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Spin-polarized electron-energy-loss spectroscopy on epitaxial fcc Co layers on Cu(001)

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The magnetism and the electronic structure of epitaxial fcc Co/Cu(001) films were investigated by spin-polarized electron-energy-loss spectroscopy (SPEELS). Strong spin-flip scattering reveals an exchange splitting of 0.8 eV and a Stoner gap of 300 meV, independent of thickness. Films above 2 monolayers (ML) thickness show ferromagnetic order at room temperature, while the monolayer Co/Cu(001) cannot be remanently magnetized above 80 K. SPEELS spectra from monolayer films are very similar to spectra from bulklike films (20 ML) and display no evidence for enhanced magnetic moments. SPEELS spectra from the bare Cu(001) substrate exhibit a gradual increase of spin-flip scattering for loss energies above 2 eV, while SPEELS data from disordered CoO show sharp loss features, which can be interpreted in terms of ligand-field theory.

I. INTRODUCTION

Spin-polarized electron-energy-loss spectroscopy (SPEELS) has become a valuable technique to probe the elementary magnetic (electron-hole) excitations at ferromagnetic surfaces. The "complete" SPEELS experiment (i.e., the use of a polarized primary beam combined with the analysis of the spin polarization of the scattered electrons), which has so far been carried out on Fe and Ni surfaces,[1–5] allows the unambiguous deconvolution of the total scattering intensity in terms of spin-flip and nonflip transitions. The substantial amount of exchange scattering and its spectral dependence observed in those experiments directly proves the existence of Stoner excitations, i.e., electron-hole pair excitations between states of opposite spin. SPEELS spectra reflect the occupied and the unoccupied parts of the spin-split electronic structure and are shown to allow the determination of the average exchange splitting from the maximum in the spin-flip scattering rate. Accordingly SPEELS yields complementary information to techniques such as spin, angle, and energy-resolved photoemission and inverse photoemission spectroscopy, which directly probe the occupied and unoccupied parts of the spin-split band structure, respectively. SPEELS can thus help to elucidate the microscopic origin of ferromagnetism.

Due to the short mean free path in the ferromagnetic metals at low kinetic energies,[6,7] SPEELS is sensitive to the magnetization of the very first few atomic layers. This surface sensitivity makes SPEELS a very useful tool for the study of ultrathin magnetic films. The ferromagnetism of ultrathin films has recently become a very active field of research.[8] One of the most important questions connected with the fundamental properties of ferromagnetism in such two-dimensional systems concerns the experimental verification of the theoretically predicted enhancement of the magnetic moments.[9,10] Since the size of the magnetic moment is intimately connected with the average exchange splitting, SPEELS allows one to follow a possible variation of the magnetic moment with decreasing film thickness.

A SPEELS experiment on a remanently magnetized sample yields the full information on all scattering channels, the long-range ferromagnetic order leading to spin asymmetries of the scattered intensity upon reversal of the primary beam polarization. But even in a state without ferromagnetic order the complete SPEELS experiment is sensitive to the presence of local magnetic moments. Exchange scattering then leads to a depolarization. This has been demonstrated, e.g., for Ni above the Curie temperature,[11] on Cu and Mo,[12] and for the antiferromagnetic compound Cr_2O_3.[13] This special capability of SPEELS allows the investigation of the magnetism of ultrathin films above the Curie temperature, i.e., in the paramagnetic state.

II. EXPERIMENT

The experiments were performed in a UHV system (base pressure $<10^{-10}$ Torr) designed for various spin-polarized electron spectroscopies.[14] The experimental arrangement is shown schematically in Fig. 1. Briefly, a transversely polarized electron beam derived from a GaAs photocathode (primary beam polarization $P_0 = \pm 26\%$) is scattered off the remanently magnetized sample surface. The scattered electrons are energy analyzed by a hemispherical electrostatic analyzer and their spin polarization is measured in a high-energy Mott detector (100 keV), which has been calibrated to high accuracy ($\pm 2\%$).[15] The total scattering angle is 90°. Specular and off-specular spectra can be taken by rotating the sample. Surface cleanliness and structure are characterized by Auger electron spectroscopy (AES) and low-energy electron diffraction (LEED).

The substrate used was a 2-mm-thick Cu(001) single crystal with a diameter of about 7 mm and oriented within ±1° along the [001] surface normal. The mechanically polished crystal (down to 1 μm) was cleaned by Ne^+-ion sputtering and annealing cycles until no residual contaminants could be detected by AES (i.e., $<1\%$) and a sharp 1×1 pattern with very low background was observed in LEED.

The epitaxial Co/Cu(001) layers were grown by evaporation from a high-purity Co rod heated by electron bombardment. The evaporator was enclosed in a water-cooled jacket. During evaporation the pressure always stayed below 6×10^{-10} Torr. Evaporation at a rate of about 0.5 to 1 atomic layers per minute onto the Cu(001) substrate, held at room temperature, resulted in very clean films, with a contamination (mainly C) on the percent level, as judged by AES. A clear and sharp 1×1 LEED pattern was observed.

The growth of Co/Cu(001) has been thoroughly investigated by a number of authors.[16–22] Despite minor differences, the general picture of the growth mode is quite clear: Co grows on Cu(001) in a well-ordered fcc structure in a layer-by-layer fashion for substrate temperatures between 300 and 450 K. Although most studies[16–19,21,22] claim to see layer-by-layer growth for all investigated coverages [0–20 monolayers (ML)], a recent study by Li and Tonner,[20] using angle-resolved x-ray photoelectron scattering, came to the conclusion that the growth for coverages below 2 ML proceeds via the formation of two-layer-thick islands. For coverages of more than 2 ML they found the Co films to grow in a perfect layer-by-layer mode. Detailed LEED I/V measurements[17] revealed a small contraction of the interlayer spacing in the Co films (the top layer is contracted by 6%, and the inner layers by about 3%), i.e., the films grow in a slightly tetragonal distorted fcc structure.

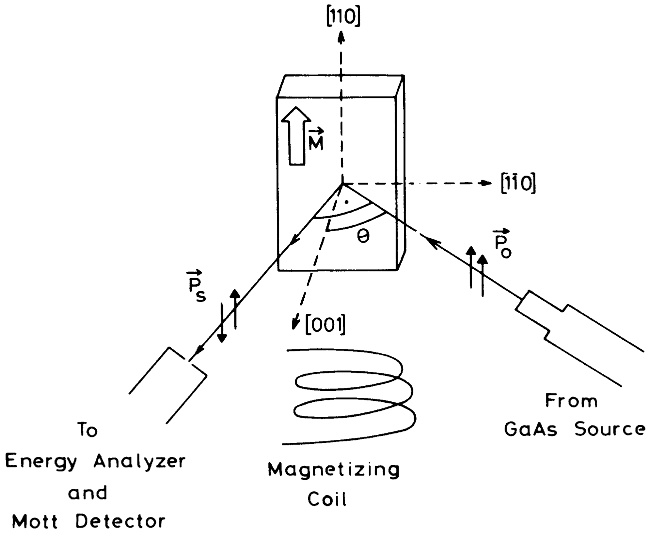

FIG. 1. Schematic of the experimental setup.

