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The energies and life times of magnons in bulk iron and one-monolayer Fe film

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ABSTRACT

The dynamic magnetic susceptibility is calculated for bulk bcc iron, a supported monolayer Fe/W(110) and corresponding free standing Fe monolayer. The calculational approach is based on linear response density functional theory formulated within the framework of Korringa–Kohn–Rostoker Green's function method. The calculated susceptibilities are analyzed in terms of dispersion and damping of spin-waves. The results for bulk Fe are in good agreement with experiment and previous theoretical studies. In contrast to the bulk phase, the spin-wave excitations in thin films are well defined in the whole two-dimensional Brillouin zone. It is shown that a non-magnetic substrate changes strongly both the dispersion and damping of the spin-waves.

1. Introduction

The excitations changing the magnetization of the system are of fundamental importance in the physics of magnets [1]. The excitation spectrum determines the thermodynamics of the magnetic materials and the magnetic transition temperature. The magnetic excitations (MEs) contribute to the electronic specific heat and the electrical and thermal conductivity. They couple to charge degrees of freedom [2], control the hot electrons' mean free path [3] and can even provide a coupling mechanism in high temperature superconductors [4].

The inelastic scattering of neutrons is a standard technique to probe bulk MEs [5], but due to the small interaction cross-section, it is ineffective in the case of nanostructured materials. Recently new experimental methods have been successfully applied to study MEs in nanostructures. They are based either on scanning tunneling microscopy [6] or on spin polarized electron energy loss spectroscopy (SPEELS) [7]. The latter technique, due to small penetration depth of electrons, probes the surface layers of the sample. These experiments offer a unique opportunity to study the exchange coupling in nanosystems and the features of the damping related to reduced dimensionality. The spin-dynamics in nano-systems is of strong practical importance in connection with the need for ever faster magnetic storage technologies.

Until now, the main body of theoretical studies on MEs have been based on adiabatic treatment of magnetic degrees of freedom, e.g. in the frozen magnon approximation [8–10]. The method utilizes density functional theory and does not involve adjustable parameters, yielding dispersion relation $\omega_0(\boldsymbol{q})$ of spin-waves (SWs), i.e. collective low-energetic modes. In this approach the presence of single particle excitations leading to particle–hole triplet states (so called Stoner excitations) is neglected. The Stoner excitations are pronounced mostly at higher energies, corresponding roughly to the exchange splitting of electron states, but they can, however, have non-vanishing contribution also in the SW energy region and lead to the damping of the SW states. These phenomena are captured in the calculations of wave-vector and frequency-dependent magnetic susceptibility $\chi(\boldsymbol{q},\omega)$, where SWs and Stoner states are treated on an equal footing. The position of the singularities of χ in the complex energy plane determines energies and life times of MEs. The imaginary part of χ is probed directly in the scattering experiments.

It was predicted that the breaking of translational symmetry in nanostructures should lead to the renormalization and enhanced damping [11] of the SW states. Therefore, for these systems, the use of a dynamical approach is essential. Up to now the study of the dynamic magnetic susceptibility of magnetic thin films was based on the use of model Hamiltonians [11]. *Ab initio* calculations of χ within the framework of linear response density functional theory (LRDFT) [12] have been so far performed only for few simple bulk materials [13]. It is important to extend the LRDFT studies of MEs to the film geometry. Although the model Hamiltonian studies provide important information on general relation between parameters of the Hamiltonian and calculated observables, the self-consistent DFT calculations are inevitable for the realistic description of complex systems since the excitations of magnetic systems depend sensitively on the detailed properties of the underlying electron structure.

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In this paper, we report a first attempt of LRDFT calculations for a magnetic film. The implementation of the LRDFT approach has been performed within the Korringa–Kohn–Rostocker (KKR) Green's function method [14]. The implementation allows to treat both bulk and 2D systems. The formalism is summarized in Section 2. In Section 3 we consider bulk bcc Fe, Fe monolayer (ML) on W(110) and the corresponding free standing Fe ML and reveal the role of low dimensionality in the properties of MEs.

