First-principles calculation of structural and magnetic properties for Fe monolayers and bilayers on W(110)

X. Qian and W. Hübner
Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany
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Structure optimizations were performed for 1 and 2 monolayers (ML) of Fe on a 5-ML W(110) substrate employing the all-electron full-potential linearized augmented plane-wave method. The magnetic moments were also obtained for the converged and optimized structures. We find significant contractions (~10%) for both the Fe-W and the neighboring Fe-Fe interlayer spacings compared to the corresponding bulk W-W and Fe-Fe interlayer spacings. Compared to the Fe bcc bulk moment of $2.2\mu_B$, the magnetic moment for the surface layer of Fe is enhanced (i) by 15% to $2.54\mu_B$ for 1 ML Fe/5 ML W(110), and (ii) by 29% to $2.84\mu_B$ for 2 ML Fe/5 ML W(110). The inner Fe layer for 2 ML Fe/5 ML W(110) has a bulklike moment of $2.3\mu_B$. These results agree well with previous experimental data. [S0163-1829(99)02347-4]

I. INTRODUCTION

Magnetic thin films on metal substrates demonstrate fascinating phenomena such as the preferential orientation of the magnetization normal to the film plane, enhanced low-temperature surface magnetization, and the pronounced effects of magnetism on the electrical conductivity. There has been considerable effort[1–21] in studying the atomic structures and magnetic properties including magnetic moments and the orientation of the magnetic easy axis for Fe thin films on W(110). It is especially interesting to study these properties for 1 and 2 monolayers (ML) of Fe on W(110) due to the pseudomorphic layer-by-layer growth of the film when the Fe coverage θ is below 2 ML and to the possible magnetization reorientation from in-plane to perpendicular for $1 < \theta < 2$. Both bulk Fe and W are bcc structures with lattice constants of 2.86 and 3.165 Å, respectively. It was found that Fe thin films grow pseudomorphically up to 1.2 ML (Refs. 14 and 21) on the flat W(110) surface and up to 1.8 ML (Ref. 1) on a vicinal surface. Significant structural relaxation in the vertical lattice spacings for the Fe thin films is expected arising from the large lattice mismatch (~9%) between the film and the W substrate. However, the exact amount of relaxation and the magnetic moments for the Fe overlayers need to be clarified because of the conflicting results between the experiments[8,9,15] and with previous theoretical predictions.[4] On account of (i) the pseudomorphic growth of the Fe thin film, and (ii) the transitions of both the atomic structures and magnetic properties already in the ultrathin (<2 ML) regime, it is feasible to employ the *ab initio* method to investigate these properties.

Earlier experimental work done by Albrecht *et al.*[8,9] with low-energy electron diffraction (LEED) on 1 ML Fe on W(110) substrate showed that the Fe-W interlayer spacing is contracted by 13% to 1.94 Å compared to the bulk W(110) interlayer spacing of 2.238 Å. The magnetic moment for the top Fe layer is enhanced to $2.53\mu_B$ measured by Torsion oscillation magnetometry (TOM).[10] However, recent work done by Tober *et al.*[15] using photoelectron diffraction (PED) for 1 ML Fe on W(110) yielded a Fe-W interlayer spacing of 2.07 Å, a relaxation of 7.2% only. Earlier *ab initio* calculations by Hong, Freeman, and Fu[4] showed that the Fe-W interlayer distance is dramatically reduced by as much as 16% to 1.88 Å. Because of this strong inward relaxation, the magnetic moment of the overlayer Fe is only $2.18\mu_B$, which is very close to the bulk bcc Fe moment of $2.2\mu_B$. Recent calculations by Batirev *et al.*[22] showed that the Fe-W interlayer spacing is contracted by 3.1% with respect to the average theoretical bulk bcc(110) Fe and W interlayer spacings. The magnetic moment for the Fe layer is $2.17\mu_B$ similar to the bulk value. Because of these differences between the experimental results and also with the calculations, a detailed theoretical investigation is presented to illuminate the incoherent data on structural and magnetic properties of Fe monolayers on W(110).

