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Magnons in complex systems from TDDFT

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Introduction

The properties of excited magnetic states are of great importance in the fundamental and applied magnetism. Their spectrum determines the thermodynamical properties of magnets, including the Curie temperature [11]. The excitations contribute to the electronic specific heat [5] and the electrical and thermal conductivity, couple to charge degrees of freedom [14], and can provide a coupling mechanism in high temperature superconductors alternative to phonons [10]. The control of spin dynamics and its attenuation are the central problems in the rapidly growing field of spintronics [9].

Until now the main body of theoretical studies on magnetic excitations has been based on the adiabatic treatment of magnetic degrees of freedom [8]. The approach describes correctly long wavelength *spin-waves* (*magnons*), i.e. collective modes associated with the coherent precession of atomic moments. It captures most of the physics of non-conducting materials, but is of limited validity in the metallic magnets, because it neglects the presence of particle-hole states with opposite spin called *Stoner excitations*. The Stoner states are pronounced mostly at the energies corresponding roughly to the exchange splitting of electron states, but they can have non-vanishing contribution also in the lower energy region where the magnons appear. The resulting hybridization leads to the attenuation of spin-waves. The effect is called *Landau damping*, influences qualitatively the spin dynamics of metals and cannot be described in the adiabatic theories.

The damping can be captured in calculations of wave-vector and frequency dependent transverse magnetic susceptibility $\chi(\mathbf{q}, \omega)$, where spin-waves and Stoner states are treated on an equal footing. The dynamic method became particularly powerful after the parameter free linear response density functional theory (LRDFT) [7] was formulated. Such calculations are, however, very demanding both from the point of view of algorithmic complexity and computer resources and for a long time they were restricted to simple bulk systems. [13, 15]

Recently, we have developed a novel efficient numerical scheme allowing to evaluate the spin susceptibility of complex magnets and applied it to study energies and life-times of magnons in complex bulk phases [1, 4] and ultrathin films [2, 3].

The spin-wave attenuation is determined by fine properties of Stoner continuum. The first principle approaches based on the calculation of transverse magnetic susceptibility are indispensable in the consistent description of spin dynamics in real materials.

Below we outline briefly the formalism and discuss closer several case studies of spin flip dynamics in complex magnets.

Method

Linear response time dependent density functional theory allows to compute the generalized susceptibility in the following two step procedure. [7, 12] We focus on the transverse magnetization dynamics. First, one considers the *Kohn-Sham susceptibility*

$$\chi^{\pm}_{\mathrm{KS}}(\mathbf{x},\mathbf{x}',\omega) = 2\sum_{km}\left(f_k^{\uparrow\downarrow} - f_m^{\downarrow\uparrow}\right)\frac{\phi_k^{\uparrow\downarrow}(\mathbf{x})^*\phi_m^{\downarrow\uparrow}(\mathbf{x})\phi_m^{\downarrow\uparrow}(\mathbf{x}')^*\phi_k^{\uparrow\downarrow}(\mathbf{x}')}{\omega+\epsilon_k^{\uparrow\downarrow}-\epsilon_m^{\downarrow\uparrow}+i0^+}. \tag{1}$$

giving the retarded response of the formally non-interacting Kohn-Sham system. $\phi_j(\mathbf{x}\alpha)$'s and ϵ_j's denote respectively KS eigenfunctions and corresponding eigenenergies. $f_j \equiv f_T(\epsilon_j)$, where $f_T(\epsilon)$ is the Fermi-Dirac distribution function. The induced magnetization densities described by the Kohn-Sham susceptibility modify the exchange-correlation potential, giving rise to a self-consistent problem: the induced densities contribute to the effective fields and are, simultaneously, induced by it. The self-consistency is reflected in the second step of the formalism

$$\chi^{\pm}(\mathbf{x},\mathbf{x}',\omega) = \chi^{\pm}_{\mathrm{KS}}(\mathbf{x},\mathbf{x}',\omega) + \int d\mathbf{x}_1\chi^{\pm}_{\mathrm{KS}}(\mathbf{x},\mathbf{x}_1,\omega)K_{\mathrm{xc}}(\mathbf{x}_1)\chi^{\pm}(\mathbf{x}_1,\mathbf{x}',\omega). \tag{2}$$

The last equation is referred to as "susceptibility Dyson equation", because of its characteristic form. χ is the physical (enhanced) susceptibility of the system. The exchange-correlation kernel, K_{xc}, is defined as a functional derivative of exchange-correlation potential with respect to the density

$$K^{ij}_{\mathrm{xc}}[\langle\hat{\sigma}(\mathbf{x})\rangle](\mathbf{x},\mathbf{x}',t-t') = \frac{\delta v^i_{\mathrm{xc}}(\mathbf{x},t)}{\delta n^j(\mathbf{x}'t')} \tag{3}$$

evaluated at the ground state values of electronic and magnetic densities.

