# Origin of Spin-Dependent Asymmetries in Electron Transmission through Ultrathin Ferromagnetic Films

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We present theoretical calculations of exchange asymmetries in the transmission of electrons through ultrathin films of ferromagnetic Fe. The results account nicely for the magnitude of the asymmetries observed by Pappas *et al.* in photoemission studies of Cu covered by an ultrathin film of Fe. We argue that exchange asymmetry in the transmissivity of the Fe film, rather than the spin dependence of the electron mean free path, is responsible for the effects reported by these authors.

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During the past decade, various spin-polarized electron spectroscopies have been employed to probe the nature of magnetism in the outermost atomic layers of crystals, and in ultrathin ferromagnetic films absorbed on substrates.[1] Quite clearly, the quantitative interpretation of data generated by such spectroscopies requires knowledge of the spin dependence of the mean free path of the electron ultimately detected. As far as we know, there are no full, reliable calculations of both the energy and spin dependence of the mean free path in the ferromagnetic metals, though the various factors which influence the spin dependence have been discussed.[2] While it does not seem necessary to invoke the spin dependence of the mean free path to obtain quantitative theoretical descriptions of some data,[3] the issue remains open and is very important in our view.

In their analysis of an elegant experiment, Pappas *et al.*[4] inferred the magnitude, energy variation, and spin dependence of the electron mean free path in an ultrathin film of ferromagnetic Fe, for electrons with kinetic energy in the 5–40-eV range above the vacuum energy. The 4-monolayer Fe film was deposited on a Cu(100) substrate, and electrons photoemitted from the $3d$ bands of the Cu substrate were examined. The photocurrent emitted normal to the surface was studied with a spin-sensitive detector; the photoelectrons with spins parallel to those in the Fe film were emitted with intensity greater than those with spin antiparallel to the Fe spins. Pappas *et al.* argued that this phenomenon had its origin in the spin dependence of the electron mean free path in the Fe; general arguments suggest[2] the mean free path for up-spin electrons (spins parallel to the majority spins in the ferromagnet) should be larger than that for down-spin electrons, a result consistent with the data of Ref. 4.

In this paper, we present theoretical calculations which suggest a different origin for the spin asymmetries in the photoemission data just discussed. As the excited electron propagates through the Fe film, of course, it engages in multiple scattering off the Fe atoms. There is a spin dependence of the cross section for elastic scattering of an electron from an Fe atom, with origin in its exchange coupling to the Fe moment. This spin dependence is responsible for the exchange asymmetries observed in spin-polarized electron diffraction from ferromagnetic materials.[3,5,6] It will also produce spin-dependent asymmetries in the transmissivities of an ultrathin film, with origin in the spin dependence of the *elastic*-scattering amplitude, rather than that of *inelastic* loss processes. We have calculated the exchange asymmetry of the transmissivity of a 4-monolayer Fe film to find very good agreement for both the magnitude and energy variation of the photoemission anomalies reported in Ref. 4. We thus argue that the data are consistent with the notion that the electron mean free path displays very little spin dependence in Fe, in the energy regime explored in Ref. 4. Our calculations also suggest additional measurements would prove most useful.

We model the Cu substrate as jellium, with a (complex) inner potential whose real and imaginary parts are consistent with those used in the low-energy electron-diffraction (LEED) literature.[7] The Fe film is a set of monolayers arranged to form an fcc stack, with lattice constant equal to that of Cu. This choice of lattice constant is appropriate for an epitaxial overlayer. We assume the inner potential of the ultrathin Fe overlayer is the same as that of the Cu substrate; the imaginary part of the inner potential of Fe is spin independent. We thus have no detailed description of the interface between the two materials in our analysis. At the electron kinetic energies of interest, which lie between 10 and 50 eV above the Fermi energy, we believe this approximation will not have serious consequences.

The Fe atoms are represented by spin-dependent, spherically symmetric potentials placed within muffin tins, as in earlier analyses of spin-polarized low-energy electron-diffraction (SPLEED) data from the Fe(110) surface.[3] The potentials, kindly provided to us by Fu and Freeman, are taken from the study of a seven-layer Fe(110) film by these authors.[8] We use the spherically

 

symmetric portion of the potential of the middle plane of their film, which can be viewed as accurately representing that of an atom in the bulk of Fe. We are thus assuming the magnetic moment of each layer in our model film is identical to that of bulk Fe, which is surely an approximation but one difficult to remove reliably, given our present state of understanding of the detailed geometrical and magnetic structure of such ultrathin overlayers. The single-site $t$ matrices used in our multiple-scattering analysis of the transmissivity of the film are calculated by means of the fully relativistic method developed by Feder.[5]

We proceed by "shooting" an electron at the Fe film from within the Cu substrate along the normal to the interface, and we then calculate the amplitude of the beam transmitted through the film, as a function of energy, spin orientation, and kinetic energy.

