Spin Wave Signature in the Spin Polarized Electron Energy Loss Spectrum of Ultrathin Fe Films: Theory and Experiment

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We present theoretical studies based on the use of realistic electronic structures, which conclude that in the spin polarized electron energy loss spectrum of Fe and of ultrathin Fe films a strong signature of spin waves should appear for energy losses in the range of 250 meV and below. New experimental data we present show that indeed the spin asymmetry in the loss spectrum increases dramatically in this regime, as expected from its presence. [S0031-9007(99)08710-4]

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Currently there is great interest in ultrathin ferromagnetic films and, more generally, in the outer surface layers of magnetic materials. Through their study, we may test our ability to predict electronic structures and magnetic properties of new, artificially synthesized materials and to extend our knowledge of bulk magnetism to the very different surface environment. One may also explore magnetism in two dimensions and the transition to three. With ultrathin films incorporated into multilayers, exciting applications in magnetic recording have been realized, and new applications to magnetic storage are envisioned. Such applications require full knowledge of the response characteristics of ultrathin ferromagnetic films. Finally, while the remarks above and the present paper place their primary focus on ultrathin ferromagnetic films, we note that in many other systems of current interest magnetism in reduced dimensionality plays a key role. The high temperature superconductors provide an example. Thus, new experimental methods which explore the magnetism of ultrathin films and surfaces are of fundamental interest. In this paper, we present theoretical calculations and the first data which show that electron energy loss spectroscopy in its spin polarized version (SPEELS) can probe short wavelength spin waves in ultrathin magnetic films. This is thus a new technique which may be employed to probe the nature of spin fluctuations at the surface of diverse magnetic materials, and in thin films of such materials.

Theory in the area of ultrathin film and surface magnetism has focused almost entirely on ground state properties of the ultrathin films [1]. Electronic band structures from such studies do approximate the quasiparticle energies; these can be compared with data taken by techniques such as inverse photoemission [2].

In addition, such films possess a spectrum of spin excitations which control their dynamic response. Also, these enter importantly into the analysis of other phenomena. For instance, the spin dependence of the inelastic mean free path of excited electrons is controlled by their scattering from spin excitations. This enters centrally into the analysis of diverse spin polarized electron spectroscopies and also transport in such materials. Low lying spin waves in ultrathin films have been studied both by ferromagnetic resonance [3] and by Brillouin light scattering [4]. Both methods excite only modes with very long wavelengths compared to a lattice constant. Thus, they probe only properties of the film that are macroscopically describable and provide no information of a truly microscopic nature. For this purpose one needs a means of probing spin waves whose wavelength is on the scale of a lattice constant.

It has been argued that electron energy loss spectroscopy, in its spin polarized version (SPEELS), offers the means of probing such short wavelength spin waves, in principle [5]. The method, without spin analysis, has proved useful in the study of short wavelength surface phonons on diverse surfaces [6]. Theory shows the excitation cross section for spin waves is much smaller than for phonons, though in the range where their detection may be feasible [5]. In this paper, we present the first data which show clear evidence of a spin wave signal, compatible with our recent theoretical predictions [7], and new quantitative calculations which may be compared with the data, along with predictions of the spin wave spectrum of the Fe(110) film used in the experiment. The calculations employ a proper itinerant electron description of ferromagnetic Fe, based on a realistic electronic structure. The details of the model are discussed elsewhere [8].

Earlier SPEELS studies of ultrathin films or magnetic surfaces show broad loss bands, with origin in spin flip scatterings off the sample [9]. These are produced by Stoner excitations (particle-hole excitations, where a spin flip occurs) and are centered about the exchange splitting in the ferromagnetic d bands. The spin wave loss feature will occur in the same spin flip channel, at a much lower energy loss than explored in earlier studies [5,7]. It necessarily appears in the spin flip channel, by virtue of angular momentum conservation. Creation of a spin wave by the beam electron decreases the spin angular momentum of a ferromagnet by $\hbar$. To conserve angular momentum, the beam electron spin must then flip, from down (the minority spin direction) to up. Spin wave emission is thus forbidden for an up spin beam electron. We show the spin flip exchange scattering process schematically in the inset of Fig. 1.

A central question has been the relative intensity of the spin wave feature in the SPEELS spectrum, relative to the previously observed Stoner loss bands. We have addressed this in a recent quantitative theoretical analysis [7] for an electron propagating in bulk Fe. We predict the spin wave loss peak to be quite strong, as confirmed by the data reported here. In Fig. 1, for the electron energy used in the data reported below, and for the relevant momentum transfer, we show the theoretical spin flip portion of the SPEELS spectrum for an electron propagating in bulk Fe. One sees the very broad loss band associated with the Stoner excitations; the peak is at roughly 2.5 eV, with a shape similar to that found experimentally. Near 100 meV, one sees a prominent peak produced by the spin wave loss process. The structure near 700 meV in the calculated spectrum is a low lying feature in the Stoner spectrum.

