Letter

Correlations, disorder, and multimagnon processes in terahertz spin dynamics of magnetic nanostructures: A first-principles investigation

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Understanding the impact of electronic correlations and disorder is essential for an accurate description of solids. Here, we study the role of correlations, disorder, and multimagnon processes in THz spin dynamics. We reveal that a significant part of the electron self-energy, which goes beyond the adiabatic local spin density approximation, arises from the interaction between electrons and a virtual magnon gas. This interaction leads to a substantial modification of the exchange splitting and a renormalization of magnon energies, in agreement with the experimental data. Finally, we establish a quantitative hierarchy of magnon relaxation processes based on first principles.

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Magnetic nanostructures and their intricate spin dynamics have fueled remarkable developments in experimental, applied, and theoretical quantum many-body physics over the recent years. On the applied side, nanostructures, beginning with thin films [1] and proceeding down to single magnetic atoms [2], constitute the basis for spintronic and magnonic information storage and processing devices [3]. A substantial body of research has been devoted to the realization of quantum logical gates using magnetic degrees of freedom [4]. Optical magnetization switching allows to control the magnetization dynamics on the femtosecond timescale [5]. For ultrathin magnetic films, recent experiments have yielded highly resolved spectra of the electronic (using, e.g., angle-resolved photoemission [6]) and, by means of spin-polarized high-resolution electron energy-loss spectroscopy (SPHREELS) [7–9], magnonic (spin-wave) band structures across the entire Brillouin zone.

On the other hand, the current first-principles theoretical description of spin dynamics lags clearly behind these spectacular experimental developments. In this Letter we attempt to narrow this gap. The task is of considerable interest, as the spectrum of collective spin excitations (called spin waves or magnons) determines the thermodynamic properties of magnets, including the phase transition temperatures. Additionally, the excitations contribute to the specific heat as well as to the thermal and electric conductivities. Furthermore, their coupling to electronic degrees of freedom can give rise to a superconducting state [10] and, in general, influences the electronic band structure [11], leading to a finite lifetime of excited electronic states [12]. Last but not least, the damping of the spin dynamics is of paramount practical importance in spintronic applications [13] and, as we show here, constitutes an additional experimental probe sensitive to the atomic and electronic structure of nanostructures.

The density functional theory in a local spin density approximation (LSDA) is able to provide a qualitatively correct picture of electronic band structures of films and surfaces, including effects such as the formation of electronic surface and quantum well states [14], but misses important corrections arising from correlation effects [15], notably predicting an unrealistic value of the Stoner exchange splitting [16], among others.

In order to remain specific, we address Co films on different substrates for which a sufficient body of experimental evidence and theoretical studies concerning the electronic structure and spin dynamics is available [9,17,18]. Particularly, in the case of three monolayers (ML) of Co the fcc unit cell is complete and one expects to observe three magnon modes. As a matter of fact all these modes could unambiguously be resolved by the recent high-resolution experiments by means of SPHREELS [9,19]. The latter SPHREELS experiments show that the theoretical description of these systems require a substantial *ad hoc* "negative U correction" of the occupied majority spin bands, as the LSDA drastically overestimates the spin-wave energies. Additionally, it falls short in accurately replicating the magnon lifetimes, performing well for Co/Cu but inadequately for Co/Ir.

Different descriptions of the band-structure renormalization have been proposed, including three-body scattering [16] and a sophisticated dynamical mean-field treatment [20]. We have recently put forward a fully *ab initio* scheme [21]

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allowing to compute the electronic self-energy for complex systems, including two-dimensional films, within Hedin's many-body perturbation scheme. This approach allows for the calculation of a nonlocal self-energy while being numerically much less expensive than the aforementioned methods. Here, we show that in the considered Co films a substantial part of these corrections arises indeed due to the interaction of electrons with the gas of virtual magnons.

Likewise, the damping mechanism of THz spin excitations has not yet been fully understood. While the Landau damping, arising due to the interactions of collective spin waves with single-particle (Stoner) excitations, is known to be an important decay channel in conducting systems [22,23], it does not explain the entire experimentally observed magnon peak width [9], especially in the high-frequency range. Hence, other conceivable damping mechanisms may arise due to spin dynamics occurring beyond the linear-response regime (expressible in the language of multimagnon processes [24]), relativistic spin-orbit coupling (SOC) [25,26], and the presence of structural disorder [27]. None of these effects have been systematically studied within a realistic *ab initio* framework so far. Here, we introduce a methodology capable of accounting for all these effects. Thus, we provide a clear *quantitative* hierarchy of spin dynamics damping mechanisms in itinerant magnetic films dominated by the Landau mechanism and disorder with multimagnon processes contributing weakly to the magnon linewidth.

