Joint European Magnetic Symposia – JEMS 2010 IOP Publishing
Journal of Physics: Conference Series **303** (2011) 012004 doi:10.1088/1742-6596/303/1/012004

Magnon excitations in ultrathin Fe layers: The influence of the Dzyaloshinskii-Moriya interaction

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Abstract. High wave-vector magnon excitations in ferromagnetic Fe monolayer and double-layer grown on W(110) are investigated using spin-polarized electron energy loss spectroscopy. The magnon dispersion relation is obtained up to the Brillouin zone boundary. A direct comparison among different systems shows that the magnons in the Fe monolayer are extremely soft and are even softer than the acoustic surface mode of Fe(110).

By measuring the spectra in both energy loss and gain regions on a double-layer Fe film at room temperature and by reversing the sample magnetization, it is demonstrated that the magnon dispersion is asymmetric with respect to the sign of the wave-vector. The asymmetric dispersion relation is attributed to the degeneracy breaking of the magnons due to the presence of the Dzyaloshinskii–Moriya interaction.

1. Introduction

In a magnetically ordered solid the collective magnetic excitations are called magnons. Since in solids the properties are governed by collective phenomena, the investigation of the magnon spectrum would provide an access to the basic and core properties of a magnetic solid. This fact has made the study of spin dynamics in ferromagnets of fundamental interest [1, 2].

Magnon excitations are well established subjects in bulk magnets and have been intensively investigated both experimentally and theoretically over several decades [1, 2, 3, 4, 5, 6]. The experimental investigations are performed by means of various experimental techniques such as ferromagnetic resonance (FMR), Brillouin light scattering (BLS) and inelastic neutron scattering (INS). Only the later one allows a mapping of the magnon dispersion over the whole Brillouin zone in some cases, however, it cannot be employed to low dimensional systems because of its low sensitivity — due to the weak interaction of neutrons with matter—.

Since these kind of excitations in low dimensional ferromagnets are of great interest, a novel experimental technique was required to probe the magnons across the Brillouin zone. Now it is well-established that spin-polarized electron energy loss spectroscopy (SPEELS) is a powerful technique for investigating magnon excitations in ultrathin ferromagnets and ferromagnetic surfaces. The capability of SPEELS is well demonstrated by measuring the magnon dispersion relation of a few atomic layers of Co and Fe films [7, 8, 9].

In nanoscale magnetism, the investigation of the magnons with a wavelength of about (or less than) a nanometer is of great interest, since new physics will come into play in these systems. One particular example is that in low dimensional magnets the spin fluctuations can, in principle, become so strong that ferromagnetic ordering will not take place at finite temperatures. In two dimensional systems this is known as Mermin–Wagner theorem, which will happen only in an ideal isotropic Heisenberg ferromagnet. In real ferromagnets the anisotropy fields contribute and thus real two dimenstional ferromagnetic objects do show ferromagnetic order at a finite temperature. However, the spin fluctuations remain the key point to explain most observed phenomena in low dimensional systems such as reduced Curie temperature (known as finite-size effect). Here, the fundamental questions are: if the magnons in real two-dimensional ferromagnets do exist, is it possible to see their signature of excitations in SPEELS spectra? How does the magnon dispersion relation look like in such a system?

In this contribution we present the results of magnon excitations in a prototype two dimensional magnetic system: one atomic layer of Fe grown on W(110). The choice of the Fe monolayer on W(110) is due to the fact that the system shows relatively good structural and morphological properties. The Fe monolayer possesses a high thermodynamic stability and no intermixing with the substrate. The magnon dispersion relation is measured along the $\bar{\Gamma} - \bar{H}$ of the Brilluin zone. A direct comparison among the different systems shows that the magnons in the Fe monolayer are extremely soft and are softer than the acoustic surface mode of Fe(110).