The experiments cited above [9] employ both a polarized beam and a spin detector to isolate the spin flip contributions to the SPEELS spectrum. These are referred to often as “complete experiments.” As we have seen, only down spin beam electrons may create spin waves, while angular momentum conservation forbids such processes for up spin beam electrons. Thus, to observe the spin wave loss, a spin detector is not required. One needs only a polarized beam, and then, to measure the loss spectrum in the spin wave region for two cases, beam polarization first antiparallel and then parallel to the sample magnetization. The difference between the two loss spectra (more precisely the spin asymmetry, defined below) will contain the spin wave loss feature. If the spin wave loss is the only feature in its energy range, the spin asymmetry should be 100%. In practice, other losses are indeed present, so it will be reduced. If the asymmetry remains large, as the data presented here show, one need not utilize a Mott detector for the scattered electrons. Since Mott detectors are highly inefficient, the spin wave signal will be enhanced substantially by its absence. On real samples, one also has a quasielastic background as well, present by virtue of finite energy resolution in the beam, in combination with imperfections in the sample surface. This influences the data presented below significantly, as we shall see.

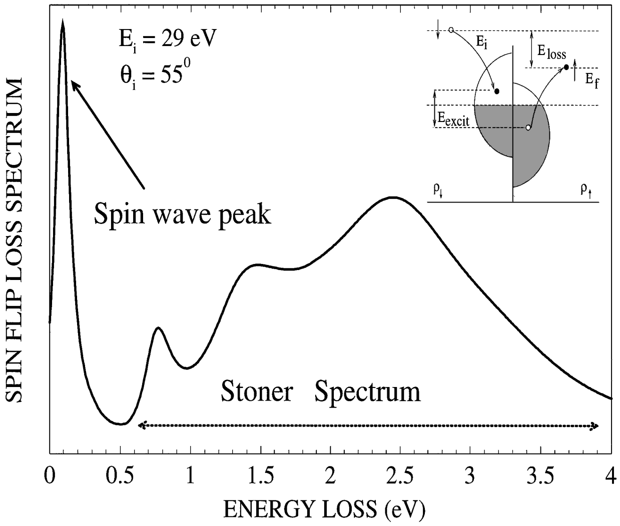

FIG. 1. The calculated spin flip loss spectrum for incident energy $E_i = 29$ eV and scattering angle $\theta_i = 55°$. The inset shows schematically the scattering exchange spin flip process.

We have employed a polarized incident beam to measure the spin asymmetry A in the SPEELS spectrum at low loss energies, from a five layer ferromagnetic Fe film grown on a W(110) substrate. The asymmetry $A = (I_\downarrow - I_\uparrow)/(I_\downarrow + I_\uparrow)$, where $I_{\downarrow(\uparrow)}$ is the loss intensity for the case where the beam polarization is antiparallel (parallel) to the substrate magnetization. The sample has magnetization parallel to the surface and to the $(\bar{1}10)$ direction. The scattering plane is normal to the magnetization and to the sample surface. The direction between the incident and scattered beam is 90°, and we have explored the SPEELS spectrum and its spin asymmetry for angles of incidence with respect to the surface normal of between 50° and 75° for two beam energies of 19 and 29 eV. In all spectra, we see a dramatic increase in spin asymmetry as we move down into the spin wave loss regime. This provides the first experimental evidence for the presence of spin wave losses.

We show such data in Fig. 2(a) for the beam energy 29 eV and angle of incidence of 55°. We show the spin averaged intensity, defined as $(I_\downarrow + I_\uparrow)/2$, and the asymmetry A. Residual magnetic stray fields from the film and the surroundings affect the electron trajectories in the source and the analyzer. These lead to a deterioration of the resolution of ~80 meV by about 30% when averaging. In Fig. 2(a), the spin averaged intensity is shown on a logarithmic scale. We see the strong quasielastic peak centered at zero loss energy. At higher loss energies, we see the gradual onset of scattering from particle-hole excitations. The elongated crosses are the asymmetry A, plotted on a linear scale. At the larger loss energies, we see the low energy end of the Stoner spectrum, which decreases with decreasing loss energy. The dramatic rise below 300 meV is the spin wave signature. This peaks at roughly 200 meV, an energy substantially above that of the spin wave loss feature in Fig. 1. While the data provide clear and unambiguous evidence for the presence of strong spin wave losses, we are in fact unable to resolve the spin wave peak. The drop in A below 200 meV is caused by the presence of the quasielastic scattering, which drives A downward by virtue of its influence in the denominator in its definition. In addition, there is in fact a negative contribution to the numerator in the quasielastic region. This has its origin in quasielastic exchange scattering, whose sign and nature are different from the asymmetry