Correlated ground state. The electronic structures are obtained using a first-principles Green's function method [28], fully taking into account the effects associated with the semi-infinite substrates and, if necessary, the disorder on the level of the coherent potential approximation (CPA).

For Co grown on the Cu surface a major deficiency of the LSDA is the resulting location of the occupied majority states too far below the Fermi level. In turn, this yields a too large Stoner exchange splitting between majority and minority bands. While the experimental value for the exchange splitting for similar systems is reported to be around 0.8 eV [29] the LSDA predicts values between 1.7 and 2 eV depending on the position in the Brillouin zone. As shown below, this leads to substantial overestimations of magnon energies. The origin of this shortcoming is that the LSDA systematically fails to reproduce important correlation effects in the band structure of magnetic 3d transition metals [30], influencing the values of the bandwidth and exchange splitting, as well as the presence of satellite states. In order to account for these effects, one must evaluate the electronic self-energy, e.g., using Hedin's approach [many-body perturbation theory (MBPT) framework] [21,31], evaluating selected classes of Feynman diagrams. In particular, the possibility of an electron or hole decay associated with the emission of a virtual magnon accounting for the conservation of the spin angular momentum ("electron-magnon interaction"), turns out to be essential in the description of 3d magnets [21]. For bulk materials, the reduction of the exchange splitting has been shown to originate from the Coulomb-hole screened-exchange (COHSEX) self-energy [32] or the introduction of the electron-magnon interaction [33]. The latter effect has been also studied on the model level by Edwards and Hertz [34,35]. While being state of the art, such calculations are computationally

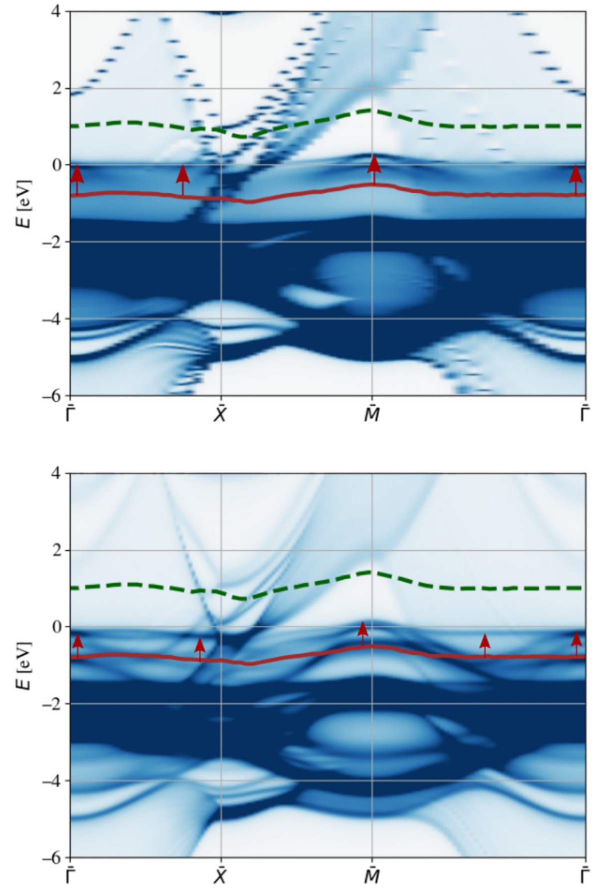

FIG. 1. Electronic spectrum of majority spin carriers in three layers Co on a Cu(100) substrate. Top: Result within the LSDA including the electron-magnon interaction (ordered system). Bottom: Result within the LSDA + U with $U = -1.6$ eV including disorder. The red/green solid/dashed line represents the highest almost horizontal band for majority/minority carriers within the LSDA. Arrows indicate the shift of the majority band through the electron-magnon interaction (or the "negative U correction").

demanding and so far have hardly been applied to complex solids and nanostructures. In our recently proposed computational scheme [21], we successfully approximate the corresponding series of ladder diagrams in Hedin's theory [36] with less expensive response functions and kernels available in the time-dependent density functional theory [22] allowing us to address systems as complex as Co films considered here. The upper panel in Fig. 1 shows the impact of spin fluctuations on the band structure of the 3-ML Co/Cu(001) film. The magnon-electron interaction shifts the occupied majority bands towards the Fermi energy. The energy shift of the majority bands amounts to approximately 0.8 eV, in agreement with results from hcp cobalt [16,30]. The quasiholes in the majority-spin channel acquire a finite lifetime being dressed now in the gas of virtual magnons. The impact of the spin fluctuations on the minority band is much weaker due to fewer electron partners in the unoccupied spin-up channel for the exchange of the magnons. It turns out that the shift of the band (but not the hole lifetimes) can indeed be modeled upon the application of $U = -1.6$ eV (in a standard LSDA + U calculation) on the 3d bands of Co (cf. lower panel in Fig. 1).

