Spin-Dependent Electron Attenuation by Transmission through Thin Ferromagnetic Films

D. P. Pappas, K.-P. Kämper, B. P. Miller, and H. Hopster
Department of Physics and Institute for Surface and Interface Science, University of California, Irvine, California 92717

D. E. Fowler, C. R. Brundle, and A. C. Luntz
IBM Almaden Research Center, San Jose, California 95120

Z.-X. Shen
Stanford Electronics Laboratories, Stanford University, Stanford, California 94305
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Spin-polarized photoemission spectra at low photon energies from ferromagnetic ultrathin Fe layers on Cu(100) show a substantial polarization of the Cu 3d peaks. This is attributed to spin-dependent attenuation in the Fe overlayer. Values of the spin-dependent mean free path at low electron energies are obtained.

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Electron inelastic mean free paths (IMFP's) and electron attenuation lengths in solids are of fundamental importance in electron spectroscopies of solid surfaces. The generally applied method of measuring attenuation lengths is the overlayer method, where the attenuation of a substrate peak is measured as a function of overlayer thickness d and fitted with an exponential decay $e^{-d/\lambda}$, where λ is the attenuation length. For the purpose of this work elastic scattering is neglected, so λ can be identified with the IMFP.[1] A large amount of data have been accumulated for many materials (mainly by x-ray photoemission spectroscopy and Auger electron spectroscopy) and is often displayed as a "universal curve" with λ as a function of electron energy, even though there is a considerable spread in the data and λ is material dependent.[2] For low electron energies (<40 eV) there are little data available for transition metals and the increase in the IMFP at low energies, as displayed by the universal curve, is experimentally not well established. This has become of importance recently in conjunction with spin-polarized secondary-electron spectroscopy, where the magnetism of the first 2–3 atomic layers dominates the spin polarization.[3,4]

In addition, in ferromagnetic materials the IMFP can be expected to be spin dependent.[5] This has been a matter of uncertainty and controversy in spin-polarized electron spectroscopy for a long time. The experimental evidence for a spin-dependent IMFP comes from more indirect measurements, e.g., the enhancement of secondary-electron polarization,[6] or, more directly, spin-polarized electron-energy-loss spectroscopy (SPEELS).[7]

In this paper we report on the first measurement of spin-dependent attenuation lengths for electrons by determining the polarization and attenuation of the Cu 3d UV-photoemitted electrons after they have passed through ultrathin ferromagnetic Fe films on Cu(100). In previous spin-polarized electron spectroscopy studies (e.g., threshold photoemission[8] or secondary-electron spectroscopy[4]) the relative contributions of the substrate and overlayer could not be determined. In the present energy-resolved experiment, however, the origin of the electrons is known since the Cu electrons are marked by their kinetic energy, and the Fe background can be subtracted out.

If the IMFP is different for spin-up and spin-down electrons, one expects that the individual spin currents can be described by

$$I_{+,-} = \tfrac{1}{2} I_0 e^{-d/\lambda_{+,-}}, \qquad (1)$$

where it is assumed that one starts from an unpolarized intensity I_0 (Cu 3d in this case). SPEELS experiments have shown that the energy-loss rate is larger for spin-down than for spin-up electrons.[7] Therefore, one expects $\lambda_+ > \lambda_-$ and $I_+ > I_-$, i.e., a positive (majority) spin polarization.

From Eq. (1) one obtains for the total (spin-averaged) intensity

$$I(d) = \tfrac{1}{2} I_0 (e^{-d/\lambda_+} + e^{-d/\lambda_-}). \qquad (2)$$

If λ_+ and λ_- are not too different (as turns out to be the case) this double-exponential decay is experimentally indistinguishable from a single exponential,

$$I(d) = I_0 e^{-d/\lambda} \text{ with } \lambda = \tfrac{1}{2}(\lambda_+ + \lambda_-); \qquad (3)$$

λ can therefore be identified with the spin-averaged IMFP. Spin-averaged attenuation lengths were determined using the Ar I, Ne I, He I, and He II lines at 11.8 eV, 16.8 eV, 21.2 eV, and 40.8 eV, respectively, from a resonance lamp. Spin-polarized photoemission experiments were performed on the 6-m TGM beam line 1-2 at the Stanford Synchrotron Radiation Laboratory.

