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# Spin-polarized electron energy loss spectroscopy on Fe(100) thin films grown on Ag(100)

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**Abstract**

We report sharp spin-dependent energy loss features in electron scattering from bcc Fe(100) thin films grown on Ag(100). Majority spin features are observed at ∼1.8 and 2.5 eV energy loss, and a minority spin feature is observed at ∼2.0 eV energy loss. The majority spin peaks are attributed to spin-flip exchange scattering from the Fe films. The minority spin peak is attributed to non-flip exchange scattering with an energy corresponding to the separation between occupied and unoccupied minority spin bands. The observed peak energies compare favourably with electronic structure calculations for Fe and with spin-resolved photoemission and inverse photoemission results.

## 1. Introduction

Spin-polarized electron energy loss spectroscopy (SPEELS) was developed in the 1980s and has become a valuable technique for probing Stoner excitations [1–8] and, more recently, spin waves [9]. In so-called spin-flip exchange scattering, an incident electron of a given spin occupies an empty state of the target material and an electron of opposite spin is excited and detected. This process produces a Stoner excitation and gives SPEELS its sensitivity to the occupied and unoccupied parts of the spin-split electronic structure of materials. Consequently, SPEELS is a complementary technique to spin-resolved photoemission and inverse photoemission, which more directly probe the occupied and unoccupied spin-split band structure, respectively. SPEELS studies have been performed using unpolarized electron sources with scattered electron spin detection [1], with spin-polarized sources without scattered electron spin detection [2, 3], and with both spin-polarized sources and scattered electron spin detection [4–8]. In general, these studies report rather broad featureless data in both energy and angle dependence, and this has been attributed to non-conservation of the perpendicular momentum component in the scattering process [7], nonuniform exchange splitting throughout the Brillouin zone (BZ) [2], and umklapp scattering together with the structure of interband densities of Stoner states in the material [10]. Only in the study of $Cr_2O_3$ [11] and bcc

Co/GaAs(001) [3] are sharp loss features reported. However, a subsequent study of fcc Co/Cu(001) [7] did not observe any sharp energy loss features.

It is useful to describe the SPEELS experiment in terms of the four basic spin-dependent scattering processes. These are divided into flip and non-flip scattering for incident electrons with spins parallel or anti-parallel to the majority spin orientation. For flip scattering, an incident electron occupies an empty state just above the Fermi energy and transfers its energy to an electron of the opposite spin. This electron may now escape the material and be detected. This process results in an electron–hole excitation with opposite spins for the electron and hole; a configuration that is identical to a Stoner excitation. Non-flip processes can occur via both direct and exchange scattering. For dipole scattering, the incident electron scatters from long-range electric field fluctuations associated with elementary excitations of the target, and the scattering process occurs well outside the target. Here it is assured that the incident and scattered electron are the same, and this is a direct scattering event. Shorter-range scattering, where the electron penetrates into the material before being inelastically scattered (impact scattering), produces both direct and exchange non-flip scattering. These are distinguished by the incident electron being the same or different from the scattered electron, respectively. It has been shown that direct non-flip scattering preserves the polarization of the incident beam [12].

For an initially unpolarized electron beam, the polarization of the scattered electrons can be written as $P = \frac{(F^{\uparrow}-F^{\downarrow})+(N^{\uparrow}-N^{\downarrow})}{F^{\uparrow}+F^{\downarrow}+N^{\uparrow}+N^{\downarrow}}$, where $F$ and $N$ refer to flip and non-flip scattering, and the arrows refer to the spin of the scattered electron. Since direct non-flip scattering preserves the incident beam polarization [12], we will only consider non-flip exchange scattering. Then $N^{\uparrow}$ results in an electron in a previously unoccupied majority spin state and should be negligible because of the relatively low density of empty majority spin states for a ferromagnetic Fe(100) thin film [14]. A similar argument suggests that the probability of $F^{\downarrow}$ should be small, because this involves the occupation of a previously unoccupied majority spin state. This leaves $F^{\uparrow}$ and $N^{\downarrow}$ as the only remaining non-negligible scattering processes. Consequently, majority spin features in the energy loss spectrum are associated with spin-flip scattering, and these features occur at energies separating occupied majority spin bands and unoccupied minority spin bands. Minority spin features, however, are associated with non-flip exchange scattering where an incident minority spin electron occupies an empty minority spin state above the Fermi energy, producing a minority spin scattered electron. These features will be observed at energies separating occupied and unoccupied minority spin bands. In this paper, we present the experimental results of spin-polarized electron energy loss spectroscopy on bcc Fe thin films grown on an Ag(100) crystal, where we see sharp energy loss features in both the majority and minority electron spectra. We interpret the spin-dependent energy loss features in terms of spin-flip scattering for the majority spin features and non-flip exchange scattering for the minority spin peak, containing information about the spin-dependent band structure of the films. These results have important implications on the interpretation of SPEELS and on its use in understanding spin-dependent band structure for surfaces and thin films.