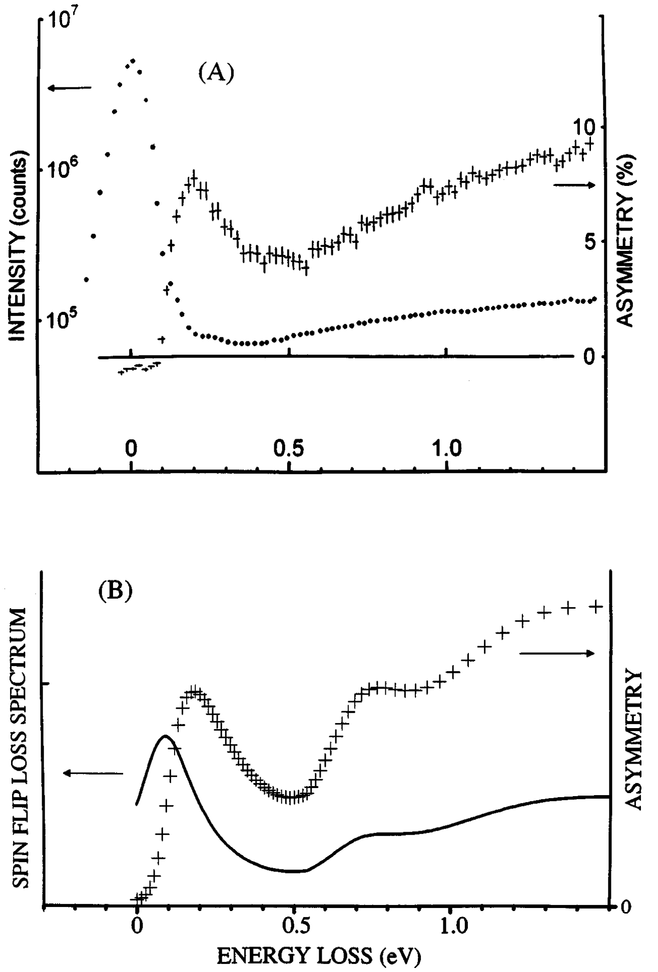

FIG. 2. (a) SPEELS intensity (dots) and scattering asymmetry (crosses) data at $E_i = 29$ eV and $\theta_i = 55°$. (b) Theoretical simulation of the scattering asymmetry A (crosses) and the spin flip SPEELS spectrum (solid line).

in the spin wave region. With improved resolution the quasielastic background will be reduced in the spin wave regime, allowing full resolution of the spin wave structure. The maximum value of the asymmetry at the spin wave peak is roughly 7%. The data for A have not been corrected for the partial polarization of the source, which is in the range of (25–30)%. To correct for this one should multiply A by a factor of 3 to 4, so in fact the true asymmetry in the spin wave regime is very large indeed.

In Fig. 2(b), we show a simulation of A using the spin flip loss spectrum in Fig. 1. The solid curve is the spectrum in Fig. 1, broadened appropriately, and plotted on the energy scale of Fig. 2(a). The spin wave peak lies at 100 meV, as in Fig. 1. The crosses are A, calculated by fitting the measured average intensity empirically, then using the definition of A given above. The absolute scale used for A here is arbitrary. We see the spin wave peak shift up to 200 meV, in very good accord with the data. The feature at 700 meV discussed earlier appears as a shoulder in the theory, and is not evident in the data. We see from the solid curve in Fig. 2(b) this is a broad structure of modest strength in the loss spectrum, which appears more prominent in A because of the reduction in the spin wave regime produced by the quasielastic scattering. Recall that our theoretical SPEELS spectra are for an electron propagating in bulk Fe, not the ultrathin film, so this feature may not survive in a complete theory.

In an ultrathin film, the spin waves will be standing wave modes, rather than the plane waves found in the bulk. A resolved loss spectrum of spin waves will thus exhibit structure if Landau damping is not too severe, as opposed to the single peak present in the calculations presented above. To explore this, we have extended the analysis of spin waves in ultrathin Fe films in Ref. [8] to the case of the (110) film studied here. Our ground state for the (110) film, generated through the scheme used earlier, has a magnetic moment distribution in very good accord with that provided by *ab initio* descriptions of Fe(110) films [10].

