔR}(ω) e^{-i q ΔR}.
\]

(10)

Employing the Lehmann representation of the Green function

\[
G^r(r, r'; ω) = \frac{1}{N} \sum_k \sum_m \frac{\phi_{km}^∗(r) \phi_{km}^*(r')}{ω - ε_{km} + i η \text{sgn}(ε_{km} - ϵ)}
\]

(11)

with the energies \(ε_{km}\) of the corresponding Bloch states, the Fermi energy \(ε_F\), and a positive infinitesimal \(η\), one obtains for the diagonal element of the \(GT\) self-energy

\[
Σ_{qm}(ω) = \frac{i}{2π} \int_{−∞}^{∞} dω' \sum_{k,n_1,n_2,n_3,n_4} T^{σσ'}_{n_1n_2,n_3n_4}(k, ω')
× \sum_{m', m''} \frac{U_{qm,n_1}^{σσ'} U_{q-km',n_2}^{σσ'} U_{qm,n_3}^{σσ'} U_{q-km',n_4}^{σσ'}}{ω - ω' - ε_{q-km',m''} + i η \text{sgn}(ε_{q-km',m''} - ϵ)}
\]

(12)

where the summation over \(m'\), in principle, runs over the infinitely many single-particle eigenstates, but in the present case the summation is effectively restricted to the ones which have been used for the construction of the Wannier basis.

The \(T\) matrix exhibits poles at the spin excitation energies along the real-frequency axis. The collective spin-wave excitations produce particularly poles at low energies, which complicates a straightforward frequency integration. The integration along the real-frequency axis can be avoided by employing the method of analytic continuation \([49, 50]\). To this end, Eq. \((12)\) is analytically continued to the imaginary axis and evaluated there for a set of imaginary frequencies forming a mesh along this axis. This is achieved by replacing \(ω → iω\) and \(ω' → iω'\), formally changing the prefactor \(-i/(2π)\) to \(1/(2π)\). In this way, one avoids the real-frequency axis where the quantities show strong variations. Along the imaginary axis, the functions are much smoother, which enables an accurate sampling and interpolation of the functions with relatively coarse frequency meshes, also simplifying the frequency convolution of Eq. \((12)\). At the end of the calculation, the self-energy has to be analytically continued back to the real-frequency axis. We employ Padé approximants for the \(T\) matrix (Thiele’s reciprocal difference method \([51]\), which allows an analytic frequency convolution with \(G\), and also for the self-energy, yielding the self-energy on the whole complex frequency plane. In this approach, the self-energy is effectively represented by a sum over poles with analytically determined complex positions and weights. However, it is well known that this extrapolation can lead to spurious features in the self-energy if one of the effective poles happens to lie close to the real frequency axis. To avoid such unphysical results, we make use of an averaging procedure where, for each set of values \(Σ\) (\(ω'\)), we evaluate about 1000 different Padé approximants with input values randomly varied in the range \([-10^{-8} h\omega, 10^{-8} h\omega]\) and average over them.

The evaluation of Eq. \((12)\) can be expensive. Therefore, we exploit a few symmetries to accelerate the computation. The first is a symmetry in frequency space which the \(T\) matrix inherits from the propagator \(K\) \([11]\), \(T_{n_1n_2,n_3n_4}(k, -iω) = T_{n_1n_2,n_3n_4}(k, iω)\). This leads to \(Σ_{mn}(k, -iω) = T_{mn}(k, iω)\), where \(T\) denotes the sum of the products \(TUUUU\) over \(n_1, n_2, n_3,\) and \(n_4\). Hence, we can restrict the frequency mesh to the positive imaginary axis and utilize this symmetry for the frequency integration from \(-∞ \sim ∞\). Furthermore, we exploit spatial symmetry in the evaluation of the \(k\) summations. Equation \((12)\) contains a sum over all \(k\) vectors. We restrict this summation to the extended irreducible Brillouin zone (corresponding to the irreducible zone that is created by the subset of symmetry operations that leave the \(q\) vector invariant \([26]\), the so-called little group). The contribution of the symmetry-equivalent \(k\) points is obtained by a symmetrization procedure employing the symmetry transformation matrices of the Wannier set. In the same way, we can accelerate the computation of the two-particle propagator \([Eq. (3)]\), which involves a \(k\) summation, as well \([11]\).