## 2. Experimental procedure

The main components of the SPEELS experiment are shown in figure 1. The electron source is a commercial low-energy electron diffraction (LEED)/Auger electron gun. The data were acquired using an incident kinetic energy of 300 eV. After scattering from the sample, the electrons are collected in a hemispherical electron energy analyser (Physical Electronics Model 3057) with multichannel electron detection. For spin-resolved work, the multichannel detector has a central hole that allows passage of the energy-analysed electrons into the electron optics for the mini-Mott detector. In this case, the high voltages on the channel plates are turned

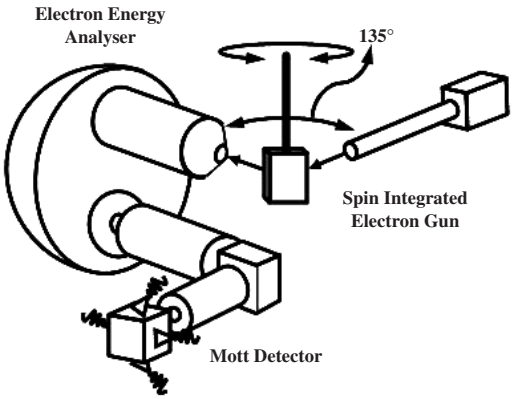


**Figure 1.** Schematic of the experimental SPEELS system.

off and the channel plates and anode assembly become part of the first lens stack, directing the electrons into a 90° spherical sector. The 90° sector is run at relatively high pass energy: energy resolution is determined by the hemispherical analyser pass energy and the electron source. The 90° sector allows the simultaneous resolution of spin-polarization both normal to the sample and in the plane of the sample perpendicular to the plane containing the emission direction and the sample normal. After the 90° sector, the electrons travel through another lens stack, into the mini-Mott detector. In the mini-Mott, the electrons are accelerated to ∼25 kV, with four channeltrons positioned horizontally and vertically used for electron counting. More details of this system may be found in [13]. For the SPEELS measurements, the total scattering angle is fixed at 135°, with the electrons incident at 67° with respect to the sample normal. The overall energy resolution for the SPEELS measurements (electron source and energy analyser) is about 0.6 eV or better at full width at half maximum. The data acquisition of experiments was carried out at $2 \times 10^{-10}$ Torr.

Films of Fe grown on Ag(100) have been characterized in previous studies using many techniques [14–18]. These studies have established the growth of bcc Fe(100) with a perpendicular easy axis of magnetization for films thinner than about six monolayers (6 ML) and an in-plane easy axis (along [010]) for thicker films at room temperature [14, 16]. The precise growth mode remains somewhat controversial, with some studies finding Stranski–Krastanov growth with island growth occurring after 3 ML of layer-by-layer growth [17], and some reporting layer-by-layer growth of up to 6 ML at room temperature [18]. The Ag crystal was cleaned using $Ar^+$ sputtering followed by annealing to 450 °C. Fe films are deposited using electron-beam evaporation. The films have a p(1 × 1) LEED pattern and no observable contamination, as judged from x-ray photoelectron spectroscopy (XPS) measurements. Because of the reactive nature of the Fe films, the SPEELS data is acquired in less than 4 or 5 h, after which the Fe is sputtered off and a fresh film is produced for additional measurements. Here we focus on in-plane magnetism, which can be achieved for bcc Fe films more than for 6 ML thick films. We have concentrated our measurements on 20 ML Fe films, and we use the ratio of Fe 2p and Ag 3d photoemission peaks as a thickness monitor. Due to the long photoelectron mean free paths for these core-level electrons at the photon energies used, it is not possible to distinguish between layer-by-layer growth and cluster growth for the Fe films. However, results from earlier studies and our magnetic measurements establishing an in-plane easy axis for the samples studied clearly suggest that we are in the thicker bcc film regime, where cluster growth of the Fe is likely and in-plane magnetization occurs.

