Magnons in a Ferromagnetic Monolayer

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We report the first observation of high wave vector magnon excitations in a ferromagnetic monolayer. Using spin-polarized electron energy loss spectroscopy, we observed the magnon dispersion in one atomic layer (ML) of Fe on W(110) at 120 K. The magnon energies are small in comparison to the bulk and surface Fe(110) excitations. We find an exchange parameter and magnetic anisotropy similar to that from static measurements. Our results are in sharp contrast to theoretical calculations, indicating that the present understanding of magnetism of the ML Fe requires considerable revision.

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Quasiparticles play a fundamental role in nature. In magnetism, elemental magnetic collective excitations (magnons) are essential for explaining magnetic ordering [1,2] and electron and spin dynamics [3]. The magnons are of great importance also for modern spintronic devices [4–7]. Of particular interest are high wave vector excitations that are determined by exchange interaction, and occur on the scales of femtoseconds and nanometers [8–10]. However, the magnon excitations in a ferromagnetic monolayer (FML) have been never studied experimentally, even though the spin dynamics of FML belongs to one of the most fundamental problems of magnetism. The experimental techniques enabling magnon investigation either do not have the required monolayer sensitivity, as in inelastic neutron scattering (INS) [11], or probe only a small region of the momentum space close to the Brillouin zone center, as in Brillouin light scattering (BLS) [12,13], and ferromagnetic resonance (FMR) [14,15] experiments. Even inelastic scanning tunneling spectroscopy (ISTS), recently adopted to a magnon investigation in ultrathin films [16,17], cannot probe the surface states selectively due to a lack of in-plane momentum resolution.

The magnetic excitations in two-dimensional spin systems have been studied theoretically for many years [1,18–26]. According to the Mermin-Wagner theorem, a two-dimensional spin system with an isotropic and short-range interaction cannot exhibit any long-range magnetic order at finite temperatures [27]. However, arbitrarily small anisotropies or dipolar interactions are, in turn, sufficient to stabilize long-range magnetic order [18,28], and the FMLs reveal substantial Curie temperatures [29]. Yet, the experimental magnon spectrum in such spin systems is unknown.

In this Letter, we report the first observation of the high wave vector magnon excitations in the ferromagnetic Fe monolayer (ML). Using spin-polarized electron energy loss spectroscopy (SPEELS), we measured the magnon dispersion in pseudomorphic 1 ML Fe epitaxially grown on W(110). We find that the exchange and magnetic anisotropy constants are similar to that obtained from static measurements on vicinal W(110) [30]. We show that the magnons in the Fe ML are much softer than in the bulk Fe, and the surface Fe mode. Surprisingly, the measured magnon energies in 1 ML Fe are much smaller than theoretically predicted [22,25,26]. This discrepancy, related to the strong magnon softening, indicates that the present understanding of the magnetism of the ML Fe is not complete. Our results support the hypothesis that 1 ML Fe/W(110) may not be a simple ferromagnet, as usually assumed.

