PHYSICAL REVIEW B VOLUME 51, NUMBER 17 1 MAY 1995-I

Empty electronic states in magnetic thin films: Fe on Au(100), Ag(100), and Cu(100)

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(Received 28 November 1994)

Iron films with thicknesses ranging from 5 to more than 20 monolayers (ML) have been deposited onto Au(100), Ag(100), and Cu(100) and investigated by spin-resolved inverse photoemission spectroscopy. Analysis of surface features corresponding to image states indicates a substrate-dependent overlayer structure and surface quality. Measurements confirm that Fe grows in the bcc phase on Au(100) and Ag(100) and in the fcc phase on Cu(100). Above 15 ML also Fe/Cu(100) films display bcc character. Thick ($\approx$15 ML) bcc films on all the substrates present a remanent in plane magnetization with easy axis along the Fe [010] direction as well as bulklike empty states with majority and minority spin character. Instead, fcc films are not in-plane magnetized. A different behavior of the minority and majority peaks' evolution with film thickness in Fe/Au(100) versus Fe/Ag(100) thin films is also found.

I. INTRODUCTION

Empty electronic states in ferromagnets are particularly interesting, since they contain the unbalanced minority-spin holes, that make up the magnetic moment. In classical 3d ferromagnets there are less unoccupied than occupied d bands, resulting in less overlapping states in the empty region of the spectrum. It is then experimentally simpler to picture out the dispersion relation of the exchange split minority and majority bands. These states may be efficiently probed by inverse photoemission spectroscopy (IPE), which is a well established experimental technique providing direct information about unoccupied bands in the vicinity of the Fermi level E_F.[1,2] IPE measurements on 3d metal have been performed by various authors.[3–12] The spectra are often dominated by bulk derived transitions, but emission associated with surface states (induced either from the crystal potential[7,11] or the image Coulomb potential,[9–12] is also observed.

An obvious extension of the technique is the spin-resolved inverse photoemission. In fact, by employing a spin-polarized electron beam, it is possible to gain direct information on the spin character of the bands and on the ferromagnetic exchange splitting (for a review on spin-resolved IPE, see Ref. 13). The spin-polarized inverse photoemission investigation of Ni(110) dates back to 1982;[3] it was soon followed by measurements on Fe samples.[4,5] Such pioneering studies were dealing with bulk-like properties, while more recently surface related properties have also been investigated.[7,10–12]

While all of the studies quoted above have been performed on bulk materials, in the last years is clearly emerged the technological as well as fundamental relevance of interface, thin film, and multilayer magnetism.[14,15] As a consequence, a lot of research on the properties of magnetic-nonmagnetic metal interfaces is going on. In particular, iron growth on noble-metal substrates is a widely studied topic. Owing to the almost perfect lattice parameter matching, iron is found to grow in the bcc structure on both Au(100) (Refs. 16–19) and Ag(100) (Refs. 20–26) with the same lattice spacing as bulk bcc Fe, the overlayer lattice being rotated by 45° with respect to the substrate lattice. On Cu(100), instead, the surface square mesh with a reduced spacing favors the growth of fcc Fe (Refs. 27–33), which is the equilibrium bulk phase above 900 °C.

In this frame, IPE measurements on thin films of 3d metals grown on nonmagnetic substrates,[34–38] or vice versa,[39,40] have been performed. In these studies, however, unpolarized electron sources were used, so that the IPE spectra were not spin resolved. Therefore no direct experimental access to the spin character of the bands was obtainable, and this and similar magneticlike information had to be inferred in an indirect way.

In this work, which follows recently reported studies on Fe/Ag(100) thin films,[41,42] we investigate the electronic structure of Fe/Au(100), Fe/Ag(100), and Fe/Cu(100) films by means of angle- and spin-resolved isochromat IPE, for different thicknesses ranging from 5 to more than 20 monolayers (ML, 1 bcc Fe ML = 1.44 Å, 1 fcc Fe ML = 1.8 Å). The films have been deposited and investigated at room temperature. Our measurements confirm that Fe grows in the bcc phase on Au(100) and Ag(100) and in the fcc phase on Cu(100). Above 15 ML also Fe/Cu(100) films display bcc character. Moreover Fe/Au(100) films display a remanent in plane magnetization with easy axis along the Fe [010] direction as well as bulklike empty states with majority and minority spin character. This holds true also for relatively thick ($\approx$15 ML) bcc films grown also on the other substrates. Instead, thin (<6 ML) bcc Fe/Ag(100) and fcc Fe/Cu(100) films are not in-plane magnetized. Intermediate thickness (7–10 ML) Fe/Ag(100) films present a reduction of the exchange splitting with respect to the bulk value.

