

## Large Wave Vector Spin Waves and Dispersion in Two Monolayer Fe on W(110)

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We present the first surface spin-wave (SW) dispersion measurements up to the surface Brillouin zone boundary of a two monolayer Fe film on W(110) by using spin-polarized electron energy loss spectroscopy. Pronounced features of SW peaks are observed in the spectra at room temperature. We found that the SW energies in the Fe film are strongly reduced compared to spin waves in bulk Fe and to theoretical predictions. Our results suggest that this reduction is caused by the reduction of exchange interaction within the 2 ML Fe on W(110) as compared to bulk Fe.

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The large-wave-vector spin waves (SWs), as magnetic excitations, are entirely determined by the microscopic exchange coupling. Truly microscopic spin dynamics can be revealed in the magnetic systems. These high energy spin excitations are not only essentially important for the spin dynamics in low dimensional magnetic systems, which is very interesting and a forefront of magnetism research today [1–5], but they have also received renewed attention because of the suggestion that high-temperature superconductivity may result from the electronic coupling to SWs [6]. Recently, photoemission data of the Fe(110) surface provided some evidence of a quasiparticle mass enhancement due to electron-SW coupling [6]. Moreover, new applications for even smaller and faster devices in magnetic recording and storage also require full knowledge of the dynamical spin motion in low dimensional systems.

It has been demonstrated that spin-polarized electron energy loss spectroscopy (SPEELS) is a unique method to explore large-wave-vector SWs and their dispersion in thin magnetic films [7]. For ultrathin Fe films on W so far only the signature of SWs has been detected [8]. In the SPEELS measurements on this system only broad humps were visible that did not show an obvious sign of dispersion [7–11]. However, for Fe ultrathin films on W prominent SWs loss features and a distinct dispersion were theoretically predicted [1,2]. These data are essentially important to furnish corroborating evidence for the spin- and electron-dynamics theory [1,2,6]. For measuring large-wave-vector SWs in bulk Fe, inelastic neutron scattering (INS) is a powerful method [12,13]. It is, however, impractical to explore SWs in ultrathin films. Limited by these difficulties, we have very little knowledge on the large-wave-vector SWs in Fe films. In this Letter we present the first full surface SW dispersion measured on 2 ML Fe on W(110). In the spectra the SW excitations appear as pronounced and well-defined as loss features that show a clear dispersion at room temperature. The most interesting observation is that the measured SW energies are much smaller than what has been theoretically predicted [1] for

this system or experimentally measured for bulk Fe. The surface SW energy remarkably coincides with the characteristic energy observed in Fe(110) and provides independent evidence for their suggestion of the electron-SW coupling [6].

The experiments were performed in an UHV chamber with a base pressure below  $5 \times 10^{-11}$  mbar. Two ML Fe(110) films were grown by electron beam evaporation from a Fe rod onto a clean W(110) single crystal at room temperature (RT). The Fe-film thickness was calibrated by monolayer period oscillations in medium energy electron diffraction. The uncertainty was below 10% of the film thickness, as cross-checked by the absolute signal of the Kerr ellipticity measured *in situ* on each Fe film. Morphological and magnetic properties of the pseudomorphic 2 ML Fe films on W(110) were consistent with the literature [14–16]. After the preparation, the Fe film was magnetized along the  $[1\bar{1}0]$  direction in the surface plane. The measurements were taken in the remanence state. The scattering plane was chosen to lie along the [001] direction, perpendicular to the magnetic easy axis. Hence, we probed the properties of the SWs along the  $\bar{\Gamma}$ - $\bar{H}$  direction.

We used our recently developed high-performance SPEELS [17]. The degree of spin polarization  $\mathbb{P}$  of the electron beam was  $0.65 \pm 0.08$ . Electron energy loss intensity spectra were recorded for the spin of the incident electrons *parallel* ( $I_{\uparrow}$ ) and *antiparallel* ( $I_{\downarrow}$ ) to the spin of majority electrons inside the film [7–10]. The total energy resolution was between  $\Delta E = 20$ –30 meV depending on the scattering conditions. The wave-vector transfer parallel to the sample surface is given by the scattering geometry:  $\Delta K_{\parallel} = k_f \sin(\theta_0 - \theta) - k_i \sin\theta$ . Here,  $k_i$ ,  $k_f$  are the magnitude of the wave vectors of the incident and scattered beam, and  $\theta$  and  $\theta_0$  are the angles between the incident and sample normal and between the incident beam and outgoing beam, respectively [see inset in Fig. 1(a)].