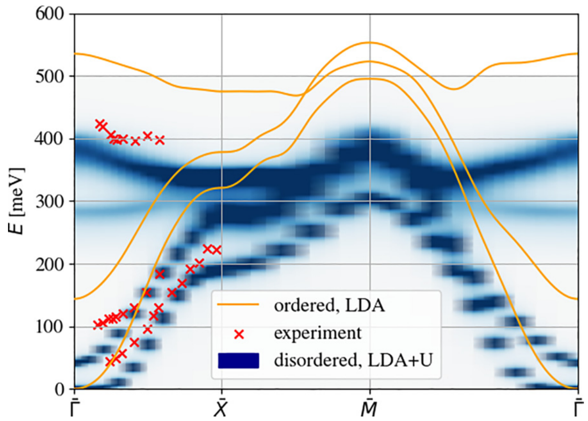

FIG. 2. Magnonic spectrum of Co/Cu(100) calculated within the LSDA (orange lines) and with the LSDA + U and disorder (blue background). The red crosses are experimental results [19].

While being methodologically limited, the latter approach allows, nevertheless, to determine the band structure self-consistently which is still beyond the computational reach of the current many-body approach (LSDA + $\mathcal{V}_{e-m}$). Consequently, we choose to use the LSDA + U ground state as the basis for the calculation of the magnon spectra as they are are sensitive to the details of the band structure close to the Fermi energy, where the self-consistency is particularly important.

Interestingly, in the Co/Cu system (but not Co/Ir), the LSDA + U correction results in the ferromagnetic ground state becoming unstable. At first glance this is not surprising, as the shift of the bands towards the Fermi level results in long-range exchange interactions between magnetic moments with oscillating sign. However, this contradicts the experimental findings. This hints at a missing element in the theoretical description. According to the experimental evidence [37,38] both films feature a certain degree of disorder, generally less significant for Co/Cu compared to Co/Ir. The primary effect of disorder in Co/Cu is the smearing of majority bands below the Fermi level as shown in the lower panel of Fig. 1, the finite lifetime arising from the collisions of electrons with the lattice imperfections, and the consequent stabilization of the ferromagnetic ground state. Nevertheless, the smearing is comparable with the electronic lifetime acquired due to the exchange of virtual magnons. Thus, it is likely that both disorder and electron-magnon scattering are decisive in the description of Co/Cu. However, as we shall show, it is crucial in the description of the spin-wave *damping* in Co/Ir.

The impact of spin fluctuations on the band structure of the 3-ML Co/Ir(001) film is significant as well (see Supplemental Material Note II [39]). However, the impact of spin fluctuations cannot justify the value of $U = -1.6$ eV necessary for reproducing the experimental magnon energies. The contrast between these two seemingly similar films placed on the Cu and Ir substrates reveals the rich many-body physics yet to be unveiled for nanostructures.

Spin-wave energies and Landau damping. Figure 2 shows the spin-wave dispersion for Co/Cu calculated from a Heisenberg ferromagnet with exchange couplings obtained from the magnetic force theorem [40]. While the LSDA leads to magnon energies which are much too high (as also discussed in Refs. [9,19]), the inclusion of the "negative U correction" and disorder in the system leads to a good agreement with the experimental results obtained by means of SPHREELS [9,19]. Note that the result for the disordered system in Fig. 2 has four magnon modes while in the experiment only three modes were observed. Theoretically, this is expected, as the magnetic Co atoms are spread across four layers in the disordered system (cf. Supplemental Material Note I [39]). The spectral density of the almost dispersionless mode at $E \approx 300$ meV is much smaller than for the other modes, which we suspect to be the reason for its absence in the experimental data. A closer inspection of Fig. 2 reveals a very low spin stiffness. We believe that this is a remnant of the magnetic instability encountered when considering the ordered system within the LSDA + U. In the latter case the magnon energies are negative close to $\bar{\Gamma}$ as also shown in the Supplemental Material Note II [39]. The situation is similar for Co/Ir with a few exceptions. Most notably, the ferromagnetic ground state is stable also after the negative U correction.

