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Ferromagnetic Fe on Cu(001) throughout the fcc-like phase: arguing from the viewpoint of the electronic structure

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Abstract

The scientific enthusiasm for ultrathin Fe films on Cu(001) has now lasted for more than 20 years. Is there ferromagnetic iron with a face-centred cubic (fcc) structure? Does ferromagnetism in Fe hinge on the body-centred cubic (bcc) structure? In this contribution, we try to establish that the electron system gives evidence of ferromagnetic behaviour with fcc-like electronic bands. We examine a crystal-induced surface state, which is characteristic of fcc surface order. Furthermore, we compare electronic signatures of fcc and bcc: the d-band exchange splitting, image-potential-state energies and the work function. We conclude that, from the viewpoint of the electronic structure, Fe on Cu(001) is found to be ferromagnetic throughout the fcc-like phase. This result raises a new question: how much deviation from the relaxed fcc order is acceptable without losing the electronic signature of fcc?

1. Fe/Cu(001)—a test case

Understanding and ultimately predicting the interplay between crystal structure, electronic states and collective phenomena such as ferromagnetism is one of the primary goals of solid state physicists. Unfortunately nature decided to challenge us by bestowing three different crystal structures on the three d-band ferromagnets in existence. At room temperature iron is crystallized in a body-centred cubic (bcc) structure, cobalt is hexagonal close-packed, while the crystal structure of nickel is face-centred cubic (fcc). To compare the three consecutive elements, experimentalists strive to artificially crystallize iron, cobalt and nickel in a common structure. Pseudomorphic growth on a substrate with the desired crystal structure is one of the approaches pursued.

Bulk iron, for example, undergoes a structural phase transition at 1183 K from bcc (α -Fe) to fcc (γ -Fe) and a magnetic phase transition from ferromagnetic to paramagnetic at $T_C = 1044$ K. The lattice constant $a = 3.65$ Å of γ -Fe is remarkably close to the lattice constant of Cu at room temperature with $a = 3.61$ Å [1]. Consequently the interest

in iron films grown on copper sparked the hope of stabilizing ferromagnetic fcc iron at room temperature.

Due to the uniquely complex structural behaviour and its correlation to the magnetic properties, the interest in iron films grown on the Cu(100) surface never completely abated during the last decades [2–9]. After a wealth of experimental and theoretical studies a consensus has been reached that the magnetic property of iron deposited on Cu(100) at room temperature runs through three different coverage regimes. Up to four monolayers (ML) the Fe film is ferromagnetic throughout with the easy axis of magnetization perpendicular to the film plane (regime I). The easy axis is still perpendicular to the surface between 5 and 10 ML (regime II), but only the top layers are ferromagnetically coupled while the layers below show antiferromagnetic ordering. It has been found that the magnetic structure of the underlying fcc Fe on Cu(100) in regime II most likely is a spin-density wave [7]. At still higher coverage (regime III) the film becomes ferromagnetic bcc iron with an in-plane magnetization direction. The coverage regime from 5 to 10 ML (regime II) shows an fcc structure in the body of the film [10] though there is an ongoing discussion whether

the crystal structure in regime I and the surface layers of regime II are bcc-like rather than shear-deformed fcc.

In a recent resumé by Hammer *et al* [11], Fe/Cu(001) was identified as a test case for the understanding of epitaxially grown magnetic thin films. The authors indicate that, while LEED analysis (LEED = low-energy electron diffraction) for these films may not unambiguously exclude the one or the other structure model, the scanning tunnelling microscopy (STM) studies by Biedermann and coworkers [5, 6] tilt the scales in favour of body-centred-cubic ferromagnetic iron in regimes I and II, at least at low temperatures. With this the authors of [11] conclude the part of the crystallographers and place the ball in the court of electronic structure and magnetism. We accept.

