

Spin-dependent electron scattering in ferromagnetic Co layers on Cu(111)

E. Vescovo, C. Carbone, and U. Alkemper

Institut für Festkörperforschung des Forschungszentrums Jülich, D-52425 Jülich, Germany

O. Rader, T. Kachel, and W. Gudat

Berliner Elektronenspeicherring-Gesellschaft für Synchrotronstrahlung mbH (BESSY), Lentzeallee 100, D-1000 Berlin 33, Germany

W. Eberhardt

Institut für Festkörperforschung des Forschungszentrums Jülich, D-52425 Jülich, Germany

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By means of spin-resolved photoemission we have studied the spin and energy dependence of electron scattering in ferromagnetic Co overlayers epitaxially grown on Cu(111). Taking advantage of the tunability of the synchrotron radiation, the kinetic energy of the electrons propagating inside the solid could be varied between 7 and 50 eV. For all the coverages studied we found that the attenuation length is larger for majority-spin electrons than for minority-spin electrons. A comparison with theoretical models indicates that the observed spin-dependent effects originate from inelastic rather than elastic-scattering processes.

I. INTRODUCTION

In a ferromagnetic material the electron mean free path (MFP) is expected to be spin dependent so that an unpolarized electron beam traveling through such a material will progressively acquire a spin polarization along its way. Already more than 15 years ago spin-resolved photoemission experiments, without energy analysis, provided evidence of these kind of phenomena.¹ It was, for example, found that the very low-kinetic-energy electrons (secondary electrons) generated by photoemission from ferromagnets, exhibit spin polarization considerably larger than the one expected from the bulk band contribution to the magnetization, i.e., the primary electrons are effectively spin filtered through the cascade of elastic- and inelastic-scattering events. These kind of experiments clearly demonstrate that the MFP of electrons traveling inside a ferromagnet can be spin dependent. However a quantitative analysis of this kind of data is hindered by the fact that the secondary electrons are a complex product of many scattering events.

Recently, by means of energy-resolved photoemission experiments, spin-filtering effects have been observed also in the *elastic* transmission of unpolarized electron beams through thin films of ferromagnetic materials.²⁻⁴ In principle, these types of experiments have the advantage of providing very detailed information on the spin-dependent electron MFP which should allow one to discriminate between the various types of possible scattering events. It is, in fact, possible to determine the dependence of the electron MFP not only with respect to the spin but also as a function of the kinetic energy and direction of the electrons traveling in the solid and eventually of the thickness of the ferromagnetic layer.

Qualitatively, the observed spin-dependent effects have often been interpreted by means of the spin dependence of the *inelastic* electron mean free path (IMFP). However, parallel to the experimental investigations, the study of the

electron-scattering processes has been also the subject of considerable theoretical effort.⁵ Various models⁶⁻⁹ have been considered which emphasize different types of spin-dependent scattering mechanisms. Recently, it has been argued⁹ that some of the transmission spin-filtering experiments just mentioned could be quantitatively reproduced by a theoretical model taking into account only the *elastic* contribution of the total scattering cross section. This calculation predicts strong and nonmonotonical dependence of the elastic MFP both with respect to the kinetic energy of the electrons and with respect to the thickness of the ferromagnetic layer. While the comparison of the few available experimental data with this model is extremely encouraging, these predictions have not yet been verified. The purpose of the present study is to provide a set of experimental results collected on a sufficiently large electron-kinetic-energy region and film thickness range such that a close comparison with the theoretical models is made possible.