The interpretation of experiments on the magnetism of ultrathin, epitaxial films has to be based on a reliable and reproducible thickness calibration. The apparent differences in the experimentally observed thickness dependence of the Curie temperature (T_C) of Co/Cu(001) films[18,22,23] might actually be due to different thickness calibrations. For the calibration we use the 656- and 716-eV lines of Co and the 840- and 920-eV lines of Cu. To correct for the different sensitivity of AES for Co and Cu we incorporate a sensitivity correction factor S into the normalized ratio R

$$R=\frac{I_{\mathrm{Co}\ 716\ \mathrm{eV}}+I_{\mathrm{Co}\ 656\ \mathrm{eV}}}{(I_{\mathrm{Co}\ 716\ \mathrm{eV}}+I_{\mathrm{Co}\ 656\ \mathrm{eV}})+S(I_{\mathrm{Cu}\ 840\ \mathrm{eV}}+I_{\mathrm{Cu}\ 920\ \mathrm{eV}})},$$

where the correction factor S, representing the ratio of the AES intensities of pure bulk Co and Cu, respectively, has been taken from the reference AES spectra of Ref. 24:

$$S=\frac{I_{0,\mathrm{Cu}\ 920\ \mathrm{eV}}+I_{0,\mathrm{Cu}\ 840\ \mathrm{eV}}}{I_{0,\mathrm{Co}\ 716\ \mathrm{eV}}+I_{0,\mathrm{Co}\ 656\ \mathrm{eV}}}=0.73\ .$$

The film thickness d (in ML) is then calculated from the measured values for R, using the equation

$$R=1-e^{-d/\Lambda}\ ,$$

where the "attenuation length" Λ (in ML) was calculated from an average value for the mean free path ($\lambda=15$ Å) at the kinetic energies used from Ref. 25: $\Lambda=\lambda\cos(42°)/(1.77\ \text{Å})=6.3$ ML (1.77 Å represents the thickness of a single monolayer[17] and 42° is the average acceptance angle of the cylindrical mirror analyzer used for AES).

Simultaneous evaporation on a quartz-crystal microbalance was used to check the above thickness calibration. The quartz thickness monitor was calibrated against AES ratios using a former thickness calibration for Fe/Cu(001) films, determined by optical interferometry.[7] The two independent calibrations agree well with each other within ±15%. For future reproducibility the thickness values used in the following are those obtained directly from the AES spectra.

Remanent magnetization of the epitaxial Co/Cu(001) layers was achieved by magnetizing them by a field pulse from a small coil placed directly under the measuring position. The easy axes of the magnetization in Co/Cu(001) were shown to be the in-plane ⟨110⟩ axes.[18,26,27] Therefore the Cu(001) single crystal was oriented with its [110] axis parallel to the quantization axis defined by the spin polarization of the primary electron beam, which coincides with the direction of the magnetic field of the magnetizing coil. To minimize any spurious effects due to beam deflections upon reversal of the magnetization, the pulse current was kept as low as possible.

III. SPEELS ON BULKLIKE fcc Co/Cu(001) LAYERS

Photoemission studies of the evolution of the band structure of epitaxial layers of transition metals with increasing thickness have shown that in many cases the development of the bulk band structure is almost completed upon the deposition of only 5 monolayers [e.g., in the case of Ni/W(110) (Refs. 28 and 29) or Ag/Cu(001) (Refs. 30 and 31). Spin-resolved photoemission studies of Co/Cu(001) films also lead to the conclusion that the electronic structure of 5 ML is already bulklike.[32,33] Accordingly, the electronic structure of 20 ML Co/Cu(001) can be safely assumed to represent the electronic structure of fcc Co. We thus employ SPEELS on a (20 ML Co)/Cu(001) film to investigate the spin-resolved electron-hole excitations from bulk fcc Co.

We analyze our experimental data following the framework given by Venus and Kirschner.[2] The basic quantities measured are the intensity asymmetry A upon reversal of the incoming beam polarization, the polarizations of the scattered electrons in the two cases of the polarization of the incident electrons being oriented parallel and antiparallel to the sample magnetization, and the total scattering intensity I. To eliminate experimental asymmetries we additionally take the same measurements after reversal of the magnetization. From those quantities we calculate the four basic spin-dependent partial scattering rates for flip ($F^{\uparrow},F^{\downarrow}$) and nonflip ($N^{\uparrow},N^{\downarrow}$) scattering, where the ↑ and ↓ refer to the spin of the incoming electrons.[2]

Figure 2 shows those rates as a function of energy loss for (20 ML Co)/Cu(001) together with the corresponding spectrum of the intensity asymmetry A. The data were taken 20° off specular, using a primary energy of $E_P=25$ eV. The energy resolution of the experiment is approximately 300 meV. The off-specular geometry is used to suppress dipolar scattering in favor of impact scattering, in order to see exchange effects in a more pronounced fashion. Unlike the SPEELS data obtained by Idzerda *et al.* on bcc Co/GaAs,[34] all four partial rates and the asymmetry do not show any sharp structures. The *elastic* region is dominated by nonflip scattering. Going into the *inelastic* regime the flip rates (mainly $F^{\downarrow}$) rapidly increase with increasing loss energy. (The flip and nonflip intensities are given on the same absolute intensity scale in the figures.) For loss energies from about 0.5 to 1.5 eV the flip-down rate $F^{\downarrow}$ is approximately equal in magnitude to either of the two nonflip rates. Thus spin-flip scattering plays a very important role in this range of loss energies. The rate for spin-flip scattering of incoming spin-up electrons ($F^{\uparrow}$) is always the smallest (but nonzero) partial rate. The large difference in the flip rates for incoming spin-up and spin-down electrons clearly reflects the spin-split band structure in the vicinity of the Fermi energy (E_F), the general picture emerging from those spectra not being unlike the previous SPEELS data for Fe(001) (Refs. 2 and 3) and for Ni(110).[4] Thus the new SPEELS data for fcc Co support the previous interpretation of SPEELS data in terms of Stoner excitations. The flip-down rate $F^{\downarrow}$ exhibits a broad shoulder at a loss energy of $E_{\text{loss}}\simeq 0.8$ eV, corresponding to the average exchange splitting Δ_{ex} of fcc Co, in accordance with spin-resolved photoemission data of Co/Cu(001).[33,35] This is similar to the case of Fe (Refs. 2 and 3) where the partial flip-down rate displays a shoulder at $E_{\text{loss}}\simeq\Delta_{\text{ex}}\simeq 2$ eV. In Ni, however, the effect is much more pronounced and the partial flip-down rate shows a clear maximum at $E_{\text{loss}}\simeq\Delta_{\text{ex}}\simeq 0.3$ eV. The difference was attributed to the different spin-split electronic structures of Ni and Fe, respectively. While in Ni all majority-spin d states (spin-up) are occupied (so-called strong or saturated ferromagnet), in Fe one finds spin-down and spin-up d states above the Fermi level (so-called weak or unsaturated ferromagnet). Thus, taking only d electrons into account, the flip-up rate in a strong ferromagnet like Ni should vanish. Even in an unsaturated ferromagnet like Fe flip-up Stoner excitations within the d band are only possible for a nonzero momentum transfer $q\neq 0$. Flip-up Stoner excitations are, however, in all ferromagnets possible upon the involvement of *sp* electrons, i.e., in the form of electron-hole pairs consisting of a d hole of given spin and an electron in a free-electron-like state of opposite spin. The importance of such free-electron-like Stoner excitations for the interpretation of SPEELS data on Fe (Ref. 2) has been stressed by Penn and Apell.[36,37]

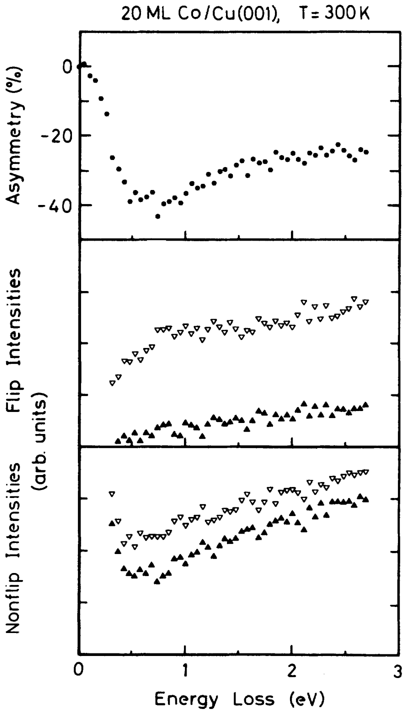

FIG. 2. Intensity asymmetry (upper panel), flip intensities (middle panel: ▲, $F^{\uparrow}$; ▽, $F^{\downarrow}$), and nonflip intensities (lower panel: ▲, $N^{\uparrow}$; ▽, $N^{\downarrow}$) for a 20 ML fcc Co/Cu(001) film at $T=300$ K (▲, incident spin-up electrons; ▽, incident spin-down electrons).