2. Integrating techniques versus local probes

Four years ago the picture for the crystal and magnetic structures of epitaxially grown iron on Cu(001) seemed consistent again. It was concluded that the integrating techniques LEED and surface x-ray diffraction were not able to distinguish unequivocally between the bcc-like $p4g(2 \times 2)$ reconstruction and the fcc-like $p2mg(2 \times 1)$ reconstruction [12, 11, 10]. With the assumption that mobile domain boundaries hide the exact atomic configuration at elevated temperature, the localized bcc-like reconstruction of the film in regime I and the top layers in regime II could only be resolved with STM at very low temperatures [12]. Certainly the amount of bcc-like reconstructed nanocrystals within the iron films seemed to fluctuate with temperature, preparation conditions and film thickness.

At a first glance the experiments measuring the magnetism of the thin iron films do not contradict the idea of ferromagnetic iron intrinsically tied to a bcc-like configuration. Studies with the magneto-optic Kerr effect, magnetization-induced second-harmonic generation, spin-polarized metastable de-excitation spectroscopy, spin-polarized appearance-potential spectroscopy and x-ray absorption spectroscopy agree on the general three-regime picture described in the previous section [2, 13, 14, 4, 15–18].

However, while the iron films for these experiments may have been prepared under slightly deviating conditions and have been analysed at different temperatures, all these experiments show comparable behaviour of the magnetism of thin iron films grown on Cu(001). What is more, no variation in the magnetization was found throughout the entire thickness range of regime II, that could not be ascribed to the spin-density wave of the underlying Fe layers. This raises the question whether ferromagnetism in iron is truly irrevocably connected with local bcc-like reconstructions. Incidentally all these techniques measuring the magnetization employ only non-local probes.

Only recently did investigations with ion beam triangulation, also an integrating technique, yield strong evidence in favour of an unreconstructed fcc surface structure for iron coverages up to 10 ML, with a possible exception of 3 ML iron, where the structure seems more distorted than at other coverages [9]. The authors indicate, however, that at lower

temperatures reconstructions consistent with the STM studies appear. It must be underlined here that so far no studies of magnetism *and* local structure investigated under identical conditions are available. Therefore a direct correlation between the amount of reconstructed bcc-like domains in the iron film and the intensity of a magnetic signal could not be shown.

In the following we will try to establish that, most interestingly, the electrons also seem to view the structure of the iron films differently, namely as ferromagnetic with fcc-like electronic bands.

3. Probing the surface electronic structure with inverse photoemission and two-photon photoemission

Since crystal structure, magnetic order and electronic states are intimately connected, we gain information about ultrathin Fe films deposited on Cu(001) via the identification of particular states and their spin dependence with the help of two techniques.

Inverse photoemission (IPE) [19, 20] and two-photon photoemission (2PPE) [21, 22] probe unoccupied electronic states above the Fermi energy E_F within the surface region of the sample. With spin resolution, these techniques are also sensitive to surface magnetic properties. The information depth is, in principle, limited by the inelastic mean free path of electrons in the solid at a given electron energy. In our case, the kinetic energy is below 15 eV and therefore the probing depth is a few atomic layers only. As a consequence, bulk as well as surface-derived states are accessible. Measuring particular states, such as surface states with wavefunctions concentrated at or even outside the outermost atomic layer, these states and their spin dependence provide information about the topmost layer.

In IPE experiments, unoccupied electronic states are detectable via radiative transitions into these states when the surface is bombarded with electrons. With the use of spin-polarized electrons for excitation, the majority and minority states are populated independently [23]. The experimental energy resolution ranges typically from 700 to 300 meV [24]. Consequently, the measured linewidths may be dominated by the apparatus function rather than revealing the intrinsic linewidths. Nevertheless, exchange splittings of some tens of meV are detectable because the two partial spin spectra are measured separately.

In 2PPE experiments, unoccupied electronic states between E_F and the vacuum level E_V are populated by electrons from below E_F via a laser pumping pulse. With a second laser pulse, electrons in these intermediate states are excited to energies above E_V and detected after passing through an electron energy analyser. In comparison with IPE, 2PPE has a superior energy resolution and, therefore, provides the means to measure intrinsic linewidths, which are only of the order of some tens of meV. Since our 2PPE set-up is equipped with a spin-polarization detector, the majority and minority spin components of the respective states can also be separated [25, 26].