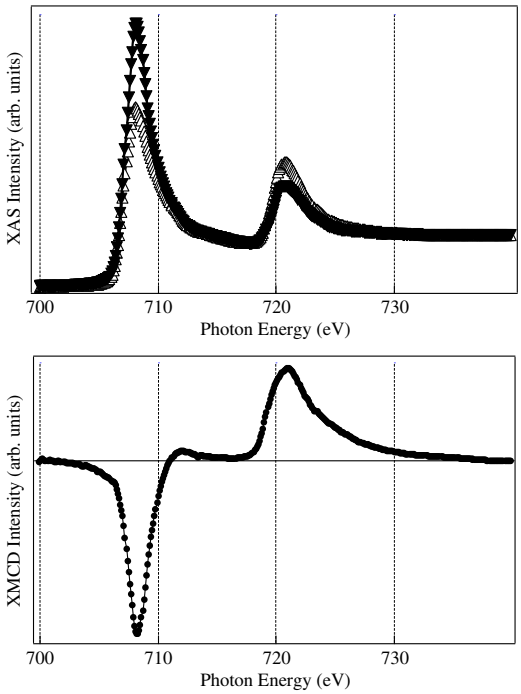


**Figure 2.** (a) Fe L-edge x-ray absorption spectra (XAS) for parallel (unfilled upward triangles) and antiparallel (filled downward triangles) orientations of the photon helicity and sample magnetization, and (b) x-ray magnetic circular dichroism (filled circles) derived from the XAS results in (a).

The magnetic character of the films has been confirmed using x-ray magnetic circular dichroism (XMCD) acquired using beamline 4ID-C at the Advanced Photon Source at Argonne National Laboratory [19]. Fe L-edge x-ray absorption and XMCD spectra for a typical Fe film are shown in figure 2. The sample was magnetized parallel and antiparallel to the [001] direction between successive energy loss spectra using a pulsed magnetic field applied with a nearby coil. The measurements were carried out at room temperature. In these measurements, the photon helicity is fixed (for example, right circular polarization) and, by reversing the magnetization direction of the sample, the relative orientation of the sample magnetization and photon helicity is varied from parallel (unfilled upward triangles) to anti-parallel (filled downward triangles). The average of these two spectra is the usual x-ray absorption spectrum and the difference is the XMCD spectrum (lower panel). In this case, selection rules determine that the spectra are dominated by $2p \rightarrow 3d$ transitions and therefore contain information about the empty d-like density of states. The difference in intensities seen for the two orientations of sample magnetization and photon helicity is related to the difference in majority and minority spin holes in the absorbing material. The large dichroism signal indicates that the unoccupied d-states are predominantly minority spin in character. (This data has not been corrected for incomplete photon polarization or for imperfect alignment of the sample magnetization and photon helicity. Both corrections would increase the dichroic signal.) Thus it is clear that the XMCD data complement not only SPEELS measurements, but spin-polarized photoemission and inverse photoemission as well. A comparison of XAS and XMCD with inverse photoemission and spin-polarized inverse photoemission is detailed in [20].

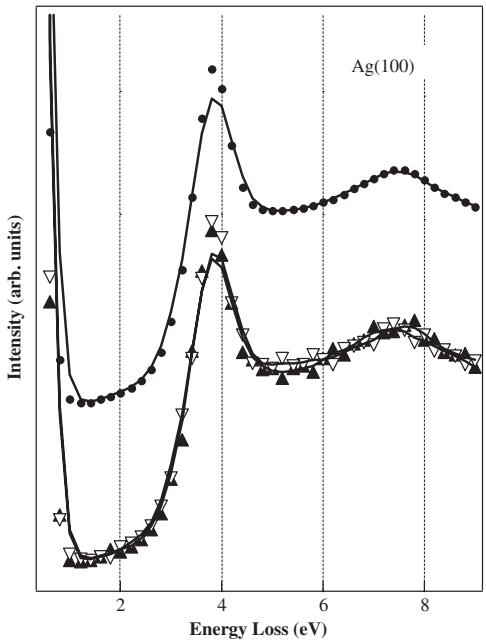


**Figure 3.** Experimental SPEELS results for clean Ag(100). Majority spin and minority spin data are indicated as upward and downward triangles, respectively. The circles indicate the total intensity. The data were acquired with a primary beam energy of 300 eV. We observe no difference between majority and minority spin spectra, as expected. The solid lines are guides to the eyes.

