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Electronic and magnetic properties of the fcc Fe(001) thin films: Fe/Cu(001) and Cu/Fe/Cu(001)

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All-electron total-energy local-spin-density studies of the electronic and magnetic properties of fcc Fe(001) as overlayers or sandwiches with Cu(001) were undertaken in order to understand the following: (1) the surface (interface) magnetism of fcc Fe(001), (2) the effect of nonmagnetic Cu on the magnetization of Fe and (3) the effect of the reduced coordination number on the magnetic coupling of Fe layers near the surface and interface. From our systematic studies of (i) one and two layers of Fe on Cu(001) and (ii) one and five layers of Fe sandwiched by Cu, it is concluded that the Fe magnetic moment is enhanced on the surface (to $2.85\mu_B$) and the surface (interface) Fe layer is predicted to couple ferromagnetically to the subsurface (subinterface) Fe layer (in contrast to the antiferromagnetic behavior in the bulk fcc Fe). The effect of the nonmagnetic Cu overlayers decreases slightly (by $0.25\mu_B$) the magnetic moments of Fe at the Fe/Cu interface from those of the free-standing surfaces, indicating the persistence of the two-dimensional magnetization at the interface. Magnetic hyperfine fields are compared among various magnetic states; the interface Fe atoms are found to experience a larger hyperfine field than the inner layers for the magnetic ground state due to the retention of antiferromagnetic coupling between Fe layers away from the interface. Electronic charge-density, work-function, and single-particle spectra are presented and discussed. The calculated energy dispersions agree well with a recent photoemission measurement by Onellian *et al.*

I. INTRODUCTION

The study of the metastable states of magnetic $3d$ transition metals has been a subject of great interest. Recent progress in the fabrication and stabilization of new phases of metals has added more excitement to this area.[1] This development offers enormous opportunities for synthesizing metastable crystalline phases with novel physical properties to be specified, and new phenomena to be investigated. A prototypical example is fcc Fe which can be stabilized at low temperatures either by the formation of γ-Fe precipitates in a Cu matrix[2] or as a thin film grown epitaxially on a Cu substrate.[3]

Experimental investigations of γ-Fe precipitates in Cu and stainless steel reveal that the spin vector is oriented parallel to the $\langle 001 \rangle$ direction and that the magnetic moments in alternating ferromagnetic (001) sheets are coupled antiferromagnetically to each other with a size-dependent Néel temperature.[4–6] However, for very small-sized precipitates ferromagnetic ordering was found by Mössbauer spectroscopy[7] and susceptibility measurements.[8] While the antiferromagnetic behavior of γ-Fe precipitates at low temperatures has been well established, controversies concerning the magnetic ordering near the fcc Fe surface or at the Cu/Fe interface is not a settled issue at present.

For a decade now, the magnetic ordering of fcc Fe films has been a subject of considerable controversy: Wright[9] found that epitaxial Fe films are ferromagnetic when prepared on Cu(110); the same conclusion was obtained by Kummerle and Gradmann[10] for Fe/Cu(111), whereas, Keune *et al.*[11] found that fcc Fe(001) is antiferromagnetic when prepared on Cu(001). On the other hand, Mössbauer measurements by Halbauer and Gonser[12] concluded that the three basic orientations (111), (110), and (001), when prepared by epitaxial growth, are all antiferromagnetic. This conclusion, however, is contradicted by results of a recent measurement using

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electron-capture spectroscopy by Rau,[13] which supports a long-range ferromagnetic order at the surface of fcc Fe(111)/Cu(111).

The possible difference in the magnetic behavior between γ-Fe precipitates and the fcc Fe surface may be explained mostly by the difference in the measured lattice parameters (i.e., 3.61 Å for fcc Fe/Cu, and 3.59 Å for precipitates)[9,10,14] and the strong dependence of the bulk magnetic ordering on this small lattice-parameter difference.[15] However, little attention has been paid to surface and interface effects on the electronic and magnetic structure of fcc Fe layers near the surface or at an interface. The reduction of the coordination number and the lower symmetry may lead to physical properties which are different from those of their bulk counterparts: For example, magnetic phase transitions for Cr(001) (Refs. 16 and 17) and Gd(0001) (Ref. 18) surfaces, surface reconstruction of W(001) (Ref. 19), etc. The size dependence of the Néel temperature and magnetic ordering for γ-Fe precipitates clearly indicates the importance of surface-(interface-) related properties in accounting for the difference in their electronic and magnetic behavior. It is the purpose of this paper to ellucidate the ground-state electronic and magnetic properties of metastable fcc Fe thin films as overlayers and as sandwiches with Cu(001) using a local spin-density-functional total-energy approach.[20]