Fe/W(110) system has been for nearly two decades one of the most intensively studied film systems due to its mechanical and structural properties (see e.g. [15] and references therein) interplaying with rich magnetic behavior. Generally, the system features strong in-plane magnetic surface anisotropy with an easy axis $[1\,\bar{1}\,0]$ [16]. Despite of a substantial lattice misfit (9.4%, $a_W = 3.161$ Å, $a_{Fe} = 2.866$ Å) the first ML of Fe grows pseudomorphically with a vertical lattice relaxation [15]. The theoretical study presented below was stimulated by the SPEELS SW measurements in this system [7] which revealed an unusual softening of the exchange interaction compared to bulk bcc Fe.

2. Formalism

We consider a system with a collinear ferromagnetic ground state. The properties of spin excitations are reflected in the analytic structure of the transverse magnetic susceptibility $\chi^{\pm}(\boldsymbol{x},\boldsymbol{x}',z)$, where $z \in \mathbb{C}$ and the function is analytic in the upper complex semi-plane. It has been shown (see e.g. [17]) that within the adiabatic local density approximation (ALDA) the susceptibility can be computed from the following "susceptibility Dyson equation":

$$\chi^{\pm}(\boldsymbol{x},\boldsymbol{x}',z) = \chi^{\pm}_{KS}(\boldsymbol{x},\boldsymbol{x}',z) + \int d\boldsymbol{x}_1 \chi^{\pm}_{KS}(\boldsymbol{x},\boldsymbol{x}_1,z) K_{xc}(\boldsymbol{x}_1) \chi^{\pm}(\boldsymbol{x}_1,\boldsymbol{x}',z), \quad (1)$$

where $\chi^{\pm} \equiv \chi_{xx} \pm i\chi_{xy}$ and χ_{ij} is the element of the susceptibility matrix which connects the ith component of induced magnetization with jth component of the external magnetic field. The response of a non-interacting Kohn–Sham (KS) system is given in terms of KKR Green's function G as

$$\chi^0_{ij}(\boldsymbol{x},\boldsymbol{x}',i\omega^b_n) = \frac{1}{\beta}\sum\nolimits_{m\in\mathbb{Z}} \boldsymbol{\sigma}^i_{\alpha\beta}\boldsymbol{\sigma}^j_{\gamma\delta} G_{\beta\gamma}(\boldsymbol{x},\boldsymbol{x}',z_m) G_{\delta\alpha}(\boldsymbol{x}',\boldsymbol{x},z_{m-n}), \quad (2)$$

where $i,j = x,y,z$ and $\boldsymbol{\sigma}$ stands for the vector of Pauli matrices. $z_n = \varepsilon_F + i\omega^f_n$ and ω^f_n and ω^b_n are nth fermionic and bosonic Matsubara frequencies, respectively. Small Greek letters denote spinor components. In spite of formal resemblance to the random phase approximation method, the formalism presented above is exact up to the specification of the exchange-correlation kernel, which in ALDA reads

$$K_{xc}(\boldsymbol{x}) = -\frac{1}{\mu_B}\frac{B_{xc}(\boldsymbol{x})}{m(\boldsymbol{x})}, \quad (3)$$

where $B_{xc}(\boldsymbol{x})$ and $m(\boldsymbol{x})$ are, respectively, local values of the exchange-correlation magnetic field and magnetization density.

If one casts the spatial dependence of $\chi^{\pm}$, $\chi^{\pm}_{KS}$ and K_{xc} in a given basis, the Dyson equation takes a matrix form

$$\chi^{\pm}(z) = \chi^{\pm}_{KS}(z) + \chi^{\pm}_{KS}(z) K_{xc} \chi^{\pm}(z). \quad (4)$$

This equation can be solved by means of matrix inversion.