A second reason for the observed flip-up intensities could be the reduction of the magnetization by transverse spin fluctuations at a finite temperature, as suggested by Abraham and Hopster.[4] Because of the high Curie temperature of fcc Co this effect should be only of minor importance in the present case.

Experimentally, the $F^{\downarrow}/F^{\uparrow}$ ratios at a loss energy corresponding to the exchange splitting are not too different for Fe, Ni, and fcc Co. Whereas $F^{\downarrow}/F^{\uparrow}$ amounts to 4.4 for Fe,[2,3] the data for Ni (Ref. 4) and for fcc Co (Fig. 2) give a value of about 5.5 for $F^{\downarrow}/F^{\uparrow}$ at $E_{\text{loss}}\simeq\Delta_{\text{ex}}$. These relatively small differences indicate that Stoner excitations involving states other than the spin-split d states cannot be neglected. The apparent differences in the form of the flip-down spectrum (i.e., maximum for Ni and broad shoulder for Fe and fcc Co) may be due to the background of multiply scattered electrons. The "single-energy-loss-event" spectra are superimposed on this "secondary-electron" background, whose intensity increases with decreasing kinetic energy of the secondary electrons. For small energy losses this background is not significant. However, for larger energy losses it might no longer be ignored. For even larger loss energies, where the true secondary background starts to be the dominant contribution, care must be taken in interpreting the spectra. In this regime the whole concept of flip and nonflip scattering starts to break down: In the extreme case of only secondary electrons the measured polarization is independent of the incident electron polarization and equal to the secondary electron polarization (which, except for very low kinetic energies, approximately equals the average conduction band polarization). The usual analysis of the measured polarization in terms of flip and nonflip scattering would thus pretend larger flip-down and non-flip-up rates and smaller flip-up and non-flip-down rates ($N^{\uparrow}=F^{\downarrow}>N^{\downarrow}=F^{\uparrow}$). Experimentally one could try to suppress this adulterating background by increasing the energy of the incident electrons, i.e., by shifting the energy loss spectrum to higher kinetic energies. However, due to the strong energy dependence of exchange scattering, this would also diminish the exchange effects to be observed. This strongly limits the primary energies for which Stoner excitations can be clearly observed in a spin-polarized electron energy-loss spectrum. In addition to the multiply scattered electrons dipolar scattering also gains spectral weight with increasing loss energy, leading to an increase of the nonflip rates. Inelastic dipolar scattering is largely confined to a lobe (of angular width $\Theta=E_{\mathrm{loss}}/2E_P$) along the specular direction.[38] Experimentally, this results in a relative suppression of the exchange contribution. Once the dipolar lobe is outside the spectrometer detection cone, we do not observe any significant changes of the SPEELS spectra upon further change of the scattering angle. This is in accordance with the previous SPEELS investigations on Ni(110) (Ref. 4) and Fe(001).[2]

Assuming momentum conservation to be valid in the scattering process and *d-d* Stoner excitations to be the dominating contribution to the flip rates, the angle-resolved (and thus momentum-resolved) flip rate spectra should mirror the wave-vector-dependent Stoner density of states. Thus one would expect to see strongly angle-dependent structures in the SPEELS spectra. The previous SPEELS experiments on Ni and Fe, as well as the present study on fcc Co have, however, only revealed quite broad features without a significant angle dependence. One possible reason for this behavior might be nonconservation of the momentum component $q_{\perp}$ perpendicular to the surface in the scattering process.[4] This would cause the SPEELS spectrum to reflect momentum integrated Stoner density of states.

Qualitatively similar to the results for Ni and Fe is also the energy dependence of the asymmetry A. In the elastic regime one observes only very small and slightly positive values for A. With increasing energy loss the asymmetry changes sign and begins to fall rapidly, reaching a maximum of approximately -40% at $E_{\mathrm{loss}}\simeq 0.8$ eV. This pronounced negative asymmetry is mainly due to the difference between the $F^{\downarrow}$ and the $F^{\uparrow}$ rate. However, the difference between the two nonflip rates also contributes about $\frac{1}{4}$ of the asymmetry. For $E_{\mathrm{loss}}\geq 0.8$ eV the absolute values of the asymmetry slowly decreases with increasing energy loss.

Thus the maximum asymmetry ($\simeq -40\%$) found for fcc Co is considerably larger than the maximum values of A found for Fe [-30% (Ref. 2) and -25% (Ref. 3)], but smaller than the maximum asymmetry of -50% observed for Ni.[4] The difference between Ni and Fe was attributed to the different position in energy of the Fermi level relative to the spin-split d bands (saturated versus unsaturated ferromagnet), as discussed above. The intermediate value for $A_{\max}$ observed for fcc Co directly relates to the question whether fcc Co is a saturated or unsaturated ferromagnet. Band-structure calculations[35,39,40] predict fcc Co to be a saturated ferromagnet with an exchange splitting of approximately 1.8 eV. The predicted Stoner gap δ is on the order of $\delta\simeq 0.3$ eV. The calculated exchange splitting seems to be almost independent of the assumed crystal structure (fcc or bcc) and is distinctly larger than the experimental values, determined by means of angle-resolved photoemission spectroscopy. For hcp Co, Δ_{ex} was found to be different for states of different symmetry: $\Delta_{\mathrm{ex}}(e_g$ symmetry$)\simeq 0.85$ eV, $\Delta_{\mathrm{ex}}(t_{2g}$ symmetry$)\simeq 1.2$ eV;[41] for fcc Co, Δ_{ex} has been measured only for Δ_5 states with t_{2g} symmetry, where a value of $\Delta_{\mathrm{ex}}\simeq 1.2$ eV± 0.2 eV was found.[32,33,35] Thus the observed exchange splittings are also independent of the crystal structure. Since the SPEELS experiment seems to integrate over at least parts of the Brillouin zone, the symmetry-dependent distribution of exchange splittings might be partly responsible for the large width of the structures observed in the SPEELS spectra.