4. A characteristic fcc feature: the crystal-induced surface state at \bar{X}

Face-centred cubic crystals of noble and transition metals with a (001) surface exhibit a characteristic crystal-induced surface state SS within a gap of the surface-projected bulk-band structure. This gap is located mostly above the Fermi level, originates from a gap at the L high-symmetry point between L_2' and L_1 , and appears around the \bar{X} point of the surface Brillouin zone.

Figure 1 displays inverse-photoemission spectra for Cu(001), Ni(001), 6 ML Fe/Cu(001) and 21 ML Fe/Cu(001), taken at an angle of electron incidence of 50° relative to the surface normal. At this angle, electron states with energies of about 4 eV above E_F are probed close to the \bar{X} point. The spectrum for Cu(001) [27] is dominated by the surface-state emission SS at 3.6 eV and shows an additional surface-related feature SR originating from a surface resonance. The projected bulk-band structure for Cu(001) with the surface-state dispersions $E(k_{\parallel})$ along $\bar{\Gamma}\bar{X}$ is given as an inset in figure 1 [28]. The spectrum for Ni(001) [29] resembles the Cu spectrum closely with only a few distinct differences: minority d states (d_{\downarrow}) appear above the Fermi level, leading to a peak just above E_F , SS and SR appear at somewhat higher energies and the surface-state energy becomes spin-dependent. The exchange splitting of this sp-derived surface state SS amounts to 0.18 ± 0.06 eV.

All spectroscopic features of the Ni surface can also be found in the IPE spectra for 6 ML Fe on Cu(001) [30, 31]. The surface state on the 6 ML Fe film is exchange split by 0.92 ± 0.03 meV, which is five times as much as the surface state at Ni(001). This reflects the larger exchange splitting of the magnetic d bands in Fe compared with Ni. In addition, the minority d holes appear at higher energies and the majority d states, which are totally occupied in Ni, become partially unoccupied for Fe.

Comparing inverse-photoemission data for 6 and 21 ML of Fe on Cu(001) [27], however, reveal remarkable differences. First, minority and majority d states appear in the inverse-photoemission spectrum, yet with a larger exchange splitting for 21 ML than for 6 ML of Fe on Cu(001). We will discuss the d-band splitting in more detail in section 5. The second, more important, difference between the IPE spectra for 6 and 21 ML of Fe on Cu(001) is the existence or rather non-existence of the surface states⁴. Hence, the surface electronic structure of 6 ML of Fe on Cu(001) is fully compatible with an fcc(001) surface, while the surface resonance SR and the surface state SS are conspicuously absent for 21 ML Fe/Cu(001), which are known to have a bcc structure.

It was assumed in the literature that bcc-like reconstructed nanocrystals within the fcc-like film are responsible for the ferromagnetic behaviour for film thicknesses below the transition from region II to region III. We do, however, observe the surface state SS for film thicknesses below this transition

⁴ It must be emphasized here that the disappearance of SS cannot be attributed to a loss of order at the surface of the 21 ML film, because IPE spectra of the same film at normal emission clearly show the image-potential-state (IS) feature, a signature of a well-ordered flat surface [27].

