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Spin waves in ultrathin Co-films measured by spin polarized electron energy loss spectroscopy

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Abstract

We present spin polarized electron energy loss spectra of ultrathin Co films on Cu(0 0 1), in which spin-wave excitations appear as prominent peaks. The spin-wave dispersion can be measured up to and beyond the surface Brillouin zone boundary. For Co-film thicknesses down to 2.5 monolayers no strong thickness dependence of the spin-wave energies are observed.

Keywords: Electron energy loss spectroscopy (EELS); Electron–solid interactions; Magnons; Cobalt; Copper; Magnetic surfaces

The properties of spin waves have been extensively investigated by neutron scattering, Brillouin light scattering and ferromagnetic resonance in the past. However, the weak interaction of neutrons with spin waves prevents neutron scattering experiments on single thin films and on surfaces [1]. The investigation of spin waves in thin films by ferromagnetic resonance and Brillouin light scattering are limited to small wave-vectors of the order of 10^{-2} Å^{-1} [2]. Therefore, the region of high wave-vector spin waves in thin films and at surfaces is largely unexplored. Recently a signature of a spin wave was observed by spin polarized electron energy loss spectroscopy (SPEELS) [3]. With a new high performance SPEEL-spectrometer we were able to measure the spin-wave dispersion up to the surface Brillouin zone boundary in a thin film, consisting of 8 monolayer (ML) Co on Cu(0 0 1) [4]. To explore the influence of reduced thickness on the spin waves we have extended this work to thinner films.

Details of the SPEEL-spectrometer can be found in Ref. [6]. By the illumination of a strained GaAs-photocathode with circularly polarized light a spin-polarized electron beam is emitted [7]. After passing through the monochromator the beam is transversely polarized at the sample position. The scattering geometry is shown in the inset of Fig. 1. The intensities of the reflected electrons are analyzed with respected to their wave-vector transfer $\Delta K_{\parallel}$ *parallel* to the surface and their energy transfer. Switching the helicity of the light which is incident on the photocathode changes the spin polarization of the photoelectrons. In the following $I_{\uparrow(\downarrow)}$ denotes the intensity of the scattered electrons for the incident electrons having majority (minority) spin character with respect to the

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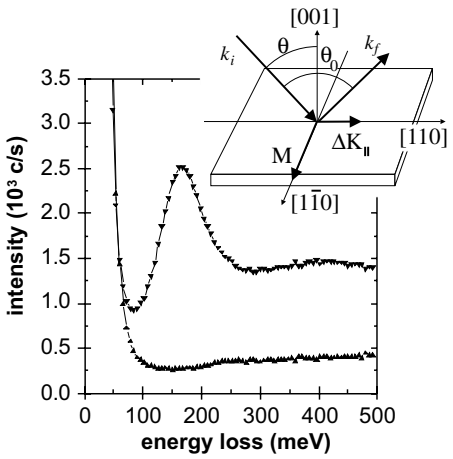

Fig. 1. SPEEL-spectra of a 5 ML Co-film for a wave-vector transfer of $\Delta K_{\|} = 0.81$ Å^{-1}. ▲(▼) mark the $I_{\uparrow(\downarrow)}$-spectrum. The inset shows the scattering geometry. $k_i(k_f)$ is the wave vector of the incident (scattered) electron, θ the incident angle, θ_0 the angle between incident and outgoing beam, and M is the magnetization of the sample.

sample. All measurements shown were performed with a primary energy of about 7 eV of the incoming electron beam. Save for a few data points at the zone boundary and in the next Brillouin zone, the angle θ_0 was kept fixed at 90°. The energy resolution was about 40 meV full width half maximum. The measuring time for one spectrum is about 30 min.

For the sample preparation, Cu was cleaned by Ar-sputtering and subsequently annealed at 820 K for 10 min. Co was deposited by molecular beam epitaxy at 300 K. The film thickness was calibrated by the monolayer oscillations measured with medium energy electron diffraction. In this study films of 8, 5, and 2.5 ML thickness are investigated. After deposition, the films of 8 and 5 ML thickness were annealed to 450 K to smoothen the surface. It is known that films thicker than 4 ML are stable against pinhole formation and strong diffusion of Cu to the surface at 450 K [5]. We found that annealing of the 5 and 8 ML film resulted in an overall increase of intensity in the $I_{\downarrow}$-spectrum compared to the $I_{\uparrow}$-spectrum, but had no influence on the spin-wave peak position in the spectrum.

