Interface Electronic Complexes and Landau Damping of Magnons in Ultrathin Magnets

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The damping of magnons in ultrathin metallic magnets is studied from first-principles. We contrast Fe/Cu(100) and Fe/W(110) systems for which the influence of the substrate on the magnon life time differs strongly. We introduce the concept of Landau map in momentum space to assess the role of different electronic states in the attenuation. The formation of electronic complexes localized at the film-substrate interface leads to hot spots in the Landau maps and enhances the damping. This finding allows tuning the attenuation of high-frequency magnetization dynamics in nanostructures.

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The life time of excited magnetic states is one of the key parameters in the design of spintronic devices. In the rapidly growing field of magnonics [1,2] the collective precession of magnetic moments, so called spin-wave or magnon, is used to transmit [3,4] information. The dissipation of the magnon energy limits the distance over which the spintronic appliances can communicate and breaks the phase relationships crucial for the spin-wave interference [5,6]. On the other hand, the damping is important for the stabilization of a new memory state after rewriting of magnetic bits [7,8]. Today's magnonic devices operate in the gigahertz band corresponding to the Larmor precession in magnetic anisotropy fields [9]. However, well defined exchange driven spin-waves with typical energies in the terahertz range exist both in bulk [10] and in nanostructured materials [11] and a possibility of utilizing them in the "terahertz magnonics" has been explored [6]. At these frequencies the main attenuation mechanism is of the Landau type [12], where the precessing magnetic moments excite spin-flip single-electron transitions, creating so called *Stoner particle-hole pairs*. Despite obvious importance of the Landau damping for both fundamental physics and applications very little is known about its material specific features, in particular, in nanostructured systems. To our best knowledge no first-principles studies of these properties have been reported so far owing to the algorithmic and computational challenges such calculations pose. On the other hand, Costa *et al.* [12] examined spin-wave properties on the basis of empirical tight-binding approach to the electronic structure and recovered a great sensitivity of the result to the parameters of the model. We resort to the parameter free linear response time dependent density functional theory [13,14] in order to make quantitative predictions regarding the nature of Landau damped spin dynamics in ultrathin films.

In their pioneering work Muniz *et al.* [15] discussed the consequences of the reduced out-of-plane periodicity of magnetic films, concluding that the Landau attenuation of the spin-waves in the films should be more severe than in the corresponding bulk materials. Another aspect of dimensionality arises when considering films grown on nonmagnetic substrates. In this case the two dimensional periodicity of the system coexists with its infinite extent in the third dimension. As a consequence, the number of the electron states of the system corresponding to a given in-plane wave-vector vector $\mathbf{k}_{\|}$ increases infinitely, forming a continuum. Muniz *et al.* pointed to the Stoner transitions involving the electronic states of the substrate as an important contribution to the Landau damping.

In this Letter we present an alternative picture of the magnetic Landau damping in nanostructures and provide means for its engineering. To substantiate our conclusions we contrast two epitaxial systems: one monolayer (ML) Fe/Cu(100) and 1 ML Fe/W(110) and explore a novel mechanism leading to a drastically enhanced Landau attenuation at surfaces. We show that the simple dimensionality arguments are not sufficient to reflect the complexity of the substrate's impact on the spin-wave damping.

A transparent *ab initio* picture of spin-waves and their Landau damping is given by the linear response time dependent density functional theory [13]. The description includes two steps and, respectively, two important physical quantities. The response of the formally noninteracting Kohn-Sham electron system to a transverse magnetic field is given by the retarded susceptibility

$$\chi^{ll'}_{\mathrm{KS}}(\mathbf{r},\mathbf{r}',\mathbf{q},\omega) = \sum_{\mathbf{k}\in\Omega_{\mathrm{BZ}}}\sum_{nn'}(f^{\uparrow}_{n\mathbf{k}-\mathbf{q}} - f^{\downarrow}_{n'\mathbf{k}}) \times \frac{\psi^{\uparrow}_{n\mathbf{k}-\mathbf{q}}(l'\mathbf{r}')\,\psi^{\uparrow}_{n\mathbf{k}-\mathbf{q}}(l\mathbf{r})^{*}\,\psi^{\downarrow}_{n'\mathbf{k}}(l\mathbf{r})\,\psi^{\downarrow}_{n'\mathbf{k}}(l'\mathbf{r}')^{*}}{\omega-(\epsilon^{\downarrow}_{n'\mathbf{k}}-\epsilon^{\uparrow}_{n\mathbf{k}-\mathbf{q}})+i0^{+}}, \qquad (1)$$