The paper is organized as follows: after the experimental Sec. II, the results are presented and discussed in Sec. III. There we consider first the case of Fe single crystals and homoepitaxial films (Sec. III A), then the growth of Fe films on noble metals, referring to published structural studies and presenting our experimental findings on the effect of the film quality and structure on IPE (Sec. III B).

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Finally, in Sec. III C, the spin-resolved spectra for Fe/Au(100), Fe/Ag(100), and Fe/Cu(100) films are presented and discussed. Conclusions are summarized in Sec. IV.

II. EXPERIMENT

The experiments have been performed in an ultrahigh-vacuum (UHV, pressure in the 10^{-11} Torr range) apparatus described in detail elsewhere.[42]

Mechanically and chemically polished Au, Ag, and Cu(100) single crystals have been cleaned by grazing incidence Ar ion sputtering (0.5–1.5 keV) and annealing (500–700 °C) procedures. The surface quality and cleanliness were monitored with standard analysis techniques, such as low-energy electron diffraction (LEED) and x-ray photoemission (XPS). The clean surfaces did not present any trace of contamination and exhibited bright LEED patterns, showing 5×20 and 1×1 reconstructions for Au(100) and both Ag(100) and Cu(100), respectively. As already reported in a very accurate IPE study of (100) noble metal surfaces,[43] it is found that the best indicators for the surface quality are the IPE features, like the intensity of the transition to image state or the depth of the emission minimum in the X_4'-X_1 gap along the Γ-Δ-X direction in the fcc Brillouin zone (BZ).

Fe overlayers have been deposited at room temperature, by means of a water-cooled evaporation cell, at a typical rate of 0.5 ML/min, as measured by a calibrated quartz microbalance. The estimated accuracy in the absolute thickness calibration is about 20%, whereas the relative values are known with a much larger precision ($\approx 5\%$). During the Fe growth the pressure remained below 3×10^{-10} Torr. High-purity films are grown as confirmed by XPS which did not show any detectable trace of contaminants. Epitaxial growth is checked by LEED. All of the data reported here refer to freshly prepared film evaporated on a clean surface.

For comparison with the results on films, a bulk Fe(100) crystal has also been measured. Following reported procedures,[44,8] before installation in vacuum the crystal underwent an initial heat treatment in 1 atm of H_2 at 400 °C for one day followed by another day at 700 °C, in order to deplete the subsurface region from contaminants. Once in vacuum sputtering-annealing cycles were performed until no contamination was detectable and the LEED showed a sharp 1×1 pattern, indicating a clean and ordered surface.

Inverse photoemission spectra are taken at room temperature in the isochromat mode. A collimated and transversely polarized (spin perpendicular to electron momentum) electron beam, with a polarization $P_0=25\pm 3\%$,[45] produced by a negative electron affinity GaAs photocathode, is sent onto the sample and the emitted photons are collected at a fixed energy $h\nu=9.6\pm 0.3$ eV,[42] while the beam energy is swept to change the initial electron state. The primary beam incidence angle is varied by rotating the sample: in the present work, however, we discuss only normal incidence data.[46]

The Fe films are magnetized in-plane along a high-symmetry direction of the Fe lattice by means of a current pulse sent through an eight-turn coil, which surrounds the sample. In our geometry, the incident beam polarization is parallel to the surface: in this way we maximize the spin-dependent interaction of the electron beam with an in-plane magnetized film. On the other hand, in such a configuration no spin-dependent effects are measurable for perpendicularly magnetized films.

Spin resolution is achieved by measuring two sets of spectra for impinging electron-beam polarization parallel or antiparallel to the sample magnetization.[13] As usual with electron-based spin-resolved spectroscopies, all the measurements have been taken in magnetic remanence.

III. RESULTS AND DISCUSSION

A. Fe(100) single crystal

For the sake of clarity, before discussing the case of thin films, we present results on bulk iron. Normal incidence IPE data from a clean Fe(100) sample are shown in Fig. 1(a). Two structures, labeled *B*1 and *B*2, are clearly seen in the spectrum just above E_F: *B*1 results from a peak at E_F cut by the Fermi function, while *B*2 is located at 1.6 eV above E_F. Moreover, a smaller feature *S* centered at around 3.8 eV superimposed to the typical IPE background due to inelastic processes,[47] is also visible. The measured spectrum is in very good agreement[48] with reported investigations on empty states in bulk iron.[8,9] At normal incidence on the Fe(100) surface electron states along the $\Gamma\Delta H$ line of the bcc BZ are probed. *B*1