To this end, we present results of a highly precise self-consistent, all-electron total-energy full-potential linearized-augmented-plane-wave[21,22] (FLAPW) local spin-density-functional study of the electronic and magnetic properties of fcc Fe(001) as overlayers on the Cu(001) substrate or as sandwiches with Cu(001). In Sec. II, the crystal structure and the calculational model used in our investigation are described briefly. Results on the magnetic structures and the magnitude of magnetic moments are presented in Sec. III, together with a comparison of total energies for possible magnetic states in both overlayers and sandwiches. Charge density and work function are given in Sec. IV. Calculated values of Fermi-contact hyperfine fields for various magnetic states, which can be used in direct comparison with Mössbauer measurements, are given in Sec. V. The single-particle spectra for fcc Fe as overlayers, which are more pertinent to photoemission measurements, are presented in Sec. VI.

II. STRUCTURE AND CALCULATIONAL MODEL

One notable feature of the Fe/Cu interface is that since Cu and Fe do not form solid solutions, a sharp interface is to be expected. Low-energy electron diffraction (LEED) studies have verified that thin Fe layers, deposited on single-crystal films of Cu, form into an fcc structure with a lattice constant that is exactly matched to that of the Cu substrate.[3,23] More recently, epitaxial growth of Fe on Cu(001) was further examined by Onellion *et al.*[24] This experimental analysis shows that Fe grows predominantly in a layer-by-layer mechanism, with thicknesses ranging from 1 to 4 monolayers stabilized by the Cu substrate. Because of its well-characterized structure, the Fe/Cu interface and fcc Fe thin films have provided a challenging opportunity for theoretical studies of the correlation between magnetism and crystal structure.

We have carried out extensive studies on the following systems: (1) one layer of Fe atop both surfaces of a five layer Cu(001) slab [1Fe/Cu(001)]; (2) two layers of Fe on either side of a three layer Cu(001) slab [2Fe/Cu(001)]; (3) case (1) but now the 1Fe/Cu(001) is further covered on each side by one layer of Cu [Cu/1Fe/Cu(001)]; (4) five layers of an fcc Fe slab sandwiched by two layers of Cu(001) on both sides [Cu/5Fe/Cu(001)]. The Fe has the fcc structure with a lattice parameter equal to that of bulk Cu (3.61 Å); the assumed stacking has atoms in the fourfold hollow site of adjacent atomic-layer planes. The Cu-Fe interlayer spacing is determined from total-energy calculations.

The local-spin-density equations are solved self-consistently by use of the FLAPW method. In this method, no shape approximations are made to the potential or charge density in solving Poisson's equation for a general potential and all matrix elements corresponding to this general potential are rigorously taken into account in all parts of space. All core electrons are treated fully relativistically, and the valence states (originating from the atomic $4s$, p, and $3d$ orbitals) are calculated semirelativistically.[25] These procedures allow a precise description of the spin density at the nucleus (Fermi-contact term) for the interpretation of hyperfine fields measured by Mössbauer spectroscopy.[26]

A total of 550 (650) augmented plane waves are used as a variational basis set for the seven- (nine-) layer films, split into two blocks using the mirror-plane reflection symmetry about the center layer of the slabs (z reflection). Within the touching muffin-tin (MT) spheres, the charge density and the potential are expanded in lattice harmonics with angular-momentum components $l \leq 8$. For the exchange-correlation potential we employ the explicit form of von Barth and Hedin.[27]

III. TOTAL ENERGY STUDIES: STRUCTURE AND MAGNETISM

A. Overlayers: 1Fe/Cu(001) and 2Fe/Cu(001)

We first examine the Fe-Cu interlayer spacing for a monolayer of Fe deposited on the Cu(001) substrate. Results based on our total-energy calculations indicate that the Fe-Cu interlayer distance is 1.75 Å, which is about 3% contracted from that of fcc Cu (1.805 Å). This total-energy calculation gives evidence that an Fe layer on Cu(001) tends to form in an fcc structure with a lattice constant sufficiently close to that of Cu. The epitaxial growth of fcc Fe on (or sandwiched by) Cu(001) has been well established by LEED.[3] As we have demonstrated earlier, there is a tendency for the formation of strongly enhanced magnetic moments for $3d$ transition metals as overlayers or sandwiches with noble metals.[28] The monolayer of Fe on Cu(001) is no exception; a moment of $2.85\mu_B$ is developed on the Fe site with a spin-polarization energy lowering that amounts to 0.70 eV. Furthermore, a moment of $0.04\mu_B$ is induced on the adjacent Cu site. Apparently, the hybridization between Fe and Cu is much smaller than expected. As we will discuss

later, this enhancement is related to the localized interface states near E_F.