where ω and $\mathbf{q}$ are the frequency and the wave-vector of the magnetic field. The response is determined by the electronic transitions between occupied ($f = 1$) majority spin states ($\sigma =\uparrow$) with crystal momentum $\mathbf{k} - \mathbf{q}$, and empty ($f = 0$) minority ($\sigma =\downarrow$) states with momentum $\mathbf{k}$. They are called Stoner excitations. The momenta belong to the two dimensional Brillouin zone (Ω_{BZ}) of the system, index l labels the layers of both the film and the substrate,

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while $\mathbf{r}$ varies within the given layer. $\psi^{\sigma}_{n\mathbf{k}}$ stands for the Bloch state of band n and $\epsilon^{\sigma}_{n\mathbf{k}}$ is the corresponding Kohn-Sham energy. The induced magnetization described by the Kohn-Sham susceptibility modifies the exchange-correlation potential which gives rise to a self-consistent problem: the magnetization contributes to the effective magnetic field and is, simultaneously, induced by this field. The self-consistency is reflected in the second step of the formalism, a Dyson-like equation

$$\chi^{ll'} = \chi^{ll'}_{\mathrm{KS}} + \sum_{l_1 l_2} \chi^{ll_1}_{\mathrm{KS}} K^{l_1 l_2}_{\mathrm{xc}} \chi^{l_2 l'}. \tag{2}$$

All quantities entering the equation are represented by matrices in a basis of orthonormal functions centered at the atomic positions; in our calculations we employ a basis combining Chebyshev polynomials and spherical harmonics. The exchange-correlation kernel, K_{xc}, describing the evolution of the effective Kohn-Sham potential, is based in our work on the adiabatic local density approximation [13,14]. The solution of this equation, $\chi^{ll'}(\mathbf{q}, \omega)$, is the true magnetic susceptibility of the system and includes information about the collective spin-flip modes (spin-waves). The latter excitations are identified as the peaks in the imaginary part of the susceptibility and signify a strong absorption of energy by the system. The frequency corresponding to the maximum of the peak, $\omega_0(\mathbf{q})$, is identified as the magnon energy (dispersion relation), whereas the full width at half maximum (FWHM) of the magnon peak is interpreted as the inverse life time of the excitation [11]. The finite life time of the spin-waves originates from their hybridization with the continuum of the single-electron Stoner excitations, appearing as singularities of the Kohn-Sham susceptibility χ_{KS} in Eq. (1).

In Fig. 1 we present the calculated magnon spectra for the two Fe films considered. In both cases we compare the supported monolayers with their freestanding counterparts. We note, first, that for the freestanding film the attenuation of the spin-waves is weak. This behavior is in a drastic contrast to the case of the bulk bcc Fe where both experiment and theory [10] show that only small momentum spin-waves are well defined. Second, the presence of the substrate leads to the modification of the magnon dispersion and increases the Landau damping. This latter trend is consistent with the conclusions of previous works [12]. However, a remarkable difference between the influence of substrate in Fe/Cu(100) and Fe/W(110) systems can be seen.

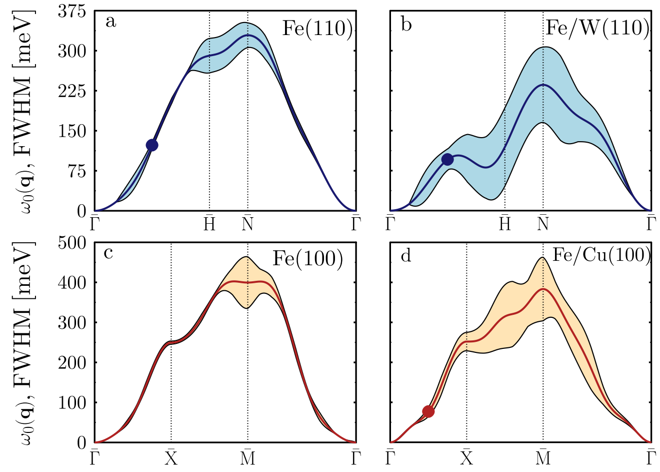

FIG. 1 (color online). Magnon spectra in iron films. Thick lines denote the dispersion relation, $\omega_0(\mathbf{q})$, and the width of the shaded area corresponds to the full width at half maximum on the energy axis. The Stoner spectrum contributing to the damping of marked magnons (●) is analyzed in Fig. 2.