The existence of a Stoner gap δ should show up directly in the SPEELS data in the form of a definite threshold for spin-down Stoner excitations. The statistical uncertainties, however, make it difficult to extract a value for δ directly from the partial rates in Fig. 2. The threshold should also show up in the asymmetry as well as in the spin polarization of the scattered electrons P_S/P_0 [called depolarization in the following: for the calculation of P_S/P_0 the polarization of the scattered electrons P_S was taken to be $\frac{1}{2}$ of the difference between the polarization of the scattered electrons in the two cases of the polarization of the incident electrons parallel and antiparallel to the magnetization $P_S=\frac{1}{2}(P_S^{\uparrow}-P_S^{\downarrow})$]. Figure 3 shows those two independent quantities for small energy losses, together with the total scattering intensity. The asymmetry as well as the depolarization start to rise rapidly at $E_{\mathrm{loss}}\simeq 0.27$ eV. This distinct increase of A and P_S/P_0 is not solely due to the falloff of the elastic intensity. At $E_{\mathrm{loss}}\simeq 0.27$ eV the quasielastic intensity and the "true" inelastic scattering intensity (as judged from the "background" on which the elastic peak is superimposed) have about equal magnitude. If the increase of A and P_S/P_0 were due to the falloff of the elastic intensity one would expect A to have already reached 50% of its maximum value $A_{\max}$ at this loss energy, whereas the depolariza-

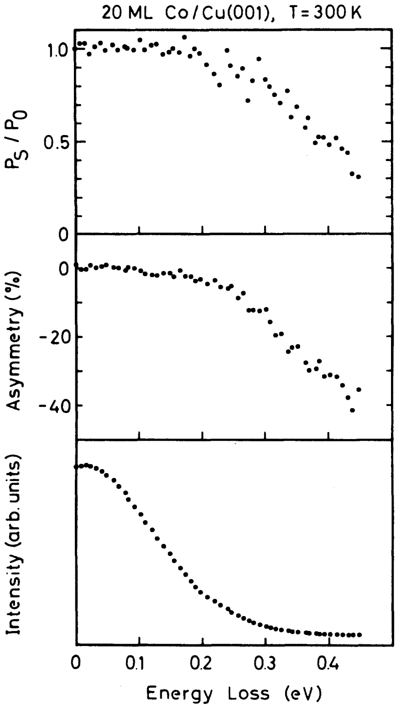

FIG. 3. Spin polarization of the scattered electrons normalized to the primary beam polarization (upper panel), intensity asymmetry (middle panel), and total scattering intensity (lower panel) for (20 ML Co)/Cu(001) at $T=300$ K for small energy losses.

tion should have reached a value of $\frac{1}{2}(P_S/P_{0,\max}+1)$ at this loss energy, since the polarization of the elastically scattered electrons is equal to the polarization P_0 of the incident electrons. Those values for A and P_S/P_0 are, however, only reached for somewhat higher loss energies. This indicates the existence of a threshold at a loss energy of $E_{\text{loss}} \simeq 0.27$ eV. A value for the Stoner gap δ of fcc Co of about 300 meV is thus consistent with the present SPEELS data and agrees well with spin-resolved photoemission data on fcc Co.[33,35]

IV. SPEELS ON ULTRATHIN Co/Cu(001) FILMS

The thickness dependence of the Curie temperature of ultrathin Co/Cu(001) films is subject to a still continuing controversy: While Pescia and co-workers[19,23,26,27,42] find ferromagnetism at room temperature for the very monolayer Co/Cu(001) (grown at room temperature), Schneider *et al.* observe ferromagnetic behavior at room temperature only for films with thicknesses of more than approximately 2 monolayers (both for films grown at 450 K and for room temperature grown films).[18] In contrast, a very recent investigation by Mankey, Kief, and Willis[22] reports the Curie temperature to depend on the growth temperature: For growth at 150 and 300 K films thicker than 1.5 ML are ferromagnetic at room temperature, whereas for growth at 450 K T_C was found to be considerably lower. The difference was attributed to a segregated Cu overlayer on the surface of Co/Cu(001) films grown at 450 K. Schneider *et al.* did not, however, observe Cu surface segregation for growth at 450 K.

Remanent magnetization shows up directly in the SPEELS data as a nonvanishing asymmetry. In our experiments Co/Cu(001) films with a thickness of 2 ML or less could not be remanently magnetized at room temperature. Upon cooling to 80 K (the lowest temperature accessible in our experiment) a film with a thickness $d=1.6$ ML did show a clearly nonvanishing asymmetry, while the monolayer could still not be magnetized remanently. We thus could perform the complete experiment down to a thickness of 2 ML. The SPEELS spectra of films with a thickness of 4 and more monolayers were recorded at room temperature, while the data on thinner films (i.e., $d<4$ ML) were taken at 80 K. For our room-temperature-grown Co/Cu(001) films and the thickness calibration as described above, the thickness dependence of the Curie temperature agrees with the T_C versus thickness relation of Schneider *et al.*[18] According to this relation the measuring temperature used by us is in all cases sufficiently below the Curie temperature ($T/T_C \leq 0.3$).

Not too surprisingly, the SPEELS data of ultrathin Co/Cu(001) films consisting of four or more atomic layers are almost identical to the SPEELS spectra taken on the thicker films ($d \simeq 20$ ML), described in the preceding paragraph. This behavior agrees well with the results of recent spin-resolved photoemission measurements,[32,33,35] which found the electronic structure of 5-ML-thick Co/Cu(001) films to be already bulklike. Furthermore, these authors claim that even 2-ML films show essentially the same spin-polarization spectra of the photoemitted electrons. On the basis of the current understanding and interpretation of SPEELS spectra in terms of *d-d* Stoner excitations one would thus expect similar SPEELS spectra even for (2 ML Co)/Cu(001).

Such data for a 2-ML film are shown in Fig. 4. They indeed look very similar to the data of Fig. 2. The spectra of the partial rates and the asymmetry display almost the same shape as the corresponding bulk spectra. In particular, we do not observe any significant spectral shifts, e.g., in the flip-down channel or in the asymmetry. At most, the shoulder in the flip-down scattering rate and the maximum of the asymmetry both exhibit a barely visible shift towards lower loss energies upon comparison to the 20-ML data. This implies an essentially unchanged (or possibly even somewhat smaller) exchange splitting. Thus (2 ML Co)/Cu(001) do not show any sign of enhanced magnetic moments. Looking more closely at the asymmetry and the depolarization P_S/P_0 in the vicinity of the elastic peak, the spectra do not reveal any change of the threshold loss energy for the flip-down rate, i.e., a Co/Cu(001) film consisting of 2 monolayers is a saturated ferromagnet with the same Stoner gap $\delta \simeq 0.3$ eV as bulk fcc Co.

For films consisting of less than 4 monolayers the absolute magnitude of the asymmetry, however, decreases with decreasing thickness. The asymmetry measured for a 2-ML-thick film (see Fig. 4) is reduced by about a factor

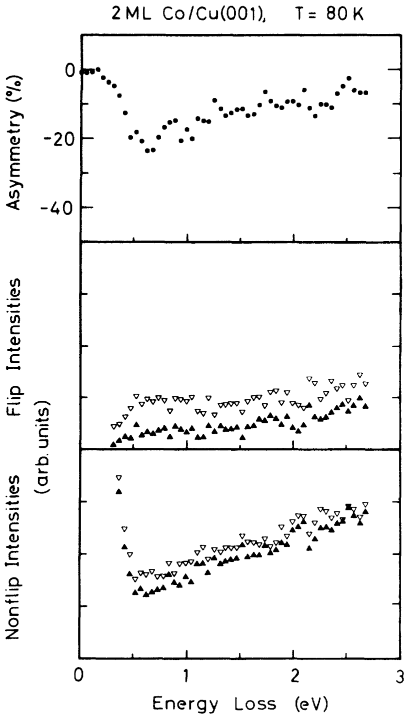

FIG. 4. Intensity asymmetry (upper panel), flip intensities (middle panel: ▲, $F^{\uparrow}$; ▽, $F^{\downarrow}$), and nonflip intensities (lower panel: ▲, $N^{\uparrow}$; ▽, $N^{\downarrow}$) for a 2 ML fcc Co/Cu(001) film at $T=80$ K (▲, incident spin-up electrons; ▽, incident spin-down electrons).

of 2 compared to the bulk data. Inspection of the partial scattering rates reveals that a major cause of this asymmetry reduction is an assimilation of the flip-up and the flip-down rate. In addition one observes a relative increase of the nonflip scattering rates with decreasing thickness, effectively reducing the asymmetry too. However, since the change of the ratio F/N $(F=F^{\uparrow}+F^{\downarrow}, N=N^{\uparrow}+N^{\downarrow})$ is only quite moderate ["bulk" fcc Co, $F/N \simeq \frac{2}{3}$; (2 ML Co)/Cu(001), $F/N \simeq \frac{1}{2}$], the enhanced nonflip scattering accounts only for a small part of the total reduction of the asymmetry.