In this paper we report on the results of spin-, angle-, and energy-resolved photoemission experiments performed from a Co layer epitaxially grown on a Cu(111) substrate. The Cu 3*d* electrons excited by UV light are used as an internal unpolarized electron source whose kinetic energy can be easily varied by changing the incident photon energy. In their path towards the surface of the solid, these electrons are forced to cross the ferromagnetic Co layer whose thickness can be also varied. The electrons emitted along the normal of the sample are collected and their spin polarization is measured. We have explored the behavior of the electron attenuation length (AL) in Co layers of thicknesses between 4 and 20 Å for electron-kinetic energies between 7 and 50 eV. As a function of the kinetic energy the spin dependence of the AL becomes appreciable only for electron-kinetic energies below 30 eV. Furthermore, at every photon energy (i.e., electron-kinetic energy), a monotonical increase of the electron-spin polarization is observed with increasing thickness of the Co overlayer. Since for elastically transmitted electrons a non-

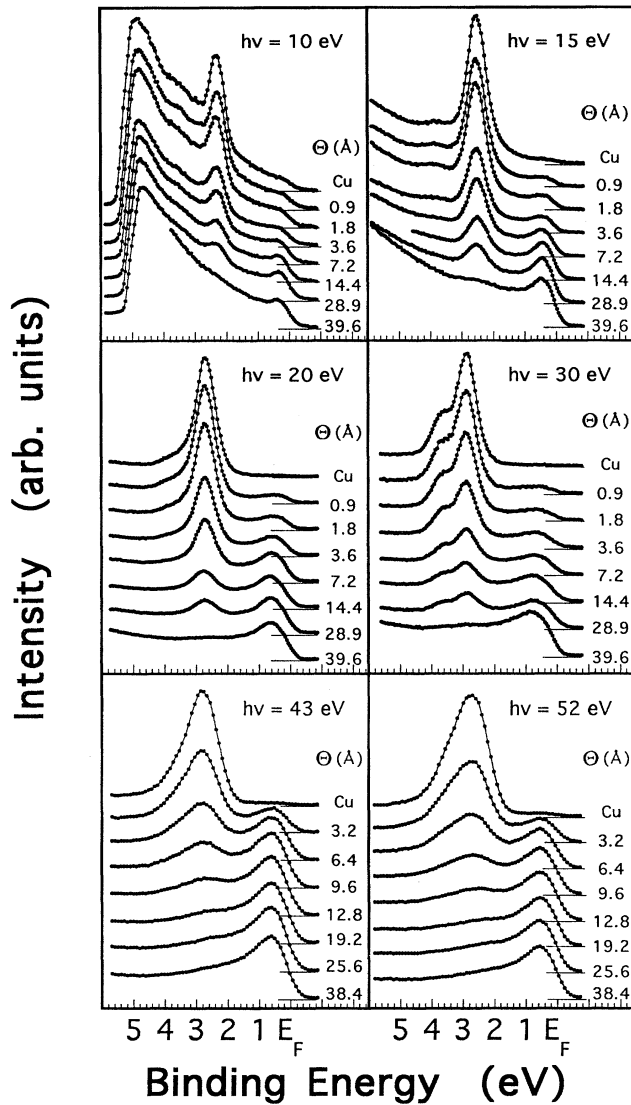


FIG. 1. Spin-integrated photoemission spectra of Co/Cu(111) as a function of Co coverage. The spectra were taken in normal light incidence and normal electron emission geometry at various photon energies. The spectra are normalized to the incident photon flux.

monotonical scattering cross section has been calculated, these results indicate that in the case of Co/Cu(111) the spin-filtering effects observed in the transmitted electrons should be attributed mostly to the inelastic rather than the elastic-scattering events.

II. EXPERIMENTAL AND DATA ANALYSIS

The electron AL inside a material can be obtained by measuring the intensity reduction experienced by an electron beam while crossing a film of that material. If the film is prepared as an overlayer deposited on a substrate, by means of photoemission it is sometime possible to evaluate the AL by monitoring the intensity of a certain emission characteristic of the substrate as a function of the overlayer thickness.

At this point it is useful to point out that the attenuation of an electron beam in a transmission experiment is the result of both *elastic*- and *inelastic*-scattering events. This fact renders the quantitative analysis of this type of data quite difficult because the elastic part of the attenuation does not need to follow a simple exponential decay. Usually the difficulty is removed simply by neglecting the elastic-scattering events. However this assumption is particularly difficult to justify *a priori*, especially in the case of transmission through an epitaxial overlayer. In the following we will first analyze the data in terms of the usual IMFP parameter (λ), i.e., we will neglect the elastic-scattering events. Then this assumption will be justified *a posteriori* when we will see that the spin-filtering effects predicted considering only elastic scattering are very different from the ones we have found, thereby suggesting that in the present case the inelastic-scattering events are the dominating.

