Hot-electron scattering length by measurement of spin polarization

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A new method of determining the hot-electron scattering length l is provided by the measurement of the spin of photoemitted electrons from a thin film on a substrate of dissimilar electron-spin polarization (ESP). Results have been obtained for Ni, $l = 10.8^{+2.5}_{-1.7}$ Å for electrons 5.4 ± 0.3 eV above E_F, and for Cu, $l = 10.0^{+2.6}_{-1.7}$ Å for electrons 5.2 ± 0.5 eV above E_F. The agreement between our result for Ni and measurements where electrons were "marked" by their kinetic energy rather than spin indicates the absence of any thickness-dependent depolarization mechanism in the photoemission process. A nonzero ESP is observed in very thin Ni films showing that ferromagnetism occurs already in films of one or two layers average thickness. The similarity between l for Cu and Ni is unexpected in terms of the random-k approximation and points out the importance of the scattering-matrix elements.

Crucial to the interpretation of photoemission data from solids is the relative importance of bulk and surface contributions. The depth of origin of the photoemitted electrons is determined by the penetration depth of the light ($\sim 1/\alpha$, where α is the optical-absorption coefficient) and by the electron mean free path for inelastic scattering l. Normally in metals $\alpha l \ll 1$ and the origin of the electrons is limited by l. If the initial state is localized over a few atomic sites, like d electrons in transition metals, l is a measure of the thickness of material tested in photoemission studies.[1] We report here a new way to determine the hot-electron attenuation length, i.e., the photoelectron escape depth, which complements the existing measurements and estimates of l.[2-4] The experiment uses electron-spin polarization (ESP) to mark the electrons. Results are reported for photoelectrons near photothreshold in Ni and Cu.

Ni was chosen because the interpretation of magnetic photoemission data is of particular interest. The band theory of magnetism predicted a photon energy dependence and a sign of the ESP from Ni[5] and Co[6] that was not observed in the experiments. It has been suggested that a magnetically dead layer exists at the surface[7,8] or that there is an antiferromagnetic coupling of the surface spins to the bulk.[9] A reliable determination of l allows us to ascertain the importance of such surface effects in explaining the discrepancy between the band theory and the ESP results.

Cu was chosen because its electronic structure is very similar to Ni except that the d bands are full which reduces the density of states in the neighborhood of the Fermi energy E_F. The inelastic electron-electron scattering is customarily characterized by an energy-dependent mean free path $l(E)$.[3,10] Kane[11] has shown that ignoring the k dependence of the scattering might be a reasonable first approximation (random-k approximation). In this model, l is predicted to be shorter in metals with a high density of states near E_F. This prediction is tested by comparing the l of Cu with the l of Ni.

The principle of the measurement is that as successively thicker layers of a metal are deposited on a substrate with a different spin polarization, the measured ESP gradually approaches that of the deposited metal because photoelectrons from the substrate are scattered and no longer emitted. The escape probability of an electron photoexcited at a distance x beneath the surface and travelling at an angle θ to the surface normal is proportional to $e^{-x/l(E)\cos\theta}$, if both photoexcitation and escape are isotropic.[10] In the present experiments, electrons are excited to energies slightly greater than the work function φ, and $\cos\theta \approx 1$.

The polarization P of the total photocurrent I is made up of contributions from the substrate with ESP P_1 and the deposited metal with ESP P_2 as follows:

$$PI(E) = P_1 I_1(E) e^{-x/l(E)} + P_2 I_2(E)\,(1 - e^{-x/l(E)}) \,, \quad (1)$$

where $\varphi < E < \hbar\omega$ and

$$I(E) = I_1(E)\, e^{-x/l(E)} + I_2(E)\,(1 - e^{-x/l(E)}) \,.$$

In principle, we can determine both $l(E)$ and the ratio of the internal photocurrents $I_1(E)/I_2(E)$ from a measurement of P as a function of x. However, I_1/I_2 can also be determined from the photoelectric yield of thick films taking into account φ and optical data. At the interface of Ni and Cu, the reflectance is less than 2% ($4.6 < \hbar\omega < 5.7$ eV) and $1/\alpha$ is ~100 Å.[12] From yield measurements we obtained $I_1/I_2 = 1.0$. We observed fluctuations of I_1/I_2 of ~15% with different films. Variation in the yield of polycrystalline evaporated films has also been observed by others.[13] This is expected to cause some scatter in the $P(x)$ curve.