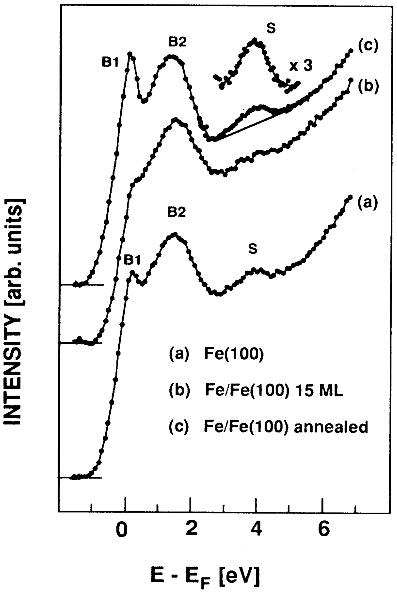

FIG. 1. Normal incidence inverse photoemission spectra taken at $h\nu=9.6$ eV from the following iron samples: (a) Fe(100) single crystal; (b) 15 ML thick Fe film homoepitaxially grown onto Fe(100); (c) the same film after annealing at 500 °C. In the topmost spectrum, the surface feature *S* at 3.8 eV above the Fermi level is also shown enlarged by a factor of 3 after a linear background subtraction.

and $B2$ correspond to bulk transition to majority and minority spin states close to the high-symmetry point H (H'_{25} majority and minority levels, respectively). This attribution, which follows directly from the Fe band structure (see Ref. 8 for details), relies also upon spin-resolved IPE.[5] In isochromat measurements the electron momentum is not constant in the spectrum, so that the transitions giving rise to the two structures occur in slightly different places of the BZ, at different **k** values. Therefore the energy separation between $B1$ and $B2$, namely 1.6 eV, yields the exchange splitting at the H point δE_{ex} only after having taken into account the bulk Fe band dispersion: the extrapolated value is in very good agreement with the known value of 1.8 eV.[8]

The peak S at higher energy is identified as a surface feature: similar structures are frequently present in IPE spectra from metal and are due to transitions into image potential states.[1,2] As noted above, the presence of such peak is a clear indication of clean and well ordered surfaces.[43] Its energy position fits very well with the reported value for Fe(100) surface,[9] while its width is mainly due to the experimental resolution.

In order to make comparison to the case of thin films, a 15 ML thick Fe film was deposited on the clean Fe(100) surface at room temperature: the results are shown in Fig. 1(b). The epitaxial film does not order as well as the bulk, which is reflected in the weakness of the image surface state and the lower intensity of the majority band at the Fermi level, $B1$. Similar findings have been reported in the spectroscopy of both occupied[22] and unoccupied[35,42] states in Fe/Ag(100) films. In these cases too, the increasing of the background and the variation of the relative intensities of the peaks observed in the spectra with respect to bulk Fe have been interpreted in terms of poor crystalline quality of the film. The present measurements on homoepitaxial films give a very direct confirmation of such findings. Order can be reestablished by annealing the film (500 °C for about 10 min), as shown in Fig. 1(c). We see indeed that the annealed films present sharp structures as well as a strongly reduced inelastic background, even with respect to the clean substrate. This result indicates that the best way to prepare a clean and well ordered Fe surface is by homoepitaxy followed by annealing. Note that while the different surface quality strongly influences the IPE spectra, the clean substrate and annealed film LEED patterns are undistinguishable, at least with qualitative observations. When considering Fe films grown on noble metals, however, the annealing procedure for improving the order of the film cannot unfortunately be followed. In fact, in this case heating the film produces sizeable interdiffusion, as seen, for instance, in IPE from annealed Fe/Ag(100) films.[49] Actually, detailed structural analysis indicates that some degree of noble-metal segregation takes place already at room temperature.[19,25,33]

B. Fe films on noble metals: Quality and structure

Here we consider the structure and quality of the films deposited on noble metal (100) surfaces. We discuss our results on the basis of the large amount of data on the Fe films growth, showing how structural and ordering effects are visible in spectra. Our measurements add new information giving further experimental support to the picture of film growth arising from literature.

During the iron deposition, we find the following evolution of the LEED patterns. In the case of Fe/Au(100) films, the 5×20 pattern of the clean surface turns to a bright 1×1 upon the deposition of 0.2–0.3 Fe ML. Then, it remains very sharp, as the one from the substrate, indicating an excellent epitaxy and film quality. In fact, the growth has been found to proceed layer-by-layer, with one layer of Au floating on top of the growing Fe film.[16,19] The Au overlayer lowers the surface energy of the film, preventing island formation and giving rise to a well ordered structure. When the substrate is Ag(100), the LEED pattern remains quite sharp during the growth of the first 2–3 Fe ML, even if the spots become larger than those of clean Ag. Then, the pattern becomes more and more diffuse and the spots get very large. Above 8 ML, the LEED pattern improves again, even if it is not as sharp as that of clean Ag. This qualitative behavior agrees very well with previously reported quantitative studies,[25] indicating that the films are not very flat and the growth occurs by agglomeration of Fe atoms into islands, with valleys in between which are progressively filled as Fe deposition goes on. In Fe/Cu(100) films, we find a LEED pattern very similar to that of the clean substrate up to more than 10 ML, then it becomes quite diffuse with large background and broad spots above 15 ML. Quantitative investigations indicate that at the beginning (<5 ML) fcc Fe island agglomeration as well as Cu segregation occurs,[32,33] then fcc films are layer-by-layer grown up to more than 10 ML,[32] and eventually a three-dimensional bcc island appears and dominates the growth process resulting in less ordered bcc thick films.[32]