One could think of several possible reasons leading to a reduction of the asymmetry with decreasing thickness. The assimilation of the flip rates, i.e., the observed increased flip-up scattering, could, of course, be due to an effectively reduced macroscopic magnetization. The reduction of the Curie temperature of ultrathin ferromagnetic films with decreasing thickness leads to a reduction of the magnetization at a given temperature. However, since we used different measuring temperatures (300 K for $d \geq 4$ ML, 80 K for $d \leq 4$ ML) the reduced temperature T/T_C [on the basis of the $T_C(d)$ relation from Ref. 18] is approximately the same for the 20-ML film (Fig. 2) and the 2-ML film (Fig. 4) and yet the $F^{\uparrow}/F^{\downarrow}$ ratio is quite different. This is even more surprising, since the experimentally determined temperature dependence of the magnetization $M(T)$ of Co/Cu(001) in the monolayer regime exhibits a surprisingly small reduction of the magnetization up to at least 400 K.[19,26] Thus an explanation of the observed amount of flip-up scattering in terms of a reduced magnetization does not seem very plausible in the case of the (2 ML Co)/Cu(001) films, as well as for the bulklike Co films.

A process that reduces the macroscopic magnetization would be the existence of magnetic domains. Investigations of the domain structure in ultrathin Co/Cu(001) films using electron microscopy with spin-polarization analysis have, however, shown that the films are single domain.[43,44] A breakup of the magnetization into several large domains could only be achieved by means of an ac-demagnetizing field. On the basis of these results it does not seem very likely that the reduction of the asymmetry is caused by domain formation in the ultrathin Co/Cu(001) films.

A possible reason for the decrease of the asymmetry due to increased nonflip scattering might be an enhanced roughness of films with a thickness below 4 ML. Such an increase of the surface roughness would lead to a broadening of the dipole scattering lobe and thus to a relative increase of the nonflip scattering intensity in off-specular geometry. A possible increase of the surface roughness for very thin films is corroborated by the investigations of Li and Tonner,[20] who found the first two monolayers to grow in 2-ML-thick islands.

Also scattering contributions from the Co/Cu(001) substrate, increasing with decreasing Co thickness, could possibly contribute to the observed decrease of the asymmetry. The SPEELS spectrum of the clean Cu(001) surface (see Fig. 6), however, reveals only a very small scattering intensity for energy losses less than 2 eV.

A much more interesting explanation would be a change of the electronic structure with decreasing thickness. Previous photoemission studies of epitaxial Fe and Ni films on metal substrates found the three-dimensional d-band structure to develop in the thickness range from 2 to 4 ML,[28,29,45] exactly in the thickness range of interest in the present case.

The most obvious cause for an assimilation of the flip-up and the flip-down rate would be the appearance of (unoccupied) majority-spin states above the Fermi level. However, this reason can be excluded, since the SPEELS spectra for small energy losses below 0.5 eV are very similar to the "bulk" data shown in Fig. 3 and in particular do not reveal any change of the threshold energy δ with decreasing thickness.

Spin-resolved photoemission experiments have shown a very weak dependence of the electronic structure on thickness attributed to the tetragonal distortion of the Co/Cu(001) layers.[33,35] This slight tetragonal compression of the Co layers perpendicular to the surface[17] can be expected to become less significant with increasing thickness.[35] The spin-resolved photoemission spectra reveal a small shift of the minority-spin states towards lower binding energies with decreasing thickness. The influence of such a shift on the SPEELS spectrum would mainly consist in a reduction of the amount of spin-flip scattering within the d bands, whereas the nonflip scattering rate would be much less affected. The observed small shift of the minority-spin states could thus be partly responsible for the small relative increase of the nonflip scattering intensity compared to the flip scattering rate.

Another implication of this small shift in energy of the minority-spin *d* states concerns the relative importance of *d*-electron Stoner excitations and so-called free-electron-like Stoner excitations, i.e., electron-hole pairs consisting of a *d* hole of given spin and an electron in a free-electron-like state of opposite spin. The concept of such free-electron-like Stoner excitations was introduced by Penn and Apell.[36,37] In their theoretical analysis of the experimental SPEELS data of Fe (Refs. 1 and 2) they found free-electron-like Stoner excitations to be of similar importance as *d*-electron Stoner excitations despite the much larger density of unoccupied *d* states compared to the density of empty *sp* states. The comparison of the SPEELS data of Fe, Ni, and the bulklike fcc Co/Cu(001) films in the preceding paragraph also suggests that spin-flip scattering involving *sp* states substantially contributes to the SPEELS spectra. The shift of the minority-spin states towards lower binding energies would certainly not lead to a reduction of the partial scattering intensity corresponding to free-electron-like Stoner excitations. More likely, the amount of spin-flip scattering between *d* states and *sp* states would increase due to the increased occupation of the *d* band. Thus the large amount of flip-up scattering observed for (2 ML Co)/Cu(001) might be another indication for the importance of free-electron-like Stoner excitations in SPEELS.

Due to their delocalized nature free-electron-like states are expected to show a stronger thickness dependence than the more localized *d* states. Free-electron-like states should be, in particular, more subject to the quantization of the wave-vector component $k_\perp$ perpendicular to the film plane. This quantization of $k_\perp$ should induce additional structure in the *sp*-like part of the density of states. This might possibly involve an increase of the number of empty *sp*-like states, leading to enhanced scattering cross section for free-electron-like Stoner excitations. However, without the backing of a detailed theoretical calculation, this explanation of the enhanced flip-up scattering in the ultrathin Co/Cu(001) films remains rather speculative.

It would be very interesting to follow the development of the partial scattering rates down to even smaller thicknesses, like, e.g., the very monolayer Co/Cu(001). Upon reaching this thickness range of about 1 ML (or less) even the band structure of the quite localized *d* electrons should be fully two dimensional, i.e., the binding energies of the *d* states should depend only on the wave-vector component parallel to the surface $k_\parallel$. The angle-resolved SPEELS experiment possibly integrates over $k_\perp$. The wave-vector component $k_\parallel$, however, is conserved in the scattering process. Thus one would expect to observe a sharpening of the structures related to *d*-*d* Stoner excitations in the SPEELS spectra.

