Excitation of spin waves at the Fe(100) surface by spin-polarized electron scattering

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Inelastic scattering of low-energy electrons off Fe(100) surfaces shows a strong spin dependent energy loss feature in the range of 100–350 meV due to the excitation of spin waves. The highly asymmetric line shape is attributed to the excitation of a continuum of bulk spin waves due to nonconservation of perpendicular momentum in the scattering process.

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The spin dependence of electron scattering in ferromagnetic materials and at interfaces is of fundamental interest. Spin dependent electron scattering processes are the basis of spin-polarized transport properties in spintronics^{1,2} devices, e.g., in giant-magnetoresistance devices. Spin-polarized electron scattering experiments are the most direct way to probe these processes. Spin dependences in elastic and inelastic scattering events have been studied in some detail in the past using spin-polarized electron scattering. In particular, spinpolarized electron energy loss spectroscopy has revealed the importance of Stoner excitations as a source of spin dependent energy losses.^{3,4} The energy range of Stoner excitations is on the order of the exchange splitting (on the order of 2 eV in Fe or Co). The Stoner excitations lead to a large spin dependence of the energy loss rate, with spin-down electrons having higher loss rates than spin-up electrons. The other type of fundamental magnetic excitations, besides Stoner excitations, are collective spin waves. In bulk materials, spin waves have been studied extensively by inelastic neutron scattering. Well-defined spin waves are found at long wavelength with the spin wave branches merging into the Stoner continuum further out into the Brillouin zone.^{5,6} Spin waves have also been extensively studied using Brillouin light scattering for small k (long wavelength spin waves). However, electron scattering remains the only feasible probe for the study of short wavelength spin waves at surfaces. The smaller energy scale of spin waves ($\approx 100 \text{ meV}$) compared to Stoner excitations makes them important as a possible source of spin dependence in magneto-transport phenomena. The interaction of magnetization with electrical currents has been predicted theoretically for some time^{7,8} and has recently been observed in magnetic nanostructures.⁹

There has never been any evidence reported for spin wave losses using conventional electron energy loss spectroscopy. Mills and co-workers have performed theoretical calculations of spin wave excitations, and they predicted that spin wave signals, although small, should indeed be observable.^{10,11} The first evidence was reported in recent spin-polarized electron energy loss spectroscopy (SPEELS) experiments on Fe layers on W(110)¹² The crucial point is the use of spinpolarized electrons. Spin waves can only be excited by incoming spin-down electrons, thus giving rise to a strong spin asymmetry of the scattering intensity. This situation is similar to the earlier detection of Stoner excitations, where only spin resolved experiments allowed unambiguous detection.^{13,14} Most of the previous SPEELS experiments did not have a sufficient energy resolution to resolve the spin waves. In this paper we report the observation of a well-defined loss structure well below the Stoner spectrum with large spin asymmetries at small energy losses due to the excitation of spin waves at the surface of thick Fe(100) films.

The experiments were performed in an ultrahigh vacuum chamber with a base pressure at 10^{-10} Torr. The electron spectrometer consists of cylindrical sectors as monochromator and analyzer. The monochromator is of double-pass type, and the analyzer is a rotatable single-pass sector. The spin-polarized electron source is a standard GaAs source, based on a highly p-doped GaAs(100) wafer, treated with Cs and O₂ to achieve a negative electron affinity. The preparation of the GaAs photocathode is done in a small separate chamber, which is connected to the main chamber by a gate valve. After preparation, the photocathode can be moved into the spectrometer by a linear transfer mechanism. The light source used is a 810-nm diode laser. The light is circularly polarized by a Pockels cell, which can be switched between left and right circular polarizations by a programmable highvoltage power supply. The spin polarization of the photoemitted electrons is longitudinal. Polarization values were around 25% as measured by a Mott detector in a different chamber on identically prepared sources using material from the same GaAs wafer. Figure 1 schematically shows the scattering geometry used in the experiments. After passing



FIG. 1. SPEELS scattering geometry. The electrons in the incident beam are polarized parallel to the beam direction and are incident onto the Fe scattering sample which is magnetized in-plane.

through the monochromator the electrons are scattered off the sample, which is on an xyz manipulator and can be rotated. The sample magnetization in these Fe films is in-plane. The spin polarization of the electron beam is not completely aligned with the magnetization. The data were corrected to take this into account.

The samples used in this study were thick Fe(100) films grown on GaAs(100) substrates following standard growth procedures.^{15,16} Before growth, fresh GaAs substrates were heat cleaned to 600 °C and then Ne ion sputtered at beam energies of 1 keV and then 0.5 keV for several minutes. The substrates were then once again annealed to 600 °C before film growth. Fe films of several hundred monolayers were deposited on the substrates using molecular beam epitaxy from an e-beam-heated Fe source. Deposition rates were kept to 2 Å/min and monitored by a quartz crystal microbalance. The films were grown at an elevated substrate temperature of 150 °C to reduce island formation. Pressures during film growth were below 5×10^{-10} Torr. Once grown, the films were remanently magnetized by a current pulse through a coil placed close to the sample. The films were magnetized in-plane along Fe $\langle 100 \rangle$, which is the easy axis.