The asymmetry in the transmissivity, $A$, is here defined as

$$A=\frac{T(\uparrow\uparrow)-T(\downarrow\uparrow)}{T(\uparrow\uparrow)+T(\downarrow\uparrow)},$$

where $T(\sigma,\sigma')$ is the transmissivity when the beam electron has spin in direction $\sigma$ and that in the substrate is $\sigma'$. For forward scattering, such as that considered here, reflection symmetry requires that $T(\uparrow\uparrow)=T(\downarrow\downarrow)$, and also that $T(\uparrow\downarrow)=T(\downarrow\uparrow)$, so spin-orbit effects (included fully in our calculations of the single-site $t$ matrices) leave the final transmissivity unaffected. The spin dependence thus arises exclusively from exchange, for this special geometry.

Particularly near the lower-energy regime of the range we consider a proper calculation of the energy variation and absolute magnitude of the transmissivity requires incorporation of the image potential into the analysis. Unfortunately, in the energy range of interest, the results of such a calculation will be sensitive to the detailed manner in which the image potential is "rounded off" to join the inner potential of the substrate. It is quite difficult to address this question, without LEED data in hand which explores fine-structure resonances associated with the substrate of interest.[9] In the present case, we have no such data on hand. However, since the asymmetry $A$ involves $T(\uparrow\uparrow)$ and $T(\downarrow\uparrow)$ for electrons of precisely the same energy, as long as we neglect the spin dependence of the image barrier outside the crystal, we expect the correction to be small in the asymmetry, which is a ratio of transmitted intensities all calculated at the same energy.

For overlayers with thickness between 1 and 5 monolayers, we display the energy variation of the asymmetry in Fig. 1. At each energy in this range, the asymmetry increases monotonically with overlayer thickness. As the number of layers is increased, we see the development of a broad maximum near 12 eV. Also, $A$ falls off monotonically with energy, at the higher energies in the range covered by this figure. Pappas *et al.* have measured, by

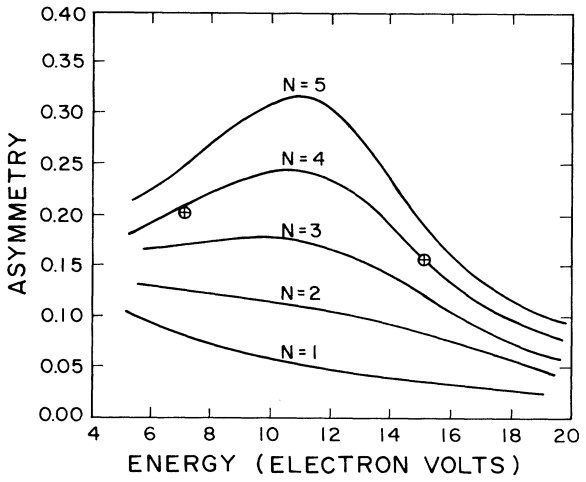


FIG. 1. The asymmetry $A$ in the transmissivity of ultrathin Fe films, model as described in the text. The curves are labeled by the number of Fe layers, $N$. The electron energy is measured from the vacuum level.

the means outlined earlier, the asymmetry at three photon energies, 14, 22, and 44 eV. These have been converted to electron kinetic energies, and two of the points fall within the range of Fig. 1. The experimental values for the asymmetries measured at 14- and 22-eV photon energy have been placed in Fig. 1. The third measurement at a kinetic energy of 37 eV gives $A=5\%$, while theory for 4 monolayers provides a value very close to this. Above 20 eV kinetic energy, we find a more complex behavior for $A$. There is structure, dependent strongly on film thickness, in the few eV above the threshold at 21 eV where the first Bragg beams emerge on the vacuum side of the sample. We comment on this further below.