As already mentioned above, we presently refrain from combining Eq. \((1)\) with the fully dynamical GW self-energy. Such a study is deferred to a later work. Instead, we combine it with the LSDA solution whose exchange splitting has been corrected in such a way that the Goldstone condition (the spin-wave energy must vanish in the limit \(k → 0\)) is fulfilled. To be more precise, we introduce a parameter \(Δx\), with which the spin-up and spin-down states are shifted with respect to each other, i.e., \(ε_{km}^{↑↓} → ε_{km}^{↑↓} ± Δx/2\). (A positive \(Δx\) decreases the exchange splitting.) The parameter \(Δx\) is varied until the spin-wave excitation energy vanishes in the long-wavelength limit. The so-corrected single-particle system has been shown \([23]\) to be similar to a self-consistent solution obtained with the COHSEX self-energy \([33]\), the static limit of GW. The corrected LSDA Green function can thus be understood as an approximation to the COHSEX Green function, which is then further renormalized with the \(GT\) self-energy.

We introduce another parameter to achieve partial self-consistency in the Green function. The application of the \(GT\) self-energy does not only introduce lifetime broadening effects but also leads to an energetic shift of the electronic bands. In general, this entails that the Fermi energy has to be readjusted to ensure particle number.
The correction is tantamount to shifting the exchange-correlation potential by the same amount $v_{xc} \rightarrow v_{xc} + \Delta v$. Thus, the non-interacting reference system already contains information about the renormalized system through the shift $\Delta v$, which can be regarded as some level of self-consistency. (Since the Fermi energy depends on the quasiparticle energies in the whole irreducible wedge of the Brillouin zone, the shift has to be calculated self-consistently until the Fermi energy and $\Delta v$ converge.)

This approach solves the aforementioned two problems in a natural way.

The modification of $v_{xc}$ changes the single-particle energies accordingly so that, in summary, the latter are adjusted by $\xi_{km}^{\uparrow} \rightarrow \xi_{km}^{\uparrow} + \Delta v \pm \Delta_x / 2$, which shifts the self-energy argument in the opposite direction, effectively undoing the shift from before, $\Sigma(\omega - \Delta v) \rightarrow \Sigma(\omega)$. The $\Delta v$ correction does not affect the Goldstone condition, which has been used to fix $\Delta_x$, since only the differences of the single-particle states (of opposite spins) enter the evaluation of the spin excitations energies, and not their absolute values. We note that the two parameters, $\Delta_x$ and $\Delta v$, are determined from exact physical constraints, so they do not imply a deviation from the “ab initio-ness” of the method.

With the aligned chemical potential, Eq. (13) can alternatively be written as

$$\Im G^\sigma(\omega, \mathbf{k}) = \sum_m \Im \left\{ \omega - \epsilon_{km}^\sigma - \Sigma^\sigma_{km}(\omega - \Delta_v) \right\}^{-1}. \quad (13)$$

Since the corrected energies approximate COHSEX quasiparticle energies, no subtraction of the expectation value of the exchange-correlation potential as in one-shot $GW$ calculations is needed in this case.

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$$\Im G^\sigma(\omega, \mathbf{k}) = \sum_m \Im \left\{ \omega - \epsilon_{km}^\sigma - \Sigma^\sigma_{km}(\omega - \Delta_v) \right\}^{-1}. \quad (13)$$

where the single-particle energies $\epsilon_{km}^\sigma$ are assumed to contain the two parameters $\Delta_x$ and $\Delta v$ as described above. (We note that the parameter $\Delta v$, in Eq. (14) seems to have no effect because the term $-\Delta v$ de facto cancels the $\Delta_x$ in $\epsilon_{km}^\sigma$, but it should be remembered that $\Delta v$ shifts the Fermi energy, too. We also note that it might be surprising that a spin-independent shift of the exchange-correlation potential and, thus, of the Kohn-Sham eigenvalues should have an effect at all, but one has to remember that quasiparticle energies correspond to total-energy differences and are therefore defined absolutely.)