II. METHOD

It is well known that magnetic properties depend strongly upon the atomic structures of the thin films. Therefore it is necessary first to obtain an optimized structure for these systems. Three different slabs: (i) 5 ML W(110) clean substrate, (ii) 1 ML pseudomorphic Fe overlayer on each side of 5 ML W(110) substrate, and (iii) 2 ML pseudomorphic Fe overlayers on each side of 5 ML W(110) substrate were studied. The schematic picture of 2 ML Fe on 5 ML W(110) is shown in Fig. 1. The bare W(110) substrate was studied to test our theoretical accuracy since reliable experimental results are available and theoretical calculations are abundant.

These calculations were performed employing the WIEN97 code.[23] This program is based on the density-

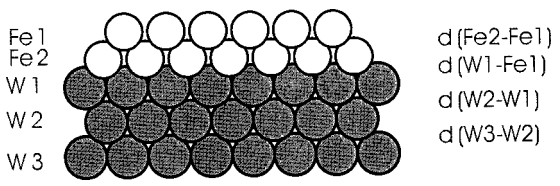

FIG. 1. Schematic picture of 2 ML Fe on 5 ML W(110) (upper half of the slab only).

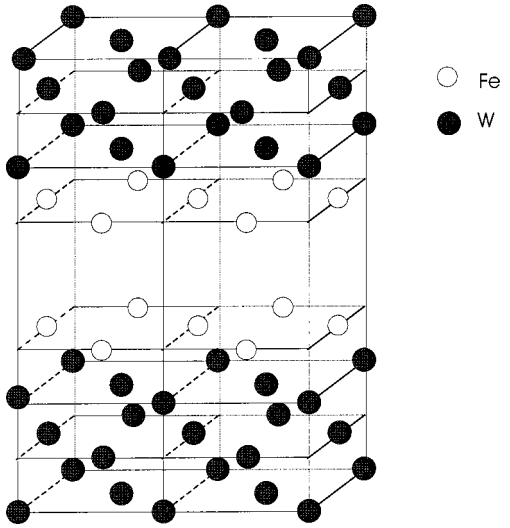

FIG. 2. Unit cells of the 1 ML Fe on each side of 5 ML W(110).

functional theory (DFT) and adopts the full-potential linearized augmented plane-wave (FP-LAPW) method. It has the additional capability of computing atomic forces,[24–26] which makes the structure optimization much more efficient compared to the total energy only calculations. The FP-LAPW method adopts different representations for wave functions, charge density, and potential inside the muffin-tin sphere and in the interstitial region. The spherical harmonics were expanded up to $l=10$ inside the muffintin and to $l=4$ for the interstitial in the present calculations. Spin-polarized calculations were carried out in order to determine the magnetic properties. Spin polarization was implemented in the WIEN97 code adopting local spin-density approximation (LSDA) with two separate spin densities. Two sets of Kohn-Sham (KS) orbitals for the two spin components were obtained, and two sets of KS single-particle equations were solved. The scalar relativistic calculations including the *velocity* and the *Darwin* terms were adopted for the calculations. Spin-orbit coupling was not included for the present calculations. The shallow $5p$ states were treated as semi-core, i.e., as local orbitals, thereby ensuring the flexibility of the basis functions to closely represent these low-lying p orbitals. Pulay corrections[25,27] to the Hellmann-Feynman forces were calculated, which makes the structure optimization highly accurate. The improved tetrahedron method[28] was used for the integrations.

Figure 2 shows the unit cells for the calculations. The structure optimization for the slabs was done by giving an initial guess of the interlayer spacings based on the optimized structure of Fe/Mo(110). The direction and degree of relaxation for the vertical interlayer spacings depend on the magnitude and sign of the forces present. The in-plane lattice constant for the slab was fixed and taken from the bulk calculations and will be described later. This is due to the in-plane two-dimensional translational invariance and the fact that there is only one atom on each layer in the unit cell. As described in our previous paper,[29] eight vacuum layers were incorporated in the supercell to separate the slabs in order to minimize any Coulomb and exchange interactions. Furthermore, slabs are symmetric with respect to the central substrate layer to avoid any charge accumulation on the surfaces. Thus the contribution to the total energy from the electric-dipole interaction between the supercells is negligible compared to the contributions from within the supercell. In addition, only real wave functions are needed for the calculations because of the presence of inversion symmetry. The Fe layers on each surface are ferromagnetically coupled. The spin-polarized calculations were applied.