The low Curie temperatures of Co/Cu(001) films in the monolayer regime prevent a SPEELS experiment with a remanently magnetized sample, which would reveal the full information on the four partial scattering rates. A special virtue of the complete SPEELS experiment is, however, its ability to differentiate between spin-flip and nonflip scattering even in the case of an unmagnetized sample, such as an antiferromagnet[13] or a nonmagnetic material like Mo.[12]

The measured depolarization P_S/P_0 of the scattered electrons directly translates into a separation of the total scattering intensity I into the flip (F) and nonflip (N) contributions:

$$I=N+F,\quad F=\tfrac{1}{2}I(1-P_S/P_0),\quad N=\tfrac{1}{2}I(1+P_S/P_0)\,.$$

Figure 5 shows the depolarization P_S/P_0 and the partial scattering rates N and F for Co/Cu(001) films consisting of 20, 2, and 0.9 monolayers Co. In the case of the 20 ML of Co film the depolarization quickly drops for loss energies larger than the threshold energy $\delta \simeq 0.3$ eV and reaches a very shallow minimum at a loss energy of $E_{\text{loss}} \simeq 0.8$ eV, corresponding to the average exchange splitting. For larger loss energies P_S/P_0 is approximately constant with a value of $P_S/P_0 \simeq 0.3$. This shallow minimum in P_S/P_0 translates in a broad shoulder in the spin-flip scattering rate F.

For 2 ML of Co the picture is essentially the same. P_S/P_0 quickly drops with increasing energy loss until it reaches a shallow minimum at $E_{\text{loss}} \simeq 0.8$ eV. However, the plateau of the depolarization for loss energies larger than 0.8 eV is at a value of $P_S/P_0 \simeq 0.5$ and thus considerably larger than for the 20 ML film. This reduced depolarization reflects the already discussed relative increase of the nonflip scattering intensity (possibly due to increased surface roughness) for ultrathin films. The form of the partial rates spectra remains, however, essen-

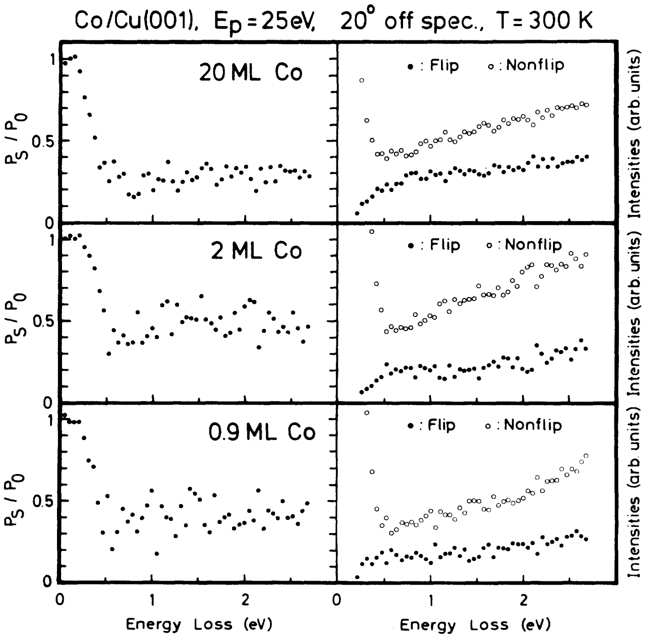

FIG. 5. Depolarization P_S/P_0 (left column) and total flip and nonflip scattering intensities (right column) for Co/Cu(001) films consisting of 20 ML Co (upper part), 2 ML Co (middle part), and 0.9 ML Co (lower part) at $T = 300$ K.

tially unchanged.

This is still valid in the case of the (0.9 ML Co)/Cu(001) film. Again, we observe the rapid decrease of the depolarization and a plateau for loss energies larger than $E_{\rm loss} \simeq 0.8$ eV. The depolarization in the plateau is even somewhat more effective for (0.9 ML Co)/Cu(001) ($P_S/P_0 \simeq 0.4$) than for (2 ML Co)/Cu(001). The shape of the flip and nonflip intensity spectra is still very similar to the 20 ML spectra. In particular, we do not observe any significant spectra shifts or sharper structures in the SPEELS spectra (except for the possible shift of the shoulder in the flip rate towards smaller loss energies, already mentioned above for the case of the remanently magnetized 2 ML film).

This observation could lead to different conclusions. If the experimentally observed spin-flip scattering is due to a significant amount to *d-d* Stoner excitations, the exchange splitting of the monolayer Co/Cu(001) is not enhanced over the bulk value. The broadness of the corresponding shoulder in the flip rate is, however, even more puzzling than for the SPEELS data of bulk samples. The essentially unchanged form of the energy-loss spectra and the absence of any sharpening of the spectral features might be taken as an indication to question the present usual interpretation of the SPEELS data of *d*-band metals in terms of a dominating *d-d* scattering contribution to the total scattering intensity. Instead, the *sp-d* scattering contribution might be higher than previously assumed. Such scattering between free-electron-like states and *d* states is consistent with almost structureless spectra and might wash out sharper structures due to *d-d* scattering.

For the interpretation of SPEELS data from epitaxial films as thin as 1 ML, it is of considerable importance to be able to clearly differentiate between the scattering signal from the epitaxial film and any additional contribution due to scattering from the substrate. The inelastic mean free path λ of electrons with a kinetic energy of 25 eV in a *d*-band metal like Co is approximately 2.5 ML.[7] Since the total scattering angle in our experiment is fixed at 90°, one would expect to see an appreciable substrate contribution to the total scattering intensity only for films thinner than approximately $\lambda/2\sqrt{2}$, i.e., for films of 1 ML of Co or less.

Such behavior is demonstrated in Fig. 6, which shows the depolarization and the partial scattering rates for (20, 2.2, and 0.5 ML Co)/Cu(001), together with the data for the clean Cu(001) surface. Note that the energy loss scales in Fig. 6 extends to $E_{\rm loss} = 8.5$ eV, instead of 3 eV as in the preceding figures. Most surprisingly, spin-polarized electrons scattered from the clean Cu(001) surface are quite effectively depolarized for larger energy losses. This corresponds to a high amount of spin-flip scattering for loss energies of more than 2 eV. For small energy losses up to $E_{\rm loss} \simeq 2$ eV one observes almost pure nonflip scattering. At a loss energy of $E_{\rm loss} \simeq 2$ eV both partial rates, the nonflip as well as the spin-flip scattering rate start to rapidly increase with increasing energy loss. The threshold energy of $E_{\rm loss} \simeq 2$ eV clearly reflects the *d*-band structure of Cu. Electron-hole excitations involving occupied *d* states in Cu require a minimum energy of 2 eV. The experimentally observed spin-flip scattering from the clean Cu(001) surface for $E_{\rm loss} \geq 2$ eV can only be interpreted in terms of free-electron-like Stoner excitations between *d* states and unoccupied free-electron-like states. A previous SPEELS study on Cu(001) (Ref. 12) investigated only the scattering rates for the specular geometry, for which the authors did not find any appreciable amount of spin-flip scattering. It is, however, well known from previous SPEELS studies on, e.g., Ni,[5] that exchange effects are swamped in the specular direction by direct scattering. The quite surprising amount of *sp-d* exchange scattering on Cu(001) observed in the present study unequivocally demonstrates the importance of free-electron-Stoner excitations for the general interpretation of SPEELS data from *d*-band metals.

