# Ultrathin nickel films on Cu(001): Loss of strong ferromagnetism

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The unoccupied electronic states in ultrathin Ni films on Cu(001) have been studied by spin- and angle-resolved inverse photoemission. Although the *d* holes in a strong ferromagnet such as Ni are expected to have exclusively minority character, our inverse-photoemission results for a 6 ML film of Ni on Cu(001) provide evidence of significant majority contributions from empty *d* bands. We examined carefully that the majority intensity is not caused by a reduced magnetization due to a lower Curie temperature as compared with the bulk case or by a modified magnetic structure of the film. Hence, it is attributed to a band structure different from the bulk situation, i.e., the majority *d* bands are not entirely occupied. As a consequence, ultrathin Ni films on Cu(001) do not represent a strong ferromagnet.

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## I. INTRODUCTION

Ultrathin Ni films on Cu(001) represent one of the most studied magnetic thin-film systems.[1,2] They show unique magnetic properties such as unusual spin-reorientation transitions. With increasing film thickness, the easy magnetization axis changes from in plane to out of plane at about 7–10 monolayers (ML) and back to in plane at about 40 ML.[3–6] Only few spin-integrated photoemission studies on the electronic structure of Ni/Cu(001) are available so far. One of them traces the development of the electronic states with increasing Ni thickness,[7] another deals with Ni/Cu multilayers, addressing the interaction between two Cu quantum-well states.[8] Previous work with spin-integrated inverse photoemission (IPE) dealt with Ni/Cu(001) and trilayer structures of Ni/Cu/Ni (Ref. 9) and Cu/Ni/Cu.[10] There, among other things, the existence of discrete quantum-well states in thin Ni films on Cu(001) was shown. However, the main interest was in the electronic states of the Cu layer, and no spin resolution was used. But in fact it is the spin polarization of the electronic states which governs the magnetic properties of the thin-film structures and thus determines the magnetic response of possible devices.

In the present study, the unoccupied states of ultrathin Ni films on Cu(001) are investigated by spin- and angle-resolved inverse photoemission.[11,12] The experiment shows that the Ni *d* feature, which results from indirect, non-$\boldsymbol{k}$-conserving transitions into empty *d* states, appears in both the minority and the majority-spin channels, even though with a clear surplus for minority spin. This is at variance with expectations from the Ni bulk band structure, which shows an exclusive minority character of the *d* holes.[13] This minority character for bulklike Ni was experimentally confirmed by inverse-photoemission measurements of Ni(110),[14] Ni(001),[15] and thick in-plane magnetized Ni films with (001) surface.[16] For comparison, ultrathin Co films on Cu(001) show the expected high spin asymmetry just above the Fermi level, despite a comparable bulk band-structure situation as for Ni.[17]

The electronic structure of ultrathin Ni films on Cu(001) has been the subject of a number of theoretical studies in recent years.[18–24] In ultrathin Ni films of a few atomic layers, in general, the magnetic moment and the number of *d* holes is predicted to be enhanced at the surface due to dehybridization effects and reduced at the Ni/Cu interface due to proximity effects. The latter is also true for monolayer films of Ni on Cu(001).[25] The shape of the 3*d* partial density of states (DOS) is found to be modified, compared with the DOS of bulk Ni, for overlayer thicknesses of a few monolayers. There is, however, no indication of a significantly increased majority density of states at and above the Fermi level. In all calculations, the majority 3*d* band is completely filled for ultrathin films, as it is for bulk Ni.[26,27]