Time-dependent density functional theory (TDDFT) is capable of natively describing one of the dominating magnon decay channels in metallic magnets, the Landau damping. It involves the collision of the collective spin wave with single-particle spin-flip excitations, the Stoner excitations. Our scheme [22] involves the solution of the *susceptibility Dyson equation* $\chi^{\pm} = \chi^{\pm}_{\mathrm{KS}} + \chi^{\pm}_{\mathrm{KS}} K_{\mathrm{xc}} \chi^{\pm}$ where χ_{KS} is the Kohn-Sham susceptibility and K_{xc} represents the exchange-correlation kernel. The use of KKR Green's function in the TDDFT calculations allows to describe the impact of the truly semi-infinite nonmagnetic substrate on the spin-wave Landau damping. In the specific considered systems, the impact turns out to be small, due to the weak hybridization of substrate and film states. From the imaginary part of the susceptibility we extract the width of the magnon peaks utilizing a Lorentzian fitting function (the same method was used for disorder-induced damping discussed later). The Landau damping is believed to be dominating in conducting nanostructures (except for half metals [41]) and reproduces the SPHREELS data for Co/Cu [9]. However, in the case of Co/Ir(001), the damping rate for optical terahertz magnonic bands is clearly underestimated [9]. This hints at another important spin-wave attenuation mechanism operative in ultrathin magnetic films.

A careful analysis of the experimental results, in particular the magnon damping, obtained at room temperature and at 10 K showed nearly the same results, indicating the negligible role of temperature-induced magnon decay in this temperature range. This is not surprising, as the Curie transition in the films considered here occurs far above the room temperature [42] and at significantly lower temperatures there are not many thermally excited magnons which could contribute to the decay processes. Correspondingly, our theory targets the low-temperature regime.

Non-Landau magnon decay channels. Only a few studies on other than Landau magnon damping have been published thus far. For instance, some research has been conducted at an analytical level regarding scattering on impurities [43–45]. Here, we quantify them in an *ab initio* scheme. Conceivable non-Landau damping channels are depicted schematically in Fig. 3 and discussed in detail in the following.

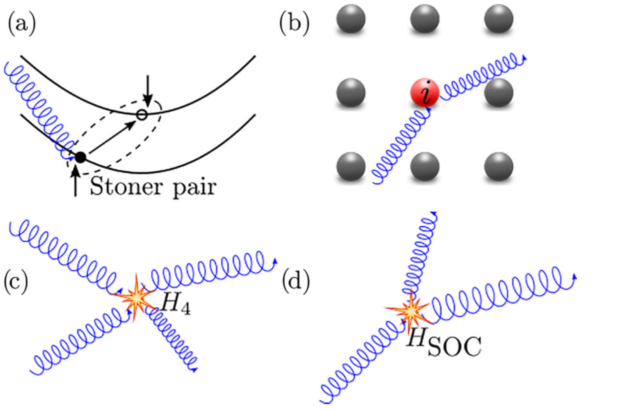

FIG. 3. Magnon (depicted as blue arrows) damping mechanisms in itinerant electron ferromagnets. (a) Landau damping: The magnon is absorbed by an electron which is excited in the process. (b) Damping through disorder: The magnon scatters on a crystal impurity i. (c) Four-magnon processes: Two magnons scatter on each other. (d) Three-magnon processes: One magnon decays into two magnons.

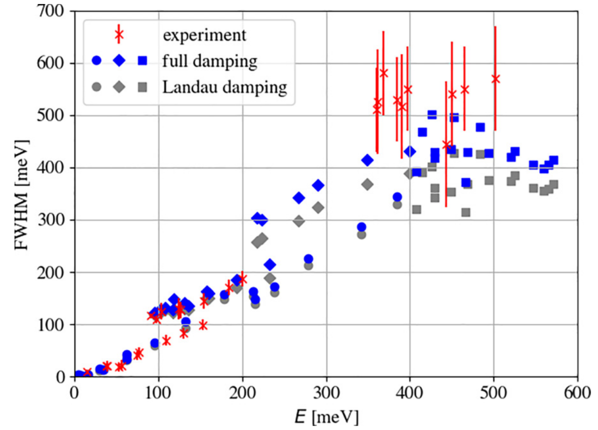

FIG. 4. Magnon damping along $\bar{\Gamma}$-$\bar{X}$ and $\bar{\Gamma}$-$\bar{M}$ due to Landau (ordered, LSDA + U) and non-Landau mechanisms (disordered, LSDA + U) in Co/Ir. The Landau damping (gray markers) and the full damping (including Landau, disorder, and multimagnon processes) depicted with blue markers are compared to experimental data [9]. Different magnon modes are represented by different marker shapes.