In Fig. 1(a)  $I_{\uparrow}$  and  $I_{\downarrow}$  SPEELS spectra are shown for a parallel momentum transfer of  $0.7 \text{ \AA}^{-1}$  with a kinetic electron energy  $E_{\text{kin}} = 4 \text{ eV}$ . We have corrected the inten-

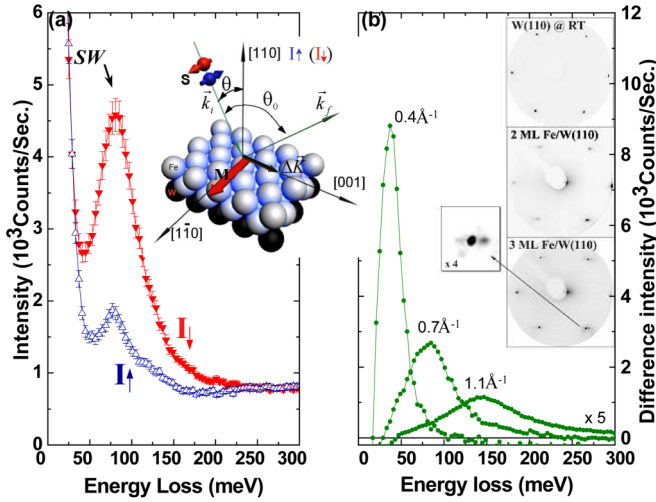


FIG. 1 (color online). (a) SPEELS intensity spectra  $I_1$  ( $\blacktriangledown$ ) and  $I_↑$  ( $\triangle$ ) for  $\Delta K_{||} = 0.70 \text{ \AA}^{-1}$ . (b) Difference spectrum  $\Delta I = I_1 - I_↑$  for 0.4, 0.7, and  $1.1 \text{ \AA}^{-1}$ , respectively. Insets in panel (b) are LEED images for the bare  $W$ , and for a 2 and 3 ML Fe film with  $E \approx 61 \text{ eV}$ .

sities for the incomplete polarization  $\mathbb{P}$  of the source. A pronounced peak at about 80 meV loss energy shows up in the  $I_1$  ( $\blacktriangledown$ ) spectrum while a much smaller peak is observable in the  $I_↑$  ( $\triangle$ ) spectrum at the same position. We attribute the peak in the  $I_1$  spectrum to the creation of SWs by the scattered electrons. The conservation of the total angular momentum during the scattering process forbids the excitation of SWs for incoming electrons having majority spin character ( $I_↑$ ) [7–11,18]. This spin selective excitation process distinguishes a SW loss feature from other nonmagnetic excitations. Since the magnetization of the 2 ML Fe film is not fully saturated in the remanent state at RT, the SW peak is also seen in the  $I_↑$  with small intensity overlapping with the residual oxygen vibrational excitation peak. By calculating the difference  $\Delta I = (I_1 - I_↑)$  of the two spectra with opposite spin orientation of the incident electron, we can strongly reduce the influence of the nonmagnetic excitations to the spectra. In Fig. 1(b), the difference spectrum for  $0.7 \text{ \AA}^{-1}$  is shown with those for  $0.4$  and  $1.1 \text{ \AA}^{-1}$ . A clear shift of the SW towards higher energies for higher wave vectors is evident.

In Fig. 2 a series of normalized intensity and difference spectra for various  $\Delta K_{||}$  are shown. To obtain high SW intensities at small wave vectors and to explore SW close to the surface Brillouin zone (SBZ) boundary, two kinetic energies  $E_{\text{kin}} = 4 \text{ eV}$  ( $\Delta E \approx 20 \text{ meV}$ ) and  $6.75 \text{ eV}$  ( $\Delta E \approx 30 \text{ meV}$ ) are used. In Fig. 2(b) a well-defined peak emerges as a shoulder from the elastic peak for  $\Delta K_{||} \leq 0.5 \text{ \AA}^{-1}$ , which becomes more pronounced in the range from  $0.6 \text{ \AA}^{-1}$  to  $1.1 \text{ \AA}^{-1}$ . At higher  $\Delta K_{||}$  the SW intensities become weaker and the SW peaks broader. The small peaks at around 70 meV and 130 meV observable in both spin channels are vibrational excitation peaks of residual amounts of oxygen and hydrogen [19], respectively. These