X-ray magnetic circular dichroism (XMCD) measurements indicated that the density of 3*d* holes is indeed reduced compared with bulk Ni for ultrathin Ni films on Cu(001).[28] The reduction amounts to about 20% at submonolayer coverage and about 5% at 5 ML. Above 4–5 ML, the increase in the 3*d* hole density starts to saturate. The authors state that, within error limits, it can be considered as constant. The presented data are, however, also compatible with the conclusion that saturation is reached not until about 15 ML. In particular, according to these data, a 6 ML film may not be considered as bulk. The saturation implies that the band structure of Ni stabilizes. As a further XMCD result, a decrease in spin and orbital moments in 4 ML Ni films was reported.[29] This result was later clarified with the conclusion that the magnetic moments are constant and close to the bulk value from 4 to 20 ML layers of Ni.[30]

In summary, from theoretical as well as experimental studies so far, there is only little indication that a 6 ML Ni film on Cu(001) shows a *d* band structure different from bulk Ni(001). Though we cannot determine the number of *d* holes in our **k**-resolved IPE experiment, we found evidence of majority *d* holes in 6-ML-thick Ni films on Cu(001). In this contribution, we will show that the band structure in a 6-ML-thick film is modified compared with the band structure of bulk Ni. The majority intensity just above $E_F$ in IPE spectra of ultrathin Ni films on Cu(001) cannot be explained as intensity from the Cu substrate nor by other transitions in the Ni film such as discrete quantum-well states. Furthermore, it is not caused by specific structural or magnetic film properties, e.g., a reduced magnetization due to the lower Curie temperature or the decay in magnetic domains with different orientations of the magnetization. Consequently, we found

 

evidence that the majority *d* bands are not entirely occupied in 6-ML-thick Ni films. Hence, ultrathin Ni films on Cu(001) do not represent a strong ferromagnet.

## II. EXPERIMENT

Spin-resolved IPE is used to measure the spin-dependent electron states above the Fermi level. In our setup,[31] a standard GaAs photocathode emits a beam of low-energy electrons with defined energy, momentum, and spin polarization $(33 \pm 3\%)$.[14,32] After passing a 90° electrostatic deflector, the spin-polarization direction is perpendicular to the propagation direction of the electron beam. With these transversally polarized electrons, parallel alignment between electron-spin polarization and sample magnetization can be achieved for normal electron incidence on a sample, which is remanently magnetized in the surface plane. After impinging on the sample, the electrons may decay via radiative transitions into lower-lying unoccupied states. The emitted photons are detected by a Geiger-Müller counter at an angle of 70° with respect to the incident electron beam. The counter is energy selective because of the use of iodine as filling gas and $SrF_2$ as entrance window. The energy resolution of this band-pass-type detector can be tuned, within limits, by varying the temperature of the entrance window.[33] The overall energy resolution, combining the detector band pass width and the electron energy distribution, is reduced from 0.4 eV full width at half maximum (FWHM) at room temperature to 0.3 eV at 80 °C.[34,35] The IPE spectra represent the photon intensities as a function of the kinetic energy of the incoming electrons. The energy scale of the spectra is referred to the Fermi energy of the sample. The spectra shown have been normalized to hypothetical 100% spin polarization.[36]

For magnetometry of the sample within a probing depth equivalent to IPE, we use spin-polarized secondary electron emission (SPSEE).[37–39] The low-energy cascade electrons, also called true secondaries, emitted from magnetic samples are known to be spin polarized.[40] The actual value of the spin polarization is related to the spin-dependent band structure of the material and is, for a given material, proportional to the sample magnetization.[39] In our experimental setup, the secondary electrons are excited by primary electrons from a fine-focus electron gun as described elsewhere.[32] The secondaries are collected by an electron optics specially designed to act as a filter for low-energy electrons below about 10 eV. No further energy analysis is done. Unlike the setup described in the literature, we determined the electron-spin polarization with a Mott-type detector[41–43] with a Sherman function of $15 \pm 3\%$.[44]