Fig. 1 displays the $I_{\uparrow}$- and $I_{\downarrow}$-spectra of a 5 ML film with a wave-vector transfer of $\Delta K_{\|} = 0.81$ Å^{-1} as a function of the energy loss (the surface Brillouin zone boundary, $\overline{X}$, is at 1.23 Å^{-1}). The intensities shown in Fig. 1 (as well as in Fig. 2) are corrected for the incomplete spin polarization of

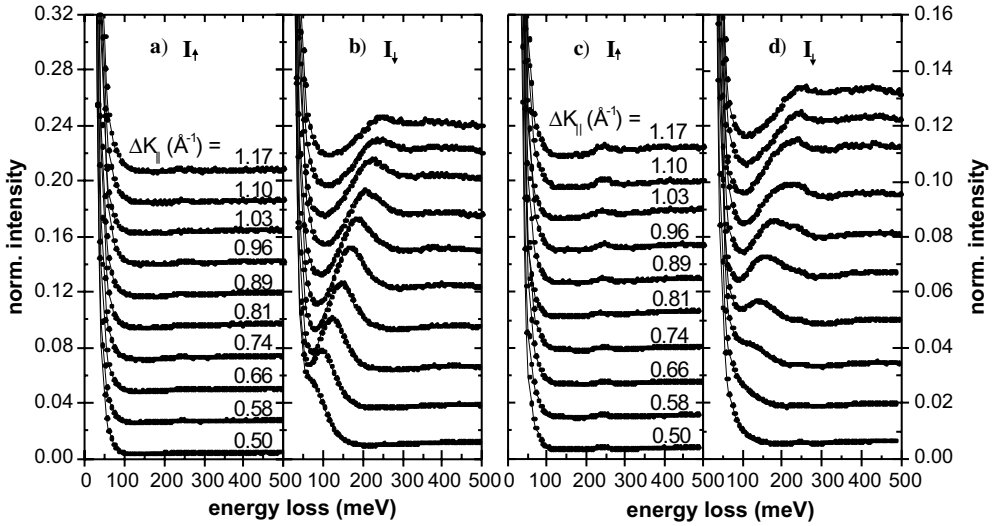

Fig. 2. Series of normalized intensity SPEEL-spectra of a 5 ML (a) $I_{\uparrow}$ and (b) $I_{\downarrow}$ and a 2.5 ML (c) $I_{\uparrow}$ and (d) $I_{\downarrow}$ film. The numbers on the spectra denote the wave-vector transfer. The surface Brillouin zone boundary is at 1.23 Å^{-1}. Adjacent spectra are displaced with a vertical offset of 0.02 for (a) and (b) and 0.01 for (c) and (d).

the incident electron beam ($P = 71 \pm 10\%$ to $79 \pm 10\%$). A pronounced loss feature at about 160 meV is visible in the $I_\downarrow$-spectrum, which is absent in the $I_\uparrow$-spectrum. We attribute the peak in the $I_\downarrow$-spectrum to the excitation of spin waves. The creation of a spin wave reduces the magnetization of the sample and, therefore, changes its angular momentum. The conservation of the total angular momentum requires that the creation of a spin wave is only possible for an incident minority electron and an emerging majority electron.

Fig. 2 shows SPEEL-spectra of a 5 ML (Fig. 2a,b) and a 2.5 ML (Fig. 2c,d) Co-film for different wave-vector transfers. All spectra are normalized with respect to their diffuse elastic peak and displaced along the ordinate with respect to each other. For small $\Delta K_\parallel$ the spin waves appear as a shoulder in the elastic peak of the $I_\downarrow$-spectra and shifts to higher energies with increasing $\Delta K_\parallel$. Note that the absolute spin-wave intensity drops by almost 2 orders of magnitude over the plotted $\Delta K_\parallel$ range. Due to the normalization with the diffuse elastic peak, which decreases by about 1.5 orders of magnitude in the same range, this drop is largely suppressed in Fig. 2. At about 240 meV a small peak is visible in $I_\uparrow$ as well as in $I_\downarrow$, which is a vibrational loss peak of CO adsorbed on the surface. The spectra of the 8 ML film and further details are given in Ref. [4]. For further analysis the difference of the $I_\uparrow$- and $I_\downarrow$-spectra was fitted by a gaussian, representing the spin-wave loss peak, and a second order polynomial for the spin dependent background. The errors due to the fitting in the determination of the peak position are estimated to be below ±10 meV. The energy positions of the spin-wave peaks determined by the fit are plotted in Fig. 3 as a function of wave-vector transfer for all film-thicknesses investigated. Despite some scattering in the data, there is a trend that for wave vectors in the middle of the surface Brillouin zone the spin-wave energies are somewhat lower for lower film thicknesses. Nevertheless at the zone boundary there is no thickness dependence of the spin-wave energies within the experimental uncertainty.

In the following paragraph the spin-wave dispersions will be discussed within the nearest neighbor Heisenberg model. The dispersion relation obtained in this model for the surface mode of a semi-infinite crystal [8] is

$$E(\Delta K_\parallel) = 8JS(1 - \cos(\Delta K_\parallel a_0)). \quad (1)$$

J is the exchange coupling constant, S is the magnitude of the spin per (primitive) unit cell, and $a_0 = 2.55$ Å is the nearest neighbor distance. Using JS as fitparameter the fit of Eq. (1) to the 8 ML data yields a value of $JS = 15$ meV. The fit curve is drawn in Fig. 3 as solid line. We estimate the error in the value of JS due to systematic errors to be less than 1 meV. The value of JS is in perfect agreement with the available neutron data for fcc bulk Co of $JS = 14.7 \pm 1.5$ meV [9]. The dispersion curve derived from the Heisenberg model for a 2 ML film, using the same value $JS = 15$ meV, is shown as dashed line in Fig. 3. [1] In the region between Brillouin zone origin and zone boundary this dispersion curve is below that of the surface

