

Spin-wave excitation observed by spin-polarized electron energy loss spectroscopy: a new method for the investigation of surface- and thin-film spin waves on the atomic scale

R. Vollmer^{a,*}, M. Etzkorn^a, P.S. Anil Kumar^a, H. Ibach^b, J. Kirschner^a

^aMax-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

^bInstitut für Schichten und Grenzflächen ISG3, Forschungszentrum Jülich, D-52425 Jülich, Germany

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Abstract

Spin-polarized electron energy loss spectroscopy (SPEELS) is a unique tool to determine the spin-wave dispersion of ultrathin films and surfaces over the whole Brillouin zone. Ultrathin Co films on Cu(001) show well-defined spin-wave loss peaks in the SPEEL spectra. For 3 ML Fe films on 1 ML Co/Cu(001) the spin-wave peaks are much broader compared to the Co case but show dispersion. In 5-ML-thick bcc Fe films on W(110), a very broad feature assigned to spin-wave excitation was observed as well, which, however, does not show significant dispersion in the investigated range of large wavevectors.

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1. Introduction

The dynamic properties of thin films are currently topics of growing interest. In a number of theoretical papers, ab initio calculations of the spin-wave dispersion of the three-dimensional (3D) ferromagnets within the adiabatic approach have been performed for bulk [1,2] as well as for thin films/monolayer geometry [3–5]. As pointed out by Mills et al., the calculated spin-wave energies using the adiabatic approach are valid only in the limit of small wavevectors [6,7]. Calculations within the adiabatic approach are not able to describe the damping of spin waves, which, however, for large wavevectors is expected to be so large that most of the optical spin-wave branches in thin films do not stand out as well-defined peak in the spin-wave spectrum [8]. A full dynamic ab initio theory of spin waves is computationally so demanding that up to now, it has been performed only for bulk Fe and Ni [9]. To test the validity of the different approximate theories, experimental results on

these large wavevector spin waves are therefore highly desired. Such spin waves can be investigated by inelastic neutron scattering for bulk materials and multilayers [10]. However, the investigation of surfaces and single ultrathin films with this method is currently out of reach. Spin waves in thin films and at surfaces were investigated by ferromagnetic resonance or by Brillouin light scattering, but these methods are limited to small wavevector spin waves below about 0.01 \AA^{-1} .

The single-particle Stoner excitation spectrum has been investigated during the last two decades by spin-polarized electron energy loss spectroscopy (SPEELS) [11–14]. Theory predicts spin-wave excitation in the SPEEL spectra as well [15–17]. However, due to insufficient energy resolution in these early experiments, no evidence of spin-wave excitation was found. A signature of spin-wave excitation was reported for the first time in Ref. [18]. Recently, we demonstrated that large wavevector spin waves show up as well-defined peaks in the SPEEL spectrum of thin films up to the surface Brillouin zone (SBZ) boundary [19].

In this paper, we compare the SPEEL spectra in the spin-wave region of Co films on Cu with those of Fe films on Cu(100) and W(110). A very different behavior was found for the Fe films compared to that of the Co films [19–21].

* Corresponding author. Tel.: +49-345-5582750; fax: +49-345-5511223.

E-mail address: vollme@mpi-halle.mpg.de (R. Vollmer).