In Fe/Cu(100), the impact of the substrate is moderate and the spin-waves are well defined in the whole Brillouin zone whereas in the case of Fe/W(110) attenuation increases severely. Around points $\bar{H}$ and $\bar{N}$ in the Brillouin zone the spin-wave energy is comparable with the damping expressed in energy units. Indeed, the strongly damped collective precession has been observed experimentally at the zone boundary in Fe/W(110) system [11]. We emphasize, however, that even for Fe/W(110) the region in the Brillouin zone featuring the spin-wave disappearance effect is small compared to the bulk case. Also for films with several monolayers the experiment [16] clearly shows that the spin-wave damping is weaker than in the bulk case that is in full agreement with our calculations (not shown).

An important insight in the spin-wave properties of the film-substrate system can be gained from the analysis of the susceptibility Dyson equation (2). Our calculations show that the small magnetic polarizability of the substrate leads to the effective decoupling of the magnetic film from the substrate. Retaining in Eq. (2) only blocks of the susceptibilities, which correspond to the magnetic film ($l = l' = \mathrm{Fe}$), does not alter the dispersion or damping noticeably. This implies that the spin procession associated with spin waves is confined in the film region. The film and substrate form one common system and valence electron eigenstates are spread in both the film and the substrate. Nevertheless, only the Stoner pairs formed by the electronic states substantially present in the film region can contribute to the damping. Although the three dimensional character of the substrate leads to the formally infinite increase of the number of Stoner transitions in the film-substrate system, the presence of each of the states in the film region decreases. The resulting influence of the substrate depends on film-substrate hybridization and is strongly material specific.

In order to assess the contributions of different electronic states to the damping, we introduce the concept of *Landau map*; cf. Fig. 2. We focus on a particular magnon with momentum $\mathbf{q}_0$ (indicated in Fig. 1) and compute the $\mathbf{k}$-resolved spectral density of Stoner transitions with momentum $\mathbf{q}_0$ and energy $\omega_0(\mathbf{q}_0)$. As discussed above, only the Fe-Fe block of the Kohn-Sham susceptibility is considered, since it describes the Stoner excitations responsible for the Landau damping. The difference between the maps of the free and supported films is dramatic. In the