As a very simple and direct test of the film structure, we used absorbed current spectroscopy (ACS),[50,51] in which an electron beam is sent onto the sample and the target current is measured as a function of the electron energy. This technique has been successfully applied to test the electronic structure of Fe(100),[51] thus providing reliable data to compare with. Figure 2 shows the target current to Fe(100) sample, noble-metal(100) samples, and Fe/noble-metal(100) films, for normal incidence and normalized to the current emitted from the GaAs photocathode. Data have not been corrected for the transport electron optics transmission, which, being very low at low energy and constant above 8 eV, accounts for the general shape of the curve. Superimposed are small features, of the order of 10% of the total signal or even less, which are characteristic of the target atomic structure.[50] The ACS I/V curves for all the clean noble metals are very similar and structureless (at least in this scale), as characteristic of the (100) surface of a fcc solid. On the contrary, in the case of a bcc sample, such as the Fe(100) single crystal, deeper modulations are seen, in agreement with previous results.[51,52] This remarkable difference between ACS from fcc and bcc solids allows us to clearly distinguish between fcc and bcc films.[52] Fe overlayers onto Au(100) and Ag(100) present bcc character, as clearly visible in Fig. 2 for the 15 ML thick films

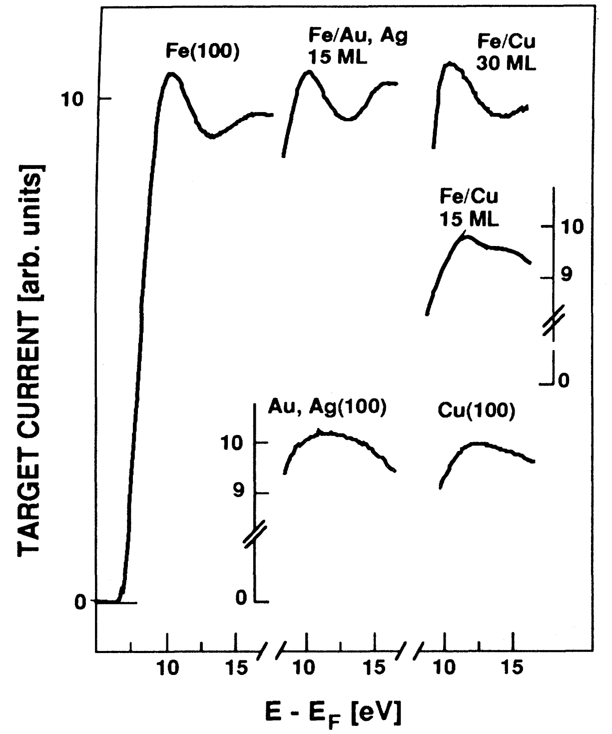

FIG. 2. Comparison of adsorbed current vs electron energy spectra from Fe(100), noble metals (100) surfaces, and Fe films deposited onto such surfaces. Only one set of spectra is presented for gold and silver since they yield identical results. The curves, normalized to the current emitted from the GaAs photocathode, refer to rough data not corrected for the electron optics transmission.

(thinner films behave in the same way). Thin films grown on Cu(100), instead, show an I/V curve very similar to the fcc substrate. The curve shape starts to change above 10 ML evolving towards the bcc-like curve. A predominant fcc character is still seen at 15 ML (see Fig. 2), while for larger thickness bcc films develop, as directly shown by the curve referring to 30 ML thick films. Our ACS measurements, then, support the above picture of Fe growing in the bcc phase on both Au(100) and Ag(100), and in the fcc phase on Cu(100). In this last case, thick bcc films can also be grown, the fcc to bcc transition thickness being around 15 ML. This value is slightly larger than the one reported in the literature (namely 11 ML).[32] A shift towards larger thicknesses in the transition due to CO contamination has also been observed:[32] however no trace of contaminants was found in our UHV deposited films. We believe that the origin of the discrepancy must be looked for in the different absolute thickness calibration in different experimental setups. On the other hand, the fcc to bcc transition can also show up in different ways when using different techniques. Moreover, in a very recent photoelectron diffraction study the growth of clean Fe/Cu(100) fcc films up to 15 ML has been reported,[53] in agreement with our result.