Next, we examine how the electronic and magnetic structure of this ferromagnetic Fe monolayer is modified by an additional Fe layer co-adsorbed on Cu(001). For bulk fcc Fe, the stable magnetic structure is antiferromagnetic (AFM), and the ferromagnetic (FM) phase may exist as a metastable state depending on the lattice constant in question.[15] Thus, in order to study the surface magnetism of fcc Fe, the immediate question is whether AFM coupling persists near the surface. This can only be answered through total-energy calculations by comparing the spin-polarization energy between different magnetic states. Our calculation shows that, similar to bulk γ-Fe, both AFM and FM coupling between Fe layers are lower in energy than the case of paramagnetic coupling and so both can exist for γ-Fe bilayers at the surface. However, in sharp contrast to bulk γ-Fe, we find that the FM state has the lowest total energy (the total energy is lower by 0.2 eV than the AFM state). In other words, there is a magnetic phase transition for γ-Fe near its surface, i.e., a reduction of coordination number tends to favor the FM coupling.

In Fig. 1 we present the calculated spin densities for the FM and AFM states on the (110) plane of 2Fe/Cu(001). These plots display the anisotropic character of the spin densities; the spin polarization is more pronounced and extended into the vacuum regions for the FM state. The integrated spin densities within the touching muffin-tin (MT) spheres are listed in Table I. Large moments of $2.83\mu_B$ and $2.58\mu_B$ are found for the surface and subsurface Fe layers of FM 2Fe/Cu(001). A comparison between 1Fe/Cu(001) and FM 2Fe/Cu(001) results shows a similar magnetization on the surface layer in both cases.

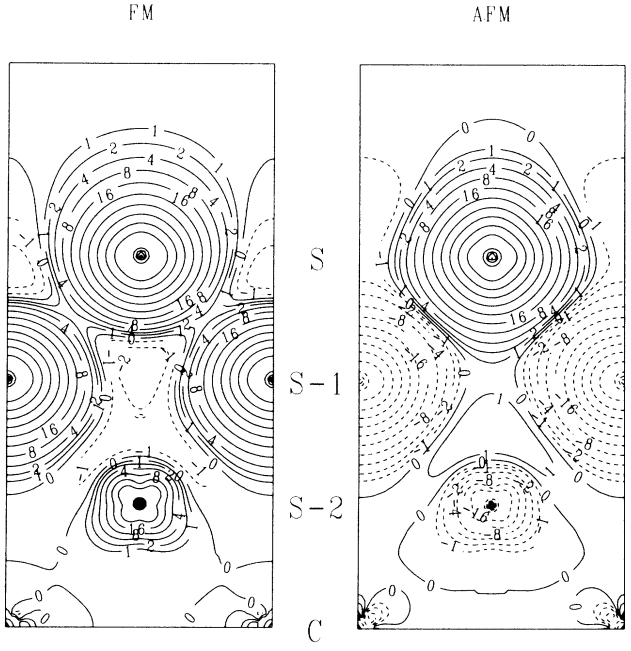

FIG. 1. Spin density of a 2Fe/Cu(001) slab in the (110) plane perpendicular to the surface in units of $10^{-3}e$/a.u.^{3} for the ferromagnetic (FM) and antiferromagnetic (AFM) states.

TABLE I. Layer-by-layer magnetic moments (in μ_B) and the energy lowering (in eV) relative to the paramagnetic state for the Fe overlayers and sandwiches.

	Magnetic moments (μ_B)			ΔE (eV)
	S	S-1		
1Fe/Cu(001)	2.85			−0.70
2Fe/Cu(001)				
FM	2.85	2.60		−0.79
AFM	2.38	−2.22		−0.58
	Magnetic moments (μ_B)			ΔE (eV)
	IS	IS-1	IS-2	
Cu/1Fe/Cu(001)	2.60			−0.48
Cu/5Fe/Cu(001)				
(+ + −)	2.68	2.31	−1.45	−0.78
(+ − +)	2.31	−1.16	1.76	−0.69
(+ + +)	2.60	2.23	1.99	−0.62

This indicates that localization plays the same role in determining the size of the magnetic moment as the magnetic exchange coupling between Fe layers. For the metastable AFM state, smaller moments (but comparable to that of bulk Fe) of $2.38\mu_B$ and $-2.22\mu_B$ are found for the surface and interface layers.