Figure 4: Band structures of (a) Fe, (b) Co, and (c) Ni. The spectral functions [Eq. (14)] are shown in light blue to green. The LSDA band structure is shown as red lines for comparison. The lifetime broadening decreases in the order of increasing $d$ occupancy (Fe→Co→Ni). The spin and particle-hole asymmetries in the lifetime broadening are clearly seen.

...
and 25 meV have been determined self-consistently. The 18 lowest eigenstates in each spin channel. The Bril-
tering diagram shown in Fig. 3 may be described as an
to the spin asymmetry of the density of states. The scat-
strongly than the minority ones. This can be attributed
effects: the majority valence states are broadened more
excited, thus reducing the phasespace of spin excitation.

We also observe a clear spin asymmetry of the lifetime
is due to the decrease of the number of available minority

d,n and fcc Ni. The LSDA band structure is the uncorrected

groups), they decrease again. The reason for this is the

IV. CALCULATIONS

Figure 4 shows the \( k \)-resolved spectral functions ac-
garding to Eq. (14) in light blue to green together with
the LSDA band structure as red lines for bcc Fe, fcc Co,
and fcc Ni. The LSDA band structure is the uncorrected

The linewidths disappear at the Fermi energy and grow
rapidly for lower and higher energies. Beyond a binding
energy of around 4 eV (negative energies in the dia-
grams), they decrease again. The reason for this is the
restriction of the self-energy to the \( GT \) diagram. This

diagram is expected to give the low-energy scattering
contribution because of the low energies of the magnon
excitations contained in the \( T \) matrix. At higher bind-
ing energies, other diagrams, which are neglected in the
present study, should become increasingly important, for
example, the \( GW \) diagram.

The lifetime broadening is strongest in bcc Fe and
weakens in the order of \( d \) occupancy (Fe→Co→Ni). This
is due to the decrease of the number of available minority
conduction states, into which a majority electron can be
excited, thus reducing the phase space of spin excitations.
We also observe a clear spin asymmetry of the lifetime
effects: the majority valence states are broadened more
strongly than the minority ones. This can be attributed
to the spin asymmetry of the density of states. The scat-
tering diagram shown in Fig. 3 may be described as an
incoming majority hole at 1, which excites an electron-
hole pair at 2 in the minority channel. The majority
hole and minority electron then propagate through the
system, successively scattering with each other until the
minority electron-hole pair recombines at 4. The corre-
related propagation of the electron and the hole (having
opposite spins) embodies the spin-flip excitations in the
form of Stoner excitations and, due to their correlated
motion, also of the collective spin-wave excitations. The
probability for the pair formation and the pair propaga-
tion process depends on the number of available majority
valence and minority conduction states, which is large in
this case, see Fig. 5 left. (We note that the states also
have similar orbital character.) On the other hand, an
incoming minority hole would have to combine with ma-

The probability for the pair formation and the pair propaga-
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this case, see Fig. 5 left. (We note that the states also
have similar orbital character.) On the other hand, an
incoming minority hole would have to combine with ma-

There is evidence for a particle-hole asymmetry as well,
the majority channel. A similar but less pronounced renor-
alization effect is also seen in the minority channel of
Co. Here, an explanation very similar to the one just
given would predict the lifetime broadening of major-
ity hole (minority electron) bands to be larger than the
broadening of majority electron (minority hole) bands,
agreement with the spectra.

In iron, and to a lesser degree in Co, the many-body
renormalization is so strong that the quasiparticle char-
acter is virtually lost between -2.5 and -0.5 eV in the
majority channel. A similar but less pronounced renor-
alization effect is also seen in the minority channel of
iron at around 0.5 eV above the Fermi energy. The \( GT \)
self-energy thus leads to a substantially more complex
spectral function than the quasiparticle band structure of
a \( GW \) calculation, let alone the mean-field band structure
of Kohn-Sham DFT. In the case of the former, one often
only plots the energy-renormalized quasiparticle bands
and drops the lifetime broadening altogether, which is
legitimate because the main self-energy poles of \( GW \) are
typically energetically far away from the valence and low-
lying conduction bands. The quasiparticle approxima-
tion is therefore a good one. To be more precise, the
main \( GW \) self-energy poles describe the coupling of the
electrons and holes to plasmons (emission and absorp-
tion of plasmons), whose energies \( \approx \epsilon_P \) are typically
in the order of tens of eV. The poles, thus, appear well sepa-
ated from the valence (conduction) band energies \( \epsilon_{km} \) at
around \( \epsilon_{km} \pm \epsilon_P \) for valence and conduction states, respec-