In these calculations, generalized gradient approximation[30] (GGA) exchange potential and scalar-relativistic treatment were used in agreement with our earlier calculations on Mo substrate. Generally speaking, we did not find any significant improvement of GGA exchange potential over (LSDA) potential. Following the procedure described previously,[29] the theoretical bulk W lattice constant was determined to be 3.205 Å, 1.3% larger than the experimental value of 3.165 Å. It is known that GGA corrects overbinding, but sometimes leads to an excessive increase in the lattice parameter for heavy atoms such as W. Nevertheless, this theoretical value was used as the in-plane lattice spacing in our subsequent slab calculations. The theoretical bulk Fe lattice constant was found to be 2.834 Å, 0.9% smaller than the experimental result of 2.86 Å. The muffin-tin radii were chosen to be 1.27 and 1.164 Å for W and Fe atoms respectively in the slab unit cells. Convergence was achieved when the total energy and charge differences between two consecutive iterations are less than 5×10^{-5} Ry and $1\times10^{-4}\,e/(\text{a.u.})^3$, respectively. The structure optimizations were done when the force on each atom is less than 1 mRy/a.u. The magnetic moments were calculated as the differences between the spin-up charge and spin-down charge for these converged

TABLE I. Structural results. (The layer spacings are given in Å. The relative changes as compared to the bulk W layer spacing are given in parentheses. The percentage of Fe-Fe contraction is relative to the bulk Fe-Fe interlayer spacing.)

	$d(\text{Fe}_2\text{-Fe}_1)$	$d(\text{W}_1\text{-Fe}_1)$	$d(\text{W}_2\text{-W}_1)$	$d(\text{W}_2\text{-W}_3)$
5 ML W(110)			2.173(−4.1%)	2.258(−0.4%)
1 ML Fe/W(110)		1.974(−12.9%)	2.263(−0.1%)	2.251(−0.7%)
2 ML Fe/W(110)	1.766(−11.9%) (Ref. 33)	2.026(−10.6%)	2.267 (0.03%)	2.272(0.2%)
W(110) (Exp.) (Ref. 32)			2.169(−3.1%)	
Fe/W(110)(Exp.) (Ref. 9)	1.82(−10%)			
Fe/W(110)(Exp.) (Ref. 8)		1.94(−13%)		
Fe/W(110)(Exp.) (Ref. 15)		2.07(−7.2%)	2.28(2.2%)	
1 ML Fe/W(110) (Ref. 4)		1.88(−16%)		

TABLE II. Magnetic spin moments.

	1 ML Fe/W(110) (μ_B)	2 ML Fe/W(110) (μ_B)
Fe(2)		2.844
Fe(1)	2.536	2.308
W(1)	−0.085	−0.104
W(2)	−0.000	−0.004
W(3)	−0.000	−0.006
Interstitial	−0.055	−0.04
Fe(2)(Exp.)Fe/W(110) (Ref. 10)		2.77
Fe(1)(Exp.)Fe/W(110) (Ref. 10)	2.53	

results. Orbital magnetic moment is not included in our calculations due to the absence of spin-orbit coupling. Moreover, it was previously estimated to be around 0.1 μ_B, (Ref. 31) only. The numbers of **k** points in the two-dimensional meshes are 20×20 for 5 ML W(110), 21×21 for 1 ML Fe on 5 ML W(110), and 22×22 for 2 ML Fe on 5 ML W(110). The numbers of **k** points in the irreducible part of the Brillouin zone (IBZ) (1/4 of BZ) are 110, 121, and 132, respectively. The plane-wave cutoffs (corresponding to the largest k vector in the plane-wave basis expansion) are 16.7, 15.3, and 13.2 Ry for the three slabs, respectively with 0, 1, and 2 ML Fe coverage. The kinetic energy cut offs (corresponding to the largest reciprocal-space vector for the potential expansion) are 196 Ry for all three slabs.