The above differences in film structure and order are also seen in the IPE measurements, presented in Fig. 3. As it is well known (see Ref. 43 and references therein), IPE spectra from the clean noble-metal substrates (Fig. 3, lower panel) are dominated by bulklike transitions occurring in the first few eV above E_F (region I in the following). Moreover, transitions to image states are seen in all the surfaces at higher energies (region II in the following), namely at 4.7, 3.9, and 4.0 eV for Au(100), Ag(100), and Cu(100), respectively, in agreement with previous results.[43] When Fe films are deposited on the substrates, the spectra change completely, as shown in Fig. 3, central panel, referring to 15 ML films. We consider first the energy region I. In Fe/Au(100) and Fe/Ag(100) the spectra are very similar to the one observed in bulk Fe(100) (cf. Fig. 2), and thus interpreted in terms of the same $B1$ and $B2$ transitions towards majority and minority spin states near the H point. As in the case of the homoepitaxial film (also shown in the inset of Fig. 3) $B1$ is reduced in intensity with respect to bulk Fe, indicating not completely

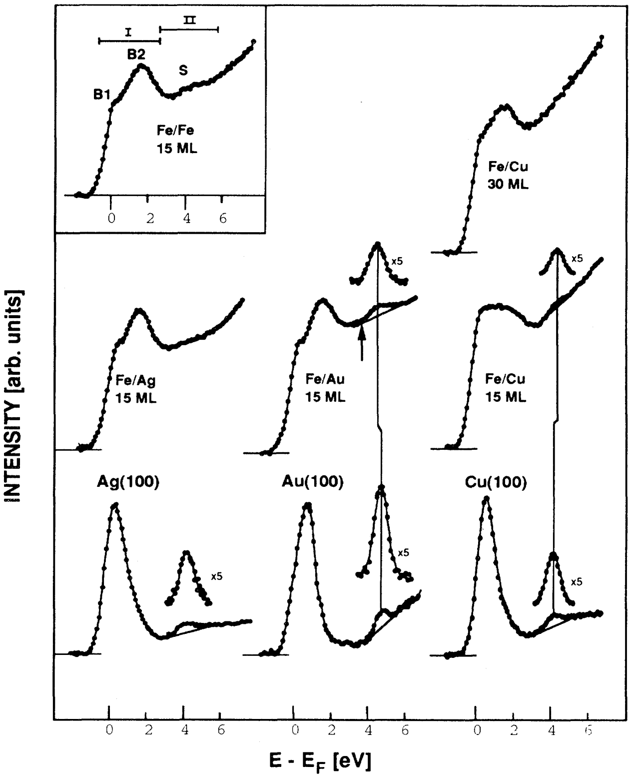

FIG. 3. Normal incidence inverse photoemission spectra from clean (100) noble-metal surfaces and from Fe/Ag(100), Fe/Au(100), and Fe/Cu(100) films. The image state peaks are also shown enlarged by a factor of 5 after a linear background subtraction. The larger intensity of the Au(100) image state is due to the fact that the spectra are presented normalized to the same height of the main peak: in Au(100) such peak is considerably reduced, since it corresponds to a symmetry forbidden transition (see also Ref. 43). The arrow in the Fe/Au(100) spectrum indicate the position of the image state in Fe(100). For a better comparison the spectrum relative to the unannealed homoepitaxial film [cf. Fig. 2(b)] is reported in the inset.

ordered films. As noted above, in this case the film quality cannot be improved by annealing because of Fe noble-metal interdiffusion. On the other hand, in Fe/Cu(100) films the IPE curve does not resemble the one of bcc iron and has to be interpreted in terms of fcc iron. The spectrum is found to change upon increasing Fe thickness, according to the fcc to bcc transition occurring above 15 ML, as shown by the data at 30 ML (Fig. 3, top panel) with the characteristic bcc-like shape.

Coming next to the energy region II, we use that the transition to image states, well visible in Fe/Au(100) and Fe/Cu(100), is instead not present in Fe/Ag(100). In consideration of the relationship between the intensity of such feature in IPE and surface order, this result implies that Fe/Ag(100) films are largely disordered, more than the Fe/Au(100) and Fe/Cu(100) films, which grow in a much more ordered layer-by-layer way. Moreover in Fe/Au(100) the image state energy is much closer to that of Au(100) than that of Fe(100) (indicated by an arrow in Fig. 3), confirming that the topmost layer is formed by segregated Au atoms. In Fe/Cu(100) similar conclusions cannot be drawn, since the image state energy of fcc Fe(100) is not known; however, when thick films are grown the surface feature disappears as expected for the more disordered thick bcc films.