B. Magnetism of γ-Fe sandwiched by Cu

1. Cu/1Fe/Cu(001)

Since Fe is a notorious getter, the Fe layers are often covered by a metallic overlayer, such as an additional Cu layer, to make the experimental observation of magnetization easier. On the theoretical side, it is also interesting to study the interface magnetism of thin Fe films in a sandwich environment.

We start with a discussion of Cu/1Fe/Cu(001). As in the case of an overlayer, the Cu-Fe spacings are set equal to that of fcc Cu. Surprisingly, the effect of the Cu overlayer is to only slightly reduce the moment by $0.25\mu_B$ (to $2.60\mu_B$). The retention of two-dimensional (2D) magnetism (despite the hybridization between Cu and Fe) is surprisingly very similar to that found earlier in the Au/Cr/Au, Au/Fe/Au, and Ag/Fe/Ag systems.[28]

2. Cu/5Fe/Cu(001)

As just described, the effect of the Cu overlayers is to partially reimpose the bulk boundary condition for the Fe layer at the interface. This produces a small decrease of the magnetization of the Fe layer at the Cu/Fe interface. A second and more important consequence will be how the magnetic coupling between Fe layers in multilayer films are affected by the Cu overlayers. To study this, we model the sandwich by a single slab consisting of nine layers—five layers of fcc Fe sandwiched by two layers of Cu(001) on both sides. We therefore have four possible magnetic couplings between the interface (IS), subinterface (IS-1), and center (IS-2) Fe layers; for convenience,

TABLE II. Work function (in eV) for the overlayers and sandwiches studied along with the total valence charge within muffin-tin spheres in the different layers.

Overlayers	S	S-1	S-2	S-3	Work function (eV)
1Fe/Cu(001)	6.93	10.42	10.37	10.37	5.00
2Fe/Cu(001)					
FM	7.01	7.20	10.41	10.37	4.95
AFM	7.01	7.20	10.41	10.37	5.10
Sandwiches	S	S-1	S-2	S-3	S-4
Cu/1Fe/Cu(001)	10.24	7.14	10.42	10.37	
Cu/5Fe/Cu(001)					
$(+\ +\ -)$	10.21	10.41	7.18	7.25	7.25
$(+\ -\ +)$	10.21	10.41	7.19	7.26	7.25
$(+\ +\ +)$	10.21	10.41	7.19	7.25	7.25

we adopt $(+\ +\ +)$, $(+\ +\ -)$, $(+\ -\ +)$, and $(+\ -\ -)$ to indicate various spin orientations for the sequence (IS, IS-1, IS-2). Spin-polarization energies (i.e., the difference in total energy from the paramagnetic state) are compared among these states in order to obtain the stable ground-state spin configuration. The calculated results are summarized in Table I. Of all the possible spin configurations, the $(+\ +\ -)$ state has the lowest total energy, i.e., the most stable state is that in which the IS and IS-1 layers are coupled ferromagnetically, whereas antiferromagnetic coupling is found for the IS-1 and IS-2 layers (as in bulk γ-Fe). The absolute values of spin-polarization energies (ΔE) are in the following order: $\Delta E(++-)>\Delta E(+-+)>\Delta E(+++)$. [A comparison among these three states suggests that the $(+\ -\ -)$ state is the least favored state.] Although the effect of Cu overlayers decreases the difference in ΔE between the different magnetic states from that of the overlayers (cf. Table I), the FM spin coupling between IS and IS-1 is not changed from that found for the bilayers of γ-Fe on Cu(001). Furthermore, a bulklike AFM magnetic structure appears to be retained between IS-1 and IS-2, indicating that the interface discontinuity is screened out in about 2 atomic layers.

Consider now the calculated magnitude of the magnetic moments. Results obtained from the self-consistent calculations again show that the magnetic moment is enhanced at the interface (cf. Table I). For the stable $(+\ +\ -)$ state, moments of $2.68\mu_B$, $2.31\mu_B$, and $-1.45\mu_B$ are obtained for the IS, IS-1, and IS-2 layers, respectively. It is interesting to note that the moment on the IS-2 layer is very close to that of bulk γ-Fe at the Cu lattice constant ($\simeq 1.5\mu_B$).[15] For the $(+\ +\ +)$ FM state, we have attempted to explore the possibility of the coexistence of two spin states as in bulk fcc Fe.[15] However, only one unique FM state is found for the thin five layer fcc Fe film.