The ultrahigh-vacuum system, base pressure $<10^{-10}$

Torr, consists of a 90° spherical electron energy analyzer coupled to a medium-energy retarding-field Mott detector. Low-energy electron diffraction (LEED) and Auger electron spectroscopy are used for sample characterization. The Fe layers were prepared following the prescription given in a recent paper on the magnetic properties of the films studied by spin-polarized secondary-electron spectroscopy.[9] In brief, the Fe layers are deposited with the Cu crystal at 100 K, then annealed to 300 K, and cooled down again to 100 K to record the spin-polarized photoemission spectra. During the annealing cycle the magnetization of the films was checked by measuring the spin polarization of electron-excited secondary electrons. The spin-polarized spectra in this paper were taken on films with perpendicular magnetization [<6 ML (monolayers) thick].

The Fe-layer thicknesses were determined by calibrating the Auger spectra against a quartz microbalance. The geometry factor between deposition rates on the sample and microbalance was determined by measuring the thickness of a thick film (1000 Å) on a glass slide, in the sample position, with optical interferometry. This calibration gives a factor of 2 larger thickness than the previous estimate,[9] which was based on identifying the kinks in the Auger uptake curve with the completion of atomic layers. The new calibration is in agreement with a calibration based on medium-energy electron-diffraction (MEED) oscillations.[10]

The photoemission spectra were taken in normal emission with the light impinging on the surface at 45° off normal. Figure 1 shows the results of the non-spin-polarized attenuation experiments using the lines from the resonance lamp. The attenuation was obtained by measuring the Cu 3*d* intensity after successive Fe evaporations. The Cu 3*d* peak areas were determined by numerical integration over the spectrum, subtracting a linear background drawn between the high-energy and low-energy sides of the peaks. The resulting attenuation lengths given in Fig. 1 are obtained by a least-squares fit (shown for the He II and He I data). The energies given are referenced to the Fermi energy (i.e., $E=h\nu-E_b$). The He II data (38.1-eV electron energy) show a clearly smaller MFP than the three other lower energies.

Spin-polarized spectra were recorded at 14-, 22-, and 44-eV photon energy. The energy resolution was adjusted so that count rates in the Mott detector were in the range 50–200 counts/s. Figure 2 shows representative photoemission spectra (spin-up and spin-down intensities) at these photon energies. For comparison, the intensity distribution curves (EDC) for the clean Cu surface are shown (arbitrary intensity scale). The Cu photoemission features are clearly identifiable in the Fe/Cu spectra and show the well-known dispersion of the Cu bands.

The disparity between the spin-up and spin-down Cu intensities in the 14-eV spectrum and also to a lesser extent in the 22-eV data is striking. A quantitative mea-

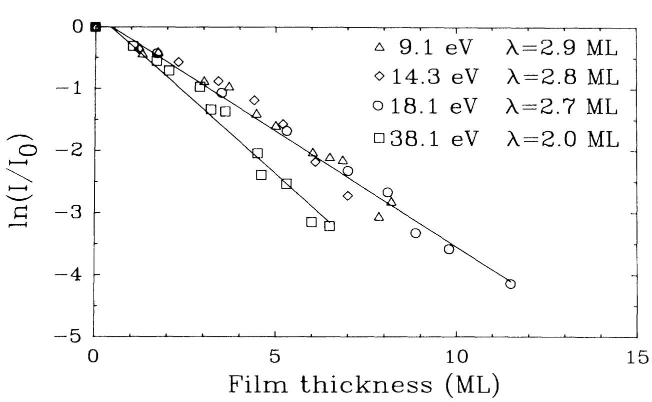

FIG. 1. Plot of the attenuation of the Cu 3*d* intensity as a function of Fe film thickness using the Ar I, Ne I, He I, and He II lines. The electron energies given are with respect to the Fermi energy. The attenuation lengths given in monolayers (ML) are from a least-squares fit of the data points (1 ML=1.8 Å).