The agreement between theory and the data available to date is thus very good. Indeed, as outlined above, we have made approximations, so that the excellent agreement at only three energies may be somewhat fortuitous. We do conclude, however, that exchange asymmetries with origin in elastic scattering are surely quite close to those observed, and also display a very similar variation with electron kinetic energy. While there surely must be some spin dependence to the electron mean free path, evidently it is modest in the energy range 5–40 eV. We suggest that data generated from electron spectroscopies which utilize electrons in this range, and most surely those at higher energies, may be analyzed without concern for this issue, at least for Fe. We note that with use of spin-independent mean fee paths, excellent theoretical accounts of the SPLEED data of Waller and Gradmann[10] over a wide energy range can be obtained.[3,11]

It would be of substantial interest to see data on the transmission asymmetries, at other electron kinetic energies. We can see from Fig. 1 that in our calculations the asymmetry does not vary in a monotonic fashion with en-

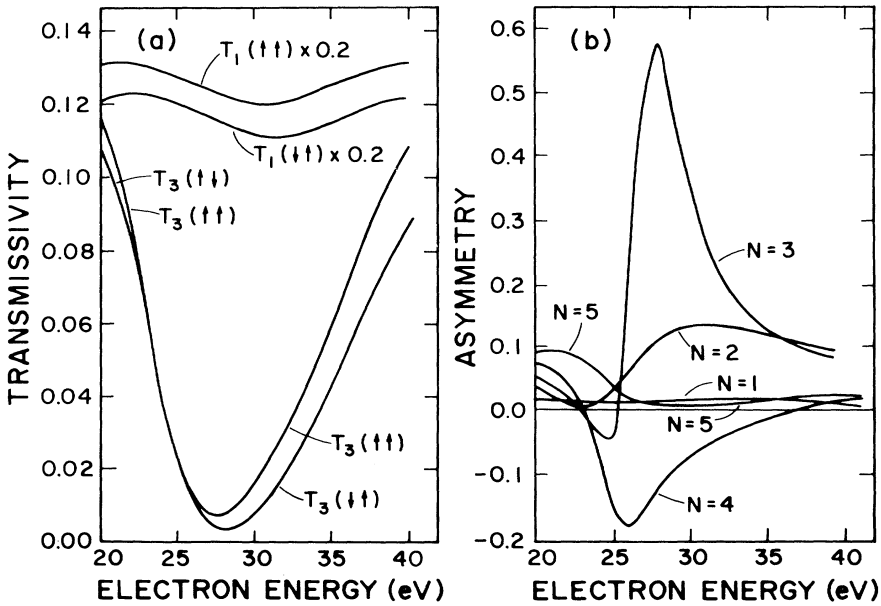


FIG. 2. (a) The transmissivities for up- and down-spin electrons, for the 1- and 3-monolayer films, as a function of energy in the range 20–40 eV. The transmissivities for the monolayer are reduced by the factor of 0.2 to ease the comparison with the three-layer results. (b) The asymmetry in transmissivity, for films that range in thickness from 1 to 5 monolayers, in the energy range 20–40 eV.

ergy. For example, for the 4-monolayer film, there is a clear maximum in the asymmetry near 12 eV. We find further and rather pronounced structure at higher energies, just above 21 eV where the first Bragg beams emerge on the vacuum side of the sample. Such structures would be absent if, as suggested in Ref. 4, the spin dependence of the excited electron mean free path controls the transmission asymmetries. Studies in this energy regime should thus allow a clear and unambiguous determination of the origin of the effect.

Above the beam emergence threshold, in the energy range just discussed, there is a minimum in the transmissivity whose depth is dependent strongly on the thickness of the ultrathin film. We illustrate this in Fig. 2(a), where for the monolayer and the three-layer film, we show the energy and spin dependence of the transmissivities. As one moves from one to three layers, the modest dip evolves into a very deep minimum; for four and five layers, the dip is less deep. In this energy range, the asymmetry in the transmissivity varies dramatically with energy as shown in Fig. 2(b), and also dramatically with overlayer thickness. It must be kept in mind that it is most difficult to prepare a sample with a precise number of monolayers; *Cu*Fe, the example explored here, has a tendency to form nonuniform films. It may thus prove difficult to synthesize samples of quality sufficiently high to explore the sensitive variation with film thickness we predict in this energy range. However, we would surely expect structure to be found in measurements near these energies. Note that the prominent peak in the 3-monolayer film asymmetry is associated with the deep minimum in the transmissivity. This peak may thus be particularly difficult to observe, and may be very sensitive to sample quality. Clearly, data taken in this energy regime should prove of great interest. On samples of suitably high quality, observation of structure in *A* in this energy regime should serve to settle the origin of the asymmetries, as remarked above.

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[2]See the discussion in Sect. 4.4.1 of Ref. 1.

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[5]See Chap. 4 of Ref. 1.

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