The preparation of the Cu(001) single-crystal substrate is identical to the procedure outlined previously.[17] The Ni films are deposited epitaxially by a commercial evaporator at a pressure below $2 \times 10^{-10}$ mbar, with the substrate kept at room temperature. The deposition rate amounts to $0.50 \pm 0.05$ ML/min, calibrated with Auger electron spectroscopy (AES).[45] After film deposition, the sample is annealed at 450 K for about 15 min to improve the film quality.[46] The pseudomorphic growth of Ni on Cu(001) results in a tetragonally distorted face-centered-cubic structure.[1,47] The Ni films exhibit the same sharp low-energy electron-diffraction (LEED) pattern as the clean Cu(001) substrate. In addition, the observed high intensity of an image-potential surface-state emission as well as the appearance of discrete quantum-well states are evidence of a very good film quality with sharp interfaces.[17] The sample is magnetized along the [110] direction, which is the direction of easy magnetization in ultrathin Ni films with in-plane anisotropy on Cu(001).

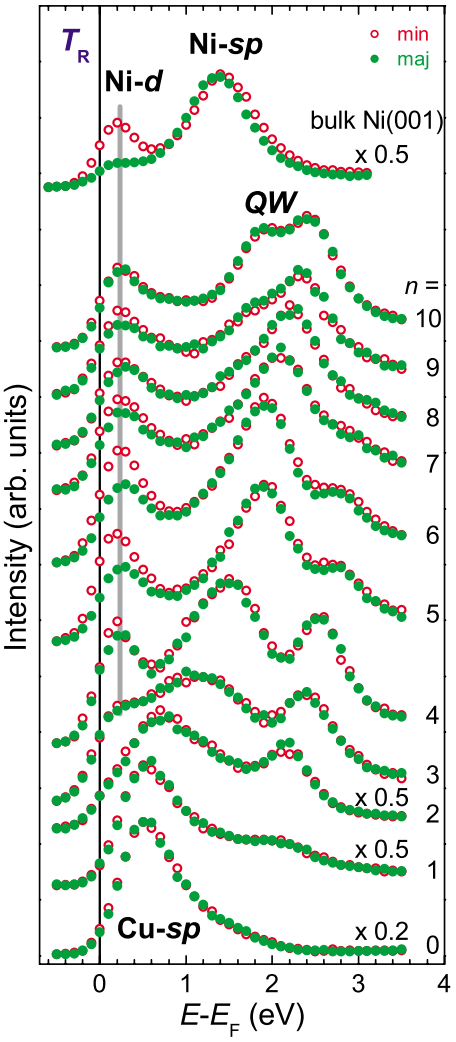


FIG. 1. (Color online) Spin-resolved inverse-photoemission spectra of *n* ML Ni on Cu(001) taken at normal electron incidence and room temperature $T_R$, showing the thickness dependence of the Ni *d* bulk feature and the quantum-well features QW. The spectra are normalized to equal background intensity. The spectra for 0, 1, and, 2 ML Ni as well as for bulk Ni(001) are reduced by the given factors. The spectra for bulk Ni(001) are taken from Ref. 15. See text for details.

## III. RESULTS

Figure 1 shows spin-resolved IPE spectra for *n* ML Ni on Cu(001), covering the range from pure Cu(001) to 10 ML Ni on Cu(001). For reasons of comparison, the spectra of a

(001) surface of a bulk Ni crystal are presented as well, taken from Ref. 15. The spectra, taken at room temperature for normal electron incidence, demonstrate the thickness dependence of the electronic states of Ni/Cu(001). Minority and majority data are represented by open and filled circles, respectively. The spectra are normalized to equal spin-averaged background intensity. For reasons of presentation, the spectra for the Cu(001) substrate are multiplied by a factor of 0.2, the spectra for 1 and 2 ML Ni as well as for bulk Ni(001) by a factor of 0.5.