2. Experiment

Details of the experimental setup can be found in Refs. [19,22]. A high-performance electron energy loss spectrometer (EELS) with geometry optimized for SPEELS has been used for the measurements presented in this paper [22]. A strained GaAs cathode generates a longitudinally spin-polarized e-beam [23,24]. For the measurements presented in this paper, the spin polarization P was between 0.53 to 0.79 ($\pm 10\%$) depending on the individual cathode. The photoelectrons pass an electrostatic monochromator with a total deflection angle of 90° . Therefore, the e-beam is transversely spin-polarized when impinging onto the sample. The scattered electrons are then detected angle and energy resolved in the sagittal plane by an electrostatic analyzer. A sketch of the spectrometer and the scattering geometry can be found Ref. [22]. The incident angle θ can be changed by rotating the crystal around an axis perpendicular to the scattering plane. The angle between incident and outgoing beam θ_0 was 90° . The wavevector transfer parallel to the surface is given by $\Delta K = k_f \sin(\theta - \theta_0) - k_i \sin\theta$, with k_i and k_f the magnitude of the wavevectors of the incident and scattered electron. SPEEL intensity spectra for the polarization of the incident beam antiparallel (I_\uparrow , majority spin) and parallel (I_\downarrow , minority spin) to the magnetization were recorded. For the spectra shown in this paper, the measured intensities are corrected for the incomplete polarization P .

Three different samples were investigated:

- (i) Co films were deposited by molecular beam epitaxy (MBE) onto Cu(001) at 300 K, forming a pseudomorphic fcc-like structure [25]. After evaporation, the Co films have been annealed to 450 K for 5 min to produce a smoother surface. With the exception of an overall increase in the difference of the I_\downarrow and the I_\uparrow spectrum, annealing did not change the results.
- (ii) 3 ML Fe on 1 ML Co/Cu(001): Low-energy electron diffraction (LEED) structure analysis found for ultrathin Fe layers on Cu(001) up to 3 ML thickness low-energy-electron diffraction (LEED) found a tetragonal structure with expanded interlayer spacings [compared to the Cu(001) interlayer spacings] which has been described as a strongly perturbed fcc structure [26]. A recent investigation by scanning tunneling microscopy found that the Fe film actually breaks up into ‘bcc’- and ‘fcc’-like stripes [27]. Ultrathin Fe films grown on Cu(001) have perpendicular anisotropy [28,29]. For our scattering geometry, an in-plane easy axis is required. Therefore, 3-ML-thick Fe films were grown on a Cu(001) substrate at 300 K, precovered with 1 ML Co. This switches the easy axis of magnetization into the surface plane along a $\langle 110 \rangle$ -direction but leaves all other properties of the film nearly unchanged [30,31]. These thin Fe films were not annealed above room temperature to avoid intermixing and pin hole formation [32].

- (iii) 5 ML Fe were grown on W(110) at about 300 K. Despite the large misfit in the lattice constants of Fe and W of about 10%, the monolayer of Fe grows pseudomorphic on W(110). When growing thicker Fe films at room temperature, partial strain relaxation sets in and leads to the formation of a regular dislocation network as observed by LEED [33]. The easy axis of the magnetization for the 5-ML Fe film on W(110) is along the $[\bar{1}10]$ direction.

All films were magnetized along their easy axis of magnetization after growth, and the scattering plane of the SPEELS was chosen perpendicular to the magnetization.

3. Results and discussion

In our recent paper, Ref. [19], we demonstrated that ultrathin Co films on Cu(001) show well-resolved spin-wave peaks in the SPEEL spectra up to the SBZ boundary of the Cu(001) surface. In Fig. 1a and b, SPEEL intensity