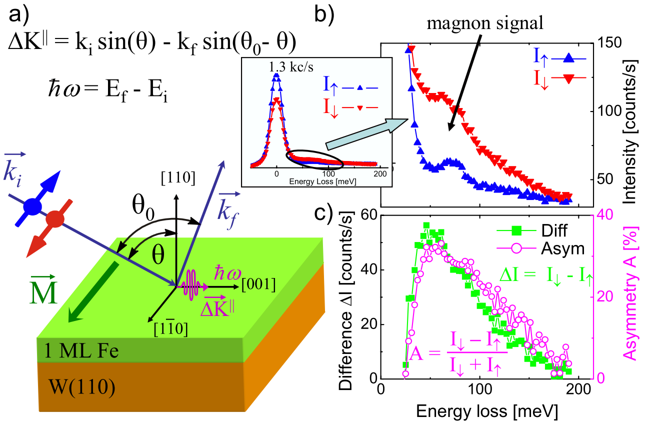

FIG. 1 (color online). (a) The geometry of our SPEELS experiment. A monochromatic spin-polarized electron beam with a polarization parallel or antiparallel to the sample magnetization ($\vec{M}$) is scattered along the [001] direction from the Fe(110) monolayer in the remanent state. The degree of polarization P, in the present case, is 0.7 ± 0.1. The scattering angle is kept at $\theta_0 = 80°$. k_i and k_f are the magnitudes of the wave vectors of the incident and scattered electrons, respectively. The inset shows the intensity $I_\uparrow$ and $I_\downarrow$ SPEELS spectra, as measured for 1 ML Fe/W(110) at 120 K, using electrons with energy $E_i =$ 3.8 eV at $\Delta K^{\parallel} = 1.0$ Å^{-1}. The energy resolution is $\Delta E_i =$ 16 meV. (b) The magnified spectra from the inset in (a). (c) The difference and asymmetry spectra clearly showing the magnon signal.

The 1 ML Fe on W(110) is a unique prototype system that has been intensively studied on both flat [29,31–34] and vicinal substrate surfaces [30,35,36]. Here, we used a flat W single crystal, prepared by cut at 0° ± 0.1° angle, with an average step width of 150 nm. We took advantage of the fact that the iron monolayer is thermodynamically stable even up to very high temperatures [32], which enables preparation of the homogenous Fe ML with good crystalline structure and morphology [29,36]. The experiments were performed in an ultrahigh vacuum (UHV) system with a base pressure of 3×10^{-11} mbar. Special care has been taken concerning the cleaning of the W crystal, which, initially performed under conditions proposed recently [37], has been improved by monitoring the thermal desorption spectra of CO. The iron layers were deposited onto a clean W(110) single crystal at room temperature (RT), and subsequently annealed at about 900 K. Prior to the SPEELS measurements, the sample was cooled down to 120 K. In order to assure that the MLs reproduce properties reported in the literature [29,31], LEED and MOKE measurements were performed after sample preparation. The pseudomorphic ML Fe is ferromagnetic below a Curie temperature of 223 K, and reveals an uniaxial magnetic anisotropy with an in-plane easy axis along the $[1\bar{1}0]$ direction [31]. The SPEELS measurements were performed using a high performance spectrometer described elsewhere [38].

The geometry of the SPEELS experiment is shown in Fig. 1(a). The spin-polarized electrons are scattered from a magnetically ordered sample, and the electron energy loss spectra are measured as a function of the spin polarization of the electron beam, and of the electron momentum transfer. The surface magnons are excited in a spin dependent inelastic electron scattering process [8–10]. The conservation of angular momentum during the scattering forbids the magnon excitation for incoming electrons of the spin polarization antiparallel ($I_\uparrow$) to the sample magnetization. Therefore, the magnon signal can be obtained by calculating the difference between the two spectra measured for the incident electrons with minority and majority spin directions.

The inset in Fig. 1(a) shows the intensity $I_\uparrow$ and $I_\downarrow$ SPEELS spectra obtained for 1 ML Fe/W(110) at 120 K. These spectra are magnified in Fig. 1(b). The difference ($\Delta I = I_\downarrow - I_\uparrow$) and asymmetry [$A = (I_\downarrow - I_\uparrow)/(I_\uparrow + I_\downarrow)$] spectra are shown in Fig. 1(c). The SPEELS spectra are dominated by a diffuse elastic peak at zero energy loss. However, there is a fine feature which arises from the shoulder of the elastic peak in the minority spectrum $I_\downarrow$, shown in Fig. 1(b), which is attributed to the magnon excitation [8–10]. In addition to the magnon feature, small peaks around 70 meV, originating from vibrational states of adsorbed oxygen, are observed in the SPEELS spectra [39,40]. Because of the high surface sensitivity of EELS [39], these vibrational peaks are easily visible, even for the weakly contaminated Fe films [40]. It is interesting that the magnon feature does not appear as a sharp peak in the intensity spectra [9,10]. This observation can be partially explained by a strong damping of the magnons in ML Fe/W(110) leading to a severe broadening of the magnon peaks [20,22,25]. Note that the magnon signal can be clearly distinguished in the difference and asymmetry spectra. For this peak a relatively high asymmetry of 30% is observed (45% after corrections due to the incomplete polarization P of the electron beam). The magnon peaks measured at lower wave vectors are obstructed by the quasielastic peak, whose sign and nature are different from the magnon excitation [8].