For the nonferromagnetic Cu(001) substrate, no spin asymmetry is observed within the experimental statistical error. The spectrum exhibits a bulklike transition between *sp* states of Cu at 0.5 eV above the Fermi level (Cu-*sp*), as well as a crystal-induced surface resonance showing up as a shoulder at its high-energy side. Both features are well known from the literature.[48,49] With increasing Ni coverage, the intensity of the Cu features rapidly decreases according to the short attenuation length of low-energy electrons in Ni. Additional spectral features appear which originate from the Ni overlayer. One feature at about 0.25 eV above the Fermi level is present independent of the Ni thickness. For a certain thickness, this feature shows up with different intensity in the two spin channels. It is attributed to indirect, non-$\boldsymbol{k}$-conserving transitions into empty *d* states,[15,50] therefore denoted as Ni-*d*, and will be discussed in detail below.

A number of additional spectral features are resolved whose energies vary with increasing overlayer thickness. They are identified as discrete quantum-well (QW) states, which are standing electron waves as a result of quantum confinement of electrons in ultrathin films.[51–55] The QW features show up from 1 ML of Ni, shift up in energy with increasing Ni coverage, as expected, and converge to the top of the Ni *sp* bands, which is at 2.74 eV above the Fermi level $E_F$ for majority spin and at 2.78 eV above $E_F$ for minority spin, respectively.[24] Three discrete QW features are clearly resolved. It has to be noted that our data are in accordance with spin-averaged IPE data of Ref. 9 except for obvious differences in the thickness calibration. Our calibration is consistent with the thickness-dependent spin-reorientation transitions and was additionally confirmed by AES. For a detailed discussion of the QW states and their spin dependence, the reader is referred to the literature.[16]

In the following, we will concentrate on the spin dependence of the Ni *d* feature. For a film thickness up to 4 ML of Ni, no spin asymmetry is detected because in this thickness range the Curie temperature of the Ni film is below room temperature.[2,56,57] The spin asymmetry is defined as the normalized intensity difference between the two spin channels: $A=(I_\uparrow-I_\downarrow)/(I_\uparrow+I_\downarrow)$. A clear spin asymmetry is observed for 5 and 6 ML, while for higher coverage the asymmetry decreases and finally vanishes due to the above-mentioned spin-reorientation transition, turning the magnetization from an in-plane to an out-of-plane orientation.[3,4,6,56,58] Since our electron beam is transversally polarized, IPE experiments for normal electron incidence are not spin sensitive to out-of-plane magnetized samples. The change in the magnetization from in-plane to out-of-plane does not abruptly occur in our data because the film thickness is not totally uniform.

It is not surprising that the spin asymmetry is only nonzero for an intermediate thickness range, where, on the one hand, the Curie temperature is higher than the sample temperature and, on the other hand, the thickness is below the critical thickness for the spin-reorientation transition. However, it comes as a surprise that one observes minority *and* majority *d*-band emission in the thickness range around 6 ML. One may have expected exclusive minority intensity for the *d*-band transition like in the spectra for bulk Ni(001) shown in the upper part of Fig. 1. There, the majority intensity is about equal to the background intensity.

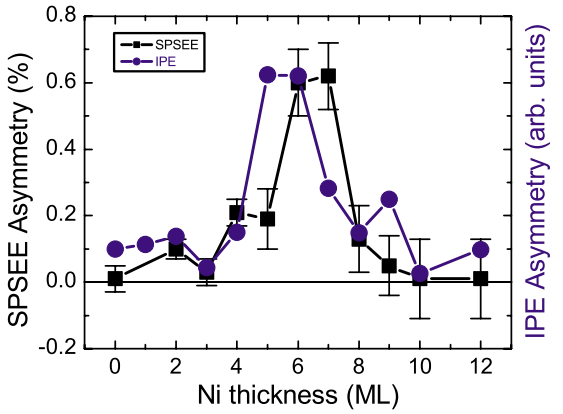


FIG. 2. (Color online) In-plane spin asymmetry $A=P\times S$ of secondary electrons (squares) as well as IPE asymmetry in the energy range 0.1 eV$<(E-E_F)<$0.3 eV, both as a function of the Ni film thickness of *n* ML Ni on Cu(001). The data were obtained at room temperature.