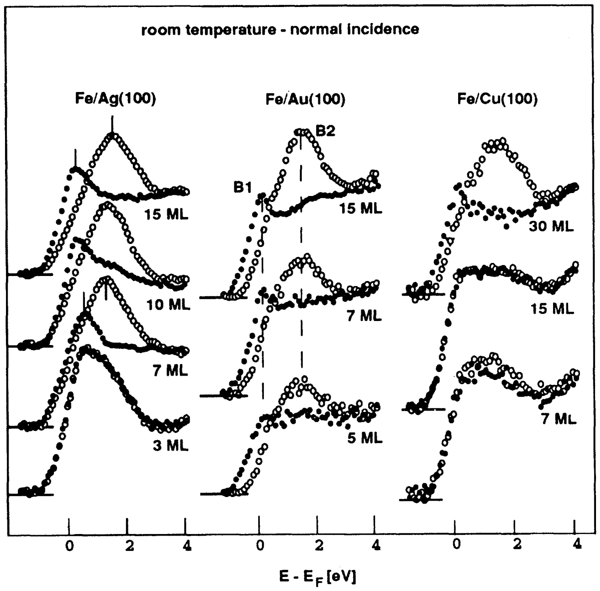

FIG. 4. Spin-resolved inverse photoemission spectra rescaled to a 100% polarized incident electron beam from Fe/Ag(100), Fe/Au(100), and Fe/Cu(100) films at different thicknesses.

C. Spin-resolved IPE

Adding spin resolution to the measurements allows to separate the transitions to majority and minority electron states, so that the structures can be directly identified and are much better resolved. Moreover new relevant information on the film magnetization is gained. In spin-resolved spectroscopies, the spin asymmetry A is defined by[13]

$$A=\frac{I_{+}-I_{-}}{I_{+}+I_{-}} ,$$

where I_{+} (I_{-}) represents the spectral intensity for primary electrons with magnetic moment parallel (antiparallel) to the film magnetization. In reality, the primary electron-beam polarization P_0 is smaller than one, so that the measured data have to be rescaled in order to reconstruct the spin-resolved spectra for a hypothetical 100% polarized beam. Such spectra, S_{+} and S_{-}, respectively, are related to the measured intensities by the correspondence[13]

$$S_{\pm}=\frac{I_{+}+I_{-}}{2}\left[1\pm\frac{A}{P_0}\right] .$$

Thus we get two spectra in which the majority and minority spin channels are completely decoupled.

The results for Fe films with different thicknesses on different substrates are collected in Fig. 4. Independently on the substrate, spectra corresponding to relatively thick bcc films (more than 10 ML on both gold and silver and more than 15 ML on copper) are very similar showing a majority spin peak at E_F and a minority one at 1.6 eV. This means that these films show a bcc bulklike behavior with the $B1$ and $B2$ features appearing in two different spin channels, as expected. Moreover, spin resolution allows us to have access to the sample magnetic properties, such as magnetic anisotropy. In fact, in reconstructing the spin-resolved spectra one has to consider the component of the beam polarization parallel to the sample magnetization. Therefore, magnetic anisotropy shows up in spin-resolved IPE taken on samples with remanent magnetization along different directions as an effective different beam polarization. The spectra in Fig. 4 have been obtained from samples magnetized along an in-plane [010] direction of the Fe lattice, which is the easy axis in bulk iron. Our results demonstrate that this holds true also in relatively thick ($\approx$15 ML) films on noble metals, as shown by data collected on samples magnetized along other directions. For instance, in films magnetized along a [011] direction the observed asymmetry in IPE spectra (not shown) is considerably reduced. This is consistent with a remanent magnetization reduced by a cos(45°) factor along the [110] direction, because of domains formation along the [010] and [001] directions, as already observed in Ni surfaces.[11] Using a reduced value for $P_0=0.25\cos(45°)\simeq 18\%$ gives rise to spectra identical to the ones of Fig. 4. The fact that the easy axis in Fe/Au(100), Fe/Ag(100), and Fe/Cu(100), relatively thick films, is along the [010] direction, as in the bulk, is a new experimental result, which is not obvious. For example in Fe/W(110) films showing bulklike electronic states, the easy axis for films thinner than 40 ML is along a $[1\bar{1}0]$ direction.[54,55]