The spin of the photoemitted electrons was measured by Mott scattering as previously described.[14] The Cu and Ni films were electron-gun evaporated

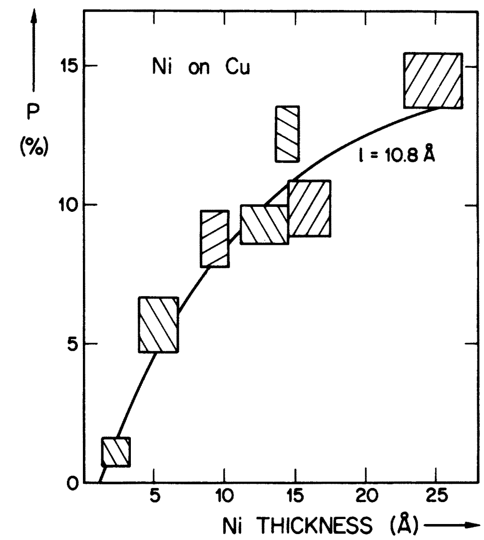

FIG. 1. Polarization P as a function of Ni film thickness is given by the rectangular fields where the vertical dimension represents the statistical uncertainty (one standard deviation) and the horizontal dimension represents the estimated uncertainty of the average film thickness. Fields with the same cross hatching are for films successively evaporated on the same Cu substrate. The solid curve is a least-squares fit of an exponential (see text) to the points.

onto substrates of 100 °C or were annealed to 100 °C. The pressure rose from a base pressure of 2×10^{-10} to 2×10^{-8} Torr during evaporation at 2 Å/sec and fell rapidly afterwards. The average thickness of the film was determined from the change in frequency of a quartz crystal with approximately 25 Hz/Å sensitivity. The uncertainty in measured film thickness (0.5–1.0 Å for a single film) is due to temperature drifts of the crystal frequency. φ was determined from Fowler plots of the yield.

The results for the escape length of Ni are shown in Fig. 1. The parameters l and x_0 in the theoretical formula $P=P_{Ni}(1-e^{-(x-x_0)/l})$ [solid curve in Fig. 1(a)] were chosen by a least-squares fit to the experimental points taking into account both the uncertainties in P and x.[15] $P_{Ni}=(15.15\pm0.3)\%$,[4] and x_0 allows for a possible nonzero intercept. We obtained $l=10.8^{+2.5}_{-1.7}$ Å (where the uncertainty corresponds to ± one standard deviation) for electrons 5.4 ± 0.3 eV above E_F.

The effect of possible magnetic dead layers on the curve is to shift it to the right as the ESP is first evident at a thickness where *magnetic* Ni is present. We obtained $x_0=1.2\pm\sim1$ Å. We cannot rule out one magnetic dead layer (2.2 Å). It appears that the second layer is already ferromagnetic and above two layers ferromagnetism is certainly present. At the temperature of our measurements of 80 °K, Liebermann *et al.*[7] observed ferromagnetism in electrolytic Ni films only at thicknesses greater than two layers. With an escape depth of 10.8 Å, only 18% of the electrons come from the surface layer. Thus one magnetically dead (e.g., paramagnetic) layer or an antiferromagnetically coupled layer cannot change the sign of the ESP.

There is of course the well-known problem of island formation in very thin films. However, Ni on Cu has been found to form continuous films from a few layers on,[16] which agrees well with our measurements of the work-function variation. The presence of islands would reduce the build up of polarization and l appears larger.

In Fig. 2 we compare our measurement at threshold to the points at higher electron energies measured by Eastman,[3] where the electrons are marked by their intensity at a given kinetic energy rather than by their ESP. From the agreement we conclude that the spin polarization is conserved as the Ni electrons travel to the surface. We cannot rule out, however, the possibility of a thickness-independent depolarization mechanism, like spin-exchange scattering from a paramagnetic surface layer. Such scattering could reduce but not change the sign of the ESP.