Since on the surface of an ultrathin film the inversion symmetry in broken, a large spin-orbit coupling can, in principle, lead to a large antisymmetric interaction —known as Dzyaloshinskii–Moriya (DM) interaction [10, 11]—. If the DM interaction is strong enough, it should change the static [12, 13, 14, 15, 16, 17] as well as dynamic [18] prosperities of the system. The consequence of the DM interaction on the spin dynamics and in particular on the magnon dispersion relation of low dimensional ferromagnets will be presented in this paper. By presenting the results of a two-atomic-layer-thick Fe film on W(110), we will show that, indeed, the DM interaction lifts the degeneracy of the magnons and causes an asymmetry in the magnon dispersion relation.

2. Experimental Details

The epitaxial Fe films with thicknesses of one and two atomic layers were deposited onto a clean W(110) single crystal at room temperature (RT) under ultrahigh vacuum (UHV) condition. Prior to film deposition the W(110) crystal was cleaned by cycles of low temperature flashing in oxygen atmosphere and a subsequent high temperature flash in UHV as it is well described in Ref. [19]. After preparation of the sample, it was annealed at about 500 K in order to improve the smoothness of the surface. Conventional UHV tools like low-energy electron diffraction and Auger electron spectroscopy were used to check the sample quality. A longitudinal magneto-optical Kerr effect measurement showed a well-ordered magnetic state of the films—a more detailed information about our sample preparation and their quality can be found elsewhere [20]—. The Fe films exhibit in-plane magnetization and a relatively large magnetic anisotropy with an effective easy axis along the $[\bar{1}10]$-direction. The SPEELS experiments were performed in the in-plane geometry, in which the polarization axis of the incoming and outgoing beam is parallel to the sample surface and thereby parallel or aniparallel to the magnetization of the film. The scattering plane was parallel to the (100)-plane.

A schematic illustration of the scattering geometry used in our experiments is shown in Fig. 1. The spin-polarized beam from a strained GaAs photo-cathode is focused on the sample surface. The intensity of the backscattered electrons is measured with respect to their energy for two different spin orientations of the incoming beam. The magnons can be excited only when the spin of incoming electrons is parallel to the sample magnetization (only by minority spins). The scattering process is commonly called as an inelastic electron scattering. In fact the scattering itself is elastic, but an apparent virtual energy loss is observed because the ejected electron stems from a lower energy level of the excited solid. This process is mediated by exchange interaction, i. e. Coulomb interaction. The wave-vector of the created magnons, $\mathbf{Q}_{\parallel}$, is provided by the wave-vector transfer of the electrons into the system, $\Delta k_{\parallel}$=k$_i \sin\theta$-k$_f \sin(\theta_0 - \theta)$. Here θ (θ_0)

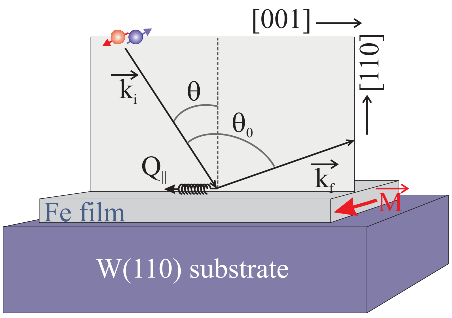

Figure 1. Schematic representation of the scattering geometry used in our SPEELS experiments.

denotes the angle between the incoming beam and the film normal (the outgoing beam), see Fig. 1.

Figure 2 shows examples of the intensity spectra measured at $\Delta k_{\|} = 0.6$ $\mathring{A}^{-1}$ on one-(a) and two- (b) atomic-layers of Fe films grown on W(110). The intensity of the backscattered electrons is plotted for the two possible spins of the incoming beam represented by $I_{\uparrow}$ and $I_{\downarrow}$ for spin up (↑) and down (↓), respectively. The spectra are dominated by the so-called quasi-elastic peak (large intensity at zero energy loss). Since only electrons with minority spin character are allowed to create magnons, the difference spectra ($I_{Diff} = \Delta I = I_{\downarrow} - I_{\uparrow}$) would provide all the necessary information. Therefore in the further data analysis we focus on the difference spectra. The magnon excitation appears as a peak in the energy loss region at a certain energy. The magnon dispersion relation can be measured by changing the wave-vector transfer. This is usually done by changing the angle of the incident beam with respect to the film normal via rotating the sample.