The photoemission experiments have been performed at the TGM1 and TGM5 beamlines at the BESSY storage ring in Berlin with the spin- and angle-resolved apparatus already described elsewhere.¹⁰ The photoemission spectra presented here have been taken in normal light incidence and normal electron emission geometry. The Cu(111) sample has been prepared by repeated sputtering and annealing cycles. The Co overlayers were evaporated under UHV conditions from a high-purity Co wire. The Co wire was heated by electron bombardment until a constant evaporation rate of about 2 Å/min was detected by means of an oscillating quartz crystal monitor located at the sample position. The base pressure of 2×10^{-10} mbar raised to about 4×10^{-10} mbar during evaporation.

The growth of thin Co films on Cu(111) has been studied by different techniques.¹¹⁻¹³ While the detailed picture of the growth mode of this system is still a matter of controversy, there is general agreement on the fact that deviation from the ideal layer-by-layer growth are certainly present in this system. We used low-energy-electron-diffraction (LEED) and Auger spectroscopy to characterize the growth mode of Co on Cu(111). The Auger spectra show that the substrate emission is rapidly reduced increasing the Co thickness. This indicates that in this system the interdiffusion is quite limited. However, has recently shown by scanning-tunneling microscopy (STM) measurement,¹² the initial state of the growth of a Co overlayer on Cu(111) is quite complicated. For low Co coverages (< 10 Å) the growth proceeds through the formation of flat fcc islands which do not coalesce. The resulting Co film is epitaxial and crystalline but granular. This growth mode explains the evolution of the LEED pattern as a function of the Co thickness. The LEED of Cu substrate displays a threefold symmetry, characteristic of the fcc Cu(111) structure. Upon Co deposition at room temperature, the LEED spots remain sharp and intense but the threefold symmetry gradually changes and becomes hexagonal at a thickness of about 6 Å. This sixfold symmetry is the results of the superposition of diffracted beams from different fcc-Co islands rotated by 60° with respect to each other.

The complex morphology of the Co films on Cu(111) is a major problem in the determination of the absolute values of the IMFP. The available STM data provide evidence of a nonlayer-by-layer growth mode but they are not enough de-

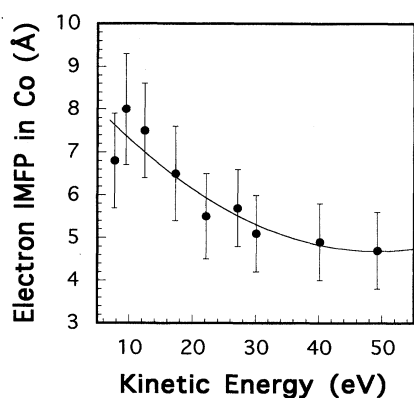


FIG. 2. Spin-averaged IMFP of electrons in Co as a function of electron-kinetic energy. The kinetic energy scale is referred to the Fermi level. The solid line is a smooth curve interpolating through the measured values.

tailed to provide an alternative growth model. Being aware of the complex growth behavior we have attempted to derive a quantitative picture of the islands formation from the ratio of the Auger signals. From our Auger data however we were unable to find a significant ($> 20\%$) deviation from the layer-by-layer growth mode. Therefore we have decided to evaluate the IMFP's assuming a layer-by-layer growth. The values derived in this analysis are thus somewhat larger than the real values due to the presence of pinholes in between the Co islands.

As a first step of our investigation of the IMFP in Co we have determined the energy dependence of the spin-averaged IMFP. A separation between spin and energy dependence is useful because spin-dependent effects are very small and relative differences between majority- and minority-spin electrons can be measured more accurately than absolute effects. Taking a series of photoemission spectra at constant photon energy for varying overlayer thickness (Θ), the attenuation of the substrate signal and the increase of the overlayer signal can be determined as a function of Θ . Examples of this kind of measurements performed at various photon energies are shown in Fig. 1. In all cases the Cu $3d$ emission is located below 2 eV binding energy and is consequently easily distinguishable from the Co $3d$ emission near E_F . Notice that the photoemission spectra converge quite rapidly to spectra without Cu emission. This indicates that the growth in the form of islands which do not coalesce is valid only for the first stages of the growth.