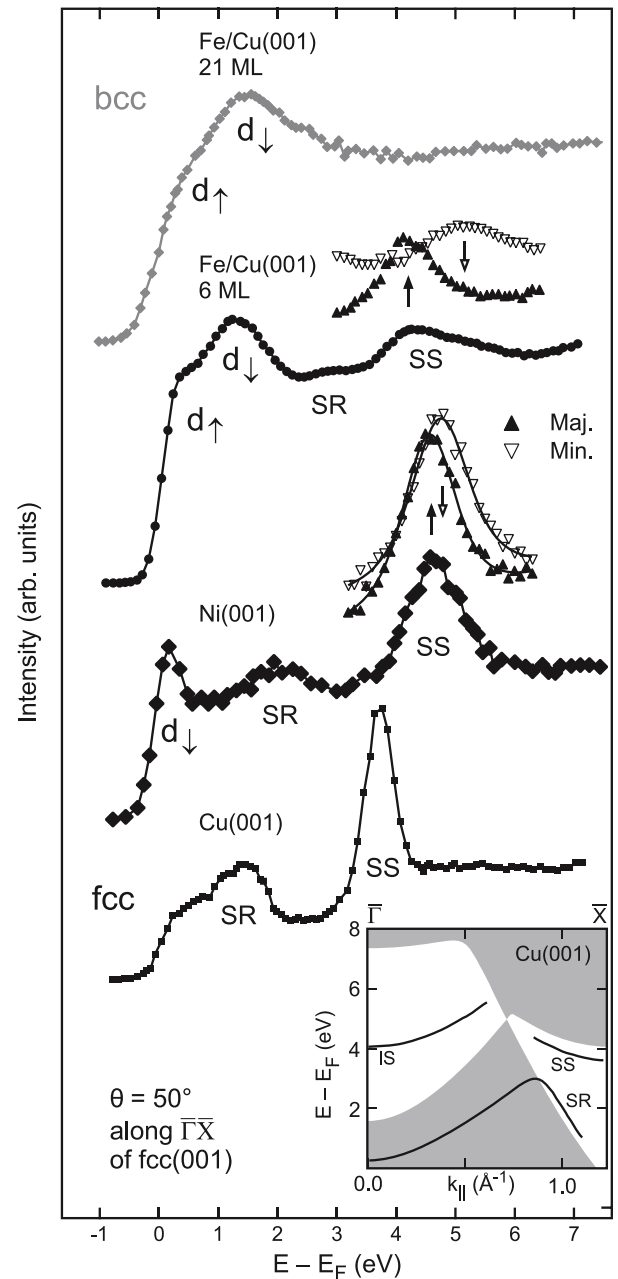


Figure 1. Inverse-photoemission (IPE) spectra for Cu(001) [27], Ni(001) [29], 6 ML of Fe/Cu(001) [30] and 21 ML of Fe/Cu(001) [27]. All data have been taken at an angle of electron incidence of 50° along $\bar{\Gamma}\bar{X}$ of fcc(001). For 21 ML of Fe/Cu(001), the iron film consists of bcc(110) crystallites, which grow in two non-equivalent orientations relative to the fcc substrate. All spectra of the fcc samples show a characteristic surface-state feature SS between 4 and 5 eV above E_F , which is absent for the bcc Fe film. The states appear in a gap of the projected bulk band structure around \bar{X} . The inset shows the situation for Cu(001) with energy gaps (non-shaded areas) and surface-band dispersions [28]: image-potential state (IS), surface state (SS) and surface resonance (SR). For Ni and Fe, the surface states (SS) exhibit a magnetic exchange splitting, which is revealed by spin-resolved measurements. d_{\uparrow} and d_{\downarrow} indicate the majority and minority d bands, respectively.

but not above. Is our observation of SS compatible with the assumption of a bcc-like film? In order to search for bcc signatures within the surface electronic structure, we