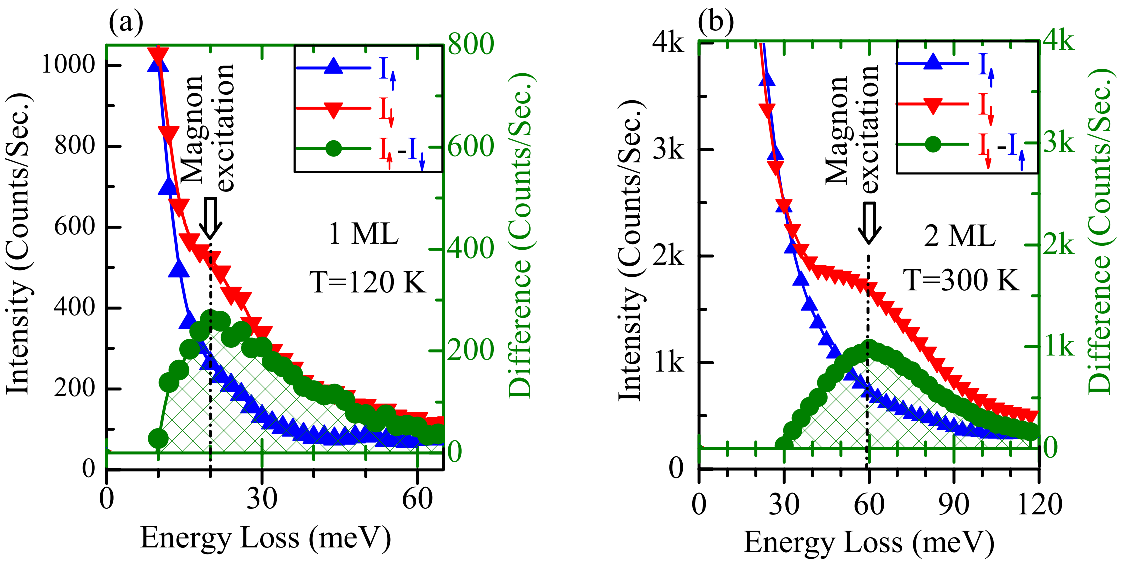

Figure 2. Spin-polarized electron energy loss spectra of one (a) and two (b) atomic layer(s) of Fe films on W(110) measured at a wave-vector transfer of $\Delta k_{\|} = 0.6$ $\mathring{A}$. All three spin up ($I_{\uparrow}$), spin down ($I_{\downarrow}$) and difference ($I_{\downarrow} - I_{\uparrow}$) spectra are shown. The spectra are recorded at 120 K (a) and 300 K (b). The energy of the incident beam was set to 3.8 eV in the case of monolayer and 4.0 eV for measuring the double-layer with an energy resolution of 11.2 meV and 13.9 meV, respectively.

3. Results and Discussion

3.1. Magnons in a monolayer of Fe

A direct comparison between the measured spectra of the Fe monolayer and double-layer, presented in Fig. 2, shows that for the monolayer Fe magnon excitations occur at much lower energies with respect to the ones in the Fe double-layer. In order to shed light onto the physics of the system, the magnon dispersion relation was measured over the whole Brillouin zone (see Fig. 3). Since the Curie temperature of the Fe monolayer is below RT (T_c^{ML}=223 K [21]), the spectra of the monolayer are measured at 120 K. For a better comparison the results of the surface mode of a thicker Fe film (with a thickness of 24 atomic layers) grown on the same substrate is also presented. As in the SPEELS experiments the electrons with very low energies are involved, therefore the technique is very surface sensitive. Hence measuring a thick film leads to a determination of the modes, which are mainly localized at the surface (acoustic surface modes). Figure 3 provides a direct comparison between the results of the monolayer Fe and the surface mode of Fe(110). It reveals that the magnon energies in the Fe monolayer are very small.