The singularities of the Fourier transform $\chi^{\pm}(\boldsymbol{q},z)$ determine the excitation energies and life times. In the simplest case of one magnetic atom per unit cell the susceptibility in the SW energy range is dominated by a single pole:

$$\chi^{+}(\boldsymbol{q},z) \approx \frac{A(\boldsymbol{q})\, e^{i\delta(\boldsymbol{q})}}{z - \omega_0(\boldsymbol{q}) + i\beta(\boldsymbol{q})}. \quad (5)$$

3. Results

Fig. 1 presents the calculated SW spectrum of bulk bcc iron compared with experimental results [5]. The agreement of the theory and experiment is excellent. Experimentally, it has been established that in the bcc Fe the well defined SW excitations are observed only for small $\boldsymbol{q}$ vectors. The short wave-length SWs are severely damped. Our calculations confirm this: for $q > 0.4$ Å^{-1} the imaginary part of the SW pole increases and for larger $\boldsymbol{q}$ the damping is so strong that the SW excitations cannot be considered as well defined.

We turn now to the consideration of Fe(110) thin films. The ΓH direction in 2D Brillouin zone (2D BZ) is perpendicular to the easy $[1\,\bar{1}\,0]$ axis; $\boldsymbol{q}$'s along this direction can be probed in SPEELS experiments. The ΓN direction is perpendicular to ΓH. The magnetic moment of Fe reads 2.44 and 3.11μ_B for the supported and free ML respectively. The interface layer of W in the supported case has negative polarization of $-0.16\mu_B$. The vertical lattice relaxations discussed at the beginning were taken into account.

In Fig. 2, the calculational results for the ΓH direction in the 2D BZ are presented. We begin with the discussion of supported monolayer Fe/W(110). The estimated SW stiffness assumes the value $D_{\Gamma H} = 278$ meV Å^2. The dispersion curve deviates strongly from simple cosine-type behavior characteristic for Heisenberg models with the exchange interaction between nearest neighbors only. Instead of a monotonous increase with increasing q, the spin wave energy decreases somewhat in the interval of q from 0.4 to 0.6 (unless differently specified wave vectors are given in units $2\pi/a_W$) staying close to 90 meV. For larger q values the SW energy increases again assuming the value of 150 meV at H point. The properties of the freestanding Fe film are essentially different. In this case the dispersion curve is close to simple cosine-form. In the low-q region the SW energies are smaller than for the supported film that gives a lower stiffness $D^{free}_{\Gamma H} = 102$ meV Å^2. The curves intersect at about $q = 0.25$. For larger q the SW energies of freestanding film exceed considerably the corresponding energies of Fe/W(110).

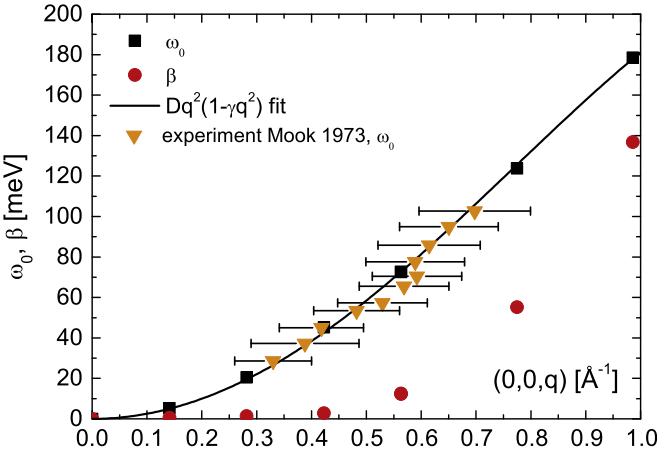

Fig. 1. SW of bulk bcc Fe. Squares represent the SW peak positions ($\omega_0(\boldsymbol{q})$), black line represents the biquadratic fit to the data with parameters $D = 252$ meV Å^2 and $\gamma = 0.28$ Å^2. Triangles stand for the experimental peak centers [5], experimental error bars are shown as well. The values of D and γ extracted from the experiment are 260 meV Å^2 and 0.47 Å^2. Circles present the imaginary part of SW poles ($\beta(\boldsymbol{q})$). The figure presents only about one-half of the interval between the center and the boundary of the Brillouin zone.