3. Discussion

Our systematic analysis shows that the reduction of the Fe coordination numbers for γ-Fe layers at the surface or interface changes the magnetic coupling between the Fe layers in the $\langle 100 \rangle$ direction from the observed bulk AFM coupling to a FM coupling between S (IS) and S-1 (IS-1) layers. This behavior is consistent with the ferromagnetic ordering for small γ-Fe precipitates (in which the total number of atoms is less than 12) in a Cu matrix.[7] This behavior can also account for the ferromagnetic ordering of the fcc Fe(110) (Ref. 9) and fcc Fe(111) (Refs. 10 and 13) surfaces [the most open surface for fcc structure is (110), whereas the (111) orientation is the most close packed], instead of invoking the dependence of the surface magnetic structure on the lattice parameters as in bulk fcc Fe. Another way to see this is that the decrease of mag-

TABLE III. Fermi-contact hyperfine fields (in kG) given by Fe layer and separated into valence- and core-core contributions. The last column gives the core polarization field per unpaired moment (in kG/μ_B).

	H_{CF} (kG)			H_{CF}/M (kG/μ_B)
	Total	Valence	Core	
		1Fe/Cu(001)		
S	−157	208	−364	129
		FM 2Fe/Cu(001)		
S	−249	116	−366	129
S-1	−316	16	−333	129
		AFM 2Fe/Cu(001)		
S	−85	219	−304	128
S-1	70	−210	280	127
		Cu/1Fe/Cu(001)		
IS	−230	102	−342	131
		$(+\ +\ -)$ Cu/5Fe/Cu(001)		
IS	−292	69	−361	134
IS-1	−237	71	−308	133
IS-2	−51	−243	192	132
		$(+\ -\ +)$ Cu/5Fe/Cu(001)		
IS	−147	160	−308	133
IS-1	−34	−188	154	133
IS-2	−69	162	−231	131
		$(+\ +\ +)$ Cu/5Fe/Cu(001)		
IS	−326	25	−351	135
IS-1	−394	−92	−301	135
IS-2	−392	−127	−269	135

netic coupling between Fe atoms or the increase of the localization of electronic states tend to favor the FM state.

Now, one more question remains to be answered: What is the magnetic ordering within the (001) layer itself? The answer is not too difficult to obtain from our calculations. As a result of the localization of the surface (or interface) states, the magnetic coupling is dominated by the nearest-neighbor interactions. We have shown that for 2Fe/Cu(001) the FM coupling between Fe layers is far more stable than is the AFM coupling. Since the interatomic distance between the Fe atoms in the (001) atomic plane is identical to that between adjacent layers for the fcc structure, it is natural to expect that a ferromagnetic ordering also forms within the (001) plane.

IV. ELECTRON DENSITY AND WORK FUNCTION

To gain a better understanding on the surface and interface formation, we list in Table II the total valence charge within the touching MT spheres. Starting from the case of the overlayers, several insights are obtained from our FLAPW calculations: (1) The effect of the surface is seen from the decrease of the number of electrons belonging to surface atoms (i.e., the electrons are transferred into the vacuum region to screen the discontinuity introduced by the surface formation); (2) the short-range screening effect manifests itself in the constant number of electrons within the Cu MT spheres just one layer from the Cu/Fe interface; (3) there is an increased number of electrons (mostly p-like) within the Cu MT spheres at the interface, coming mostly from the tails of the d wave functions centered at adjacent Fe sites. Further, because of the hybridization between surface and subsurface Fe layers for 2Fe/Cu(001), the number of electrons at the surface Fe site is increased slightly from that of 1Fe/Cu(001). A very similar charge distribution is found for the Cu/Fe/Cu sandwiches (cf. Table II): A decrease of the d-like and p-like charges within the Fe MT spheres (due to the closed d shell of Cu), accompanied by an increase of the p-like density for Cu at the interface, compared with their bulklike counterparts (with the center layer of the slabs as references).

Because of the spillout of the electrons into the vacuum region, this gives rise to the surface dipole layer, which consequently determines the work function (Φ). The calculated Φ values are 5.0, 4.95, and 5.10 eV for 1Fe/Cu(001), FM 2Fe/Cu(001), and AFM 2Fe/Cu(001). It is interesting to note that because of the larger magnetic