In Fig. 3, for several wave vectors in the surface Brillouin zone, we show spectral densities of spin wave excitations, for a five layer (110) film such as employed in the experiments reported here. These are calculated from the wave vector and frequency dependent susceptibility $\chi_{+-}(l, l'; \mathbf{q}_{\|}, \Omega)$ defined in Eq. (2.22) of Ref. [7]. Here $\mathbf{q}_{\|}$ is a wave vector in the surface Brillouin zone. The spectral density function $\rho_{l,l} = (1/\pi)\,\mathrm{Im}\{\chi_{+-}(l, l; \mathbf{q}_{\|}, \Omega)\}$ measures the square of the amplitude of the spin excitations of wave vector $\mathbf{q}_{\|}$ and frequency Ω in layer l. In the enumeration scheme, $l = 1$ is the surface layer. The third panel from the top is for

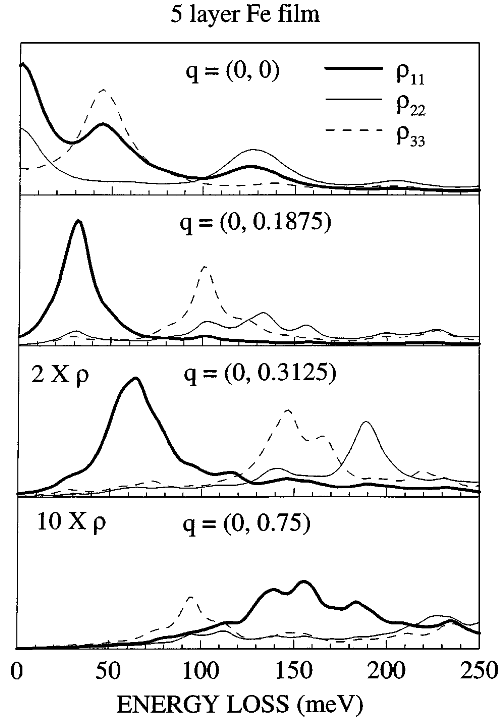

FIG. 3. Diagonal spectral functions $\rho_{l,l}$ of a five layer Fe(110) film for several wave vectors $\mathbf{q}_{\|}$.

a wave vector very close to that used in the experiment. In the spin wave regime, the spectral densities displayed here, when combined with multiple scattering theory [5], provide a description of the SPEELS spectrum in the spin wave loss regime, though a complete theory of the SPEELS spectrum, including the Stoner region, requires a more elaborate theory yet to be developed fully for the ultrathin film [7].

For the case $\mathbf{q}_{\parallel} = 0$, we see the zero frequency Goldstone mode, followed by additional standing spin waves. A five layer film of localized spins has five standing wave modes for each $\mathbf{q}_{\parallel}$. Here, at $\mathbf{q}_{\parallel} = 0$ the Goldstone mode is followed by only three discernible modes; the highest frequency of these (near 220 meV) is heavily Landau damped and shows as only a weak, broad feature. The fifth mode, which should be of higher frequency, is evidently so heavily damped it does not appear in the spectrum. As $\mathbf{q}_{\parallel}$ increases, the Goldstone mode moves up in frequency and hybridizes with the higher frequency standing wave modes (third and fourth panels). Note the lowest lying mode has the character of a surface spin wave, with maximum amplitude in the surface. Experiments carried out with higher resolution, through use of improved spectrometers, should resolve the individual standing wave modes. The data reported here explore the high frequency wing of the spin wave loss regime and, as we have seen, the measured asymmetry even here is very large. We remark that a full SPEELS calculation for the ultrathin film, with proper account of both the Stoner and spin wave loss regimes, requires a nontrivial extension of the analysis in Ref. [7]. We are addressing this issue currently.

The results reported here have important implications. For example, as noted above, the spin dependence of the inelastic electron mean free path is a key element in the interpretation of data taken on ferromagnetic surfaces, by various spin polarized electron spectroscopes. It is assumed widely that this spin dependence has its origin in exchange scattering of electrons from Stoner excitations [11]. A down spin excited electron may drop into an empty minority spin state in the energy bands of the sample, to excite via an exchange process a majority spin electron to the final state. In this picture, the inelastic mean free path of a down spin "hot" electron is shorter than that of an up spin electron in a ferromagnet because there are more minority spin holes. This picture overlooks the role of scattering from spin waves as a source of spin asymmetry in the mean free path; we see here that a fundamental physical principal, angular momentum conservation, makes this a source of spin asymmetry in the mean free path as well. The data reported here show clearly that scattering from spin waves is strong. Discussions of the spin asymmetry of the mean free path of excited electrons in ferromagnets must thus take due account of the role of spin waves, and not just that of the Stoner excitations.

Also, of course, experiments carried out with somewhat higher resolution than available currently can resolve the standing spin wave spectrum of ultrathin films. This will provide us with truly microscopic information on the magnetic response characteristics of these intriguing and important materials. It is our hope that the data and calculations presented here will stimulate new experimental efforts.

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