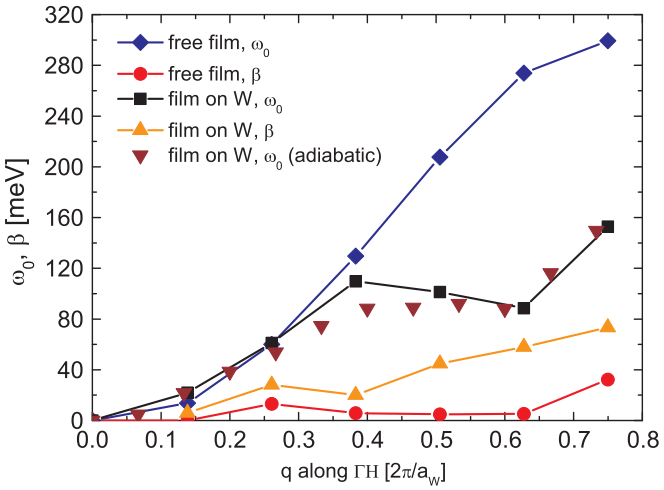

Fig. 2. Energies and inverse life-times of magnons of iron monolayer. $\omega_0(\boldsymbol{q})$ and $\beta(\boldsymbol{q})$ have the same meaning as in Fig. 1.

Also the life time of magnon states depends strongly on the presence of substrate—the damping of the SW excitations of the supported film is much stronger than in the case of free standing film. The origin of the effect can be traced back to Fe–W substrate hybridization. Similar to the case of (0 0 1) surface [18], the unsupported (1 1 0) film features much narrower bands and larger spin splitting. As a result, the density of low-energy Stoner states contributing to the damping is reduced.

The structural anisotropy of (1 1 0) surface is reflected in the anisotropy of the SW dispersion. The magnons spectrum for ΓN direction has higher energies compared to the ΓH direction. This is valid for both supported and free standing films. The presence of the substrate enhances this effect. The dispersion curve along ΓN is qualitatively similar to the curve for ΓH direction discussed above. The values of the stiffness constants are in this case $D_{\Gamma N} = 589\,\mathrm{meV\,\AA^2}$ and $D_{\Gamma N}^{\mathrm{free}} = 233\,\mathrm{meV\,\AA^2}$. Similar to the ΓH case the damping is enhanced for adsorbed film.

4. Discussion and summary

Presently, there is no experimental data for 1 ML of Fe/W(1 1 0) which can be used for direct comparison with our results. The energies we obtain are smaller than the energies observed for 2 ML [7]. This result can be explained by reduced number of neighboring magnetic Fe atoms in the case of 1 ML compared to the 2 ML film. The SW energies obtained in the model-Hamiltonian approach by Muniz et al. [11] exceed substantially our values. Also the form of the dispersion curve reported in that paper is less complex. We relate these differences to the complexity of the Fe–W hybridizations that play crucial role in the establishing of the properties of MEs. The details of these hybridizations are better captured by the parameter-free DFT calculations.

Although the damping of the SW states increases with increasing the wave vector they remain well defined throughout the whole 2D BZ. This property is in striking contrast to the bulk case. The effect of the low dimensionality on the life time of SW obtained in our calculations is opposite to the effect proposed in literature [19]. The broken translational symmetry in the direction perpendicular to the film surface destroys the conservation of the crystal momentum in this direction and should lead to the increased damping for a given in-plane wave vector. This consideration does not, however, take into account the decrease in the number of the Bloch states with the transition from a bulk system to a ML. This leads to the decrease in the number of Stoner excitations contributing to the damping.

It is also worth noting that the SW dispersion obtained within adiabatic frozen-magnon calculations is very close to the dispersion from the calculation of the dynamic susceptibility. This means that the exchange parameters obtained by the mapping of the one Fe ML on the Heisenberg Hamiltonian capture important features of the exchange processes in the film although they do not allow the estimation of the life time of the magnons.

Finally, we emphasize again the role of the non-magnetic substrate in the establishing of the properties of MEs. The hybridization of the Fe and W states modifies the electronic structure of the Fe layer leading to a more complex pattern of the effective interatomic exchange interactions between spin moments of Fe atoms and enhances damping.

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