## 3. Results and discussion

Figure 3 shows the results of SPEELS measurements from the clean Ag(100) surface. The majority spin (parallel to the magnetic field) and the minority spin (anti-parallel to the magnetic field) spectra are indicated with upward and downward triangles in the lower spectra, respectively. The spin-integrated data is shown as circles in the upper spectra, and shows peaks at ∼4 and 7.8 eV due to excitation of surface and bulk plasmons, respectively. This is in very good agreement with previously published EELS data on Ag(100) [21]. There is no distinguishable difference between the majority and minority spin spectra, indicating that the polarization is zero within experimental error over the entire energy loss region examined, as is anticipated.

On the other hand, figure 4 shows the SPEELS result for 20 ML bcc Fe films on Ag(100). The top panel, figure 4(a), shows the spin integrated spectrum, the middle panel, figure 4(b) shows the majority spin spectrum, and the bottom panel, figure 4(c), the minority spin spectrum. The bottom two panels also show the spin-integrated spectrum (dotted line) to emphasize the spin-dependent features. The spin-integrated results show a slowly varying energy dependence with a broad loss peak centred near 2.0 eV. This is in reasonable agreement with previous published electron energy loss spectra from single crystal Fe(110) [1, 4]. The spin-resolved data, however, reveal differences in the energy loss spectra for majority and minority electrons. The data reveal relatively sharp majority spin peaks at 1.8 and at 2.5 eV, and a minority spin peak at approximately 2.0 eV. These relatively sharp features have not been reported previously for SPEELS from Fe(110). The measured spin polarization in these peaks is approximately +15% in the majority peak at 1.8 eV, +10% in the majority peak at 2.5 eV, and −10% in the minority spin peak at 2.0 eV. Elsewhere in the energy loss spectrum, the spin polarization is zero.

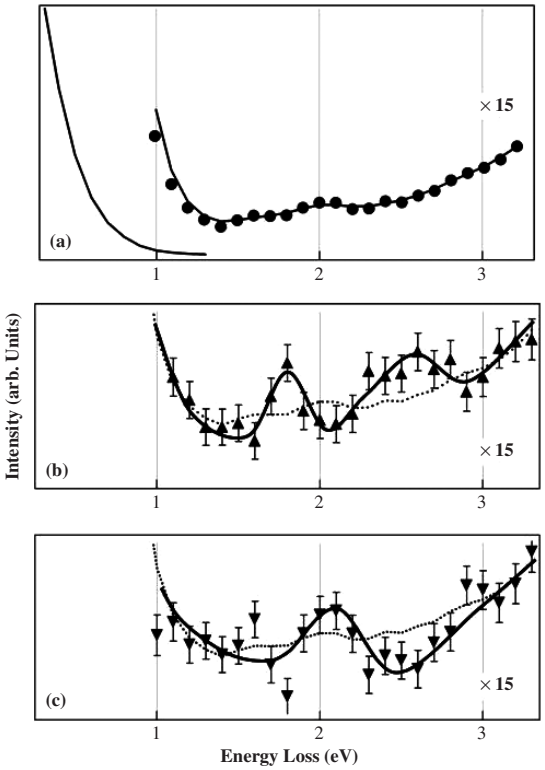


**Figure 4.** Experimental results for 20 ML bcc Fe films on Ag(100). (a) Spin-integrated energy loss spectrum with a part of the elastic feature (actual scale) on the left side of the panel. (b) Majority spin energy loss spectrum showing two peaks at ∼1.8 and 2.5 eV. (c) Minority spin energy loss spectrum showing a peak at ∼2.0 eV. The data were acquired with a primary beam energy of 300 eV. The solid lines are guides to the eyes. The dashed line in (b) and (c) is the spin-integrated spectrum, and is provided to facilitate comparison of majority and minority spin features. The spectra are enlarged (about 15 times with referrence to the actual scale) to get a clear view of each peak.