Examples

Spin-waves in half-metals

The *half-metalls* are very attractive materials for spintronic applications. We studied the relation between the half-metallicity and life-time of the spin-waves in a series of Heusler alloys [1]. We demonstrated that the acoustic spin wave mode remains practically undamped for spin-wave momenta in the entire Brillouin zone. On the contrary the optical modes feature a finite life-time changing strongly and non-monotonously with the momentum, cf. Fig. 1.

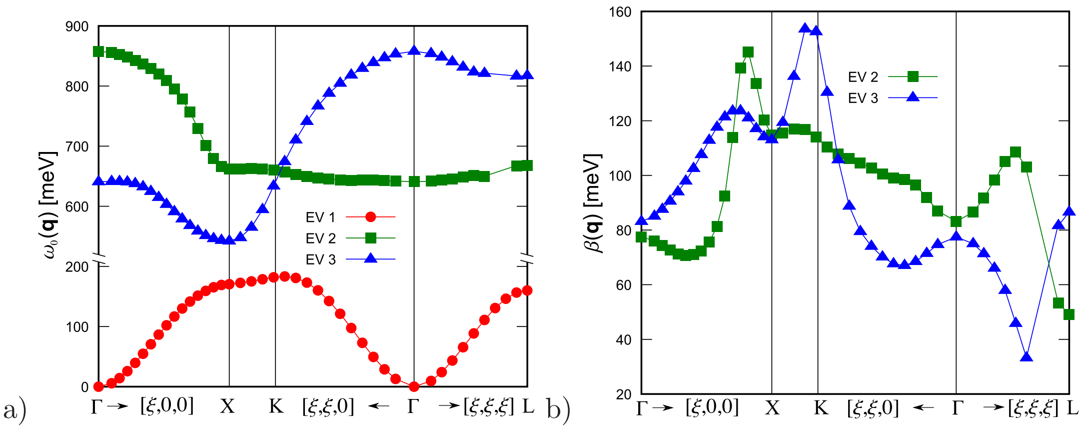

FIG. 1: Spin-waves in Co_2MnSi for different wave-vectors in the first Brillouin zone. Panel a) presents energies of the spin waves. Panel b) shows the half-width at half-maximum (HWHM) of the spin-wave peak (inversely proportional to the state's life-time) in the spectral density. The HWHM of the acoustic (EV 1) mode in this system is less than 5 meV and is not shown.

The knowledge of the life-time is however essential for applications. In the storage devices the excited states should decay as soon as possible, leaving a bit after a read-in or read-out in a steady state. On the contrary, in the inter-chip communication, the spin-wave should live as long, as it is necessary for signal to travel undistorted between emitter and antenna.

Controlling terahertz magnetization dynamics

Spintronics utilizes the spin degree of freedom to process and store information. Typical spintronic devices are operated at frequencies of the Larmor precession in magnetic anisotropy fields, which corresponds roughly to the GHz band. Only recently their spatial sizes has been reduced to the sub-μm regime. Recently, we focused on the possibility of controlling the magnetization dynamics in the THz range and on the scale of single nanometers [2]. We suggested that spatially confined (between the surface and the interface of the film) exchange driven *spin waves* could be utilized in a new generation of spintronic devices to scale down their sizes and to accelerate speed.

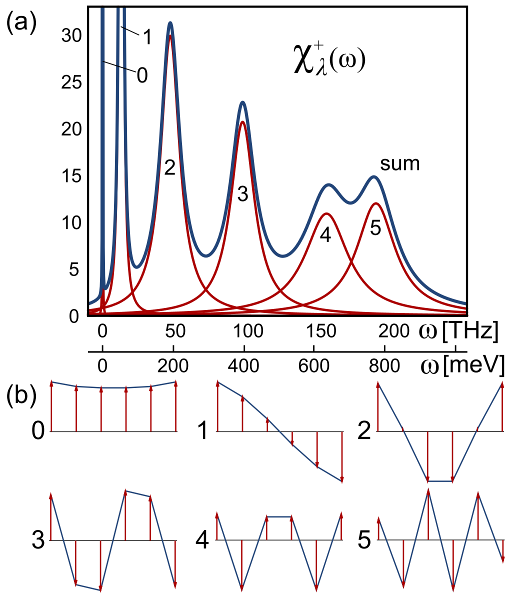

FIG. 2: (a) Spectral power of spin excitations for zero in-plane wave vector. Six Lorentzian peaks can be distinguished. They correponds to the standing spin-waves of the 6 ML Co/Cu(100) system. (b) Corresponding magnon eigenvectors. The arrows present layer-resolved transverse components of the magnetization at a certain moment in time. With time all moments precess about the z axis.