Data were taken at a 20-eV incident electron energy. Incident angles were 65° to the sample normal and scattered electrons were collected 5° off specular. The scattering plane is a (100) plane. The total energy resolution was 75 meV (full width at half maximum) with count rates of 10^4 counts per second in the energy loss region studied (100–350 meV). Data accumulation in the energy loss region required 1–2 h to achieve good statistical noise levels. During the data acquisition the beam polarization was switched at 1 Hz. Fresh Fe films were grown on top of the existing Fe films after a short sputtering cycle to remove surface contamination. All spectra were therefore taken on thick Fe films (more than several hundred Å), thus representing the (100) surface of a bulk bcc Fe crystal.

As usual in SPEELS, data are displayed as intensity (sum of the spin channels) and spin asymmetry A (normalized difference of the spin channels) defined as

$$A = \frac{1}{P_0 \cos \theta} \frac{N_{\uparrow} - N_{\downarrow}}{N_{\uparrow} + N_{\downarrow}},$$

where N_{\uparrow} and N_{\downarrow} are the measured scattering intensities for incident spin-up or down, respectively. The incomplete beam polarization (25%) is taken into account by the first term (P_0) and the angle between magnetization and spin direction by the cos θ term. The asymmetry spectrum shown in Fig. 2 shows a sharp onset at 100 meV with a peak at 165 meV and a tail that extends out to 350 meV. As expected, the measured asymmetries are negative since only incident spin-down electrons can excite spin waves. When the magnetization of the Fe film is reversed the asymmetries also reverse sign, proving the magnetic origin of the spin asymmetries.

The maximum values of the measured asymmetries are on the order of 20% and are comparable to the asymmetry seen in the peak of the Stoner continuum at much higher energy losses. Of particular interest is the highly asymmetric shape of the spin wave peak. One has to be aware that the asym-



FIG. 2. Top panel: measured spin symmetry as function of energy loss. Lower panel: the total intensity (open circles) and spin wave intensity (filled squares) (see the text).

metry can be misleading since it is a normalized quantity. An important question that arises immediately is the origin of the sharp onset. Is this due to the onset of spin wave excitations or just due to the loss features being buried by the elastic peak? To answer this question we look at the product *IA*, which is simply the difference of the spin channels, N_{\uparrow} $-N_{\perp}$. Since we expect spin waves to be excited by only spin-down electrons, we expect that a spin wave would exhibit a large excess in the energy loss intensity for incident spin-down electrons over spin-up electrons. Furthermore, IA will be unchanged by the presence of an additional unpolarized intensity contributed by elastic scattering, as it simply measures a difference. Therefore, it is a reasonable assumption that IA is proportional to the spin wave signal. This quantity is shown in the lower panel of Fig. 2, together with the total (spin averaged) loss spectrum. We see that the spin wave intensity does indeed have an onset. The intensity peak is at a slightly lower energy (150 meV) compared to the asymmetry peak (165 meV), which is due to the elastic peak.

Qualitatively, we attribute the observed broad spin wave

spectrum to excitations of a continuum of bulk spin waves. In surface scattering the momentum transfer is composed of the component parallel to the surface q_{\parallel} and the component perpendicular to the surface q_{\perp} . Using the bulk spin wave dispersion $E = Dq^2 (D = 230 \text{ meV } \text{Å}^2)^5$ the energy transfer would be $E = D(q_{\parallel}^2 + q_{\perp}^2)$. The parallel momentum transfer is given from the scattering geometry as $k(\sin \theta_{in} - \sin \theta_{out})$ where k is the magnitude of the electron wave vector. Thus, for a given geometry (i.e., q_{\parallel}) one would expect an onset energy given by Dq_{\parallel}^2 and a continuum as q_{\perp} runs through the Brillouin zone. In the experiments, q_{\parallel} was varied from 0.1 to 0.4 \AA^{-1} corresponding to scattering angles of 5° to 20° off-specular. However, the expected energies of spin wave onset for this range of q_{\parallel} 's, 2-40 meV, are far too small compared to the observed thresholds of 100 meV. Thus, the origin of the onset energy is not clear at all at present. Also, we note that we were unable to find a clear dependence of the spin wave onset or peak position over 20° of scattering angle variation. These effects might be due to significant diffuse scattering (i.e., nonconservation of q_{\parallel}) due to poor surface structure or due to surface contamination. Reference 15 suggests that Fe island formation on the GaAs(100) surface is common, and unlike Fe/GaAs(110),

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surface roughness does not decrease with film thickness. The tail toward large energies might be attributed to the increased Landau damping due to Stoner excitations which will suppress the spin wave intensities towards higher energies.

We note that the spin wave structure reported here is consistent with the previously reported spin wave "signature" seen on ultrathin (5-ML) Fe films on W(110). In going to ultrathin films one expects a quantization of q_{\perp} to become evident. In this case the continuum of bulk spin waves with q_{\perp} would be replaced by discrete peaks that correspond to standing spin waves perpendicular to the film. The detection of these standing spin waves in few-monolayer films will be a challenging goal of future experiments.¹⁷

In summary, we have shown that spin waves are excited in low-energy electron scattering off the Fe(100) surface. The data can be attributed to the excitation of a continuum of spin waves due to non-conservation of perpendicular momentum. The details, however, are not understood at present.¹⁸

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