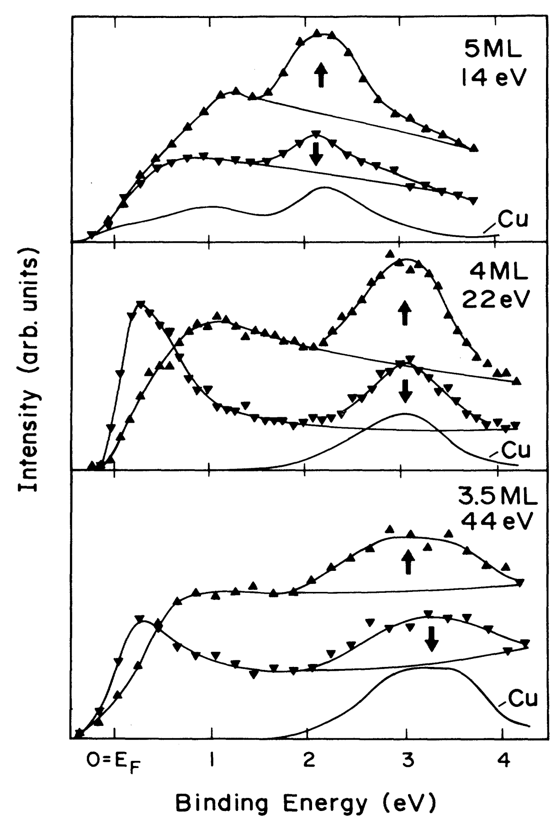

FIG. 2. Spin-polarized photoemission spectra (spin-up and spin-down intensities) of Fe films on Cu(100) at three different photon energies. Fe thicknesses are given in monolayers (ML). Also shown for comparison are the intensity spectra of the clean Cu surface (not to scale). The error in the measurements is the same as the statistical scatter in the data.

sure of the polarization of the Cu 3*d* electrons can be obtained by integrating over the peaks and calculating the net polarization

$$P(\mathrm{Cu})=\frac{I_+-I_-}{I_++I_-}=\frac{1}{2}\frac{e^{-d/\lambda_+}-e^{-d/\lambda_-}}{e^{-d/\lambda}}. \quad (4)$$

The Cu spin-up and spin-down peak areas were obtained by integration assuming a smooth background as indicated in Fig. 2.

The following net Cu polarizations are obtained as an average over several films of thicknesses around 4 ML: 20% at 14-eV, 12% at 22-eV, and 5% at 44-eV photon energy. Experiments on thick films (>12 ML) exhibit only a very small Fe peak in the region of the Cu 3*d* states at these photon energies, thus ruling out a significant contribution due to, e.g., an Fe majority-spin peak. The observed Cu polarizations depend on the Fe thickness. This rules out an induced magnetization of the Cu as an explanation for the polarization. In addition, no exchange splitting is observed in the Cu 3*d* states.

Because of the low count rates in the spin analyzers, measuring the spin polarization at many thicknesses is presently impractical. We have therefore determined the spin-dependent IMFP's by combining the spin-averaged IMFP with the polarization at one thickness, $d\sim4$ ML (for thinner Fe films the polarization effect is small, and for thicker films the Cu signal is difficult to extract from the background). Combining Eqs. (2), (3), and (4) one obtains