Taking a simple Heisenberg model with the Heisenberg spin Hamiltonian, $H = \frac{1}{2}J\sum_{i,j} S_i \cdot S_j - K_{eff}\sum_i (S_i \cdot \hat{n})^2$, one can derive the magnon dispersion relation. Here J is the exchange coupling between two spins, S, and K_{eff} is the effective magnetic anisotropy constant with an easy axis along $\hat{n}$ (the [$\bar{1}$10]-direction in this case). Fitting the experimental results with the calculated magnon dispersion relation within the Heisenberg model, $E = 4JS(1 - \cos\left(\frac{\Delta k_{\parallel} \cdot a_0}{2}\right) + 2K_{eff}S$; $a_0 = 3.165$ $\mathring{A}$, results in a very small exchange interaction (J=11±1 meV) and an effective magnetic anisotropy of $K_{eff} = 2.3 \pm 1.3$ meV [22]. These results are in reasonable agreement with the earlier theoretical and experimental works obtained using static magnetic measurements [18, 23, 24, 25, 26, 27].

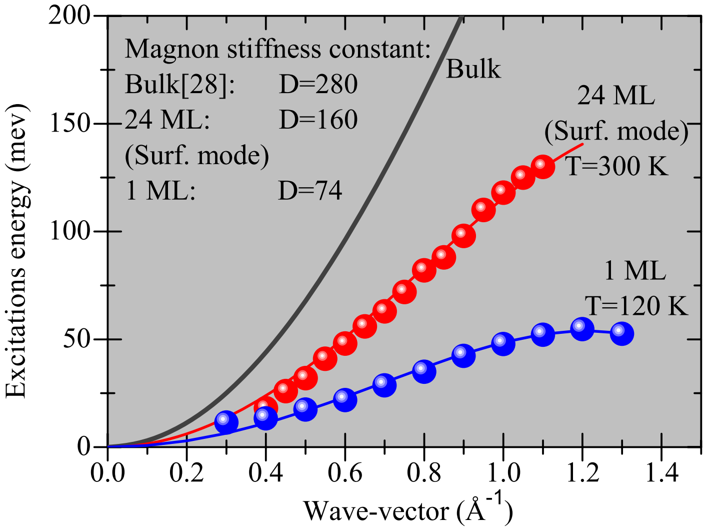

Figure 3. The magnon dispersion relation of one and twenty-four atomic layers of Fe films grown on W(110) measured along the $\bar{\Gamma} - \bar{H}$ direction of the Brillouin zone. The experiments are performed at 120 K and 300 K, respectively. The digitalized data of bulk Fe taken from [28] are also shown for a comparison. The values of magnon stiffness constants are obtained by a parabolic fit into the measured magnon dispersion and are given in meV/$\mathring{A}$.

Magnons in the Fe monolayer are even much softer than the acoustic surface mode of Fe(110). Fitting the experimental results, shown in Fig. 3, with the well-known parabolic dispersion ($E = DQ^2 + \beta Q^4$) leads to a determination of the magnon stiffness constant, D. We found that D for the monolayer system is by a factor of 2.2 smaller than the one obtained for the Fe(110) surface (D_{ML}=74 meV$\mathring{A}^2$ and $D_{surf.}$=160 meV$\mathring{A}^2$). It is only about one-fourth of the value measured for bulk Fe by INS [28]. A number of first principle calculations based on different approaches have predicted much larger energies for magnons in Fe monolayer

[23, 25, 29, 30, 31, 32, 33]. However, the theoretical results depend strongly on the parameters used for the calculations.