Considering smaller thicknesses, Fig. 4 shows that the IPE spectra evolve in a substrate-dependent way when going from thin to bulklike films. Below 15 ML, the fcc Fe/Cu(100) films present identical spin-up and spin-down spectra and no polarization effect. Since our measurements are sensitive only to the in-plane component of the sample magnetization, this result indicates that at room temperature these films cannot be remanently magnetized in the film plane. Such a behavior is a direct consequence of the magnetic anisotropy forcing the magnetic momentum to lie perpendicular to the film plane, as already reported for fcc Fe/Cu(100) films at room temperature.[28,31,32] Therefore, in these films, our measurements cannot yield new relevant information with respect to already reported non-spin-resolved IPE studies.[35,36] The very small polarization effect seen in the 7 ML spectrum is probably due to the contribution of small bcc agglomerates, which is, however, overwhelmed in thicker films by the fcc-like signal. Above 15 ML bcc-like features predominate giving rise to the above discussed bulklike behavior.

Fe/Au(100) thin films display an essentially bulklike behavior already at 5 ML, with a reduction of the polarized component in the spectra in the very thin films compatible with the reduction of the Fe signal with respect to the substrate signal. In IPE from films thinner than 5 ML, new structures related to electron confinement in the film (quantum wells states) appear.[34] Such structures as well as the empty states evolution in those ultrathin films will be discussed elsewhere.[56]

In the Fe/Ag(100) case, as already reported,[41,42] the situation is more complex. In very thin films (<6 ML) no polarization effects are seen: as discussed above for Fe/Cu(100) films, this indicates that in this system too, the magnetic moment is perpendicular to the film plane. At room temperature, a transition from perpendicular to in-plane magnetization occurs around 6 ML,[25] and we indeed see polarization effects already in the 7 ML spectra. However, the energy separation between the $B1$ and $B2$ structures in thin films is reduced with respect to the bulklike value, which is approached only above 10 ML: in turn, this implies a reduction of the exchange splitting at the H point δE_{ex} in thin Fe/Ag(100) films. Similar reduction has been found in empty states of fcc Fe/Cu(100) films.[18] This finding is at variance with theoretical calculations predicting an enlargement of δE_{ex} in ultrathin free-standing films.[57,58] Moreover, spin-resolved photoemission from the Fe/Ag(100) system shows that occupied states behave in a different way, with no change in the peak position, and thus no δE_{ex} variation, when going from very thin to thick films.[22,59] It was also suggested that our results could be due to a not properly subtracted contribution of the Ag substrate overlapping the Fe spectral features.[59] In order to put in better evidence the overlayer contribution, superimposed to possibly intense unpolarized substrate emission, one can use the asymmetry spectra. In fact, being a differentially defined quantity, asymmetry is well suited to disentangle polarized from unpolarized contributions. In general, however, since the IPE intensity contains contributions from direct transitions as well as background intensity due to inelastic processes, asymmetry data have to be interpreted with caution. This is surely the case for structures with a small energy separation (less than the structure linewidth) superimposed to a large polarized background.[10,11] On the contrary, in our case, being the peaks well separated and located very near E_F, i.e., in a region where the inelastic background is very small,[47] asymmetry data can be safely used. In particular, the energy of maxima and minima in the asymmetry curve indicates the position of the majority and minority peaks, $B1$ and $B2$, respectively. Contribution to the spectra from the unpolarized substrate does not alter the peak position, as directly checked by numerical simulations considering even the unphysical extreme case of substrate/overlayer contribution ratio larger than two.

A direct comparison between the results in Fe/Ag(100) and Fe/Au(100) films is presented in Fig. 5, where asymmetry spectra corresponding to 7 and 15 ML thick Fe films are shown. As noted above, the spectra of the 15 ML films are very similar showing a maximum-minimum separation of 1.6 eV. The 7 ML spectra, however, are substrate dependent. If the substrate is Au(100), the asymmetry spectrum is essentially equal to the one of the thicker film. There is only a small reduction of the amplitude, compatible with the reduction of the $3d$ Fe signal. On the contrary, the spectrum of the 7 ML Fe/Ag(100) film clearly shows a smaller value of the $B1$-$B2$ energy separation. Note that this reduction is directly seen in our spectra and does not rely upon any complex extrapolation and/or fitting procedure. As a matter of fact, no evidence of similar effect is seen in Fe/Au(100) films.