\[ \Delta \]
spin band energies, i.e., at around \( \epsilon \approx \epsilon_M \). In this sense, the magnon energies additionally compete with the exchange splitting. To sum up, the \( GT \) self-energy poles are very likely located in the valence (and low-lying conduction) region, giving rise to a much richer photoelectron spectrum than, for example, in \( GW \) calculations.

The plot of the \( \mathbf{k} \)- and energy-resolved spectral function allows fine details of the quasiparticle band structure to be investigated. The resonant interaction with many-body states in the valence band region (these form the pole structure of the self-energy) can give rise to anomalies in the band dispersion, as we will discuss exemplarily for a few cases in the following. Figure 6 shows selected majority valence bands of Fe, Co, and Ni close to the Fermi energy. While the LSDA bands (red lines) exhibit a very regular parabolic-like behavior, there are distinct signatures of many-body renormalization effects to be seen in the \( GT \) bands. We note that these irregularities appear in some but not all \( \mathbf{k} \) directions, which demonstrates the importance of the \( \mathbf{k} \) dependence of the self-energy. We also see in Fig. 6(a) that the effect is band-selective: only two out of three bands are affected, despite the fact that all bands have the same \( d \) or-bital character. The kinks are seen at binding energies of about 180 meV and 130 meV for the two bands, respectively. These energies are in the order of magnitude of typical spin-wave energies, which leads one to suspect that these anomalies are caused by a coupling to extended spin-wave excitations. This conjecture is corroborated by a calculation in which the self-energy is restricted to the first diagram of Fig. 3, the diagram of third order in \( W \). Since the spin-wave excitation requires the summation over a large number of diagrams (preferably infinitely many) to become well-defined, it is plausible that the third-order diagram alone cannot describe the collective spin-wave, but does contain the Stoner excitations. Indeed, the resulting quasiparticle bands do not show any anomalies. We conclude that the kinks are caused by a scattering of the quasiparticle with collective spin-wave excitations. In Fe and Co [Figs. 6(b) and (c)], similar kinks are observed, which are, however, somewhat less pronounced than in iron. Their energetic positions at 150 meV and 200 meV, respectively, are similar to the case of Fe.

In fact, there have been ARPES studies that reported kink structures in the band dispersion of iron \([11]\) and nickel \([12]\) at about 125 meV and 250 meV below the Fermi energy, respectively, thus in the same order of magnitude as in our theoretical calculation. However, in both cases, minority instead of majority bands were investigated, and we do not observe kink structures close to the Fermi energy in the minority channel. Cui et al. \([55]\), on the other hand, published ARPES spectra of iron and claimed an anomaly in a majority band at around 270 meV to originate from electron-magnon interactions. However, in Ref. \([56]\) they distanced themselves from this claim and attributed the anomaly to an inaccuracy of the line-shape analysis in case of strong lifetime broadening. Indeed, our theoretical spectra do not exhibit a dispersion anomaly in the band they have measured.

In iron and cobalt, we clearly see the onset of strong lifetime broadening when the quasiparticle bands suddenly lose intensity, and a “waterfall” structure forms. In the three bands discussed above, this happens roughly at 450 meV, 500 meV (Fe), and 250 meV (Co). Nickel, on the other hand, exhibits the weakest lifetime broadening of all. While it is still strong, it does not lead to the disappearance of the quasiparticle character as in Fe and Co. The quasiparticle bands, although substantially broadened, remain well-defined over the whole energy range. We note that the “waterfall” structures seen here are very similar to a feature often observed in ARPES spectroscopy. This feature is interpreted as a footprint of a many-body resonance, at whose energy the spectral function is expected to show non-Fermi liquid behavior. This interpretation is confirmed by the present calculations.