To relate the spin asymmetry of the Ni *d* emission to the sample magnetization, we performed SPSEE measurements of the remanent in-plane magnetization as a function of the Ni film thickness. Figure 2 shows the measured SPSEE spin asymmetry values *A* as a function of the Ni film thickness. In SPSEE, the spin polarization *P* of the secondary electrons is given by $P=A/S$ with *S* the Sherman function of the spin-polarization detector.[59] In the maximum, the spin asymmetry amounts to $0.6\pm0.1\%$. Thus, the maximum polarization of the secondary electrons from the Ni films, which have kinetic energies of a few eV, is $P=4\pm1\%$. This is in accordance with the spin polarization of secondary electrons from bulk Ni(001), which is between 2% and 10% depending on the electron energy.[37]

As an additional data set in Fig. 2, the IPE spin asymmetry of the Ni *d* emission is shown. Each data point represents an average of the spin asymmetry between 0.1 and 0.3 eV above the Fermi level, which is around the peak maximum. Note that the IPE asymmetry gives negative values because of the higher minority intensity, while the SPSEE asymmetry gives positive values due to the higher majority intensity. To compare both data sets, the IPE data have been plotted with reversed sign and scaled to the asymmetry maximum in SPSEE.

For very low coverage, the Curie temperature is below room temperature, thus the asymmetries are zero. With increasing coverage, they increase and reach their highest values between 5 and 7 ML Ni. For 8 ML Ni, the spin-reorientation transition has already started to turn the magnetization direction out of plane. For higher coverage,

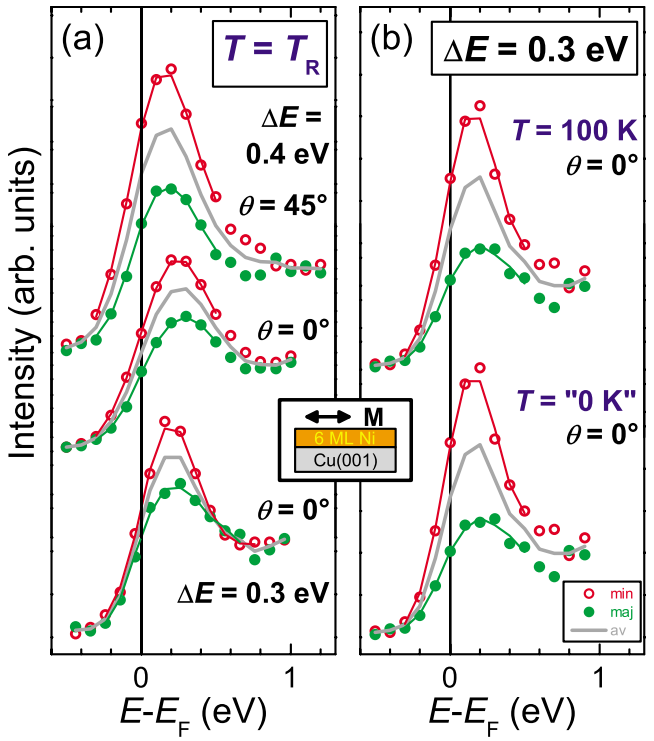


FIG. 3. (Color online) Spin-resolved inverse-photoemission spectra of 6 ML Ni on Cu(001) at (a) room temperature and at (b) $T=100$ K (open circles for minority spin, filled circles for majority spin, solid gray line for spin-averaged data). Most spectra were taken for normal electron incidence, one for $\theta=45°$. Different energy resolutions were used as indicated. The spectra in the lower part of (b) are simulated "$T=0$" spectra calculated from the spectra taken at $T=100$ K as described in the text.

the Ni films are completely magnetized out of plane, and again the asymmetries are zero. Both data sets show the same thickness dependence with a small shift of the maximum. This deviation is ascribed to slightly different layer thicknesses in the two different measurement cycles, resulting from uncertainties in the film thickness calibration by AES of about 10%. In addition, it has to be noted that, in the case of SPSEE, the thickness was increased from one measurement to the next, whereas, for IPE, a new film was prepared for each thickness. The latter procedure gives more reliable results for the film thickness than the first. Nevertheless, both measurements are compatible within the experimental error bars, and we will draw all conclusions from measurements on 6 ML of Ni only, a thickness with maximum spin asymmetry beyond dispute.