A possible explanation for the different behavior of the two interfaces can be looked for in the fact that, at variance with Fe/Au(100) films, Fe onto Ag(100) does not form well ordered overlayers. Other mechanisms can however be important. In fact, much caution is needed when dealing with thin-film magnetism, since it is a very

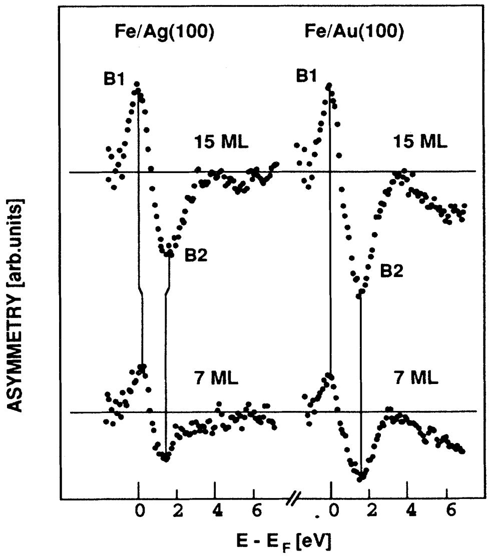

FIG. 5. Direct comparison between asymmetry spectra from Fe/Ag(100) and Fe/Au(100). The positive and negative peaks in the asymmetry correspond to the majority and minority spin bands, $B1$ and $B2$, respectively. The lines are drawn as a guide for the eye. Data have been taken at room temperature.

rich and complex subject in which many parameters are important. For instance, when performing measurements at fixed temperature (room temperature in our case), it must be considered that the Curie temperature T_C varies with thickness.[17,23,26] Therefore, reducing the film thickness corresponds to moving towards temperatures closer to T_C and then to a reduction of the sample magnetization, which can qualitatively account for the differences between the present results and those on occupied states. Approaching T_C gives rise to a **k**-dependent exchange splitting reduction,[5,60] most probably more pronounced in empty state spectroscopy (probing the Fe bands near the H point) than in occupied state spectroscopy (probing the bands near the Γ point). In fact in bulk Fe(100), occupied electron states near the center Γ of the BZ are found to be basically stationary in energy upon approaching T_C, while states with larger dispersion along the $\Gamma\Delta H$ line tend to collapse upon reaching T_C.[60] At the same time, majority and minority empty states near the H point are found to merge together with increasing temperature.[5] From the quantitative point of view, however, the situation is less satisfactory since for 7 ML films T_C has been reported to be larger than room temperature,[17,23,26] so that the above magnetization reduction mechanism seems questionable.

Another effect which can be important is the magnetic anisotropy, which in films grown on silver is different from that in films grown on gold. As discussed above, in Fe/Ag(100) films a temperature-dependent spin reorientation from perpendicular to in-plane alignment takes place. The reorientation temperature depends on the film thickness and near the transition there is a large temperature range in which the remanent magnetization is greatly suppressed.[26,61] Again, this fact can also be read, at a fixed temperature, in terms of the film thickness, so that around the transition there is a thickness region showing a reduced magnetization. In particular, at room temperature the reorientation transition takes place at about 6 ML and the reduction of the magnetization extends over a few ML.[26] An explanation of our results with reduced δE_{ex} could also be looked for in terms of the vicinity of the reorientation temperature. On the contrary, in Fe/Au(100) the magnetization lies always in the film plane and no effect is expected, in agreement with the measurements.

IV. CONCLUSIONS

In conclusion, we have presented a systematic study of the empty states in Fe films grown on the (100) surfaces of noble metals by means of angle and spin-resolved inverse photoemission. The films have been deposited and investigated at room temperature, in the thickness range from 5 to more than 20 monolayers. Information on the growth mechanism and film structure are obtained by means of adsorbed target current spectroscopy and by analyzing the surface features due to image states in IPE spectra. The measurements confirm that Fe grows in the bcc phase on Au(100) and Ag(100) and in the fcc phase on Cu(100). Above 15 ML also Fe/Cu(100) films display bcc character. The epitaxial films are found to present a poor crystalline quality with respect to bulk iron, in a substrate-depending way with increasing disorder when going from gold to copper to silver.

Experiment with polarized electrons directly show the spin character of the bands. Fe/Au(100) films display a remanent in-plane magnetization with easy axis along the Fe [010] direction as well as bulklike empty states with majority and minority spin character. This holds true also for relatively thick (≈ 15 ML) bcc films grown also on the other substrates. Instead, thin (< 6 ML) bcc Fe/Ag(100) and fcc Fe/Cu(100) films are not in-plane magnetized. Intermediate thickness (7–10 ML) Fe/Ag(100) films present a reduction of the exchange splitting with respect to the bulk value. The different behavior of the minority and majority peaks' evolution with film thickness in Fe/Au(100) versus Fe/Ag(100) thin films is clearly evidentiated and tentatively attributed to different ordering and/or different magnetic anisotropies in the films.

ACKNOWLEDGMENTS

We are grateful to L. Duo' for many useful discussions, G. L. Olcese is also acknowledged for the heating treatment of the Fe sample before insertion in vacuum. This work has been supported by the Ministero dell'Universita' e della Ricerca Scientifica through the Istituto Nazionale di Fisica della Materia.

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