III. RESULTS AND DISCUSSION

The structural and magnetic results are exhibited in Tables I and II, respectively. For the 5 ML W(110) clean substrate, we find that the top W-W interlayer spacing is contracted by 4.1% to 2.173 Å from the theoretical bulk W-W interlayer spacing of 2.266 Å in the (110) plane. This result is in good agreement with the previous FP-LAPW calculations[32] in which the same amount of contraction (4.1%) was found for the top W-W interlayer spacing with a 5 ML W(110) slab. A 3.6% downward relaxation was found for the top layer with a 9 ML W(110) slab. Our result is in disagreement with a recent calculation.[22] However, in that study, only three substrate W(110) layers were employed. The recent LEED experiment[32] yielded a contraction of 3.1% with an error bar of 0.6%. In addition to our agreement with previous theoretical and experimental data, our present result is also quite similar to the relaxation found for a 5 ML Mo(110) slab published earlier.[29] Further, we find that the second W-W interlayer distance is also slightly contracted by 0.4% to 2.258 Å. This again agrees well with earlier calculations[32] in which a 0.2% contraction was found for the 5 ML W(110) slab.

The clean W(110) substrate is nonmagnetic. The density-of-state (DOS) plot is shown in Fig. 3. Only *d*-partial DOS (PDOS) of spin down are shown since they are identical to the spin-up DOS. The inner W layer [W($S-2$)] *d*-PDOS closely resembles the bulk bcc W *d*-PDOS. The surface layer [W(S)] *d*-PDOS has a higher number of states at the Fermi level, almost double that of the W($S-2$), i.e., a less pronounced gap between the two subbands.

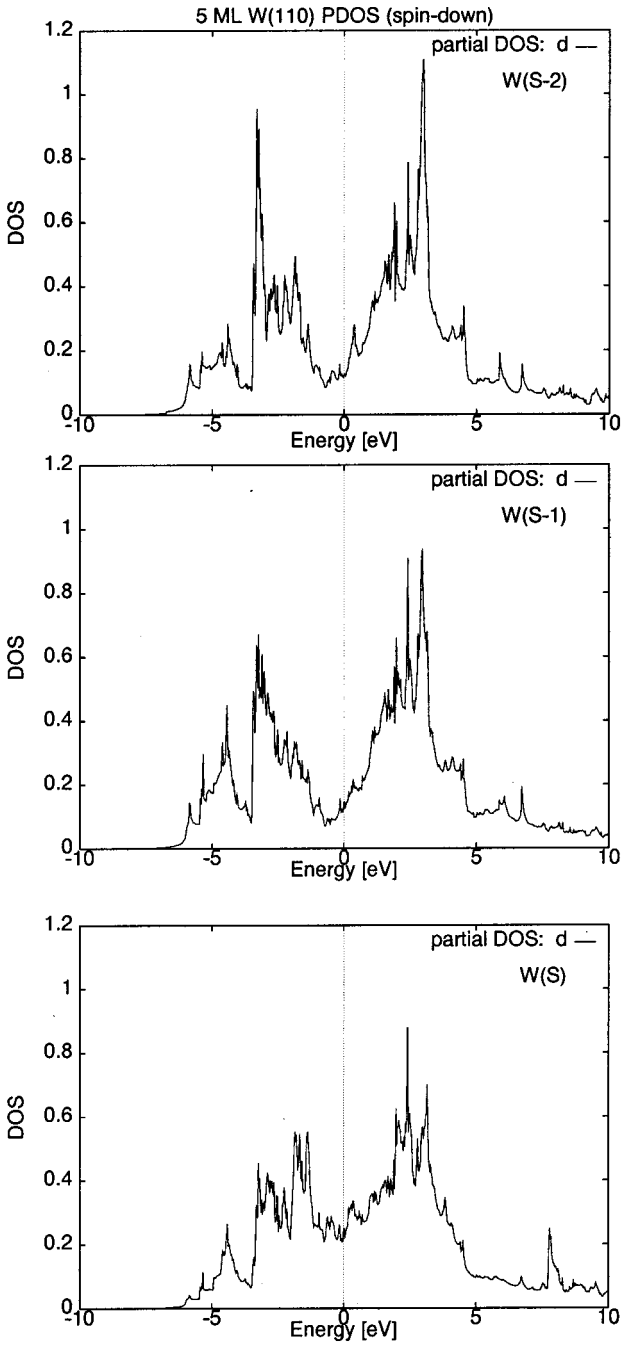

FIG. 3. Spin-down partial-*d* density of states (*d*-PDOS) for 5 ML W(110) clean substrate. S represents the surface layer, $S-1$ the layer next to the surface layer, and $S-2$ the central layer.