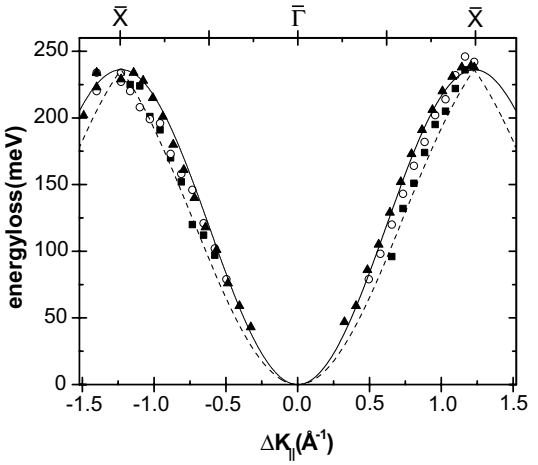

Fig. 3. Spin-wave energy vs. wave vector for the 8 ML (▲), 5 ML (○), and 2.5 ML (■) film. The solid line and the dashed line are the dispersion curves for the surface mode of a semi-infinite crystal and for a 2 ML film derived from a nearest neighbor Heisenberg model, respectively (details in the text). Some points at the surface Brillouin zone boundary and all points in the next Brillouin zone were obtained with $\theta_0 = 80°$.

[1] The Heisenberg model dispersion relation for a 2 ML film along the ⟨1 1 0⟩ direction is $E(\Delta K_\parallel) = 8JS(2 - \cos(\frac{\Delta K_\parallel a_0}{2}) - \cos^2(\frac{\Delta K_\parallel a_0}{2}))$. For thicker films the dispersion approaches quickly that of a semi-infinite crystal. The maximum energy deviation for a 8 ML film is below 1%.

mode of the semi-infinite crystal, but at the zone boundary both curves meet. That is because at the zone boundary the spin-wave amplitude is confined to the top most layers in the nearest neighbor Heisenberg model. Hence there is no difference between a semi-infinite crystal and a thin film. All the experimental data points lie between this two limiting curves within the experimental error. Thus, there is no significant change in JS as a function of the layer thickness. The fact that the spin-wave energy at the zone boundary for films of 2.5 ML is the same as for thicker films indicates that independent of the model the range of the effective exchange coupling cannot exceed the nearest neighbor distance very much.

All presented measurements were performed at 300 K. By reducing the film thickness the Curie temperature T_C of the film reduces. Therefore, the reduced temperature T/T_C is higher for lower thicknesses. For a 2.5 ML film T_C is about 620 K [10]. Extrapolating this values to higher thicknesses yields a change from about $1/4T_C$ (8 ML) to $1/2T_C$ (2.5 ML). To exclude the influence of temperature, we have performed temperature dependent measurements on a 2.5 ML film, from which the largest temperature dependence is expected. Between room temperature ($1/2T_C$) and 110 K ($1/5T_C$), the measurements did not show any significant change in the spectra, in particular the spin-wave peak position did not change. Therefore, temperature dependent effects can be ruled out for the thickness range investigated. Adiabatic ab initio calculations show an enhancement for the nearest neighbor coupling constant by 2.16 for one ML Co on Cu(0 0 1) compared to the bulk (fcc) Co [11]. The enhancement of the surface coupling constant persists for thicker films as has been calculated by Razee et al. [12]. In that paper it is shown that for a 7 ML film the intralayer exchange interaction of the surface layer is enhanced by about a factor of two compared to layers in the middle of the film. The enhanced exchange interaction of the surface is almost independent on the film thickness down to 3 ML. The interlayer exchange interaction between the surface and the sub-surface layer is also enhanced by about 30% and all other coupling constants are less affected by the surface. The magnetic moments at the surface are enhanced with respect to the bulk too, but the relative change is small compared to that of the exchange coupling and, therefore, can be neglected. An increase of the surface intralayer coupling constant by a factor of two increases the energies of the surface spin waves at the zone boundary by about 30% in the nearest neighbor Heisenberg model. When we include this enhanced surface intralayer coupling in our fitting routine, the fit deviates significantly from the data. However, a smaller enhancement of a few 10% would be compatible with the measurements. The discussion above is with the caveat that the Heisenberg model of localized spins is not really suitable for itinerant ferromagnets like Fe, Co, and Ni [13,14]. In particular the broadening of the spin-wave peaks and the suppression of optical spin-wave branches in the experimental spectra can be described only in a non adiabatic theory. The spin-wave dispersion may be affected significantly by the interaction with Stoner excitations, as well.

In conclusion we have shown that by using SPEELS one is able to measure the spin-wave dispersion up to the surface Brillouin zone boundary in ultrathin films. The spin-wave energy at the surface zone boundary is independent of the film thickness down to 2.5 ML, indicating a short range of the interlayer exchange coupling in Co. The data show no evidence of a significant increase of the intralayer exchange coupling at the surface, as was predicted by theory.

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