Figure 2(a) shows a series of normalized difference spectra taken for different $\Delta K^{\parallel}$ along the [001] direction. With an increase of the wave vector, we observe a decrease of the intensity and broadening of the magnon peaks [9,10]. By plotting the energy positions of the magnon peaks as a function of the wave vector, we obtain the dispersion relation, as shown in Fig. 2(b). Calculations performed in the frame of the itinerant electrons model predict magnon energies much higher (6 times) than those obtained experimentally [22,25]. Such a large discrepancy

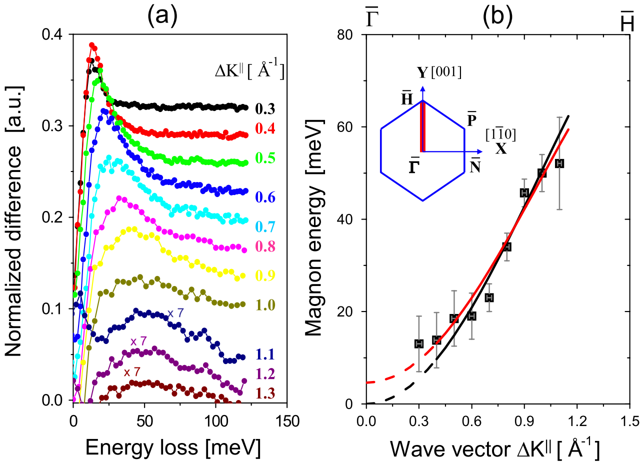

FIG. 2 (color online). (a) Series of normalized difference SPEELS ΔI spectra measured for 1 ML Fe/W(110) at 120 K for different $\Delta K^{\parallel}$ values, as denoted. The spectra are offset by 0.05 with respect to each other. Because of the low energy ($E_i = 3.8$ eV) of the incident electrons, the parallel wave vector transfer is limited to the value of 1.05 Å^{-1} (for $\theta_0 = 80°$). The higher wave vector excitations (above 1.0 Å^{-1}) are measured with a higher incident electron energy ($E_i = 6.25$ eV) and a lower resolution ($\Delta E = 18$ meV). (b) The magnon energy versus parallel wave vector transfer $\Delta K^{\parallel}$, as derived from the peak position in the difference SPEELS spectra observed in (a). Red (or gray) and black curves are fits to the dispersion obtained from the Heisenberg model with and without the anisotropy gap, respectively. The inset shows the 2D surface Brillouin zone of the bcc (110) surface with the states probed along the [001] direction.

cannot be explained only by the experimental uncertainties [25,41].