For the slab of 1 ML Fe on each side of 5 ML W(110) substrate, we find a significant relaxation for the Fe-W interlayer spacing (Table I) very similar to the case of 1 ML Fe/5 ML Mo(110) as shown in our previous work.[29] The Fe-W interlayer has a downward relaxation of 12.9% compared to the bulk W-W interlayer distance. It is in excellent agreement with the LEED experiment by Albrecht *et al.*[8,9] in which a 13% contraction was found compared to the bulk experimental W-W interlayer distance. The recent PED experiment, however, yielded a Fe-W distance of 2.07 Å (7.2%

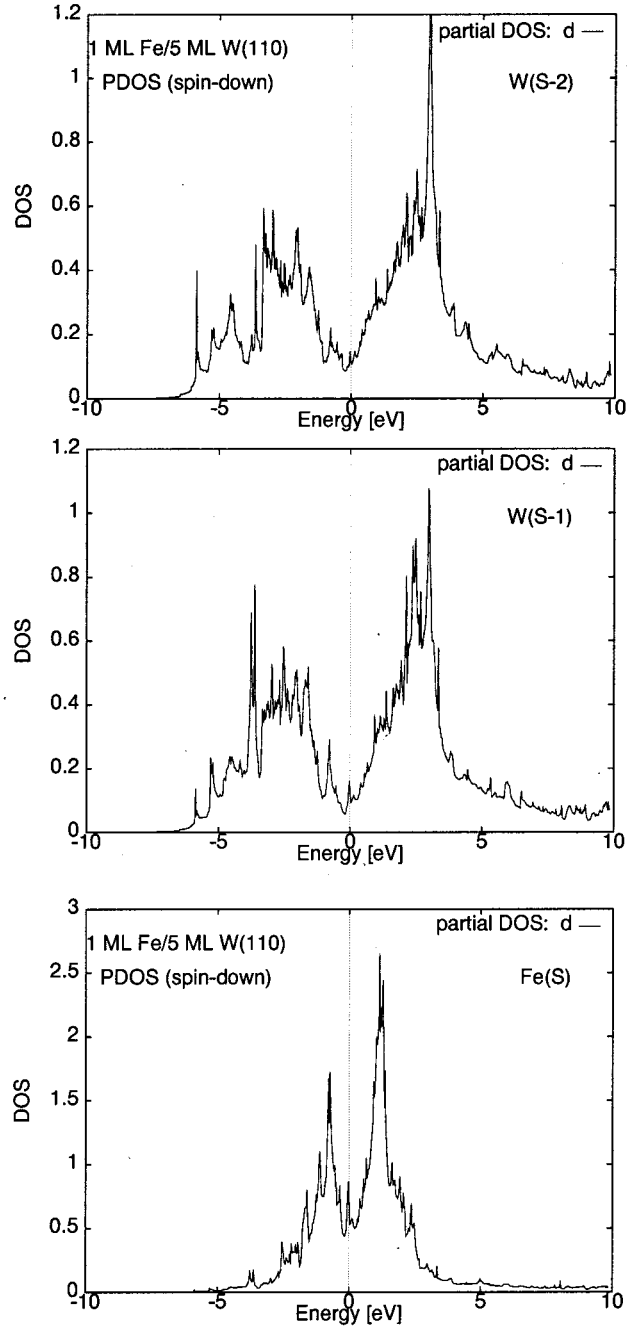

FIG. 4. Fe and W spin-up partial-*d* density of states (*d*-PDOS) for 1 ML Fe/5 ML W(110).