Due to the small hybridization between the Cu $3d$ and Co $3d$ -derived states, it is possible to decompose each spectrum of intermediate Co coverage in the sum of a clean Cu spectrum and a spectrum of a very thick Co overlayer. These decompositions are used to extract the Cu weight in the photoemission spectra I^{Cu} as a function of the Co coverage (Θ). All spectra are normalized to the incident photon flux. Within the simple assumption of layer-by-layer growth, the decrease of the Cu signal follows the curve $I^{\text{Cu}} = I_0^{\text{Cu}} \exp(-\Theta/\lambda)$ where I_0^{Cu} represents the intensity of the emission from pure Cu. For every photon energy a value of

the IMFP (λ) can then be obtained by fitting this curve to the experimental data.¹⁴

A summary of the average values of the IMFP's as a function of the electron kinetic energy obtained with this procedure applied on a large amount of photoemission spectra is presented in Fig. 2. The energy scale is referred to the Fermi level which is separated from the vacuum level by $\Phi = 4.9$ eV. The error bars in this plot are mainly due to uncertainties in the coverage of the deposited Co films of about 10%. As expected there is only a qualitative agreement between our results and the empirical universal curve.¹⁵ In particular the IMFP has a minimum at an electron-kinetic energy of about 40–50 eV and increases towards both lower or higher kinetic energies.¹⁶ However for very low electron-kinetic energies we do not observe the very rapid increase of λ predicted by the universal curve.

To obtain the spin-dependent IMFP we have measured also the spin of the photoemitted electrons. The spin analysis have been performed by Mott scattering at 100 kV onto a thin Au target. The geometry of the experimental apparatus allows the measurement only of the *in-plane* component of the spin-polarization vector. After deposition of a chosen Co coverage, the Co film was magnetized by applying a current pulse through a small coil mounted close to the sample. We

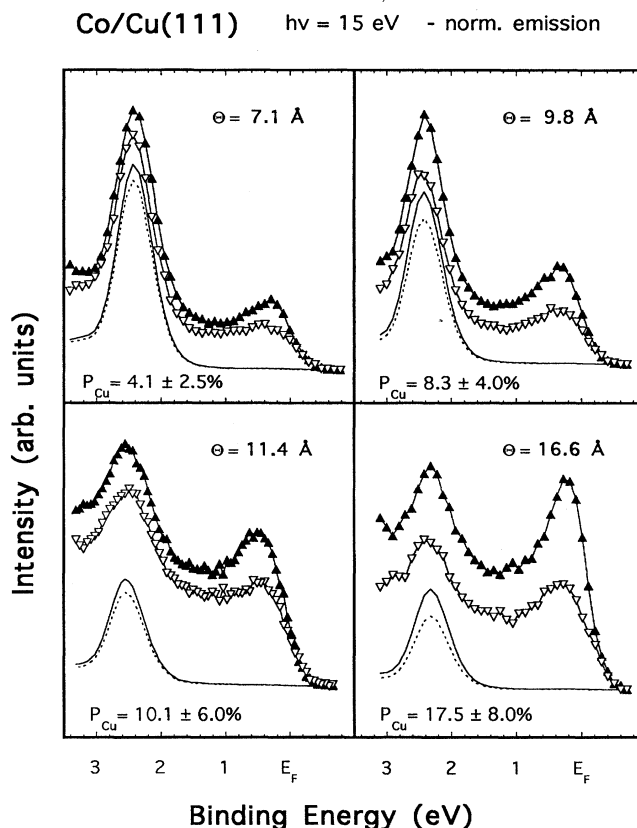


FIG. 3. Spin-resolved photoemission spectra of Co/Cu(111) for different Co coverages. The experimental geometry is the same as Fig. 1. The curves in the bottom of the figures correspond to the Cu contributions for majority (solid line) and minority-spin electrons (dashed line), respectively.