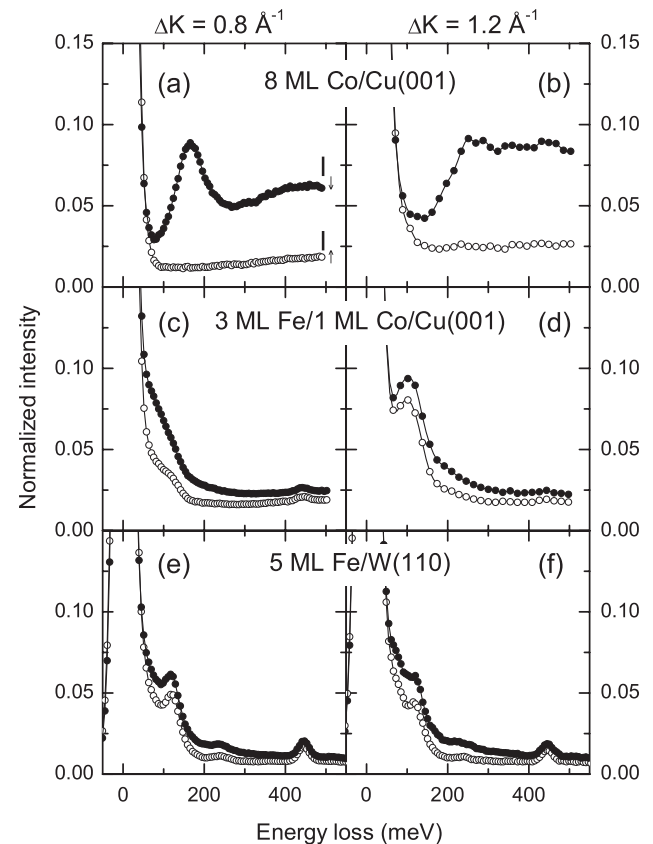


Fig. 1. SPEEL intensity spectra, \circ for I_\uparrow , and \bullet for I_\downarrow normalized to the average elastically scattered intensity peak at 0 meV energy loss: panels a and b, from 8 ML Co on Cu(001); panels c and d, from 3 ML Fe on 1 ML Co/Cu(001); and panels e and f, from 5 ML bcc Fe on W(110) at (a, c, e) $\Delta K \approx 0.8$ and (b, d, f) 1.2 \AA^{-1} , respectively. $E_0 = 7 \text{ eV}$ for all spectra. The sharp peaks at about 100/130, 240, and 450 meV are caused by vibration excitations of small amounts of adsorbed hydrogen, CO, and water.

spectra, I_{\uparrow} (\circ) and I_{\downarrow} (\bullet), of 8 ML Co on Cu(001) along the $[1\bar{1}0]$ direction for two particular wavevectors $\Delta K = 0.8 \text{ \AA}^{-1}$ ($\approx 2/3$ of the SBZ boundary) and $\Delta K = 1.2 \text{ \AA}^{-1}$ (close to the SBZ boundary at 1.23 \AA^{-1}) are shown for an energy E_0 of about 7 eV of the incident electrons. Besides the diffuse elastic peak at 0 meV energy loss, there are only the peaks in the I_{\downarrow} spectrum at about 166 meV in Fig. 1a or 240 meV in Fig. 1b, respectively, while the I_{\uparrow} spectrum is essentially flat. These peaks in the I_{\downarrow} spectrum have been attributed to surface spin-wave excitation as discussed in Ref. [19]. At large energy losses, the intensity in the I_{\downarrow} spectrum is much larger than the intensity in the I_{\uparrow} spectrum. This intensity difference is mainly caused by the creation of Stoner excitations [13,14]. The weight of the Stoner excitations relative to the spin-wave loss peak in the I_{\downarrow} spectrum depends strongly on the kinematical conditions, as can be seen by comparing Fig. 1b with Fig. 1 of Ref. [19], with negative ΔK .

In the spectra of the Fe films in Fig. 1c–f, there is no such obvious sign of a spin-wave loss peak visible in the I_{\downarrow} spectra. The I_{\downarrow} and the I_{\uparrow} spectra are dominated by vibration loss peaks of adsorbed hydrogen at 110 to 130 meV, of carbon monoxide at 240 meV, and of water at 450 meV [34]. In the foot of the diffuse elastic peak, the vibration loss peaks of carbon–Fe at about 60 meV [35] and O–Fe due to dissociated CO are hiding. The overall difference between the I_{\downarrow} and the I_{\uparrow} spectra is much smaller than in the case of the Co films which requires long measurement times, of the order of 100 min to get sufficient good statistics of the intensity difference. Because of the very high reactivity, it is therefore virtually impossible to keep the sample clean. However, we want to point out that the Fe films are only moderately contaminated despite the fact that the adsorption vibration peaks appear quite prominent in the SPEEL spectra. For example, the CO peak in Fig. 1e has only 10% of its height compared to the case when the Fe film is exposed to 4 L of CO [36].