Let us consider the magnon-magnon interaction contribution to the lifetime first. We discuss the process on the level of the Heisenberg Hamiltonian. In general, a magnon can decay into one or more magnons [24]. However, in the absence of the spin-orbit coupling (SOC), in ferromagnets, the magnons are elementary excitations and eigenstates of the magnetic system. When there is no excited gas of magnons to interact with, this channel is inactive. The observation pertains to the TDDFT as well. In half metals, under weak SOC assumption, there is no Landau damping and the spin waves do not decay [41]. An excitation of multiple magnons in a certain process corresponds to large precession amplitudes of magnetic moments and is a nonlinear effect which cannot be grasped in the linear-response theory. However, the SPHREELS involves a continuous generation of magnons caused by the electron bombardment of the sample. Thus, even at low temperatures of the experiment when there are no thermally excited spin waves, a magnon gas can form, facilitating the decay. A direct magnon-to-magnon decay cannot occur (the magnons being eigenstates) but a given magnon coupled with another magnon of the gas can decay into a new pair of magnons ("four-magnon process"). With SOC present, a magnon can furthermore decay into two magnon states with the lattice absorbing the excess angular momentum ("SOC-" or "three-magnon process").

In order to quantify these contributions we write the Heisenberg Hamiltonian as follows:

$$H = \sum_{\boldsymbol{k}} \sum_{ij} a_i^\dagger(\boldsymbol{k}) T_{ij}(\boldsymbol{k}) a_j(\boldsymbol{k}) + H_4 + H_{\text{SOC}}. \tag{1}$$

Here, the first term describes the noninteracting bosons with the torque matrix T while H_4 is the first term beyond the linearization of the Holstein-Primakoff transformation describing the four-magnon processes involving the interaction with the magnon bath [cf. Fig. 3(c)]. H_{SOC} represents the three-magnon processes enabled by the SOC [cf. Fig. 3(d)]. For the latter Hamiltonian, the exemplary Dzyaloshinskii-Moriya interaction (DMI) form is utilized. Both H_4 as well as H_{SOC} are treated as perturbations and the corresponding magnon decay rates can be calculated using Fermi's golden rule $\Gamma_{sc}^{i\to f} = 2\pi\rho(E_f)|\langle f_{sc}|H_{sc}|i_{sc}\rangle|^2$ with $sc \in \{4, \text{SOC}\}$. Here, both the initial and final state for the four-magnon process ($|i_4\rangle$ and $|f_4\rangle$) are two-magnon states. For the three-magnon process, the initial state $|i_{\text{SOC}}\rangle = \mathcal{A}^\dagger|0\rangle$ is a single magnon state ($\mathcal{A}^\dagger$ being the magnon creation operator acting on the vacuum state) and the final state $|f_{\text{SOC}}\rangle$ is a two-magnon state. A detailed description of the formalism and the results is given in Supplemental Material Note I [39]. The main observation is that the magnon-magnon-induced decay rate is orders of magnitude smaller than the Landau damping: $\Gamma_4 < 5$ meV and $\Gamma_{\text{SOC}} < 1$ meV.

Lastly, we show that this is not the case with the disorder-induced damping [27,46]. We find that the disorder-induced damping is sensitive to the shape of the magnon mode and can differently affect two modes of similar energy and momentum, leading to a "mode-selective damping" and constituting an attractive linewidth engineering approach (cf. Supplemental Material Note II [39]). The total magnon damping for Co/Ir compared with the experimental data is shown in Fig. 4. Despite the large error bars of the highest-energy mangon mode, there is a clear trend showing that the Landau damping itself cannot account for the width of the peaks. Their linewidth is clearly underestimated and the inclusion of the non-Landau damping channels, dominated by the disorder-induced scattering, accounts for a substantial part of the missing linewidth. However, the damping is still underestimated which we believe might result from the coupling of the transverse magnons to the longitudinal spin dynamics [47] arising due to the SOC but not included in this Letter.

We find the same magnitude of non-Landau damping in Co/Cu. Hence, we find that the magnon damping in Co/Cu is generally overestimated by up to 50 meV. The reason for this might be the missing damping of electronic states within the LSDA + U as mentioned before.

In summary, using a first-principles approach, we uncovered an intricate picture of spin dynamics and its damping in itinerant 3d magnetic nanostructures governed by a fine interplay between nontrivial correlation effects dominated by electron-magnon scattering and disorder. We believe that the electron-magnon interaction might be a generic effect and an indispensable ingredient in the description of all itinerant magnets. Furthermore, we established a quantitative hierarchy of attenuation mechanisms dominated by the Landau channel and disorder with the multi-magnon processes playing a secondary role.

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