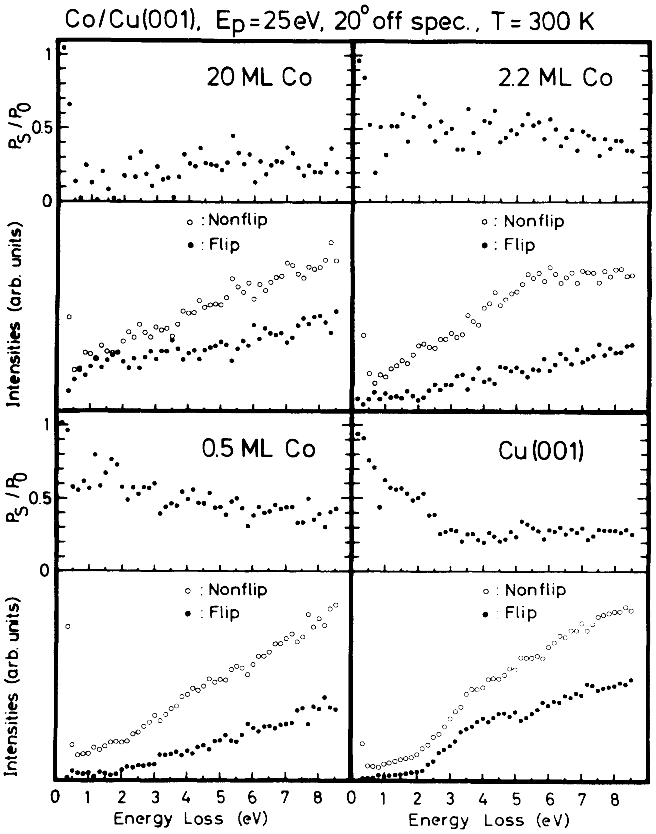

FIG. 6. Depolarization P_S/P_0 and total flip and nonflip scattering intensities for Co/Cu(001) films consisting of 20 ML Co, 2.2 ML Co, 0.5 ML Co and for the clean Cu(001) substrate at $T = 300$ K.

The data for the (0.5 ML Co)/Cu(001) film clearly exhibit a mixture of Co and Cu features. In the low-energy-loss range ($E_{\rm loss} < 2$ eV), where the scattering intensity from the Cu(001) substrate is quite small, the SPEELS data from the 0.5 ML film reflects the same features observed for the thicker Co/Cu(001) films, i.e., the rapid drop of P_S/P_0, followed by a plateaulike behavior. For larger energy losses, for which the clean Cu(001) surfaces exhibits a substantial scattering cross section, the spectra of the 0.5 ML film clearly reveal the substrate contribution in the form of a further decrease of P_S/P_0

towards larger energy losses due to the strong spin-flip scattering in the Cu(001) substrate and the clearly visible 2-eV onset in flip rate.

The SPEELS data for ultrathin Co/Cu(001) films in the monolayer and submonolayer regime give strong evidence for the existence of a local spin-split band structure, even in the absence of long-range ferromagnetic order. The (locally defined) exchange splitting in such paramagnetic films is similar to the (macroscopically defined) exchange splitting in the ferromagnetically ordered 20 ML fcc Co/Cu(001) films. We thus find no evidence for enhanced magnetic moments in the monolayer films.

The similar shape of the SPEELS spectra from films in the ferromagnetically ordered state and from paramagnetic films has implications for the general understanding of the ferromagnetism in the 3*d* transition metals: The results support the so-called local-band theory,[46,47] which predicts the existence of a locally defined spin-split band structure with short-range spin order in 3*d* ferromagnets above the Curie temperature.

V. SPEELS ON CoO

The present SPEELS results from bulklike and ultrathin Co/Cu(001) films and from the clean Cu(001) surface suggests that the generally observed broadness of the energy loss features in the SPEELS spectra from metallic systems is due to a surprisingly strong "background" of electron-hole excitations between occupied *d* states and empty *s*-*p* states, superimposed on the supposedly much sharper spectrum of the Stoner excitations between the occupied and empty parts of the *d* bands. Upon oxidation of the transition metals the *s*-*p* states are shifted to much higher energy and no longer contribute to the SPEELS spectra. A previous SPEELS investigation of the transition metal oxide Cr_2O_3 exhibited sharp loss features.[13] The SPEELS data of the ionic, large band-gap insulator Cr_2O_3 could be interpreted in terms of excitations from the spin-quartet ground state to excited doublet states of the Cr^{3+} ion via exchange scattering.

The antiferromagnet CoO ($T_{\mathrm{Neel}}=291$ K) belongs to the interesting group of the transition-metal monoxides (MnO, FeO, CoO, and NiO), which are traditionally thought to be typical examples of Mott-Hubbard insulators.[48,49] Thus the insulating nature of the transition-metal monoxides is explained by a localization of the 3*d* electrons due to a large intra-atomic Coulomb interaction. This view has been challenged, however, by attempts to describe the electronic structure of the transition metal monoxides by means of an itinerant band-structure calculation.[50,51] A more recent theory stresses the importance of excitations involving a hole in the oxygen 2*p* band for the size of the conductivity gap and the nature of the valence and conduction electron states in such compounds.[52] This controversy regarding the electronic structure of the transition metal monoxides provides additional motivation for the investigation of CoO by SPEELS.

Exposing a clean Co surface to oxygen at 300 K or higher temperature is known to lead to the formation of a CoO layer on the surface.[53,54] By heating a thick (30 ML) film of Co/Cu(001) to 300° in 10^{-6} Torr oxygen for 10 min, we prepared a thick, disordered layer (no LEED pattern discernible) of CoO.

On this CoO surface SPEELS was performed under the same conditions as in the case of the clean Co/Cu(001) films, i.e., using a primary energy of $E_p=25$ eV and taking the spectrum in off-specular geometry (20°). We note that due to the disordered structure of the CoO layer the SPEELS spectra recorded along the specular direction are very similar to the off-normal spectra. Figure 7 shows the normalized spin polarization P_S/P_0 of the scattered electrons and the flip (F) and nonflip (N) scattering rates for CoO at $T=300$ K, i.e., slightly above the Neel temperature of 291 K. However, no difference in the SPEELS spectra could be detected upon cooling the sample to 80 K.

The SPEELS data from CoO exhibit a variety of well-defined structures, similar to the case of Cr_2O_3, but in sharp contrast to the SPEELS data from the metallic Co/Cu(001) films. The most prominent feature in the flip spectrum is a strong and sharp peak at a loss energy of $E_{\mathrm{loss}}\simeq 2$ eV, which is replicated with smaller intensity in the nonflip channel. A second, equally well-defined, energy-loss peak appears at $E_{\mathrm{loss}}\simeq 0.9$ eV in the nonflip channel, while the flip rate exhibits only a much weaker and broader structure in this energy range. Additional structure in the flip spectrum consists mainly of a weak peak at $E_{\mathrm{loss}}\simeq 3$ eV, whereas the nonflip scattering intensity increases smoothly (within the statistics) for $E_{\mathrm{loss}}\geq 3$ eV.

The CoO SPEELS data in Fig. 7 are compatible with a description in terms of the ligand-field theory,[55] i.e., the energy-loss structures in the SPEELS spectrum correspond to atomiclike excitations of the Co^{2+} ion. Ligand-field theory has already been successfully used to account for the optical absorption spectrum for CoO.[56] In contrast to optical spectroscopy SPEELS directly distinguishes excitations between states of the same spin multi-

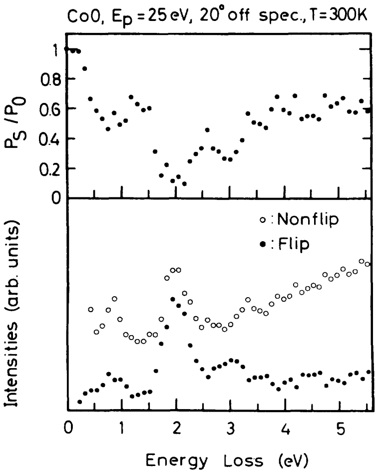

FIG. 7. Depolarization P_S/P_0 and total flip and nonflip scattering intensities for disordered CoO.

plicity from excitations between states of different spin multiplicity. Excitations between states of the same multiplicity do not change the spin polarization and thus appear only in the nonflip spectrum. In contrast, excitations between terms of different spin multiplicity are mainly by exchange scattering (since in this case direct scattering proceeds only via spin-orbit coupling) and lead to a depolarization of the scattered electrons. Since all possible transitions between the different magnetic sublevels contribute to the energy-loss spectrum, such excitations appear in the flip ($\Delta m_s = \pm 1$) and the nonflip channel ($\Delta m_s = 0$).