The magnon softening in the Fe monolayer may have different origins. The first one might be the temperature effect. Since the experimental results are obtained at 120 K, which is half of the Curie temperature of the system, this may cause the softening of the magnons. The seconded origin might be the strong hybridization with the substrate and change in the electronic structure of the film mediated by the beneath W(110) substrate. The third origin might be the influence of the DM interaction, which we will show that is very important for the Fe films grown on W(110).

Very recently, our experimental data are well reproduced by combining the first principle calculations with so-called atomistic spin dynamics simulations [34]. The authors could confirm that the temperature effect and the chemical relaxations, which influence the hybridization of the Fe film and W(110) lead to this magnon softening. According to their findings, although the DM interaction leads to an asymmetry in the magnon dispersion relation, it does not influence the overall softening too much. In the next section we will show how important is the DM interaction in the Fe/W(110) system and how does it influence the magnons.

3.2. The Dzyaloshinskii-Moriya interaction and magnons

Since the DM interaction is antisymmetric in its nature [10, 11], it would lift the degeneracy of magnons propagating along two opposite directions. In fact in the absence of the DM interaction the spin Hamiltonian is totally symmetric. By adding this interaction into the spin Hamiltonian the extended spin Hamiltonian reads as: $H = \frac{1}{2}J\sum_{i,j} S_i \cdot S_j - K_{eff}\sum_i (S_i \cdot \hat{n})^2 + \sum_{i,j} D_{ij} \cdot S_i \times S_j$. If one calculates the eigenvalues of this Hamiltonian, will find out that the cross product leads to a term, which is antisymmetric with respect to the sign of the wave-vector [23, 18]. In the other words, the DM interaction breaks the symmetry of the resulted dispersion relation, when changing Q to -Q.

In order to study the influence of the DM interaction on the magnons we have investigated the Fe double-layer in more detail [35]. The advantage of the Fe double-layer is that it is ferromagnetic at RT. This allows us to observe and analyze both magnon excitation and annihilation processes in energy loss and gain regions.

Figure 4(a) shows a set of difference SPEEL-spectra measured on a double-layer Fe film on W(110). The experiment is performed at wave-vector transfers of $\pm 0.6\mathring{A}^{-1}$ and in both energy loss and gain regions. Please note that in a SPEELS experiment the annihilation processes occur in the energy gain region. In a real experiment, which is usually done at a finite temperature the thermally excited magnons do exist in the system. The probability for having such magnons is given by the Boltzmann distribution, which depends exponentially on the temperature.

In principle, electrons with majority character can annihilate the thermally excited magnons by gaining energy. This fact leads to a peak in the majority spin channel ($I_\uparrow$) and consequently a dip in the difference spectrum at a certain energy in the energy gain region. As this effect is very small a better representation of the data is plotting the asymmetry curve that is: $I_{Asy.} = \frac{\Delta I}{I_\downarrow + I_\uparrow}$.

Figure 4(b) shows the asymmetry curves of the data presented in Fig. 4(a). The magnon excitation and annihilation processes happening in energy loss and gain regions are clearly visible. Figure 4 reveals two facts: (i) The magnon excitation for the wave-vector transfer of +0.6 $\mathring{A}^{-1}$ takes place at a slightly higher energy with respect to the one of -0.6 $\mathring{A}^{-1}$. (ii) The magnon annihilation for the wave-vector transfer of +0.6 $\mathring{A}$ takes place at a lower energy with respect to the one at -0.6 $\mathring{A}^{-1}$. These facts clearly indicate that the degeneracy of the magnons is broken by the DM interaction because in the absence of the DM interaction the excitation and annihilation energies have to be exactly the same. Although these two facts clearly demonstrate the effect of the DM interaction on degeneracy breaking of the magnons, a measurement with a reversed magnetization would be another prove. Indeed the measurements performed with