As previously mentioned, polarization in the scattered electrons can be attributed to exchange scattering, with spin-flip exchange scattering giving rise to majority spin peaks, and non-flip exchange scattering producing minority spin peaks. The majority spin peaks are therefore associated with incident electrons that occupy empty minority spin states near the Fermi energy and transfer energy to excited majority spin electrons. Beginning with the 2.5 eV majority spin peak and referring to figure 1 in [22] for the Γ–H direction appropriate to our experimental geometry, we attribute the peak to incident electrons occupying states in the $\Delta_2^{\downarrow}$ band and exciting majority electrons from the $\Delta_2^{\uparrow}$ band. This is then a primary Stoner excitation. These bands are rigidly separated along the Γ–H direction, with an energy separation calculated in [22] to be approximately 2.3 eV. This is in very good agreement with our observed value of 2.5 eV. Our observed value is also well supported by previous experimental observations, with spin-resolved inverse photoemission from Fe(100) on Ag(100) observing a minority spin peak at 1.6 eV above the Fermi level and spin-resolved photoemission attributing a majority spin feature at 1.2 eV below $E_F$ to emission from the $\Delta_2^{\uparrow}$ band near the zone centre [23]. These experimental results suggest an energy of 2.8 eV for the proposed SPEELS peak assignment. The majority peak at 1.8 eV has a similar origin. The incident electron occupies the $\Delta_5^{\downarrow}$ band and excites majority electrons from the $\Delta_5^{\uparrow}$ band. This, again,

is a primary Stoner excitation with a rigid splitting between the bands along the Γ–H direction of approximately 2.1 eV [22]. There are no experimental data with which to compare our observed peak position, but the agreement with the calculated results is reasonable. Finally, the minority spin feature observed at 2.0 eV is attributed to occupation of the $\Delta_2^{\downarrow}$ band near the zone centre by incident electrons that then excite electrons from the $\Delta_5^{\downarrow}$ band near the zone centre. The calculated energy for this transition from [22] is 1.9 eV, which is in good agreement with our observed value and consistent with the observation of a minority spin peak in inverse photoemission from Fe(100) on Ag(100) at an energy of 1.6 eV above the Fermi level [14], and from spin-resolved photoemission that shows a strong minority spin peak from the $\Delta_5^{\downarrow}$ band at 0.3 eV below $E_F$ [23].

It is important to realize that, for our experimental geometry, we are probing along the Γ–H direction in the Brillouin zone. Previous SPEELS measurements for Fe have concentrated on Fe(110) and therefore the Γ–N direction. Comparing the band structure in these directions in figure 1 of [22] suggests a possible explanation for the presence of sharp peaks in our data and broad features in previous studies. In the Γ–H direction, the band structure is relatively simple, with rigid splitting between the exchange split $\Delta_2$- and $\Delta_5$-symmetry bands. This should lead to the observation of sharp peaks in the majority spin spectra. For the observed minority spin feature at 2.0 eV, the sharpness of the peak is attributable to a high density of states in the minority $\Delta_2$ band and the majority $\Delta_5$ band near the zone centre, as born out by previous spin-resolved photoemission [23] and inverse photoemission [14] measurements. The situation for Γ–N is far more complex, with the exchange splitting varying considerably along this direction and many more possible secondary Stoner excitations. Hence, averaging over the Brillouin zone in this direction would lead to the observation of broad peaks, as has been suggested in [2]. The importance of crystallographic orientation on the observation of sharp peaks in a SPEELS experiment was previously pointed out by Idzerda *et al* [3] in their study of bcc Co along the Γ–H direction. Here they observed sharp features similar to those that we observe for iron, and they predicted the absence of sharp features along the Γ–N and Γ–P directions.

## 4. Conclusions

In summary, we have observed sharp spin-dependent energy loss features for electron scattering from bcc Fe(100) films on Ag(100). We observe both minority and majority spin features and attribute these to exchange scattering of incident electrons, with spin-flip scattering responsible for the majority spin peaks and non-flip scattering responsible for the minority spin peak. Our peak assignments are in good agreement with band structure calculations for Fe [22] and with spin-resolved inverse photoemission [14] and photoemission [23] results. The majority spin peaks correspond to primary Stoner excitations involving the exchange split $\Delta_2$- and $\Delta_5$-symmetry bands at energies of 1.8 and 2.5 eV, respectively. The minority spin peak corresponds to spin-flip scattering with the incident electron occupying the minority spin $\Delta_2$ band and excitation from the minority spin $\Delta_5$ band near the zone centre. In addition, our results emphasize the importance of crystallographic orientation in observing sharp features in a SPEELS measurement.

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