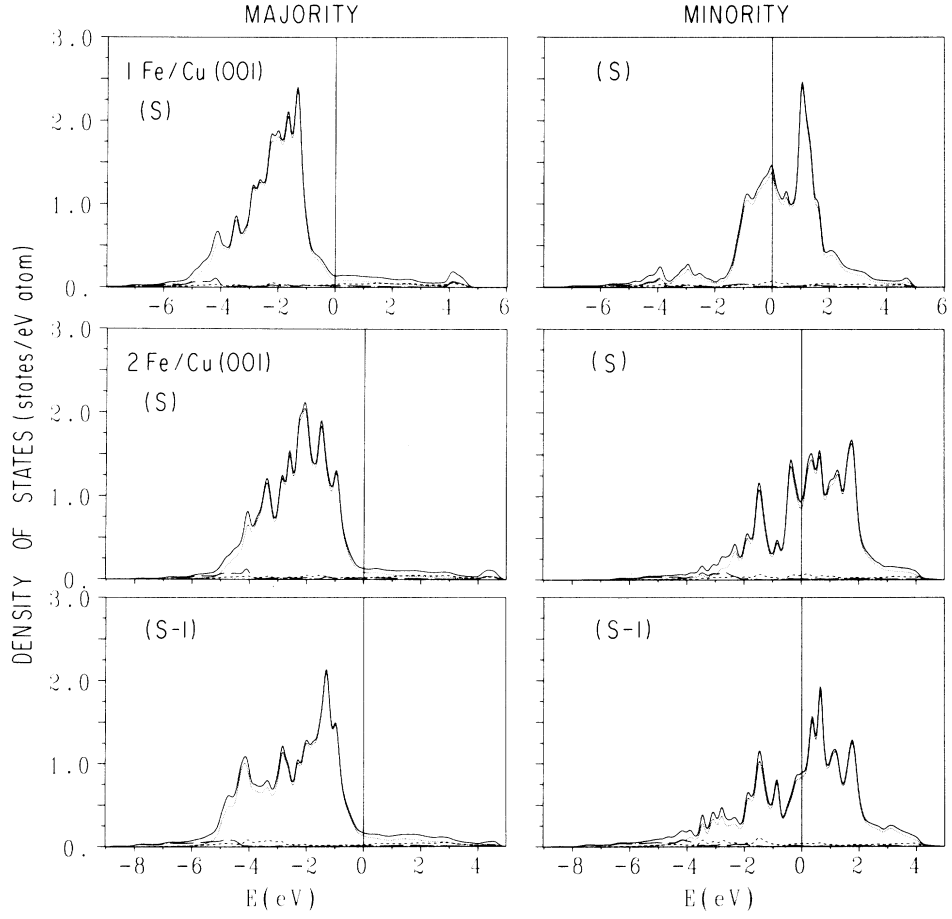

FIG. 2. Layer-projected partial density of states in units of states/eV atom for an Fe monolayer on Cu(001) (upper panel), and for two layers of Fe on Cu(001) (middle and lower panels). Dotted lines indicate d states and broken lines represent s,p contributions.

moments for FM 2Fe/Cu(001), the work function is lower than that of the AFM state, i.e., ferromagnetic ordering within the surface layer fills the antibonding states at the expense of bonding states, resulting in a surface dipole layer such as to reduce Φ relative to the paramagnetic state ($\phi = 5.4$ eV).

V. CONTACT MAGNETIC HYPERFINE FIELDS

Mössbauer spectroscopy offers a unique tool for the microscopic analysis of the electronic and magnetic properties. The electronic spin density at the nucleus is the key quantity for the interpretation of the hyperfine interaction which probes the coupling of the electronic spin to the nuclear magnetic moment. The spin density at the nucleus gives rise to the Fermi-contact field (H_{CF}) which is substantially larger than the contributions from unquenched angular momentum and dipolar fields. H_{CF} consists of two parts: (1) the dominant "negative" contribution (with respect to the sign of the local moment) from core-electron polarization, and (2) the contribution from the ($4s$) conduction electrons.

As is now well-established even for the bulk metals and their surfaces, the core contribution is proportional to the local moment[26] (because the magnetic moments originate mostly from the localized d electrons). This is seen in the constancy of H_{CF}(core) per magnetic moment ($=130$ kG/μ_B; cf. Table III). Regardless of the metallic environment, this H_{CF}(core)/M value is remarkably close (within 10%) to those of bcc Fe (001) (Ref. 29) and Fe(110) (Ref. 30) thin films. From our analysis, we find that the valence contributions are polarized *positively* by direct exchange with the unpaired d electrons for the following cases (cf. Table III): (1) at the surface or interface Fe sites (i.e., for atoms in a more open environment, although the absolute value at the interface is considerably smaller than at the surface), and (2) large contributions from Fe layers coupled antiferromagnetically with adjacent Fe layers [e.g., $(+\ +\ -)$ and $(+\ -\ +)$ states of Cu/5Fe/Cu(001), and AFM state of 2Fe/Cu(001)]. Unlike the ferromagnetic bulk materials, where the valence contribution is governed by the indirect polarization [also see the IS-1 and IS-2 layers of the $(+\ +\ +)$ state of Cu/5Fe/Cu(001)], the direct H_{CF}(valence) contribution for the AFM state tends to be large and cancel the indirect core polarization. This results in relatively small H_{CF}(total) values compared with those of the FM coupling between adjacent Fe layers. As a result, layer-by-layer Mössbauer measurements should be a direct tool for detecting these large differences. Recent measurement by Halbauer and Gonser[12] found that the interface Fe atoms experience larger hyperfine fields than do Fe atoms in the inner layers of the fcc Fe films. This rules out the possibility of the $(+\ +\ +)$ state, as is also confirmed by our total-energy calculation. However, the difference between the $(+\ +\ -)$ and $(+\ -\ +)$ states is not resolved at present using Mössbauer spectroscopy.