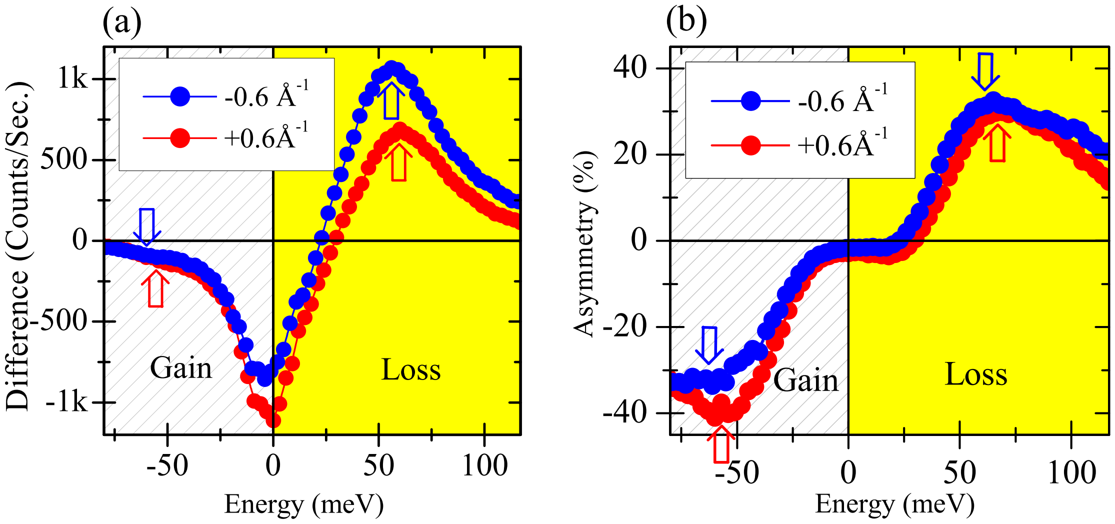

Figure 4. Difference (a) and asymmetry (b) energy loss and gain spectra measured on a two-atomic-layer thick Fe film on W(110). The sample magnetization was along [$\bar{1}$10] direction. The arrows are showing the magnon excitation (energy loss) and annihilation peaks (energy gain).

magnetization pointing along the opposite direction confirmed our findings [35].

The first theoretical prediction of the effect of DM interaction on the magnon dispersion of Fe films on W(110) was done by Udvardi and Szunyogh [18]. They examined the Fe monolayer on W(110) and showed that the magnon dispersion in the presence of DM interaction is asymmetric. More recently, theoretical calculations performed by Costa *et al.*, based on multi band Hubbard model have also shown that in the Fe films grown on W(110) the spin-orbit coupling leads to the DM interaction and influences the magnon dispersion relation [36]. The results are in very good agreement with our experimental findings.

Using a Green's function technique Michael and Trimper could also confirm that the DM interaction leads to an asymmetry in the magnon dispersion [37]. Their method allows to consider the temperature effect on the asymmetry of magnon dispersion. According to their findings the temperature dependence of the asymmetry in the magnon dispersion is more important close to the transition temperature.

4. Conclusions

In conclusion, we have measured the magnon dispersion relation of one and two atomic layers of Fe on W(110) at 120 K and 300 K, respectively. It is shown that the magnons in one atomic layer of Fe are extremely soft in comparison to the Fe bulk and to the acoustic surface magnons of Fe(110). The magnon softening is attributed to the electronic structure of the Fe monolayer on W(110) and to the finite temperature effects.

By measuring an Fe double-layer on W(110) it is demonstrated that the antisymmetric DM interaction is very important in Fe films grown on W(110). The consequence of the DM interaction on the magnon dispersion relation of a low dimensional spin systems is measured. It is shown that the DM interaction lifts the degeneracy of the magnons leading to an asymmetric magnon dispersion relation. Our results are well confirmed by resent theoretical calculations.

5. Acknowledgments

The authors appreciate the fruitful discussions with D. L. Mills, L. M. Sandratskii, P. Buczek, L. Szunyogh, P. Weinberger, A. T. Costa and R. B. Muniz.

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