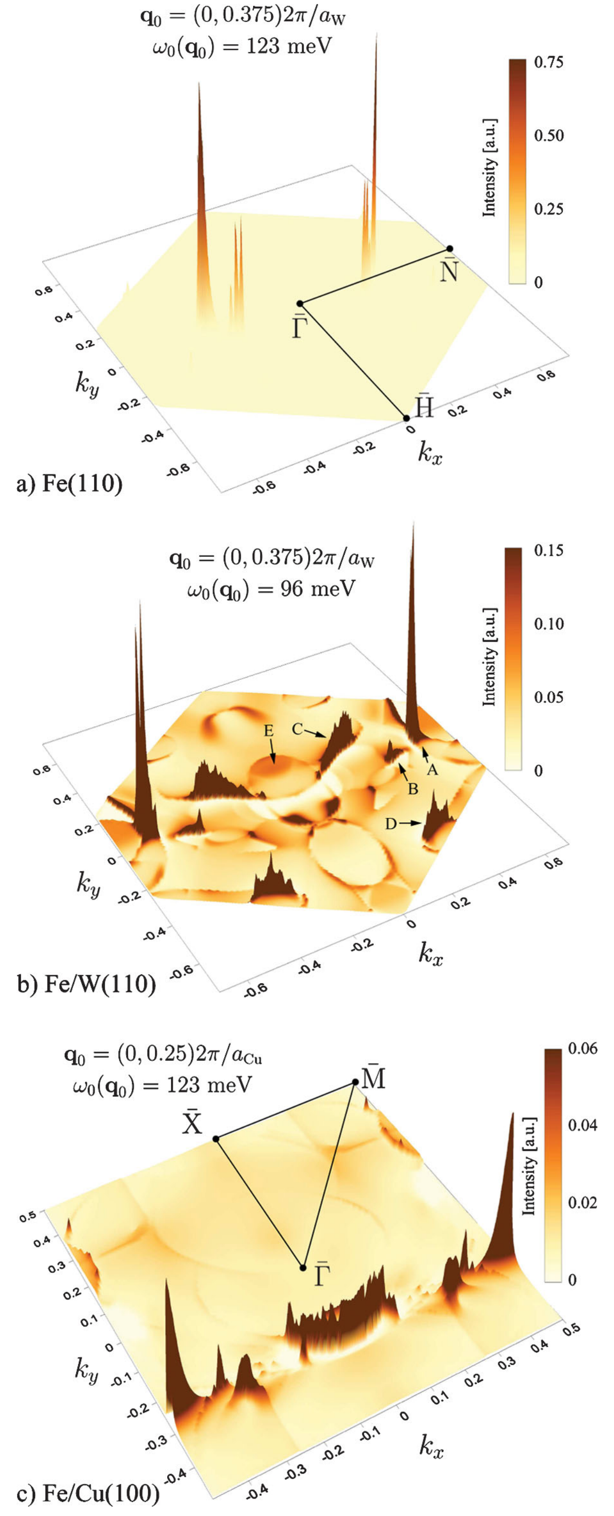

FIG. 2 (color). Intensity of Stoner transitions with momentum $\mathbf{q}_0$ and energy ω_0 in Fe layer resolved for different final $\mathbf{k}$ vectors in the first Brillouin zone. The Stoner states cause the damping of magnons indicated in Fig. 1.

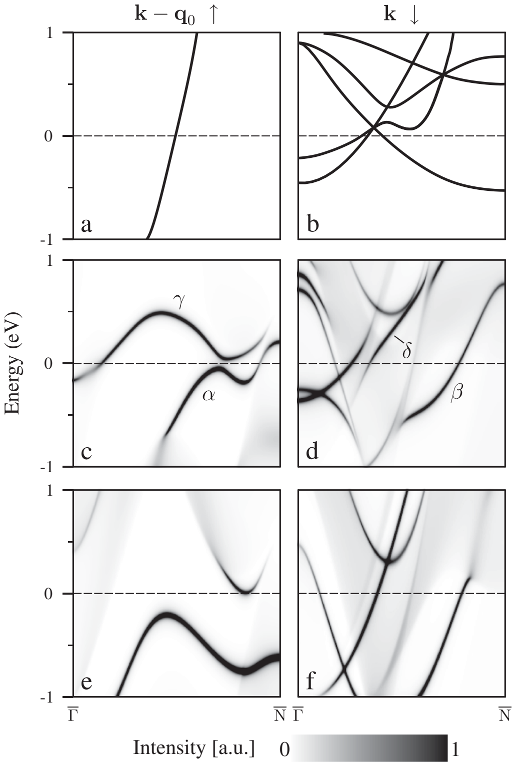

FIG. 3. Spectral density of initial (left column) and final (right column) electron states involved in the formation of Stoner pairs contributing to the damping of magnon with momentum $\mathbf{q}_0$. Final states with momentum $\mathbf{k}$ along $\bar{\Gamma}\bar{N}$ are shown. $\mathbf{q}_0 = (0, 0.375)(2\pi/a_W)$ is marked in Figs. 1(a) and 1(b). Rows from the top: freestanding Fe(110) layer, Fe overlayer of Fe/W(110), and topmost layer of clean W(110) surface.

case of freestanding film the Stoner transitions contributing to the Landau damping are strongly localized in momentum space and form a *Landau hot spot* along $\bar{\Gamma}\bar{N}$ direction in the Brillouin zone. It can be traced back to the electronic structure of the free film; cf. Figs. 3(a) and 3(b). In the energy interval of interest there are occupied spin-up states of a single s-type band and empty spin-down states of the spin-down d-type bands. The large slope of the spin-up band results in a limited possibility of forming the Stoner pairs on the energy scale of magnons.