Nevertheless, in the case of the Fe films on 1 ML Co/Cu(001), it is possible to extract the intensity due to spin-wave excitation from the SPEEL spectra. For that, the weighted differences $\Delta I = I_{\downarrow} - \alpha I_{\uparrow}$ spectra of the spectra shown in Fig. 1 is plotted in Fig. 2 in count/s normalized to the sample current in nA. The value of α is chosen such that ΔI is 0 at 0 meV energy loss. In this way, the influence of the spin dependence of the diffuse elastic intensity onto the inelastic difference spectrum is most diminished. The adsorbate vibration losses are only very weakly dependent on the spin orientation of the incident electron for the spectra of the Fe films on 1 ML Co/Cu(001) at these low E_0 of 7 eV. They almost completely cancel out in the weighted intensity difference spectra ΔI . This can be seen in Fig. 2c and d. Even the very strong hydrogen vibration excitation at about 110 meV is not visible in the ΔI spectra. For the case of 5 ML Fe/W(110), however, this is not the case. A small peak-like structure at about 110 meV remains visible in the difference spectra in Fig. 2e and f. This is caused by the

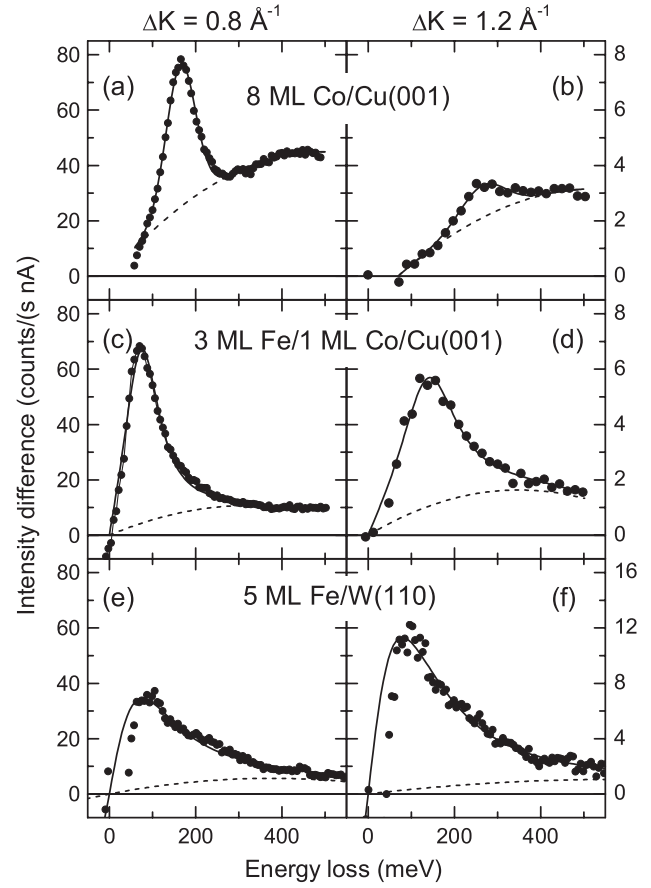


Fig. 2. Difference spectra ΔI of the spectra shown in Fig. 1 in count/s and normalized to the sample current in nA. For the exact definition of ΔI , see text. (a and b) Gaussian curves (—) fitted to the spectra with a background (---) taken into account. (c–f) A Lorentz curve instead of the Gaussian curve has been used for the fit.

dependence of the cross-section of the hydrogen vibration excitation upon the spin orientation of the incident electrons of the e-beam different from that of the elastic scattering.