We now attempt to estimate the exchange parameter for the ML Fe using a solution of the Heisenberg Hamiltonian, where only the nearest neighbor interactions are taken into account. We consider the Heisenberg Hamiltonian: $H = -(1/2)J\Sigma_{\langle i,j\rangle}S_iS_j - K_{\mathrm{eff}}\Sigma_i(S_i \cdot \hat{n})^2$, where J denotes the isotropic exchange coupling constant between spins S_i and S_j. Since our data suggest a gap in the magnon dispersion, as one may expect for a spin system with magnetic anisotropy [19,23–26], we add a term representing the effective magnetic anisotropy K_{eff}, with an easy axis along the unit vector $\hat{n}$ (the $[1\bar{1}0]$ direction). Assuming that we probe states along only the $\bar{\Gamma} - \bar{H}$ ([001]) direction, for the bcc(110) monolayer, one finds: $\hbar\omega(\Delta K^{\parallel}) = 4JS[1 - \cos(\Delta K^{\parallel} a_0/2)] + 2K_{\mathrm{eff}}S$. Here, S is the magnitude of the spin per atom, and $a_0 = 3.165$ Å is the lattice constant of the pseudomorphic 1 ML Fe/W(110). JS and $2K_{\mathrm{eff}}S$ are treated as free parameters. The points measured above 1.1 Å^{-1} showed very large errors and were omitted. Without the anisotropy, we find $JS = 12.5 \pm 1$ meV. The best fit is obtained with $JS = 11 \pm 1$ meV and $2K_{\mathrm{eff}}S = 4.6 \pm 2.5$ meV. The fits are shown in Fig. 2(b). The obtained value is in very good agreement with the JS^2 value estimated from the static analysis of the magnetic domain wall in 1 ML Fe on vicinal W(110), where the $JS^2 (S \equiv 1)$ of 14 meV (at 14 K) is reported [30]. The value obtained here is also close to the J value (8.6 meV) derived from the two-dimensional Ising model [30]. The obtained anisotropy constant $2K_{\mathrm{eff}}S$ is similar to the effective anisotropy (4.2 meV/atom at 14 K) reported in [30]. It is also in good agreement with the calculated values [25,26,42]. Our measurements of the ML dynamics provide exchange and anisotropy constants similar to that obtained from the static measurements.

Figure 3 shows the magnon dispersion for the bulk bcc Fe, and for the Fe/W(110) films of different Fe thicknesses: 1, 2, and 24 ML obtained by SPEELS [10]. Data for bulk Fe are represented by the black curve $D(\text{bulk})\Delta K^{\parallel 2}(1 - \beta \Delta K^{\parallel 2})$, with magnon stiffness coefficient $D(\text{bulk}) \approx 280$ meV Å^2, obtained from the neutron scattering measurements at RT [43]. The data for the double layer (DL) and 24 ML Fe films are measured at RT. The magnon dispersion for the 24 ML Fe/W(110) film is introduced because this sample enables a comparison between the dispersions of the bulk magnons and the surface modes on the Fe(110) surface [34]. The solid lines are guides to the eye obtained from a fit to the SPEELS data using the above formula with $D(\text{DL}) = 180$ meV Å^2 and $D(\text{surf}) = 160$ meV Å^2. For the ML Fe at 120 K, we find $D(\text{ML}) = 74 \pm 5$ meV Å^2. In the following discussion we neglect β coefficients.

For the first time, we can directly compare the magnon dispersion for the Fe systems, where the number of the nearest iron neighbors is systematically reduced, from 8 in bulk to 4 in the ML Fe. The bulk magnons have the highest energies, higher than the surface mode. The magnons in the ML are very soft. They reveal lower energy than the surface mode, and than the magnons in 2 ML Fe/W(110) film [10].