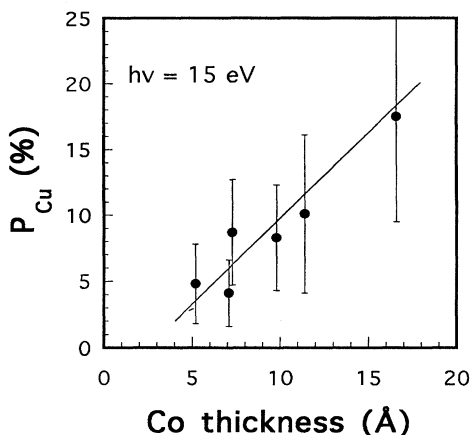


FIG. 4. Spin-polarization P_{Cu} of the Cu emission as a function of the Co coverage as derived from the spectra taken at 15 eV photon energy which corresponds to an electron kinetic energy of 12.1 eV referred to E_F . The solid line is a guide for the eyes.

found that a full remanent *in-plane* magnetization could be achieved only for overlayers thicker than 2 monolayers (1 ML=2.08 Å). This is in good agreement with previous investigations¹⁷ for this system which found that at room temperature the easy axis is perpendicular to the surface plane for one monolayer Co and turns into the surface plane at about 2 ML thickness.

Examples of spin-resolved photoemission spectra taken at 15 eV photon energy for various thicknesses of the Co overlayer are shown in Fig. 3. The majority-spin spectra are drawn as filled-up triangles, while the corresponding minority spectra are drawn as open-down triangles. A first inspection reveals that the intensity of the Cu 3*d* peaks is different in the two opposite-spin channels: i.e., the Co spin-filtering effects are detectable even though weak in some cases. This is in contrast to the observation reported for Fe/Cu(100) (Ref. 2) and for Co/W(110),³ where strong spin-filtering effects could be easily seen in the spin-resolved spectra.

We determined the Cu peaks intensity in the spin-resolved spectra by subtracting a clean Cu spectrum scaled in a way to obtain in the subtracted spectra a smooth “Co secondary background” in the region of the Cu 3*d* peaks. The subtraction has been done for the two spin directions separately. The resulting “spin-resolved” Cu spectra are shown at the bottom of each panel in Fig. 3 [continuous (dashed) line, majority (minority) spin]. The unpolarized Cu electrons acquire a small positive spin polarization when passing through the ferromagnetic Co layer. A plot of the polarization of the Cu electrons in the 15 eV photon energy spectra as a function of the Co thickness is shown in Fig. 4. Notice that consistent with the spin-filtering picture, this polarization increases with the Co thickness. This is an important result because it assures that the observed Cu polarization does not originate from spin-dependent interface hybridization of the Cu 3*d* with the Co 3*d* states.

The spin-dependent IMFP's [$\lambda \uparrow(\downarrow)$] are defined according to the formula $I \uparrow(\downarrow)(\Theta) = (I_0/2) \exp[-\Theta/\lambda \uparrow(\downarrow)]$ where I_0 is the initial intensity of the unpolarized Cu signal. Taking into account that the difference in the two spin-dependent

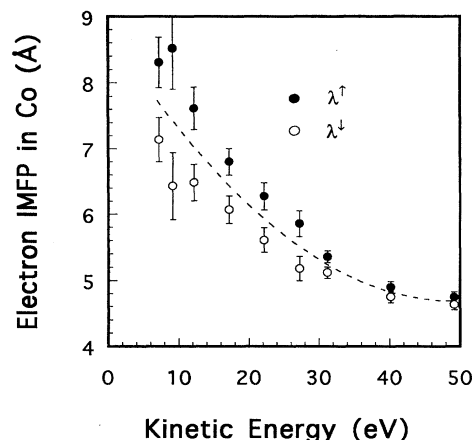


FIG. 5. Spin-resolved electron IMFP's in Co. The kinetic-energy scale is referred to the Fermi level. The dashed curve represents the spin-averaged IMFP.