As discussed in Refs. [19–21], the spin-wave loss peaks in the Co films can be fitted by a Gaussian curve drawn as solid lines in Fig. 2a and b. The dashed line indicates the Stoner excitation background as mentioned above. Only a very poor fit can be achieved with the same type of fit curve for the spin-wave peaks of the Fe films. The solid lines in Fig. 2c–f are Lorentz curves

$$I(\omega) = I_0 \frac{\omega \Gamma}{(\omega^2 - \omega_0^2)^2 + \omega^2 \Gamma^2},$$

which are fit curves with $\hbar\omega$, the energy loss, and I_0 , ω_0 , and Γ as fit parameters. The dashed lines indicate the background taken into account in the fit. First, we discuss the ΔI spectra of 3 ML Fe on 1 ML Co/Cu(001) with the corresponding spectra of 8 ML Co on Cu(001). While at low ΔK , the width of the spin-wave peak in Co (Fig. 2a) and Fe films (Fig. 2c) are comparable, the width increases very much for the Fe films at high ΔK (Fig. 2d), while for the Co

films, the increase in width is only modest (Fig. 2b) [20]. Nevertheless, the peak in ΔI shifts towards larger energies, with increasing ΔK for the 3 ML Fe on 1 ML Co/Cu(001). This is different for the 5 ML Fe film on W(110). The spin-wave feature extends up to about 400 meV energy loss in ΔI for all ΔK . By definition of ΔI , the diffuse elastic intensity does not contribute (much) to the ΔI spectrum. However, large negative peaks appear in ΔI at low energies, which we attribute to the spin dependence of unresolved vibration losses of adsorbates. While for the Co films and the Fe films on Cu(001) their intensity is quite low, they dominate ΔI for the 5 ML Fe film on W(110) at energies below about 80 meV (see Fig. 3b). The increasing slope in Fig. 2e and f is therefore strongly affected by these low-lying adsorbate vibration losses. The Lorentz fit to the data above 80 meV in Fig. 2e and f does not yield reliable values for the fit parameter I_0 , ω_0 , and Γ . ω_0 and Γ are strongly interdependent. Even without fitting, it is obvious that the spin-wave feature of Fe on W(110) is much broader than in 3 ML Fe on 1 ML Co/Cu(001) or even in Co films on Cu(001). The shape and amplitude of this spin-wave feature does not change very much with ΔK throughout the whole SBZ. However, it is not a weak feature. This can be seen in Fig. 3b where the ΔI spectrum is shown on an extended energy loss range up to 4 eV. The maximum of the Stoner excitation (here at about 1.8 eV) is nearly four times lower than the maximum ΔI at the spin-wave ‘peak’.

In Fig. 4, the energy position of the spin-wave peak maxima are plotted vs. ΔK for (a) the 8-ML Co film on Cu(001) and (b) the 3 ML Fe film on 1 ML Co/Cu(001). The measured dispersion curve can be described phenomenologically quite well with a nearest neighbor (NN) Heisenberg model $H = -J \sum_{\langle i,j \rangle} S_i S_j$. J is the nearest neighbor exchange-coupling constant and S is spin per unit cell. The solid lines in Fig. 4a are the spin-wave dispersion branches

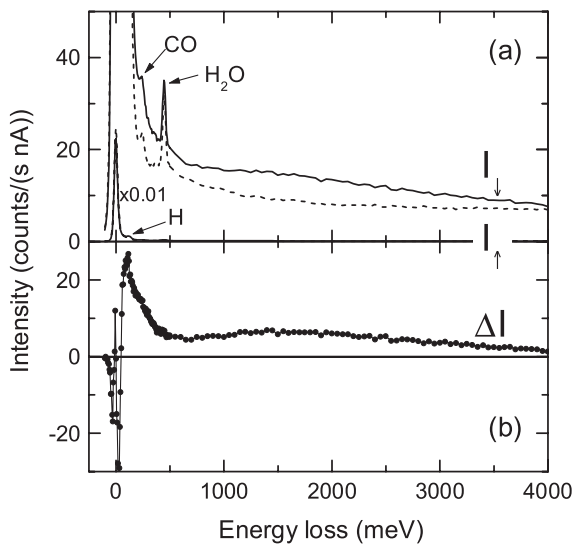


Fig. 3. (a) SPEEL spectra from 5 ML Fe on W(110) up to 4 eV energy loss at $\Delta K = 0.95 \text{ \AA}^{-1}$, $E_0 = 7 \text{ eV}$. (b) $\Delta I = I_{\downarrow} - \alpha I_{\uparrow}$.