Up to now, we have discussed kinks in the majority bands close to the Fermi energy, which we have attributed to the scattering of electrons with extended spin waves. In the following, we will investigate another band
The spectrum for $\Delta$ spectral function is largely unaffected by the parameter form of the two plots is very similar, implying that the $GT$-renormalized spectral function with $\Delta_v = 190$ meV and $\Delta_v = 0$. The LSDA bands are shown as red lines. The red crosses are fitted peak positions from experiment (data reproduced from Ref. [57]).

Figure 7: High-energy anomaly in a minority band of iron. (a) $GT$-renormalized spectral function with $\Delta_v = 190$ meV and (b) $\Delta_v = 0$. The LSDA bands are shown as red lines. The red crosses are fitted peak positions from experiment (data reproduced from Ref. [57]).

V. CONCLUSIONS

In summary, we have described a Green-function technique to calculate the electron-magnon scattering from first principles. The $GT$ self-energy is constructed from the product of the single-particle Green function $G$ and the multiple scattering $T$ matrix, which can be viewed as an effective interaction that acts through the exchange of magnons. The $T$ matrix, thus, contains the full spin excitation spectrum, comprising collective spin-wave and single-particle Stoner excitations, and, diagrammatically, it describes the correlated motion of an electron-hole pair of opposite spins. When expanded in powers of the screened Coulomb interaction $W$, the $T$ matrix can be written as a summation over ladder diagrams of ever increasing order. We have shown that a formulation consistent with the Hedin equations requires the $GT$ self-energy to include the third-order ladder diagram as the lowest order. In this way, the self-energy approximation is free of double counting with the Hartree or exchange ($GW$) diagrams.

The $T$ matrix contains the acoustic spin-wave excitations for all wave vectors, including the long-wavelength limit $k \to 0$, where the excitation energy vanishes and the $T$ matrix becomes singular with a mathematical delta peak in its imaginary part. Since such a singularity is likely to give rise to numerical difficulties, we have investigated its contribution to the $GT$ self-energy. As a result, the contribution of the long-wavelength spin-wave excitation should be very small in the problematic limit $k \to 0$. In fact, we did not encounter numerical problems related with the singularity.

The contribution of the spin-wave excitations do not vanish for finite $k$, though. Our numerical calculations have shown that it is this contribution that can lead to the formation of kinks in the dispersion of majority valence bands close to the Fermi energy in the bulk ferromagnets bcc Fe, fcc Co, and fcc Ni. Similar kink structures have already been reported in several experimental studies. Since they appear at typical magnon energies, they had been speculated to originate from the electron-magnon scattering. The present first-principles calculations confirm this conjecture.

The $GT$ self-energy gives rise to a very strong lifetime broadening of the quasiparticle bands, to the extent that in a certain energy region the quasiparticle character is virtually lost in the majority valence bands of bcc Fe and fcc Co. The minority bands, however, are much less affected by the lifetime broadening, which we have attributed to the spin asymmetry of the density of states. For a similar reasons, one also finds a particle-hole asymmetry. The spin asymmetry in the lifetime broadening explains the experimental fact that minority bands are seen to relatively large binding energies, while the majority bands disappear very soon below the Fermi energy. The strong renormalization effects also give rise to “waterfall” structures in the $k$- and energy-resolved spectral functions, in particular for bcc Fe and fcc Co.

Finally, our method can describe a band-dispersion anomaly in the minority channel of bcc Fe at a large binding energy of about 1.5 eV, which has very recently been observed in ARPES measurements [57]. Whereas such a large energy is commonly not associated with a
magnon excitation, we could elucidate\cite{35} that this band anomaly is caused by a many-body resonance formed by a superposition of Stoner excitations (an increased Stoner intensity is seen in the spin excitation spectrum at around 0.7 eV) and majority hole states that form a peak in the density of states (at around 0.8 eV).

In summary, we believe that the present work is an important step in the development of ab initio many-body methods for the treatment of magnetic materials, in which the magnon energies (collective and Stoner type), the exchange splitting, the band energies, and the electron-magnon coupling strength set different energy scales that compete with each other. Further studies in this direction might also elucidate the role of magnons in the potential formation of Cooper pairs in high-temperature superconductors.

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[59] $R = -2R^{(4)}$ in the notation of Ref. [42].