IMFP's $\lambda \uparrow$ and $\lambda \downarrow$ is small, also the total electron intensity is to a very good approximation decaying exponentially:

$$I = I \uparrow + I \downarrow = (I_0/2) \exp(-\Theta/\lambda \uparrow) + \exp(-\Theta/\lambda \downarrow) \\ \approx I_0 \exp(-\Theta/\lambda),$$

where λ is the spin-averaged IMFP previously determined (see Fig. 2). The spin-dependent AL's are then given by the formula

$$1/\lambda \uparrow/\downarrow = 1/\lambda - \ln(1 \pm P_{\text{Cu}})/\Theta,$$

where $P_{\text{Cu}} = (I \uparrow - I \downarrow)/(I \uparrow + I \downarrow)$ is the spin polarization of the Cu signal. This formula has the advantage of using the spin-averaged λ whose rather large uncertainties can be reduced averaging on many independent (spin-integrated) measurements. However even after this averaging has been performed (see Fig. 2) the estimated uncertainties in the absolute values of the IMFP are still quite large due to the systematic errors in the Co thickness. In order to be able to separate the small spin-dependent effects in the IMFP from these rather large uncertainties in the value of λ we perform the analysis of the spin-dependent $\lambda \uparrow/\downarrow$ substituting the measured spin-integrated IMFP with a smooth curve interpolating through the measured values (solid line in Fig. 2).

Using this procedure the analysis has been conducted on a large amount of spin-resolved spectra taken at photon energies ranging from 10 to 52 eV. We obtain a pair of $\lambda \uparrow$ and $\lambda \downarrow$ from each spin-resolved spectrum. The most reliable data are obtained from intermediate coverages around 4 ML, where the Co layer is already thick enough to produce a sizeable polarization and still the Cu signal is quite intense.

A collection of the average values of the spin-dependent IMFP's obtained from our data is shown in Fig. 5. In this plot the error bars represent mainly the uncertainties in the determination of the spin polarization of the Cu signal. As expected the IMFP's for electrons of opposite spin are equally split around the electron IMFP obtained from the spin-integrated spectra (dashed curve). The difference between the two spin-resolved IMFP's increases continuously

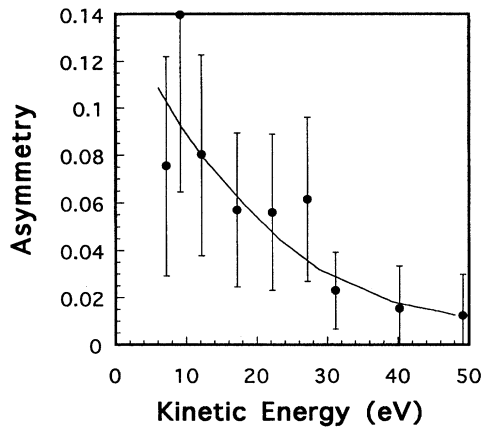


FIG. 6. Inelastic mean-free-path asymmetry A_λ as a function of electron-kinetic energy (referred to E_F). The solid line is a guide for the eyes.

when decreasing the electron kinetic energy. For electron energies below 30 eV the difference in the IMFP for the two spins becomes appreciable. The majority-spin electrons are less attenuated than the corresponding minority-spin electrons. For very low kinetic energies the IMFP of majority-spin electrons is about 18% larger than that of minority-spin electrons. For kinetic energies above 30 eV on the other side the Cu polarization is so small (below 4%) that the difference between λ_\uparrow and λ_\downarrow is within the error bars of the measurement for all spectra.

A convenient way to quantify the spin-dependent effects is to define the IMFP asymmetry $A_\lambda = (\lambda_\uparrow - \lambda_\downarrow) / (\lambda_\uparrow + \lambda_\downarrow)$. A plot of this asymmetry as a function of the kinetic-electron energy is shown in Fig. 6. While the scattering in the experimental points is quite large, the data seem to indicate a monotonous decrease of A_λ as the electron energy is increased. At very low electron energies A_λ reaches its maximum value of about 0.1. Above 30 eV the asymmetry still seems to be positive but we are not able to say whether such small effects are due to differences in the mean free path or systematic errors in the experiment.