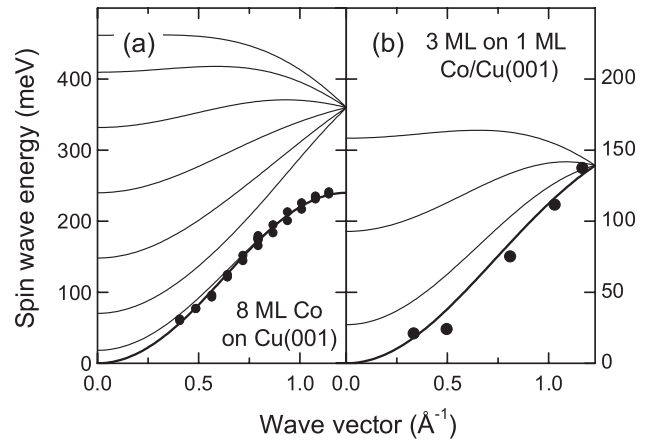


Fig. 4. Spin-wave dispersion curve of (a) 8 ML Co on Cu(001) and (b) 3 ML Fe on 1 ML Co/Cu(001). (●) The energy positions of the spin-wave peak in the SPEEL spectra. The lines are the spin-wave dispersion curves expected from a NN Heisenberg model for an (a) 8-ML-thick film with $J = 15 \text{ meV}$, $J_1 = J$ and for a (b) 4-ML-thick film with $J = 5.8 \text{ meV}$, $J_1 = 2J$.

expected from the model with $J S = 15 \text{ meV}$. The acoustic spin-wave mode is fitted quite well by the model, while there is no experimental evidence for the optical spin-wave modes [19,20].

For the 3 ML Fe film on 1 ML Co/Cu(001), the experimental dispersion curve cannot be fitted by the equivalent Heisenberg model described above. However, if we assume that the exchange-coupling constant at the surface layers J_1 is enhanced by a factor of 2, with respect to the J in the interior of the film, the acoustic spin-wave mode of the (modified) NN Heisenberg model fits quite well the experimental data with a value of $J = 5.8 \text{ meV}$ (thick solid line in Fig. 4b). Such an increased J_1 at the surface is generally expected from theoretical calculations [37]. A monolayer film shows a strongly enhanced J as well [3]. Therefore, the experimental results for the Fe film on 1 ML Co/Cu(001) is in agreement with theoretical expectations, while for the Co film, a strongly increased J_1 is not compatible with the experiment [21].

All the spectra shown above were taken with an energy of about $E_0 = 7 \text{ eV}$ of the incident electrons. The cross-section of the spin-wave excitation by electron scattering depends very strongly on E_0 . In Fig. 5a, the intensity of the spin-wave peak as derived from a Gaussian fit to the measured data is plotted vs. E_0 . For the Fe films, just the intensity difference of I_{\uparrow} and I_{\downarrow} at the peak position is used (Fig. 5b). The resolution ΔE was about 40 meV in all measurements. There is some scattering in the data, presumably because of different spectrometer transmissions for the individual measurements. Nevertheless, it is clearly seen that in both cases the cross-section peaks at low energies of about $E_0 = 7 \text{ eV}$ and drops very rapidly towards larger E_0 . The measurements shown in Fig. 5 were taken at fixed $\Delta K = -0.7$ and 0.8 \AA^{-1} , respectively. The E_0 range of high spin-wave cross-section is quite narrow, that one can think of some kind of resonance effect. However, the