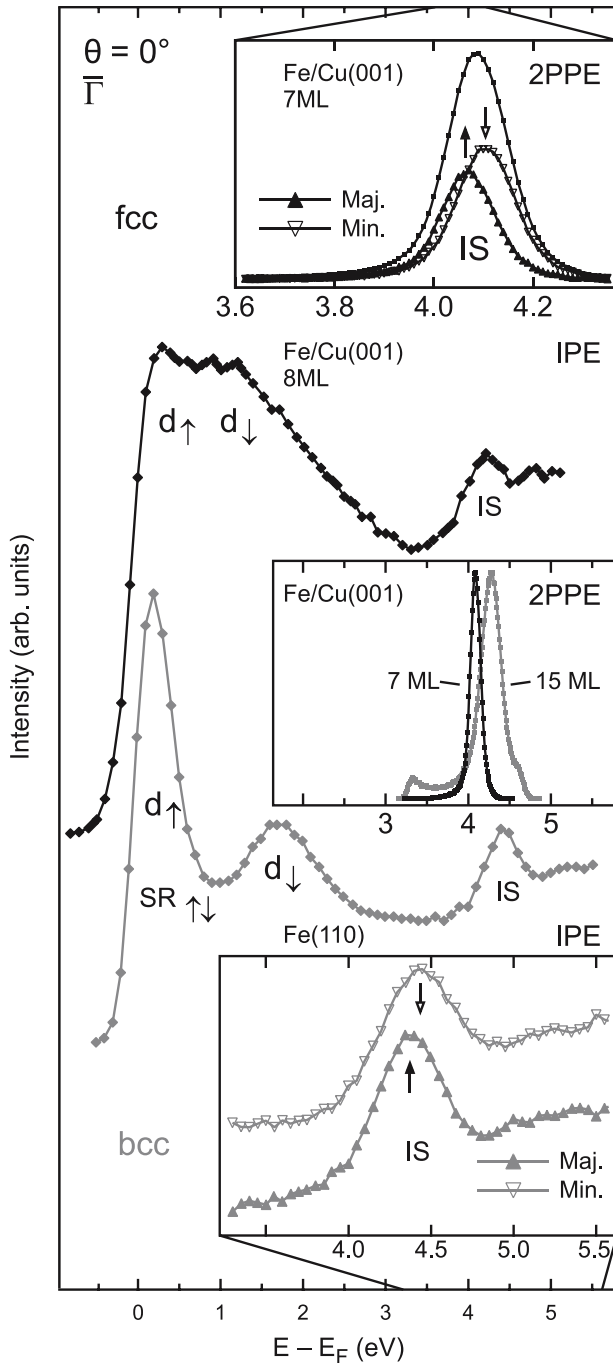


Figure 2. Inverse-photoemission (IPE) [27, 37] and two-photon-photoemission (2PPE) spectra for normal electron incidence/emission for 7 and 15 ML of Fe on Cu(001) as well as for bcc Fe(110). The spectra exhibit minority (\downarrow) and majority (\uparrow) d holes just above the Fermi level and the $n = 1$ image-potential surface state IS some hundred meV below the vacuum energy. Spin-resolved measurements reveal the exchange splitting of IS [41, 26]. Note that in the topmost inset for 2PPE the energy scale is enlarged by a factor of six, while in the bottom inset for IPE by a factor of two.

investigated energy gaps of the bcc band structure. The existence of energy gaps in the projected bulk-band structure is a necessary condition for surface states. According to structural models for substrate and film surfaces, the

relaxed α -Fe bcc(110)-like structure forms with an angle of 9.25° with respect to the fcc[100] direction [32]. The presented measurements are performed along $\bar{\Gamma}\bar{X}$ of fcc(001). Depending on the orientation of the bcc crystallites, this direction is along $\bar{\Gamma}\bar{S}$ of the bcc(110) surface Brillouin zone, or 18.5° off this direction. Early tight-binding calculations of the energy bands of a 29-layer (110) ferromagnetic bcc iron film predicted an energy gap around \bar{S} with a surface band in the energy range of interest [33]. No energy gaps were found in the respective energy range around the \bar{H} or \bar{N} high-symmetry points of bcc(110) [33, 34]. More recent band structure calculations for a 23-layer Fe(110) film along $\bar{\Gamma}\bar{S}$ confirm the existence of an energy gap but do not find a surface band [35]. In agreement with this, our measurements for the bcc films do not show any emission features in the energy range, where SS was detected for fcc(001) surfaces.

Therefore, we have a clear experimental fingerprint of an fcc(001) surface structure versus bcc(110) films. We do detect SS for films below the fcc-to-bcc transition, and no surface-state feature is observed for bcc films above the transition. In addition, the occurrence of the exchange-split surface state for film thicknesses below about 10 ML is a strong argument for the fcc structure of ferromagnetic Fe films.