Recently, Erskine and Stern[17] suggested that a spin-dependent escape depth, caused for instance by a much stronger scattering of minority spins, could explain the observed positive ESP. The positive ESP observed even in very thin films argues

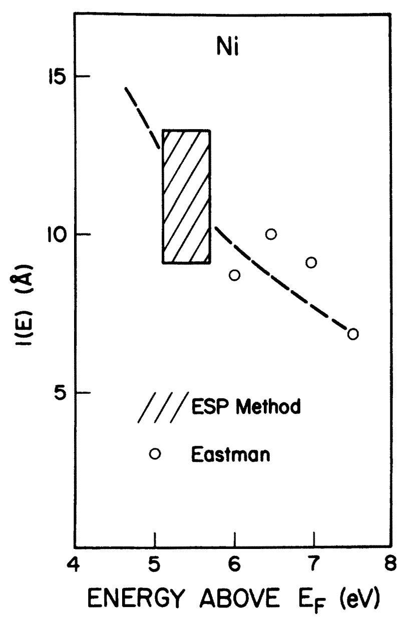

FIG. 2. Scattering length l determined by the ESP method (spin marking) is compared to values determined by the intensity method (kinetic-energy marking). The dashed curve is theoretical $l(E)$ calculated in a free-electron model and normalized to the ESP measurement.

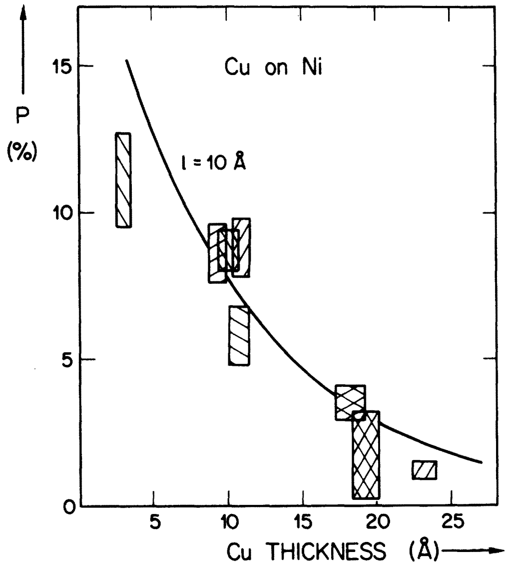

FIG. 3. Polarization P as a function of Cu thickness is shown by the rectangular fields (symbols as in Fig. 1). The solid line is a least-squares fit of an exponential to the points (see text).

against such a spin-dependent escape depth.

The results for Cu deposited on Ni are shown in Fig. 3. The solid line is a least-squares fit to the relation $P = P_{Ni} e^{-(x-x_0)/l}$ from which $l = 10.0^{+2.6}_{-1.7}$ Å for electrons 5.2 ± 0.5 eV above E_F. The solid curve reaches the ESP of pure Ni at a thickness of Cu of $x_0 = 3.3 \pm \sim 1$ Å. This suggests the possibility of a proximity effect making the first Cu layer magnetic.

The hot-electron mean free path in Cu is similar to that of Ni. This is unexpected because electron-beam attenuation measurements of $l(E)$ in Ag and Au, which are similar to Cu, give an $l = 40$ Å for electrons 5.5 eV above E_F.[2] Moreover, it contradicts the predictions of the random-k approximation. The d electrons which make up the high density of states near E_F in Ni may not be as important to the scattering as the random-k approximation predicts, because of small scattering cross sections with the s- or p-like hot electrons.[18] Theoretical studies of the mean free path taking into account the scattering-matrix elements are necessary.

Employing a measurement of electron spin we have made the first determination of $l(E)$ near photoelectric threshold in Ni and Cu. The new method is applicable to both magnetic and nonmagnetic materials, and in addition yields information on magnetic dead layers. It promises to make the new field of magnetic proximity effects accessible to experimental investigation.

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