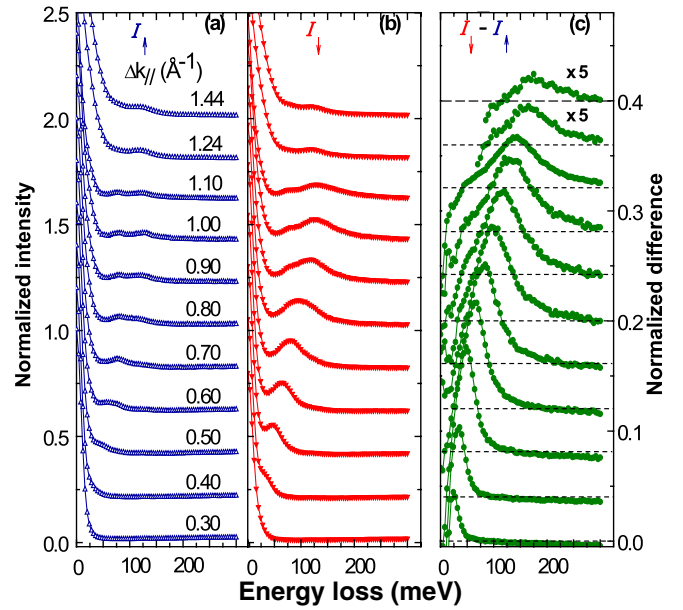


FIG. 2 (color online). Series of normalized SPEELS intensity spectra of  $I_↑$  (a),  $I_1$  (b), and normalized difference spectra (c) for  $\Delta K_{||}$  values plotted in panel (a).  $E_{\text{kin}} = 4 \text{ eV}$  for  $\Delta K_{||} \leq 1.10 \text{ \AA}^{-1}$ ,  $E_{\text{kin}} = 6.75 \text{ eV}$  for higher  $\Delta K_{||}$ . The intensity curves and difference spectra are shifted with respect to each other to allow a better comparison.

vibrational peaks do not show a dispersion and are (almost) spin independent. The normalized difference spectra are plotted in Fig. 2(c) and allow to follow the dispersion up to the SBZ boundary.

Along the  $\bar{\Gamma} \bar{H}$  direction the surface Brillouin zone boundary (SBZ) is located at  $\frac{3}{4}(\frac{2\pi}{a_0})$  ( $1.49 \text{ \AA}^{-1}$  for the 2 ML Fe film). The measured spin-wave dispersion along this direction is plotted in Fig. 3(a). The intensity of the SW peaks in the measured spectra drops very fast by 2 orders of magnitude when the wave-vector transfer increases, as shown in Fig. 3(b) by the solid triangles. The mechanism for electron-SW scattering is the exchange interaction. In Fig. 3(b), we apply the approach of DeWames [20] to calculate the differential scattering cross section arising from the local exchange potential as a phenomenological function to fit the measured data. From the reasonable agreement to the measured data we conclude that the drop in the SW intensity is mainly caused by the reduction of the cross section for higher wave vectors. Quantitative calculations are, however, needed to consider the contribution of Stoner excitations.

There are several INS measurements of SWs in bulk Fe [12,13,21,22], along the [100] direction even up to similar wave vectors as in our SPEELS measurements [13]. In the latter INS measurements on bulk Fe (with 12% Si) SW energies of up to 500 meV were found. This energy is extremely high compare to the maximum energy of about 175 meV found in SPEELS measurements on the 2 ML Fe on  $W(110)$ . In the literature, the SW dispersion is often fitted by the empirical function  $\hbar\omega(k) = Dk^2(1 - \beta k^2)$ .