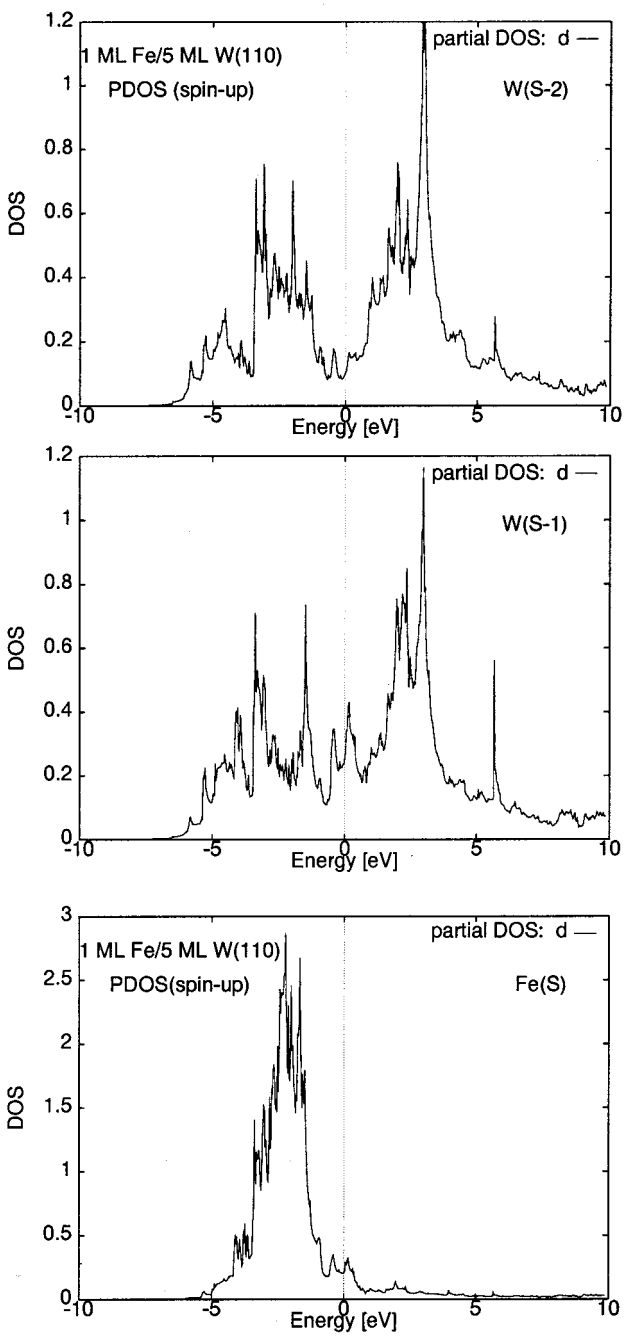

FIG. 5. Fe and W spin-down partial-*d* density of states (*d*-PDOS) for 1 ML Fe/5 ML W(110).

contraction only) which corresponds to the bond length from the hard sphere's model. The earlier calculations done by Hong, Freeman, and Fu[4] showed a much larger downward relaxation of 16% employing the FP-LAPW method. However, in their earlier calculations, not all the atoms were allowed to relax at the same time since it was not possible to compute the force on each atom. In addition to the Fe-W distance, our present calculations show that the neighboring W-W interlayer spacing is reduced slightly by 0.1%. However our earlier results on Mo(110) show a small expansion for the neighboring Mo-Mo interlayer spacing contrary to the W case here.

The magnetic moment for the surface layer of Fe is found to be $2.54\mu_B$ without orbital moment contribution, an enhancement of 15% over the bulk magnetic moment of $2.2\mu_B$ for bcc Fe. However, it is reduced by 29% compared to the moment of $3.3\mu_B$ for the Fe(110) free-standing monolayer with the same in-plane lattice parameter. In addition, our results show that the neighboring W layer acquires a small moment of $0.1\mu_B$. It is antiferromagnetically coupled to the Fe overlayer. The Torsion oscillation magnetometry (TOM) experiment done by Gradmann[10] yielded a moment of $2.53\mu_B$ for the overlayer Fe. Since the orbital moment and the induced substrate moment are both around $0.1\mu_B$ and opposite in sign, the theoretical spin moment we obtained for Fe overlayer agrees very well with the TOM experiment