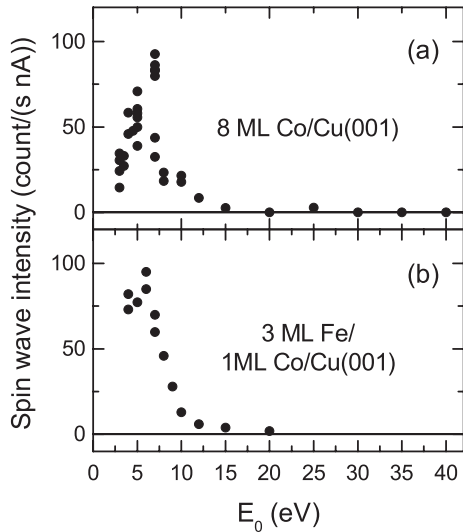


Fig. 5. Intensity of the spin-wave peak in the spectra of (a) 8 ML Co on Cu(001) at $\Delta K = -0.7 \text{ \AA}^{-1}$ and of a (b) 3 ML Fe on 1 ML Co/Cu(001) at $\Delta K = 0.8 \text{ \AA}^{-1}$.

high spin-wave cross-section range does not shift with ΔK or the scattering geometry. Theoretically, an E_0 dependence of the spin wave similar to that of phonon excitation was expected: It should strongly oscillate with E_0 , but on average, should not drop very fast towards large E_0 [16]. On the other hand, in Ref. [16], the cross-section of spin-wave excitations is estimated to be of the order of 1000 times smaller than that for phonons. At wavevector $\approx 2/3$ of the SBZ, we observe at $E_0 = 7$ eV a spin-wave peak in the I_{\perp} spectrum, with $1/10$ of the intensity of the carbon monoxide vibration loss peak at saturation coverage. Therefore, the cross-section at large E_0 is roughly within the theoretical expectations, while it is nearly two orders of magnitude above it for $E_0 = 7$ eV.

Our interpretation of the experimental results in terms of a NN Heisenberg model is entirely phenomenological. There is a good agreement of the measured spin-wave dispersion with the acoustic branch of the NN Heisenberg model but there is no experimental evidence of optical modes predicted by the Heisenberg model. The strong broadening of the spin-wave peaks in the experimental spectra is not described by the model as well. Clearly, a more elaborate theory is necessary to fully understand the physics of the spin-wave spectra. Our spin-wave energies can be compared to (ab initio) calculations of bulk spin-wave energies [1,2]. In the case of Co films, reasonable good agreement of the spin-wave energies was found when scaled to the bulk geometry [19]. A recent calculation of the Co monolayer on Cu(001) gives about 300 meV [4] for the spin-wave energy at the \bar{X} point, which is quite close to the experimental result [21]. Ab initio calculations for thicker Co layers are not yet available. The structure of Fe on Cu(001) deviates strongly from the bcc structure of bulk Fe. Therefore, a direct comparison to the results in Refs. [1,2] is not possible. The calculated energies for a monolayer Fe on

Cu(001) of Refs. [3,4] yield about 230 to 250 meV which is about 70% larger than our experimental result. Again, results for thicker Fe films are not available.