III. COMPARISON WITH THEORETICAL MODELS AND DISCUSSION

The electron IMFP inside a magnetic material is determined by different types of scattering processes, typically electron-electron, electron-magnon and electron-phonon scattering.⁵ Electron-phonon scattering can be safely neglected because it is weak and additionally it is not spin dependent.⁸ On the opposite electron-magnon scattering could be an important spin-dependent scattering mechanism but it is also generally discarded on the bases of its weakness compared to the Coulomb scattering.⁶ Due to the exchange interaction the electron-electron scattering occurs preferentially between electrons of opposite spin orientation.⁵ Following this argument in a ferromagnet as a result of the greater number of available spin-up valence electrons with respect to the spin-down, the minority-spin IMFP is expected to be shorter than the corresponding majority-spin IMFP.

With the purpose of estimating the polarization of the electron yield from a ferromagnet, Bringer *et al.*⁶ has proposed a model based on the very simple assumption $\lambda_\uparrow / \lambda_\downarrow = n_\uparrow / n_\downarrow$, where n_\uparrow and n_\downarrow are the number of spin-up and spin-down electrons per atom, respectively. This assumption corresponds to the extreme case in which the exchange contribution perfectly cancels the direct Coulomb interaction. The result is that two electrons with the same spin are effectively not interacting at all. This model is clearly oversimplified in that the spin dependence of the IMFP is by definition independent of the kinetic energy of the electrons. Moreover it overestimates the spin dependence of the IMFP. The predicted IMFP asymmetry of 0.17 for Co is in fact too large compared with the experimental one. However, the sign and also the order of magnitude are given correctly in Bringer's model. The IMFP is larger for majority-spin electrons.

More sophisticated calculations of the spin dependence of the electron mean free path in Fe, Co, and Ni have been performed by Rendell and Penn.⁷ The main improvement comes by performing the explicit calculation of the electron-scattering rates in the presence of a realistic energy-dependent electron density of states of the ferromagnet. In this way the energy dependence of the IMFP is naturally taken into account. Furthermore, in this model the energy dependence of the exchange scattering processes is also considered introducing an empirical energy dependence in the corresponding matrix elements. The results of this calculation show that in a paramagnet the effect of the exchange is indeed reducing the $e-e$ interaction with a consequent increase of the IMFP. However when this effect is combined with the different density of states for the two spins in a ferromagnet, the IMFP asymmetry becomes very small (for Co, $\text{mod}(A_\lambda) < 0.03$ between 0 and 200 eV electron energy). Furthermore in Fe and Co the spin-dependent effects increase at low kinetic energy but they have the opposite sign with respect to the intuitive arguments mentioned before. For electron-kinetic energies < 130 eV the IMFP for minority electrons is predicted to be longer than the one for majority electrons.

More pronounced spin dependence of the IMFP is found by Penn, Apell, and Girvin⁷ in another subsequent calculation in which the additional effects due to the empty part of the electronic structure very near to E_F are included. In a ferromagnet for very low kinetic energies (< 5 eV with respect to E_F) the scattering rate for minority electrons is enhanced with respect to the one for the majority electrons because there is an excess of available unoccupied minority states. This effect tends to build up a positive electron polarization ($\lambda_\uparrow > \lambda_\downarrow$) but becomes important only for very low kinetic energies when the electrons scatter into the unoccupied d states. Quantitatively the mean-free-path difference between the two spin directions is significant only for kinetic energies below 5 eV and it reaches its maximum at zero kinetic energy where the A_λ is about 0.1–0.2. This second model of Penn does show some resemblance with our experimental results. First of all the sign of the spin-dependent effects is now correctly reproduced in the calculation. Moreover also the order of magnitude and the general trend of the effect (increase of the spin dependence decreasing the electron-kinetic energy) is correct. However the experimen-

tal A_λ for Co decreases less rapidly than in Penn's calculations.