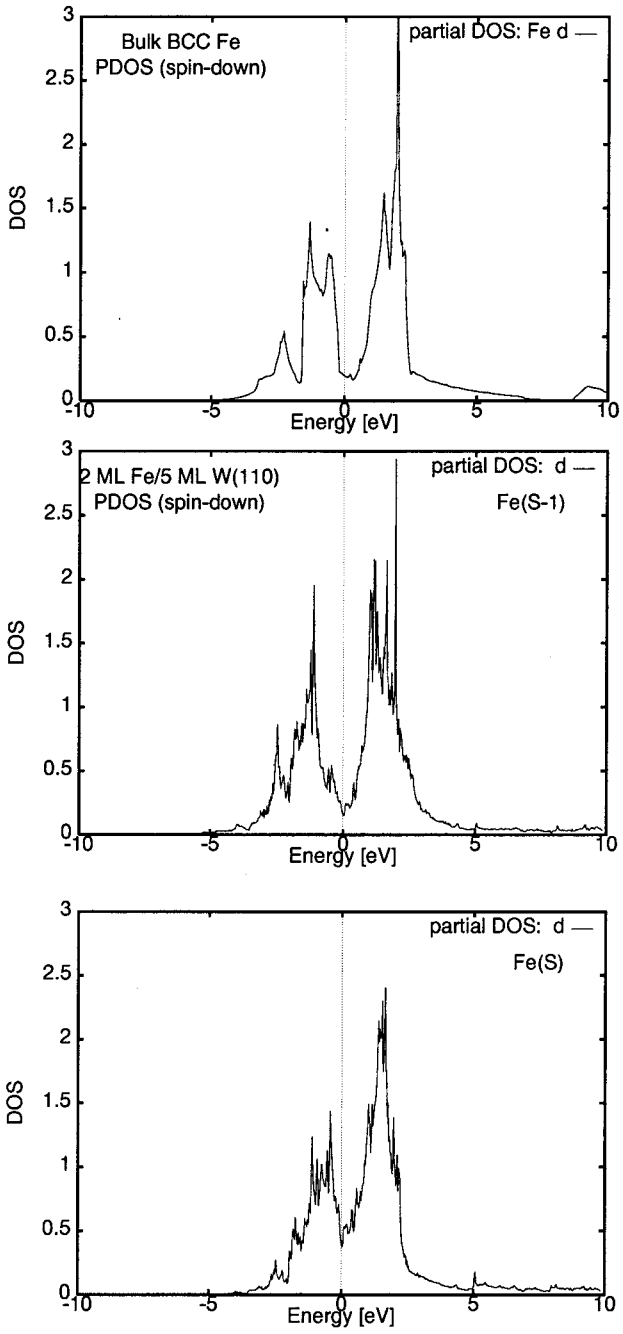

FIG. 6. Comparison of the Fe spin-down partial-*d* density of states (*d*-PDOS) for 2 ML Fe/5 ML W(110) with the corresponding bulk bcc Fe one.