VI. SINGLE-PARTICLE SPECTRA

In this section, we present the energy band dispersions of fcc Fe as overlayers on Cu(001). First, the layer-

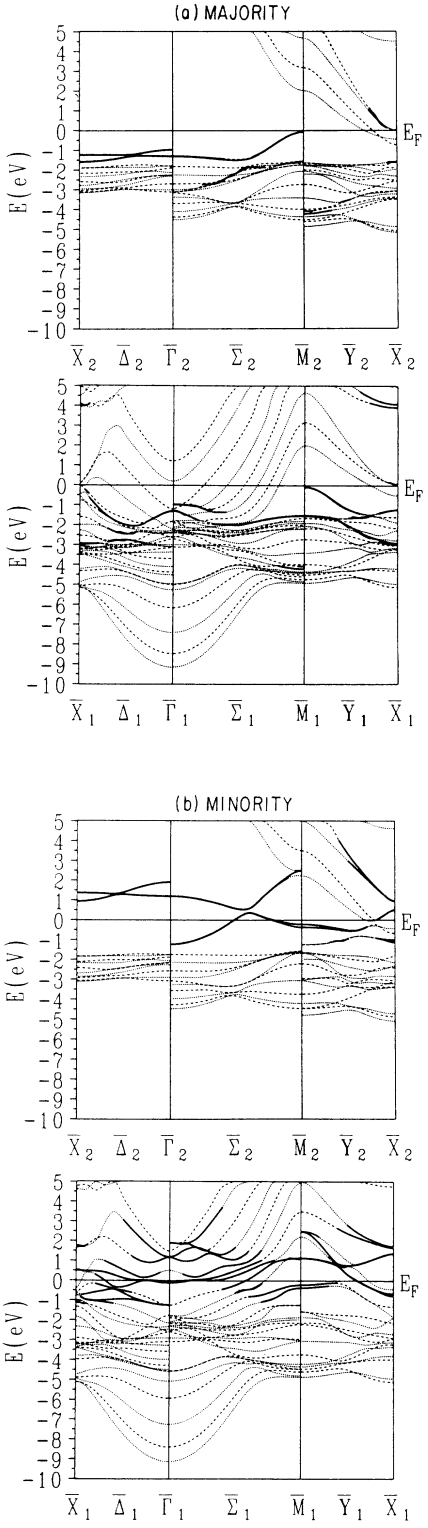

FIG. 3. Energy bands for 1Fe/Cu(001) of majority spin, and minority spin along high-symmetry directions in the 2D Brillouin zone. Top and lower panels show odd and even symmetries, respectively (see text). Dashed and dotted lines represent even and odd parities with respect to z reflection. Solid lines indicate surface states whose wave functions have more than 50% weight within the surface Fe layer.