If the continuum of the substrate bulklike states were the decisive factor in the strong Landau damping of the supported monolayer, the corresponding Landau map of Fe/W(110) would show hardly any sharp features. Instead, the Stoner transitions for the energy associated with the magnon would be available for almost any $\mathbf{k}_\parallel$ resulting in a relatively uniform filling of the map. Surprisingly, the damping of Fe/W(110) is still dominated by only slightly broadened hot spots [cf. Fig. 2(b)]. Comparing to its freestanding counterpart the number of hot spots increases. Apart from the hot spots, there appears a diffuse background with a clearly smaller intensity.

To identify the origin of these features we consider the electronic structure of the Fe/W(110) system. Figure 3 shows the $\mathbf{k}_\parallel$-resolved electronic spectral density in the surface layer. The difference between the electronic structure of freestanding and supported films is striking. In the latter case we distinguish extended colored areas corresponding to the states delocalized over the volume of the film-substrate system. Their weak intensity reflects the small probability of detecting those states in the Fe layer. In the energy gap (white regions) of the bulk electrons there appear states strongly localized in the film region seen as narrow intense lines. The analysis of these bandlike features shows that they are formed by the hybridization of the states of the Fe film and the states of the W(110) substrate confined to its surface (so called surface states [17,18]). We will refer to these hybrid orbitals as *interface electron complexes*. Their formation can be particularly clearly traced in the spin-up channel [Fig. 3(c)] where a single Fe s-band band [Fig. 3(a)] is replaced by the two flat bands as a result the hybridization with the surface states of W(110) visible in Fig. 3(e). The states of the complexes appearing at the Fermi level region yield the leading contribution to the spin-wave Landau attenuation.

In the Landau map of Fe/W(110); cf. Fig. 2(b), 50% of the spectral power is cumulated in spots A and B, which involve transitions between the interface complexes marked in Figs. 3(c) and 3(d) as α and β. Also hot spots C (transition between bands γ and δ) and D form due to such interface states. The well defined spots are responsible for 70% of the damping. Region marked with E originates from electron states in the film hybridizing strongly with the continuum of bulk states in both spin channels. It is evident that such Stoner pairs are of minor importance.

Small number of the spin-up states available for the Stoner-transitions in freestanding film leads to the weak damping of the spin waves. The situation is close to the case of half-metallic systems where the gap in the spectrum of the Stoner transitions excludes the Landau damping of low energy magnons [14]. We note that not only the damping but also the dispersion are strongly influenced by the formation of the interface complexes. The half-metallicity implies short-range exchange interaction between Fe moments [19] and results in a simple cosine-type $\omega(\mathbf{q})$ dependence, evident in Fig. 1. The interface complexes serve not only as an effective source of Stoner transitions contributing to the damping but also mediate the long range exchange coupling between magnetic moments in the film, manifesting itself as the clear dip in the magnon dispersion relation of Fe/W(110).

The observations made above allow us to interpret the large difference between Fe/W(110) and Fe/Cu(100). Cu(100) substrate does not support surface states in the Fermi level region. In contrast to the Fe/W(110) case, the calculated electronic structure of the Fe overlayer on Cu(100) (not shown) differs weakly from its freestanding counterpart. The electron states in the supported film retain basically their free monolayer character, acquiring certain energy width due to the coupling to the continuum of the substrate states. The Landau map of this system, cf. Fig. 2(c), reveals series of rather diffuse hot spots, corresponding to the transitions between those broadened Fe states. The intensity of the spots adds up to about only 20% of the total intensity of the map so the difference comparing to the free Fe(100) layer originates from the weak extended background of the map. As a result, the magnon spectrum is weakly affected by the substrate; cf. Fig. 1. In systems with well defined atomic moments and weak Landau damping the spin-wave dispersion obtained from the dynamic susceptibility is close to the one found in the adiabatic treatment of magnons; cf. Refs. [20,21].

In summary, we presented a study of Landau damping of magnons in ultrathin metallic magnets supported on a semi-infinite substrates. In order to assess the role of different electronic states to the attenuation we introduced the concept of Landau map. We conclude that contribution of the continuum of substrate states is relatively weak. As a result, the magnons in ultrathin films can live longer than their bulk counterparts. On the other hand, the surface states of the substrate can give rise to electronic complexes strongly localized at the film-substrate interface leading to a drastic reduction of the life time. These two mechanisms allow to devise metallic nanostructures with a desired damping of high-frequency magnetization dynamics.

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