Our arguments so far are based on energy gaps in the projected bulk-band structure. Let us now assume that bcc-like reconstructions form only at the surface on top of an fcc underlayer. As a consequence, the fcc bulk-band situation may be valid, although the surface layer has a bcc-like structure. Then, perhaps, the fcc surface state appears despite the surface layer with its different symmetry. This hypothesis can easily be refuted by the results for a comparable situation in the literature. The interesting case of a hexagonal surface layer on top of a substrate with quadratic order was studied for Pt(001) in its unreconstructed (1×1) and reconstructed (5×20) phase [36]. The bulk-band transitions were found to be intensity modulated by the reconstruction but their energy positions remained stationary. The surface band at \bar{X} , however, which is characteristic of fcc(001), was only observed for the unreconstructed surface. The surface layer with different surface order than the fcc(001) substrate did not show the surface state. We conclude that, in an analogous way, a bcc-like surface reconstruction on an fcc(001) substrate would quench the surface state.

To summarize our observations so far, the electronic structure of 6 ML and 21 ML of Fe on Cu(001) is distinctly different. While for 6 ML a surface state is observed, which is characteristic of fcc, the 21 ML film does not show the surface state in accordance with theoretical predictions for bcc Fe(110). This is a qualitative argument from the viewpoint of the electronic states for the fcc-like structure of 6 ML of Fe on Cu(001).

5. Signatures of fcc and bcc: d-band splitting and surface barrier

We have already seen that fcc and bcc Fe films differ qualitatively in their electronic structure. In this section, we aim at deducing quantitative differences by focusing on the

Table 1. Summary of experimentally determined image-potential-state energies relative to E_F and work functions Φ for bcc- and fcc-like Fe obtained from inverse photoemission (IPE) and two-photon photoemission (2PPE).

System	$E - E_F$ (eV)	Φ (eV)	Technique	Ref.
7 ML Fe/Cu (001)	4.1		IPE	[38]
7 ML Fe/Cu (001)	4.09 ± 0.02	4.77 ± 0.02	2PPE	This work
8 ML Fe/Cu (001)	4.15 ± 0.10		IPE	[27]
15 ML Fe/Cu(001)	4.19	4.72	2PPE	[45]
15 ML Fe/Cu(001)	4.28 ± 0.02	4.84 ± 0.02	2PPE	This work
Fe(110)	4.4 ± 0.15		IPE	[43]
Fe(110)	4.39	5.12	2PPE	[44]
Fe/W(110)	4.4		IPE	[41]
Fe/W(110)	4.32	5.02	2PPE	[45]

magnetic d bands and the surface barrier, i.e. work function and image-potential surface states (IS).

Figure 2 presents IPE and 2PPE measurements for normal electron incidence/emission. IPE spectra for 8 ML of Fe on Cu(001) [27] are compared with spectra for bcc Fe(110) [37]. It might not be unambiguous to deduce an exchange splitting from spin-integrated measurements. Nevertheless, the spectra indicate a clearly smaller d-band splitting for fcc-like films than for bcc films. For 8 ML Fe, the d-band exchange splitting was estimated to be 1.1 ± 0.25 eV [27] and, in a different work, 1.2 ± 0.1 eV [38]. For bcc Fe(110), the IPE data of figure 2 show two peaks, one just above E_F , the other at 1.7 eV above E_F . Spin-resolved IPE measurements reveal that the first consists of a majority d state and a surface resonance with both spin components [39], while the latter represents a minority d state. The majority d state is only partially empty. Therefore, the data give a lower limit of 1.7 eV for the exchange splitting in bcc films, which is significantly larger than in the fcc-like case.

The additional spectral feature found in the spectra of figure 2 is the $n = 1$ image-potential state IS. It is part of a Rydberg-like series of states, which are pinned to the vacuum level with binding energies lower than 0.85 eV [40]. The spectra obtained with spin-resolved IPE from bcc Fe films on W(110) (bottom inset of figure 2) reveal an exchange splitting of 57 ± 5 meV [41]. In the spin-resolved 2PPE spectra of fcc-like 7 ML Fe/Cu(001), shown in the topmost inset of figure 2, the image-potential state has a slightly smaller exchange splitting of 40 ± 10 meV, a trend that mirrors the d-band behaviour [42]. Of course this unambiguously tells us that the surface of the 7 ML Fe film, while different in structure from a bcc surface, nevertheless is ferromagnetic.