An extensive theoretical analysis of the electron inelastic-scattering processes with particular emphasis on the spin-flip loss events has been published by Bocchetta and Tosatti.⁷ In a spin-flip scattering event an electron of a given spin loses its energy decaying below the vacuum level and excites an electron of opposite spin above E_{vac} . These events become an effective spin-filtering mechanism in a ferromagnet where there is an unbalance between spin-up and spin-down electrons. The calculation of the e - e scattering has been performed starting from a very simplified model of the electronic structure of the ferromagnet which should roughly mimic that of Fe. Qualitatively these calculations show that taking into account the spin-flip processes can also reverse the spin of the original results of Rendel and Penn. Moreover these calculations show that one might expect a spin dependence also above 5 eV.

Recently Gokhale and Mills⁹ performed calculations to explain the spin-filtering effects found in the system Fe/Cu(100).² These calculations consider only *elastic*-scattering events. The predicted results are rather strong scattering asymmetries that agree nicely with the existing experimental data for the Fe/Cu(100) system. Due to the spin-dependent electronic structure of a ferromagnetic material, the electronic cross section for elastic scattering in the kinetic-energy range between 5 and 50 eV above the vacuum level, might contain, in fact, a pronounced spin dependence. Here the scattering processes are the same that originate the exchange asymmetry observed in the I_{00} beam of a spin-polarized-LEED experiment as a function of the kinetic energy of the electrons. The calculation is performed for an electron beam transmitted across a ferromagnetic layer. A pronounced and nonmonotonical dependence of the elastic scattering from the thickness of the layer is found. The behavior of the electron transmissivity as a function of the layer thickness is due to a superposition of spin-dependent elastic electron scattering inside the ferromagnetic material (which would be there also for an infinite system) combined with spin-dependent interference phenomena due to the scattering at the two interfaces (nonmagnetic material/magnetic material and magnetic material/vacuum) of the magnetic layer. The calculations performed for Fe/Cu(100) show that both energy and thickness dependence of the transmissivity are very strong. Although a quantitative comparison of our data with these calculation is of course inappropriate, similar effects can be qualitatively expected also for Co/Cu(111). In

particular the polarization P_{Cu} of the Cu signal in the spin-polarized spectra is expected to show oscillations as a function of the film thickness for fixed electron energy. In the case of Fe/Cu(100), at some kinetic energies it is even expected that P_{Cu} changes its sign. Similarly P_{Cu} is expected to display oscillations when, for a fixed Co coverage, the kinetic energy of the electrons is changed. We found instead that P_{Cu} increases linearly with the film thickness of the ferromagnetic Co overlayer (Fig. 4) as it is expected for inelastic scattering. On the other side increasing the electron-kinetic energy reduces monotonically the polarization of the Cu peak in the spin-resolved photoemission spectra.

When comparing our results with this kind of layer calculation some caution is mandatory. The predicted effects could in fact be difficult to detect experimentally due to the deviations from the ideal layer-by-layer growth mode. A superposition of Co islands of different heights would drastically reduce the predicted effects. However, even if the quality of the Co/Cu(111) system is not sufficient to display the spin-dependent elastic-scattering defects, our results show a pronounced spin dependence which originates from the inelastic part of the MFP. Furthermore the observed spin-dependent effects in the inelastic MFP (A_λ) are of the same order of magnitude as the one expected in the case of elastic scattering. Our results then demonstrate that any realistic calculation of the spin-dependent electron MFP is an ordered layer system should contain the contributions due to the inelastic-scattering events.

IV. CONCLUSIONS

The electron IMFP in Co as a function of electron-kinetic energy between 5 and 50 eV has been measured by means of the overlayer technique applied to the system Co/Cu(111). It is found that the IMFP displays the minimum at about 40–50 eV kinetic energy in qualitative agreement with the universal curve. For low kinetic energies the experimentally observed increase of the IMFP is much smaller than the one predicted by the universal curve.

Spin-dependent effects in the IMFP could be detected only for electron-kinetic energies below 30 eV. In Co the IMFP is larger for majority-spin electrons than for minority-spin electrons. The spin dependence of the IMFP increases monotonically towards low energies. A comparison of our results with the available theoretical models shows that the inelastic-scattering processes are the main source of the observed spin-dependent effects.

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