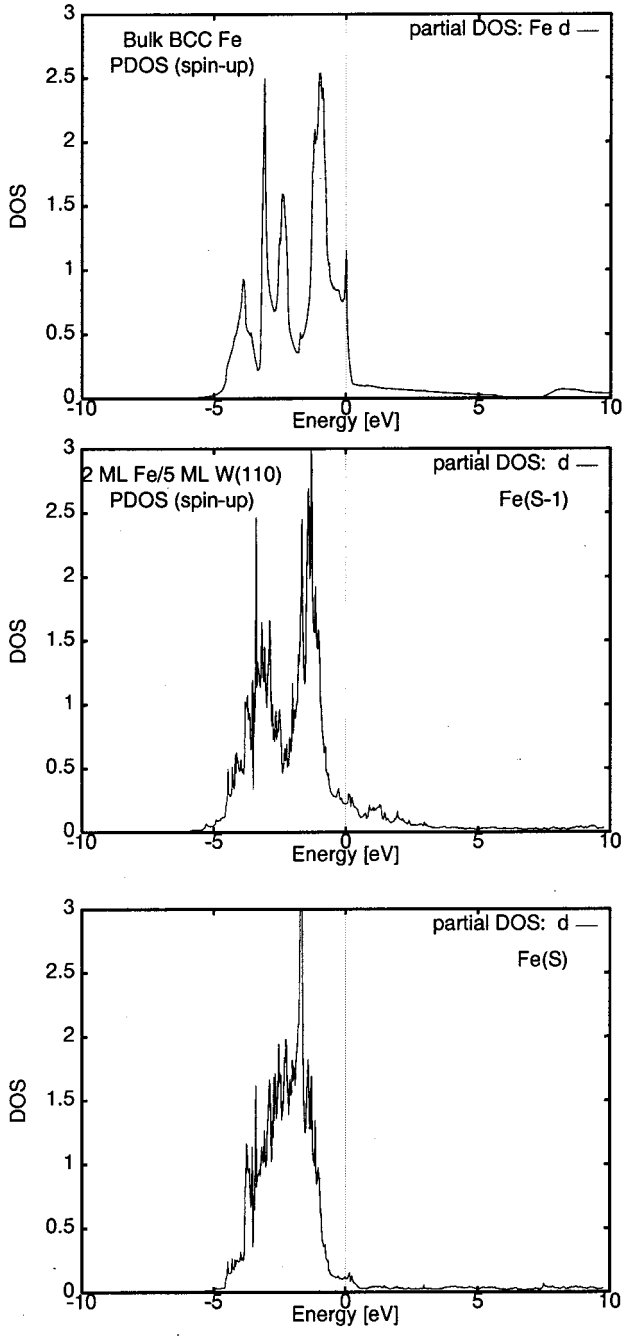

FIG. 7. Comparison of the Fe spin-up partial-*d* density of states (*d*-PDOS) for 2 ML Fe/5 ML W(110) with the corresponding bulk bcc Fe one.

since TOM measures the total moment. The earlier calculations done by Hong, Freeman, and Fu[4] showed no enhancement of the moment over the bulk value. It is probably due to the fact that their calculations yield a even larger reduction of the Fe-W interlayer distance than our present results.

Spin-down and spin-up *d*-PDOS for both the surface Fe layer and the neighboring W layers are plotted in Figs. 4 and 5, respectively. The *d*-PDOS of both spins for the inner W($S-2$) layer are very similar to the central W layer of the previous case where there is no Fe overlayer. The spin-down *d*-PDOS of the interfacial W($S-1$) layer resembles the one of the inner W($S-2$) layer. However, there are noticeable changes for the spin-up component especially when close to the Fermi surface. The small moment of the W($S-1$) layer is due to this change of *d*-PDOS. The overlayer Fe *d*-PDOS are very different to their corresponding bulk ones as shown in Figs. 6 and 7, especially for the spin-up component. Figures 6 and 7 will be discussed a little later.

For the slab of 2 ML Fe on each side of 5 ML W(110) substrate, we find both the Fe-Fe and Fe-W interlayer spacings are contracted dramatically (see Table I). The Fe-Fe interlayer distance is reduced by 11.9% (Ref. 33) from the theoretical bulk Fe value of 2.004 to 1.766 Å. The Fe-W interlayer spacing is contracted by 10.6% compared to the

bulk W-W interlayer distance. The percentage of the contractions are also very close to the Fe(110)/Mo(110) case.[29] Albrecht *et al.*[8,9] found a 10% downward relaxation for the Fe-Fe interlayer spacing compared to the bulk Fe value. Again it is in excellent agreement with our findings. Our calculations show a slight expansion $<0.2\%$ for the inner W-W interlayer spacings.

The magnetic moment for the surface layer of Fe is found to be $2.84\mu_B$, an enhancement of 29% over the bulk value of $2.2\mu_B$. It is still smaller than the moment of the Fe(110) free-standing monolayer. However, compared to the 1 ML Fe/5 ML W(110) case, the moment for the top Fe layer is increased from $2.54\mu_B$ to $2.84\mu_B$. This is probably due to the strong hybridization of Fe *d* orbitals with the ones of interfacial W layer thereby reducing the moment of the Fe layer. The second Fe layer, i.e., the interfacial Fe layer has a moment of $2.3\mu_B$ already very close to the bulk value. As in the previous case, the neighboring substrate layer also acquires a small moment of $0.1\mu_B$ and is antiferromagnetically coupled to the Fe overlayers.

The *d*-PDOS for the surface and interfacial Fe layers together with the ones of Fe bcc bulk are plotted in Fig. 6 (spin-down) and Fig. 7 (spin-up) for comparison. Basically the *d*-PDOS of the second layer of Fe are already close to the bulk ones. Consequently its magnetic moment is also approaching the bulk value. The *d*-PDOS of the surface Fe layer are different from the bulk ones particularly for the spin-up component and when close to the Fermi surface for the spin-down component.

IV. SUMMARY

The present FP-LAPW calculations resolves the discrepancies between previous experimental data and with earlier theoretical results on the atomic structure and magnetic moment of 1 ML Fe/W(110). The Fe-W interlayer spacing is significantly contracted by as much as $\sim 13\%$ compared to the bulk W-W interlayer spacing. The magnetic moment of the overlayer Fe is greatly enhanced compared to the bulk moment of bcc Fe due to the lower coordination number, but it is reduced compared to the Fe(110) free-standing monolayer because of the presence of the substrate.

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