So far, we can conclude that 6 ML of Ni on Cu(001) are within the thickness range of in-plane magnetization but show significant unoccupied majority $d$-band contributions in the room-temperature IPE spectra. How can we understand this majority intensity, clearly seen in Fig. 3(a) as measured for different experimental energy resolutions? In the following, we will propose and test several hypotheses.

(i) The unexpected majority $d$-band intensity may originate from $sp$-derived transitions of the Cu substrate or the Ni overlayer.

First, the $d$-band intensity appears at an energy distinctly different from the Cu $sp$ transition (see Fig. 1). Therefore, it cannot be attributed to remnant intensity from the Cu substrate attenuated by the Ni overlayer. Second, $sp$-derived QW states within the Ni overlayer may indeed have small intensity just above the Fermi level. At least, angle-resolved IPE data support this assumption.[16] The $n=3$ QW state seems to coincide with the Ni $d$ emission for normal electron incidence, yet with very weak intensity. However, angle-resolved data show that QW states do not contribute to the spectral intensity at the Fermi level for an electron incidence angle of 45°. Therefore, we present additional spectra for $\theta=45°$ in Fig. 3(a), which show the majority intensity as well. In conclusion, no $sp$-derived transition in Cu or Ni can explain the unexpected majority feature.

(ii) The unexpected majority $d$-band intensity may be a consequence of a reduced Curie temperature of a 6-ML-thick film compared with the bulk value.

Indeed, the Curie temperature $T_C$ of thin Ni films is known to be strongly thickness-dependent and reduced compared with bulk Ni.[2,56,57] For room-temperature measurements, the reduced temperature $T/T_C$ is enhanced from 0.46 for bulk Ni to about 0.75 for a 6-ML-thick film. This has consequences for the magnetization $M$. $M$ decreases with increasing temperature following the power law $M/M_0=(1-T/T_C)^{0.23}$ ($M_0$: magnetization at $T=0$), until the Curie temperature is reached. Although the power law is only strictly valid for temperatures close to the Curie temperature, it well describes the behavior in thin Ni films on Cu(001) also for low temperatures.[56] According to this power law, $M/M_0$ is 0.72 at room temperature and 0.93 at $T=100$ K.

We tested the hypothesis by performing IPE measurements on the Ni $d$ feature for room temperature and for $T=100$ K; see Fig. 3(b). The measurements are taken at normal electron incidence with improved experimental resolution (0.3 eV). All spectra are normalized to the same background intensity. Minority and majority data are represented by open and filled circles, respectively, the spin-averaged data by a solid line just connecting the data points. A comparison of the room-temperature data [Fig. 3(a)] with the data for $T=100$ K [Fig. 3(b)] shows a significant increase in the spin asymmetry, but even for $M/M_0=0.93$ ($T=100$ K), the majority feature is still there. We even simulated a "$T=0$" data set from the spectra taken at $T=100$ K by taking into account the slightly reduced magnetization. For this, we use an effective spin polarization $P_{\mathrm{eff}}=0.33\times0.93$ in our procedure of normalizing the measured spectra to hypothetical 100% electron-spin polarization. The result is shown in the lower part of Fig. 3(b). Again, the spin asymmetry is slightly increased, but the majority intensity is not removed.