Thus a comparison with the ligand-field theory energy-level diagrams[55] allows for a straightforward interpretation. The prominent peak at $E_{\text{loss}} \simeq 2$ eV corresponds to an excitation from the Hund's-rule spin quartet T_{1g}^4 ground state to the spin doublet excited states T_{1g}^2 and T_{2g}^2. The structure in the nonflip spectrum at $E_{\text{loss}} \simeq 0.9$ eV is due to an excitation from the ground state to the lowest lying excited state T_{2g}^4 with the same spin multiplicity. The weak structure in the flip channel at $E_{\text{loss}} \simeq 3$ eV might be attributed to an excitation into the higher lying doublet state A_{1g}^2.

Those excitations energies and their assignment agree well with optical absorption data and their interpretation in terms of ligand-field excitations.[56] Due to the spin-selection rule of optical spectroscopy the transitions from quartet to doublet states appear only as very weak shoulders in the optical absorption spectrum, which is dominated by the quartet-quartet excitations. In SPEELS (using a low primary energy and off-specular geometry), however, exchange scattering contributes strongly to the total scattering intensity. Thus the quartet-doublet excitations appear with similar spectral intensity as the quartet-quartet transitions in the SPEELS spectrum.

The importance of the exchange scattering cross section has been somehow underestimated in the interpretation of a recent unpolarized electron-energy-loss spectroscopy study of the transition-metal oxides,[57] since only quartet excited states have been used in the labeling of the energy-loss features in the SPEELS spectrum for CoO. This spectrum, however, taken with the same primary energy $E_p = 25$ eV, but along the specular direction and with a much better energy resolution (75 meV), is very similar to the present SPEELS spectrum. The authors of Ref. 57 compare their SPEELS data with spectra calculated directly via dielectric theory (i.e., only considering dipole scattering) from the optical data. The resulting spectra agree rather well with our SPEELS data, but are much too low in intensity. The authors suggest a short-range scattering mechanism, such as impact scattering, as the most probable explanation for the discrepancy. The results of the present complete SPEELS experiment on CoO give a most direct proof of this hypothesis.

The discussion of the SPEELS data from CoO has been based upon the assumption of an ionic electronic structure, i.e., the 3d electrons in CoO were considered to be well localized. Attempts to describe the electronic structure of the transition-metal oxides by means of band theory have, however, not been successful in accounting for the insulating property of CoO.[50,51] The authors suggest that this discrepancy is due to a failure of the local-spin-density-functional approximation due to the unquenched orbital angular momentum in CoO rather than a failure of band theory as applied to the transition-metal oxides. Although the interpretation of the SPEELS spectra from CoO in the framework of ligand-field theory works rather well, the data are not incompatible with the spin-resolved density of states calculated by band theory.[51] This calculated density of states exhibits quite narrow t_{2g} and e_g bands, separated by a ligand-field splitting of $\simeq 1.1$ eV, while majority-spin and minority-spin states are shifted against each other by an exchange splitting of $\simeq 2.3$ eV. This splitting compares quite well with the energies of the major nonflip and flip excitations in the SPEELS spectrum. However, the sharpness of the structures in the SPEELS spectra from CoO, as opposed to the itinerant ferromagnets, points to a high degree of localization of the d electrons in CoO, i.e., to a large (essentially unscreened) intra-atomic Coulomb interaction.

VI. CONCLUSIONS AND SUMMARY

SPEELS has been used to study the electron-hole excitations in epitaxial Co/Cu(001) films. The SPEELS data from thick, bulklike Co/Cu(001) films ($d = 20$ ML) are qualitatively very similar to the results obtained in previous SPEELS studies on Fe and Ni surfaces. In the off-specular scattering geometry exchange scattering contributes a substantial amount to the total scattering intensity. This is especially manifest in the flip-down scattering rate $F^{\downarrow}$, which represents about $\frac{1}{3}$ of the total scattering intensity for loss energies between 0.5 and 1.5 eV. Spin-flip scattering is responsible for the major part of the observed intensity asymmetry. This asymmetry exhibits a maximum value of $\simeq -40\%$ at $E_{\text{loss}} \simeq 0.8$ eV. This loss energy coincides with the broad shoulder in the flip-down scattering rate and represents the average exchange splitting in the Co film. The SPEELS data suggest the existence of a threshold at $E_{\text{loss}} \simeq 0.3$ eV. The experimental data thus support the theoretical prediction of fcc Co being a saturated ferromagnet and are consistent with a Stoner gap of $\delta \simeq 300$ meV. The comparison of the $F^{\downarrow}/F^{\uparrow}$ ratios observed in SPEELS experiments on Fe, Co, and Ni indicates that the contribution of free-electron-like (sp-d) Stoner excitations cannot be neglected in the interpretation of SPEELS spectra from 3d transition metals.

At 80 K ultrathin Co/Cu(001) films can be remanently magnetized down to a thickness of 1.6 ML. The SPEELS spectra of (2 ML Co)/Cu(001) are very similar to the data from (20 ML Co)/Cu(001). The partial scattering rates and the intensity asymmetry exhibit almost the same shape as the corresponding 20 ML spectra. They give a strong indication for an essentially thickness-independent exchange splitting, since we do not observe any significant spectral shifts. The threshold energy, representing the Stoner gap, remains unchanged as well. Thus the spin-split electronic structure of (2 ML Co)/Cu(001) is essentially bulklike. However, the intensity asymmetry decreases for ultrathin Co/Cu(001) of less than 4 ML. This reduction is mainly due to an assimilation of the flip-down and the flip-up scattering rate with decreasing thickness below 4 ML.

Similar conclusions are also valid in the case of Co/Cu(001) films in the monolayer regime, which could not be remanently magnetized. SPEELS data taken on such paramagnetic ultrathin Co/Cu(001) films reveal a large amount of depolarization. The spectral shape of the depolarization and of the total flip and nonflip scattering rates are very similar to the corresponding 20 ML spectra. We do not observe any significant spectral shift towards higher loss energies or sharper structures in the SPEELS spectra. We conclude on the existence of a local spin-split band structure, which is similar to the spin-split band structure of bulk fcc Co, even in the absence of long-range ferromagnetic order. In particular, we find no evidence for enhanced magnetic moments in such monolayer and submonolayer films.

The off-specular SPEELS spectra from the clean Cu(001) surface display a surprising amount of spin-flip scattering for loss energies larger than 2 eV. The onset at 2 eV clearly reflects the binding energy of the Cu 3*d* electrons. The spin-flip electron-hole excitations, observed for $E_{\text{loss}} \geq 2$ eV, are unequivocally due to free-electron-like Stoner excitations between the 3*d* states and *sp*-like states above the Fermi level. This clearly demonstrates the importance of free-electron-like Stoner excitations for the general interpretation of SPEELS spectra from transition metals.

In contrast to the quite broad features observed in the SPEELS spectra of the metallic Co/Cu(001) films, the SPEELS spectra from a disordered CoO surface show sharp structures. They compare well with optical absorption data and can be interpreted in terms of ligand-field theory. SPEELS directly distinguishes quartet-doublet excitations (involving spin flips) from quartet-quartet excitations (nonflip).

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