Several IPE and 2PPE studies of the $n = 1$ image-potential state have been performed on bcc Fe(110) and Fe films deposited on Cu(001) and W(110). A summary of the image-potential-state energies relative to E_F and, where determined, the work function Φ of the sample are given in table 1.

The studies agree that IS appears at around 4.1 eV above E_F for Fe films on Cu(001) in regime II. In regime III, the image-potential state shows up at about 0.2 eV higher in

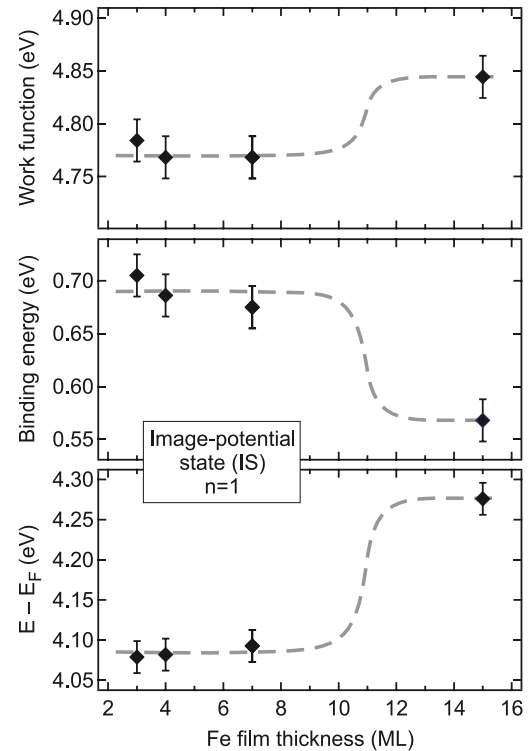


Figure 3. Work function, image-potential-state energies relative to E_F and binding energies relative to E_V for ultrathin Fe films on Cu(001) as a function of the film thickness as obtained from two-photon-photoemission measurements. The dashed lines are guides to the eye.

energy, while for bcc Fe(110) the energies are again 0.2 eV higher. The work function follows this trend qualitatively⁵. Our 2PPE spectra for 7 and 15 ML of Fe on Cu(001), presented in the middle inset of figure 2, clearly show that the $n = 1$ IS appears at distinctly higher energy relative to E_F for regime III (15 ML, bcc) than for regime II (7 ML, fcc-like).

The results of our 2PPE measurements for Fe films on Cu(001) are summarized in figure 3 as a function of the film thickness: work function, image-state energy with respect to E_F and binding energy with respect to E_V . There is a clear trend: fcc-like films have a lower work function that leads to a lower image-state energy with respect to E_F , yet with a higher binding energy. The transition from the fcc-like to the bcc-like electronic structure has an unmistakable effect on the IS appearance.

6. Conclusion

After the crystallographers have done their part, we investigated the electronic structure and the magnetism. As criteria we used the appearance of a crystal-induced surface state, the size of the d-band exchange splitting and the surface barrier. The latter was probed via image-potential states and

⁵ Please note that the experimental results of [45] show some deviation with respect to the other data. Since both their energy values and work functions deviate consistently by about 0.1 eV to lower energy, the described trend is supported by these results as well.

the work function. All results give the same message: Fe films deposited on Cu(001) at room temperature in regime II (5–10 ML) differ significantly from bcc films in regime III (more than 10 ML). We did not observe any signature from the bcc phase: our results are fully compatible with an fcc surface crystallographic order. Since it is known that the surface order differs from relaxed fcc, the results on the electronic structure raise a new important question. How much deviation from the relaxed fcc order is acceptable without losing the electronic signatures for fcc? Furthermore, the simple conclusion that only bcc-like Fe is ferromagnetic, while fcc-like Fe is not, remains questionable. From the viewpoint of the electronic structure, Fe on Cu(001) is found to be ferromagnetic throughout the fcc-like phase.

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