All the above-mentioned calculations employ the adiabatic approximation. However, for large wavevectors, this approximation becomes invalid because the interaction of the spin waves with Stoner excitations is not taken into account [8]. The ab initio calculations of the dynamical susceptibility of bulk Fe, Co, and Ni of Savrasov [9] are not based on the adiabatic approximation but are computationally very demanding. Calculations of the dynamic susceptibility based on a semiempirical tight-binding band structure have been performed for Fe films on W(110) [6,38]. In Fig. 6, the spectral density function $S(Q_{\parallel}, \Omega)$ of the surface layer of Ref. [38] is plotted for a 5-ML-thick Fe film on W(001) along the $\langle 100 \rangle$ direction for two different wavevectors as solid lines. It clearly shows, that for the small wavevector (Fig. 6a, $\Delta K = 0.44 \text{ \AA}^{-1}$), well-defined spin-wave peaks are expected, while at large wavevector (Fig. 6b, $\Delta K = 1.32 \text{ \AA}^{-1}$), only a very broad feature, with a maximum around 270 meV, appears. The dashed line in Fig. 6b represents $S(Q_{\parallel}, \Omega)$ of the center Fe layer with an even broader distribution extending down to nearly zero energy. The solid symbols are the experimental difference spectra ΔI for similar wavevectors. A quantitative agreement with the theory is not expected because $S(Q_{\parallel}, \Omega)$ is just the imaginary part of the transverse susceptibility $\chi_{+,-}(Q_{\parallel}, \Omega, 1, 1)$ and not of χ_{SPEELS} whose calculation is more complex [16,17]. However, there are qualitative differences. None of the experimental spectra of 5 ML Fe on W(110) show a maximum. The drop below

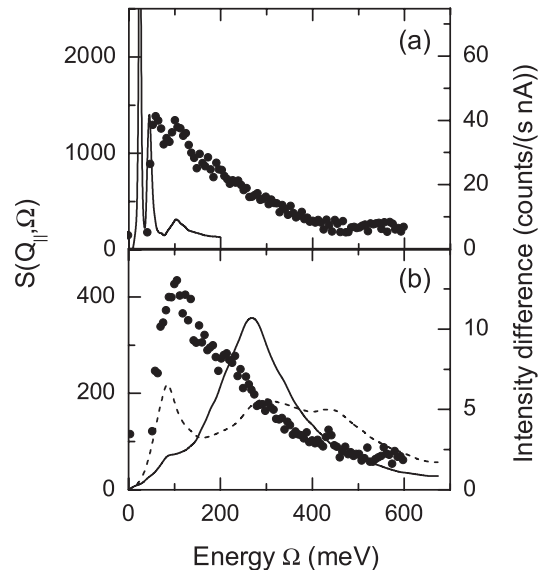


Fig. 6. Spectral density function $S(Q_{\parallel}, \Omega)$ of 5 ML Fe/W(110) along the $\langle 100 \rangle$ direction as calculated in Ref. [38] (left scale) at (a) $\Delta K = 0.44$ and (b) $\Delta K = 1.32 \text{ \AA}^{-1}$. Solid lines for $S(Q_{\parallel}, \Omega)$ of the surface layer and the dashed line for $S(Q_{\parallel}, \Omega)$ of the center Fe layer. (●) The experimental difference spectra ΔI for (a) $\Delta K = 0.5$ and (b) 1.17 \AA^{-1} .

about 80 meV is caused by low-lying vibration losses as discussed above. It might be that the spin-wave signal does not result from the first layer only, and one should then compare not only to the surface $S(Q_{\parallel}, \Omega)$ (solid line) but to $S(Q_{\parallel}, \Omega)$ of the deeper Fe layers as well. The energy range, where the sharp peaks of $S(Q_{\parallel}, \Omega)$ occur at small wave-vectors (Fig. 6a), is presently not accessible to the experiment because the (spin-dependent) low-lying vibration losses dominate the difference spectrum.

4. Conclusion

Fcc Co films on Cu(001) show well-defined spin-wave loss peaks in the I_{\downarrow} spectra up to the SBZ boundary. On Fe films on 1 ML Co/Cu(001), the spin-wave peaks are at much lower energies and are much broader compared to the case of Co films. The measured surface spin-wave dispersion curve can be described in both cases by simple NN Heisenberg models. For Fe films on W(110), no well-defined spin-wave peak can be revealed in the spectrum. Merely a broad shoulder extending up to about 400 meV independent of ΔK is observed in the difference spectrum ΔI .

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