Concerning the temperature dependence of the magnetization, it has to be noted that the thickness of the spin-reorientation transition from in-plane to out-of-plane magnetization in Ni films on Cu(001) depends also on the sample temperature.[58,60] But as this thickness slightly increases with decreasing temperature, it should not influence the spin asymmetry in the spectra when measuring at low temperature. In summary, the reduced Curie temperature of thin Ni films on Cu(001) and the temperature dependence of the magnetization cannot explain the majority intensity just above the Fermi level.

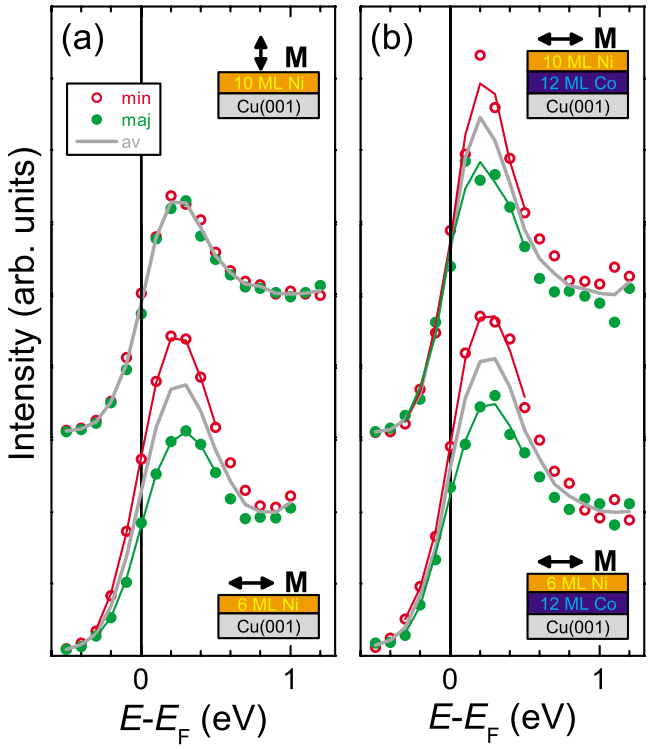


FIG. 4. (Color online) Spin-resolved inverse-photoemission spectra (a) of 6 and 10 ML Ni on Cu(001) and (b) on 12 ML Co/Cu(001) (open circles for minority spin, filled circles for majority spin, solid gray line for spin-averaged data). The spectra are taken at normal electron incidence and room temperature.

(iii) The unexpected majority *d*-band intensity may be caused by magnetic domain formation.

First, the film may contain magnetic domains with different in-plane magnetization directions. However, there is no report about such a domain formation in the literature so far. Second, the asymmetry in the IPE spectra might be reduced by magnetic domains which are magnetized out of plane. Owing to the film growth, the film is not perfectly flat and may contain islands of thicker layers[46] with out-of-plane magnetization, thereby reducing the observed in-plane spin asymmetry.

Both hypotheses have been tested in a further experiment with the idea of pinning the magnetization of a Ni film to an in-plane magnetized Co film on Cu(001). As a pinning layer, a 12-ML-thick Co film has been chosen. For this thickness, the Curie temperature is high enough to allow a magnetically saturated film, and the film quality is still good enough to provide a flat Co-Ni interface. In Fig. 4 IPE spectra of Ni films on Co [Fig. 4(b)] are compared to the spectra of Ni films, which had been deposited directly on Cu(001) [Fig. 4(a)]. The spectra for 6 ML Ni films, shown in the lower parts of Figs. 4(a) and 4(b), are quite similar. In particular, the spectra for Ni on Co do not exhibit an increased spin asymmetry. Therefore, our data give no hint of any magnetic domains of the Ni film on Cu(001), either in plane or out of plane, that are not aligned along the direction of the applied field to magnetize the sample.