$$\frac{1}{\lambda_{+,-}}=\frac{1}{\lambda}-\frac{\ln(1\pm P)}{d}. \quad (5)$$

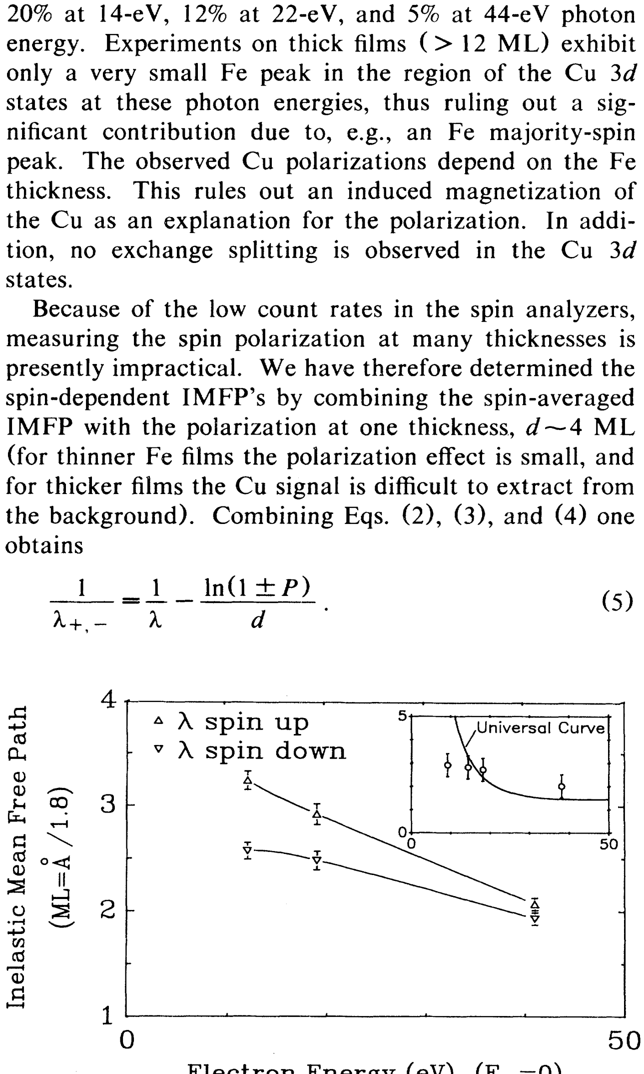

FIG. 3. Spin-dependent inelastic mean free path (IMFP) determined from the spin-polarized photoemission spectra (see text). Inset: Values of the spin-averaged IMFP compared to the universal curve. The electron energies are referenced to the Fermi energy. The error bars on the spin-averaged data reflect mainly the error in thickness calibration, while the error bars on the spin-resolved data include only the contribution from the polarization uncertainty.

The values thus obtained for the $\lambda_{+,-}$ are shown in Fig. 3 for the three photon energies used. Also shown in the inset are the values of the spin-averaged IMFP compared to the universal curve, using the parameters given for elements in Ref. 2. While there is very good agreement for the higher photon energies the point at 9-eV electron energy (from the Ar I data) clearly does not show the large increase exhibited by the universal curve. The spin-polarized data show that the increase of the spin-up IMFP towards low energies is mainly responsible for the increase of the IMFP.

These data are of interest in interpreting spin-polarized secondary-electron spectra. The secondary-electron cascade is dominated by very-low-energy electrons. The present data suggest that the mean free path is still quite small at the vacuum level (5 eV above E_F) in Fe. This explains the surface sensitivity. In addition, the spin dependence of the IMFP can explain the magnitude of the secondary polarization enhancement. In the simplest model one might assume that secondary electrons are being created uniformly with a polarization P_0 corresponding to the magnetization of the material (25% in Fe). The spin dependence of the mean free path then acts as a spin filter, preferentially allowing spin-up electrons to be emitted. The resulting polarization is then given by $P\approx P_0+A$, with $A=(\lambda_+-\lambda_-)/(\lambda_++\lambda_-)$ being the spin asymmetry of the IMFP's. From an extrapolation of the $\lambda_{+,-}$ in Fig. 3 to $E=5$ eV ($=E_{\mathrm{vac}}$) a value of $A\approx0.2$ seems reasonable. Thus one expects $\approx45\%$ for the secondary-electron polarization, in good agreement with experiment.[11]

Extending the present experiments to still lower energies and other materials will allow us to get a better knowledge of IMFP's and their spin dependence at low energies in ferromagnetic materials. While preliminary data show the Fe-thickness dependence of the Cu polarization, a larger thickness range has to be covered in order to test the underlying assumption used in the data analysis [i.e., Eq. (1)]. This is planned for future synchrotron experiments, utilizing higher photon fluxes.

In summary, we have measured the electron inelastic mean free path in ferromagnetic fcc Fe layers on Cu(100). It was found, contrary to the universal curve, that the mean free path increases only slightly towards low energies. By spin-polarized photoemission spectroscopy it was shown that the Cu 3*d* electrons become polarized by transmission through ultrathin ferromagnetic Fe overlayers due to spin-dependent attenuation in the Fe layer. The spin dependence of the electron mean free path was obtained. The increase of the mean free path at low energies is more pronounced for spin-up electrons than for spin-down electrons.

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[11]The actual secondary-electron cascade is more complicated. An electron above the vacuum level undergoing inelastic scattering can still be emitted (at lower energy). Thus the relation $P \approx P_0 + A$ is only an upper limit for the polarization. However, spin-flip scattering, which is not taken into account in this model, counteracts this reduction of P.