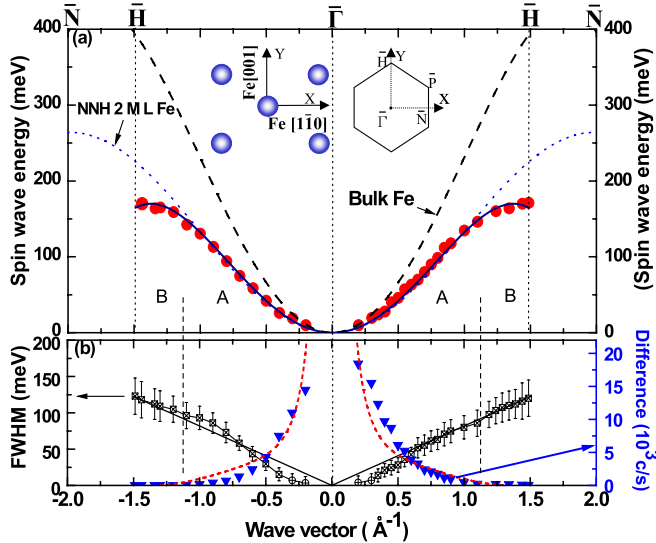


FIG. 3 (color online). (a) SW dispersion curve measured along Fe[001] for the 2 ML Fe film on W(110) by SPEELS. The dotted line is the acoustic dispersion for a 2 ML slab derived from the NN Heisenberg model with  $JS = 11$  meV [23]. The solid thick line is a fit to the data with the polynomial formula  $\hbar\omega = Dq^2(1 - \beta q^2)$ , here  $D = 180 \pm 10$  meV  $\text{\AA}^2$  and  $\beta = 0.256 \pm 0.04$   $\text{\AA}^2$ . The dashed line represents a fit to neutron data on bulk Fe from Refs. [12,13]. (b) The full width at half maximum (FWHM) and the intensity of SWs measured in the spectra are plotted together. The FWHM is corrected for the finite energy resolution of the spectrometer.  $E_{\text{kin}} = 4$  eV for region A and 6.75 eV for region B.

We have used the same function to fit our data [see also solid line in Fig. 3(a)]. The fit results in a value of  $D = 180 \pm 10$  meV  $\text{\AA}^2$  and  $\beta = 0.256 \pm 0.04$   $\text{\AA}^2$ . It is known that within the INS data the results of  $D$  and  $\beta$  vary substantially with the wave-vector range studied [21]. For the high wave-vector INS measurements [12,13] typical values of  $D \approx 280$  meV  $\text{\AA}^2$  and  $\beta \approx 0.136$   $\text{\AA}^2$  can be determined. To directly illustrate the large differences between the results obtained by the different techniques, we have included the fit curve for the neutron data as a dashed line in Fig. 3(a). One may expect differences in the dispersions measured by SPEELS and INS because of the differences of bulk and surfaces SWs. We have therefore used a Heisenberg model to estimate the size of such effects. Unlike in previous measurements on fcc and hcp Co films [7,18], we found it impossible to get an adequate fit to the experimental data with a dispersion derived from a nearest neighbor (NN) Heisenberg model. In Fig. 3(a), we plotted the dispersion curve from this model [23] (dotted line) optimized for the low energy part, which leads to an overestimation of the SW energies at large wave vectors. In bcc crystals the next nearest neighbor (NNN) exchange has a not negligible size; in bulk Fe it is about 60% of the NN exchange as shown in *ab initio* calculations [24]. In view of this result we have included the NNN exchange in our model. In order to avoid a second fitting parameter, we have fixed the ratio of NNN to NN exchange to the pre-

dicted value of 0.6. For this NNN Heisenberg model we find a reasonable agreement of the calculated bulk dispersion with the INS data and of the calculated surface dispersion of 2 ML Fe with the SPEELS data. Again, we found large difference for the fit parameters (the exchange-coupling constant) of bulk Fe and 2 ML Fe on W(110). To summarize this point: with none of the three models we used to describe our data were we able to get agreement to our results assuming bulk Fe values for the exchange-coupling parameters. Since in the Heisenberg model the difference between the surface and bulk dispersion is included, the large difference between SPEELS and INS data can be interpreted as a result of a reduced exchange interaction in the 2 ML Fe on W(110). This could be due to the temperature, since the Curie temperature is only about  $T_C = 450$  K in this system [16]. In addition, the large strain in this system [14] or hybridization effects between the Fe and W [1,2] could play a role. To address these questions experimentally, we have performed SPEELS measurements also on thicker Fe films; some results are shown in Fig. 4. For the thickest film studied (12 ML) one can assume  $T_C$  to be close to the bulk value and the lattice strain to be largely relaxed. As large wave-vector surface SWs are strongly localized at the surface, we can also neglect hybridization effects with the W. As visible in Fig. 4, the SW energies remain at the same range for the three Fe thicknesses. Thus we conclude that the reduction of the exchange interaction at the Fe surface is an inherent property. This is in sharp contrast to our observations of SW's in fcc and hcp Co films, where we found that their dispersion is well described using bulk Co exchange parameters [7,18].