The impact of the substrate on the Landau damping in ultrathin films

We considerably advanced the understanding of the way a non-magnetic substrate influences the properties of the spin waves in thin magnetic films. To get insight into the properties of magnetic excitations formed by the combination of the 2D magnetic film and

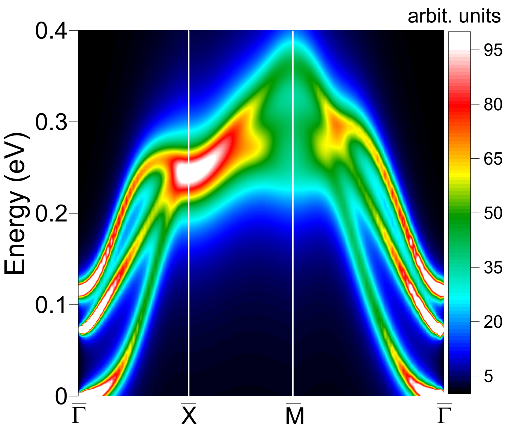

FIG. 3: Spectral intesity of spin-flip excitations in 3 ML Fe/Cu(100) system. Three branches of weakly damped standing spin-waves are clearly discernable.

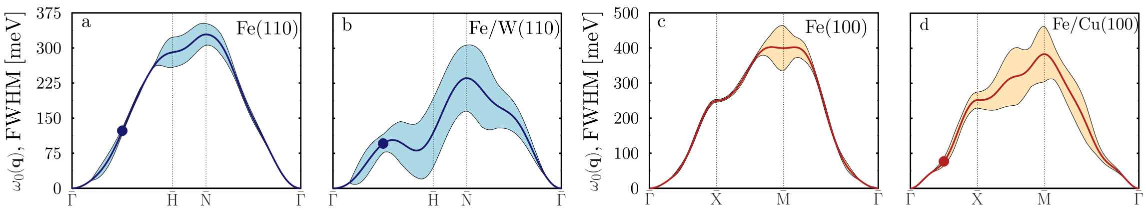

FIG. 4: Magnon spectra in iron films. Thick lines denote the dispersion relation, $\omega_0(\mathbf{q})$, and the width of the shaded area corresponds to the full width at half maximum on the energy axis. The Stoner spectrum contributing to the damping of marked magnons (•) is analyzed in Fig. 5.

3D nonmagnetic substrate we invented so-called *Landau maps* that vizualize the **k**-resolved intensity of the Stoner continuum and allow to determine the states responsible for the decay of the spin waves.

If the continuum of the substrate bulk-like 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 $\mathbf{k}_{\|}$ resulting in a relatively uniform filling of the map. Surprisingly, the damping of Fe/W(110) is still dominated by hot-spots (cf. Fig. 5b). The hot spots are responsible for 70% of the damping. The analysis of these features shows that they are formed by transition between so called *interface electron complexes*, i.e. electronic states resulting from the hybridization of the states of the Fe film and the surface states of the W(110) [6]. Region marked with E originates from electron states

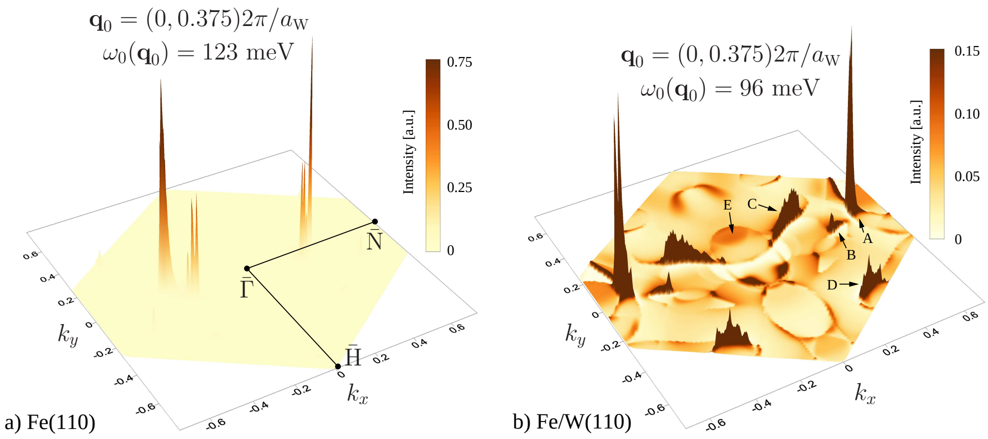

FIG. 5: Intensity of Stoner transitions with momentum $\mathbf{q}_0$ and energy ω_0 in Fe layer resolved for different final $\mathbf{k}$-vectors in the first Brillouin zone. The Stoner states cause the damping of magnons indicated in Fig. 4.

in the film hybridizing strongly with the continuum of bulk states in both spin channels. Such Stoner pairs are of minor importance.

In contrast to the Fe/W(110) case, the electronic structure of the Fe/Cu(100) differs weakly from its free-standing counterpart. Additionally, Cu(100) does not feature surface states crossing Fermi level. As a result, the magnon spectrum is weakly affected by the substrate, cf. Fig. 4.

Our current and future research will include such topics as the study of paramagnons excitations, spin-waves in non-collinear systems, magnetic excitations of clusters on metallic and non-metallic substrates.

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