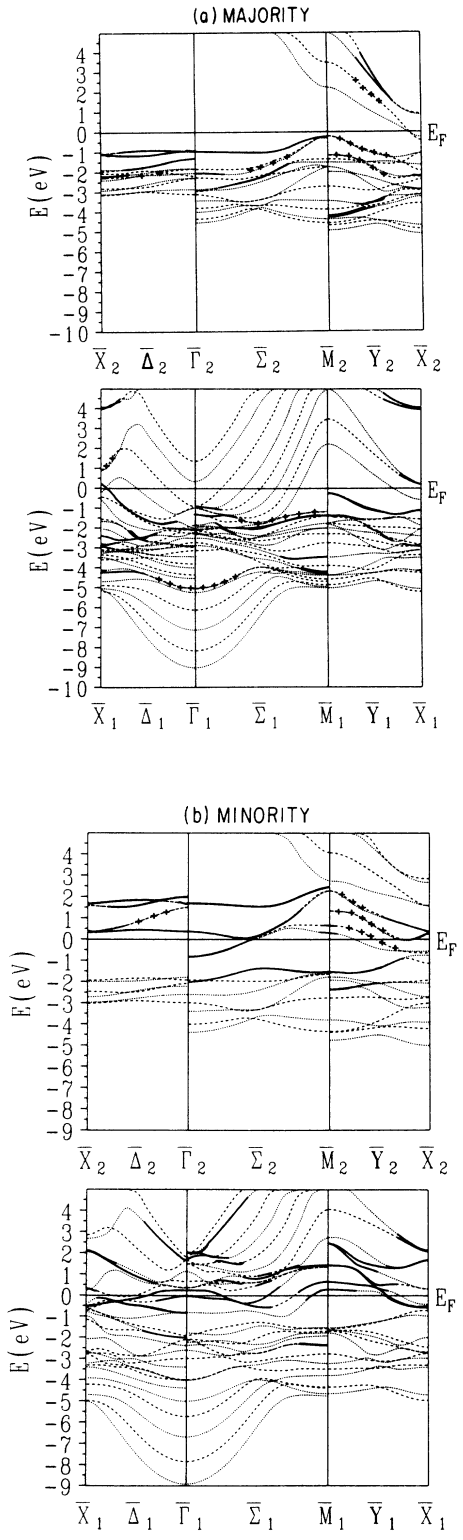

FIG. 4. Energy bands for 2Fe/Cu(001) of majority spin and minority spin along high-symmetry directions in the 2D Brillouin zone. The notation is identical to that in Fig. 3. The crosses represent the bands with wave functions localized on the subsurface Fe layer.

projected l- decomposed density-of-states (DOS) ferromagnetic Fe layers, which give the basic understanding of the surface and interface related features, are shown in Fig. 2. The relatively wide bandwidths compared with, for example, those of bcc Fe/Ag(001) indicate a stronger in-plane interaction between Fe atoms for Fe/Cu(001). Further, despite the large band overlap with the nonmagnetic Cu d band (with its center of gravity at a binding energy of 3 eV), the localized magnetic moments at the Fe sites remain essentially the same magnitude as those of Fe/Ag(001),[28] or at the surface layer of bcc Fe(001).[29] The strongly enhanced magnetic moments appear to be governed by the Fe localized surface (or interface) states. As we shall see next, these interface states have a binding energy of 1—2 eV, which are away from the Cu d band.

Figures 3 and 4 display the energy band structures along the high-symmetry lines of the irreducible 2D Brillouin zone for 1Fe/Cu(001) and 2Fe/Cu(001), respectively. The bands are sorted for clarity into even and odd mirror-reflection symmetries with respect to the plane spanned by $\hat{z}$ and the symmetry line itself. Surface states, drawn by heavy-solid lines, are defined as having more than 50% of their charge localized in the surface layer. The most prominent surface states are bands along the $\bar{\Gamma}\bar{M}$ and $\bar{\Gamma}\bar{X}$ directions in the vicinity of E_F (in particular, the odd symmetry). The magnetic exchange splitting is estimated to be 2.65 eV. Recently, these surface states have been measured and confirmed by Onellion *et al.*[31] There is, overall, good agreement between theory and experiment, except that the measured magnetic exchange splitting is slightly smaller ($\sim$0.25 eV) than predicted theoretically. This may be due to (1) temperature effects (the experiments were performed near room temperature, while the theoretical values are predicted for $T=0$ K, and (2) a manifestation of the importance of many-body effects (i.e., core-hole relaxation and correlation).

The change of band dispersion in going from the monolayer to two-layer coverage can be seen from a comparison of Figs. 3 and 4. The overall surface Fe layer derived features and dispersions for 2Fe/Cu(001) remain similar to those of 1Fe/Cu(001). However, the localization of the surface state wave functions on the surface Fe layer becomes less pronounced due to its hybridization with the subsurface Fe layer. This change of bonding also manifests itself in a reduced upward dispersion of odd symmetry bands (dominated by d_{xz} and d_{yz} orbitals) along the $\bar{\Gamma}\bar{M}$ direction. Bands with wave functions localized on the subsurface Fe layer for 2Fe/Cu(001) are shown by the crosses in Fig. 4. The effect of the Cu-Fe hybridization is seen in the increased bandwidth compared with that of the surface Fe layer (cf. Fig. 2). The magnetic exchange splittings are estimated to be 2.65 and 2.40 eV for the surface and subsurface Fe layers of FM 2Fe/Cu(001).

ACKNOWLEDGMENTS

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