The dispersion of SWs for bcc bulk Fe has been calculated using an itinerant-electron model [25,26]. More recently, *ab initio* calculations of the dynamic spin susceptibility of bulk Fe based on a time-dependent linear response density function have been published [4]. These calculations have, however, not yet been extended to ultra-thin films. For such systems two different approaches exist. First-principles relativistic calculation of SWs are avail-

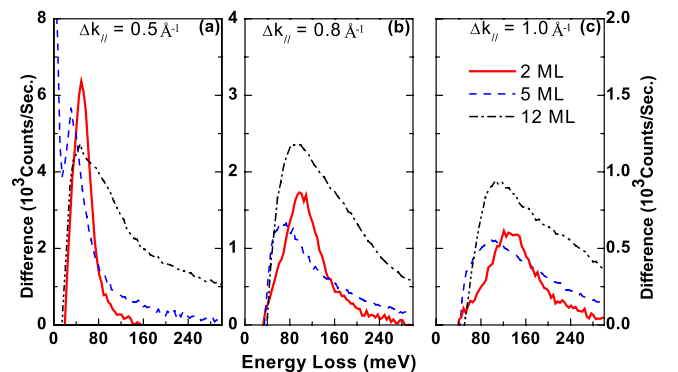


FIG. 4 (color online). (a)–(c) Difference spectra measured on 2, 5, and 12 ML Fe films for different wave-vector transfers. For all spectra  $E_{\text{kin}} = 4$  eV and  $\Delta E \approx 20$  meV are used.



able for a monolayer of Fe [5]. This theory is based on the adiabatic approximation and it was pointed out by Mills *et al.* that such a model description is not expected to be truly valid for an itinerant ferromagnetic like Fe. This is so far only possible within a tight binding model. Such calculations have been performed for ultrathin Fe films on  $W(110)$  [1,2]. In these publications, clear SW features were predicted that are relatively broad in energy due to strong damping via the decay into Stoner excitations that show an increasing SW width with increasing wave vector. This is in qualitative agreement to our observations. For example, the SW peak shown in Fig. 1 has a (corrected) FWHM of  $55 \pm 5$  meV. Its intrinsic width indicates a strong damping. As shown in Fig. 3(b) the measured FWHM also increases with the wave vector. In addition, it was predicted by this theory that the width of the SW peak increases for thicker films [1,2]. As shown in Fig. 4, we find a similar behavior, up to an almost triangle-like difference spectrum for the 12 ML film. The SW energies in the 12 ML Fe films are lower than in the 2 ML Fe but higher than in 5 ML. This nonmonotonic behavior could be related to the strain relaxation in the films [27]. It is interesting to note that a similar nonmonotonic behavior with film thickness has been predicted for the exchange stiffness in this system [1]. The absolute SW energies are, however, largely overestimated in the theoretical predictions [1,2].

We note in passing that we do not see any effect of “disappearance” of the SW intensities observed in INS measurements around  $0.3 \text{ \AA}^{-1}$  along the Fe[001] direction [22,26]. There are no clear multiplex structures which could be interpreted as proof of “optical SW mode” in the spectra of the 2 ML Fe films.

The measured SW energy scale at the SBZ boundary remarkably coincides with the characteristic energy of the renormalization feature that is attributed to electron-SW coupling observed in angle-resolved photoelectron spectroscopy measurements on a Fe(110) surface [6].

In conclusion, the dispersion of SWs in 2 ML Fe on  $W(110)$  is explored up to the SBZ boundary for the first time. Well-defined SW peaks appear in the SPEELS spectra. The large intrinsic SW peak width especially at high wave vectors and the broadening of SW features for the thicker films are in qualitative agreement with the theory [1]. The absolute SW energies predicted in the calculations are, however, much higher than in our results. The SW energies measured by SPEELS are also much lower compared to INS measurements on bulk Fe SWs. With none of our model descriptions were we able to fit the SPEELS data using exchange parameters for bulk Fe. After we have excluded substrate effects, we therefore postulate a reduction of exchange interaction at the Fe surface.

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