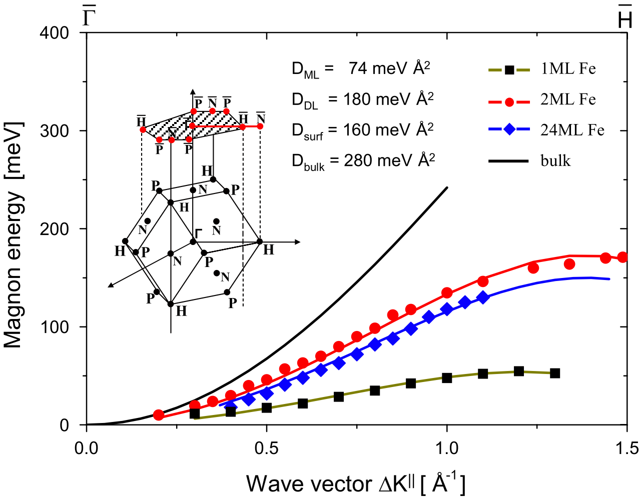

FIG. 3 (color online). Magnon dispersions for the different Fe systems. Black line denotes the parabolic magnon dispersion for the bcc Fe bulk $D\Delta K^{\parallel 2}(1 - \beta\Delta K^{\parallel 2})$, with $D \approx 280$ meV Å^2, compiled using the INS data [43]. The dispersions obtained by the SPEELS at RT for the 2 ML Fe/W(110), and for 24 ML Fe/W(110), are plotted using circles and diamonds, respectively. Squares indicate data for the 1 ML Fe/W(110) measured at 120 K. The solid lines are guides to the eye based on the same formula $[D\Delta K^{\parallel 2}(1 - \beta\Delta K^{\parallel 2})]$, with different D and β. The relation between the bulk Brillouin zone and the two-dimensional Brillouin zone of the (110) surface (shaded area) is shown in the inset.

When comparing dispersions, however, it must be remembered that the bulk and surface Brillouin zones have different sizes (see inset in Fig. 3). In addition, the 1 and 2 ML Fe/W(110) films reveal lattice constants which are about 10% larger than in the bulk Fe [29]. Moreover, we have to take into account temperature effects. For the bulk Fe at 10 K, $D(\text{bulk}) = 307 \pm 15$ meV Å^2 is measured [44]. The data for ML are measured at 120 K, i.e., at about 0.5 $T_c(\text{ML})$ of the ML's Curie temperature (223 K), and the DL Fe data are taken at RT, which corresponds to 2/3 of $T_c(\text{DL})$ (450 K) [10,29]. Assuming that the D values follow the temperature dependence of the ML and DL Fe/W(110) magnetizations, i.e., they increase by about 30% at low temperature [29], we can estimate the D(ML) and D(DL) values in the ground state. We find $D(\text{DL}) = 210$ meV Å^2 and $D(\text{ML}) = 103$ meV Å^2. Hence, the relative relation of the stiffness exchange coefficients D(bulk): D(DL): D(ML) is about 3:2:1, respectively. Such strong softening of the magnons cannot be explained only by the reduction of the nearest neighbor number of the Fe atoms derived from the Heisenberg model. One has to take into account modifications of the electronic structure of the Fe films, which, in the case of 1 ML Fe, are also related to the hybridization effects between Fe and W [22,25,26]. But even then, the predicted energies are still too large.

For the calculations of magnon energies, the ferromagnetic ground state of ML Fe/W(110) is usually anticipated [22,25]. However, recent calculations suggest a spin-spiral structure of the ML Fe/W(110) [45]. Chiral magnetic order induced by the strong Dzyaloshinskii—Moriya (DM) interaction has been observed in the antiferromagnetic ML Mn on W(110) [46]. One may expect that DM interaction should be present in the Fe monolayer as well [26,47], leading to a more exotic ground state of the ML Fe/W(110) with a net ferromagnetic moment [26]. In such a magnetically metastable spin system, the excitations of considerably lower energies are expected. The magnon softening in the ML Fe is in line with a pronounced softening of phonons [34]. A significant increase of the mean atomic displacement accompanied with the drop of the average force constant in the ML Fe/W(110) is reported [34]. For such a system, being close to stability limits, strong mutual phonon-magnon interactions cannot be excluded [48]. Alternatively, the considerable softening of magnons may also be due to the spin-charge coupling effects [49].

In conclusion, we have presented the magnon dispersion for 1 ML Fe on W(110) measured along the [001] direction at 120 K. We observed strong magnon softening in the ML Fe: The magnon energies in the ML are much smaller than those in the bulk Fe, and the Fe(110) surface. Our observations are in sharp contrast to the theoretical predictions. This fact is of fundamental importance for the understanding of the low dimensional magnetism with a large impact on a future theory.

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