For Ni films on Co, there is no spin-reorientation transition and even Ni films thicker than 8 ML are completely in-plane magnetized.[61] With this in mind, the magnetic pinning of Ni films on Co was examined by taking IPE data for 10 ML Ni on Cu(001) and for 10 ML Ni on 12 ML Co/Cu(001). The data are shown in the upper parts of Figs. 4(a) and 4(b). For 10 ML Ni on Cu(001), the magnetization is out of plane, and, therefore, the observed spin asymmetry is zero. For 10 ML Ni on Co, however, the magnetization of the Ni film is pinned to the magnetization of the Co film. Thus, both the Co film and the Ni film are magnetized in the film plane. Our data exhibit a spin asymmetry of about the same size as for 6 ML Ni on Cu(001). In particular, the Ni *d* feature still appears for majority spin. This shows that our idea of magnetic pinning works well.

In addition, the influence of a possible out-of-plane magnetization can be investigated via IPE measurements taken at off-normal electron incidence. The measurement is sensitive to the projection of the electron-spin polarization $\boldsymbol{P}$, which is perpendicular to the electron-beam direction, onto the magnetization $\boldsymbol{M}$, $\boldsymbol{P}\cdot\boldsymbol{M}$. For example, at an electron incidence angle $\theta=45°$, the measurement is equally sensitive to in-plane and to out-of-plane magnetization components. The spin asymmetry of both 6 [Fig. 3(a)] and 10 ML Ni on Cu(001) (data not shown) was found about equal to the spin asymmetry of 6 ML Ni on Cu(001) for normal electron incidence, except that the spin-averaged intensity changes with angle. In conclusion, the unexpected majority intensity of the Ni *d* feature is not caused by a mixture of in-plane and/or out-of-plane magnetized domains. Our data show that the 6 ML Ni films are remanently magnetized in the film plane in a single domain state.

(iv) Having excluded (i) to (iii), we conclude that the band structure of thin Ni films is modified compared with the bulk band structure in such a way that majority *d* states appear above the Fermi level. Owing to the existence of majority *d* holes, thin Ni films on Cu(001) do not represent a strong ferromagnet.

## IV. CONCLUSION

In summary, the unoccupied electronic states in ultrathin ferromagnetic Ni films on Cu(001) were investigated by spin-resolved IPE. It was shown that the, for the strong ferromagnet Ni, unexpected appearance of transitions into majority *d* bands in thin films is not caused by spectral intensity from the Cu substrate or from Ni quantum-well states. It is also not caused by structural or magnetic film properties, such as the reduced Curie temperature or the formation of magnetic domains. Hence, the reduced spin asymmetry of the Ni *d* feature must have its origin in a modified band structure such that the majority *d* bands are not entirely occupied. As a consequence, Ni loses its strong ferromagnetism in thin films deposited on Cu(001).

This result is surprising in the light of previous layer-dependent band-structure calculations, which do not show majority *d* holes in ultrathin Ni films. However, XMCD results indicated that the *d* hole density is only bulklike for film thicknesses larger than about 15 ML, yet without separating majority and minority contributions.[28] Our experimental approach, which probes *directly* the spin dependence of the *d*

holes, reveals a majority contribution in the *d* holes. As the shape of the *d* hole density was determined to be thickness dependent, we conclude that the spin contributions depend on the thickness as well. A comparison with measurements for ultrathin Co films on Cu(001) shows that this effect appears, if at all, much less pronounced there.[17] Obviously, there is a difference in this respect between Co and Ni films, despite the fact that both materials are strong ferromagnets in the bulk. However, Co exhibits a considerably larger exchange splitting.

We can only speculate about the origin of the observed effect. Will spin-polarized fully relativistic band-structure calculations bring an answer? These types of calculations treat spin-orbit coupling and exchange splitting on equal footing.[62] Is our understanding about the influence of electron correlations in low-dimensional ferromagnets sufficient? We hope that our results will stimulate experimental as well as theoretical work to better understand the spin-dependent electronic structure of nanoscaled ferromagnets.

## ACKNOWLEDGMENTS

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