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# Effects of interfacial relaxation on the magnetic coupling of Mn/Fe(111)

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The structural, electronic, and magnetic properties of monolayer Mn on an Fe(111) substrate are determined using the local spin density total energy full potential linearized augmented plane wave method and atomic force approach. A strong interplay between magnetism and atomic structure is found, e.g., the interfacial Fe layer undergoes a large downward relaxation by 0.59 a.u. from the ideal bcc position. As was found previously for bulk fcc Mn and Mn/Fe(001), the equilibrium Mn-Fe bond length is larger in the ferromagnetic state (4.47 a.u.) than that in the antiferromagnetic (AFM) state (4.31 a.u.). The AFM coupling between Mn and Fe is the ground state. The interplay between the interfacial magnetic coupling in the Mn/Fe(111) and the multilayer relaxation is discussed. © 1997 American Institute of Physics. [S0021-8979(97)46808-1]

The magnetic properties of ultrathin Cr or Mn overlayers on ferromagnetic materials currently receive much attention, especially in connection with the properties of the interface which may play an active role in the long- and short-period magnetic ordering in these systems.<sup>1-3</sup> Mn is of special interest, owing to the complications of structure and magnetism. As the structure and the volume of bulk Mn varies, it is found that the stable magnetic structure of the bulk Mn can be paramagnetic, ferromagnetic (FM), and antiferromagnetic (AFM).<sup>4</sup> Both FM and AFM interfacial ordering have been found in recent electronic structure calculations for Fe/Mn/Fe(001) sandwiches.<sup>3</sup> Using the full-potential linearized augmented plane wave (FLAPW) method and atomic force approach, we recently predicted a peculiar magnetic behavior for Mn overlayers on the Fe(001) substrate. A strong interplay between magnetism and atomic structure for the monolayer Mn/Fe(001) system<sup>5</sup> results in a  $c(2\times 2)$  buckling and an AFM ordering in the Mn plane ( $M=3.1 \mu_B$ ); for bilayer Mn coverage, the surface Mn layer (instead of the interfacial Mn layer) couples antiferromagnetically with the underlying Fe substrate. Our predictions for Mn overlayers on the Fe(001) substrate are supported by recent measurements using spin-resolved core level photoelectron spectroscopy.<sup>6</sup>

Since the interfacial magnetic coupling in Mn/Fe systems depends very sensitively on the details of atomic structure, calculations for Mn/Fe overlayer systems with different orientations are desired in order to reveal the effects from the change of symmetry and local coordinates. One can thus further estimate the importance of growth condition and interfacial roughness on the magnetic properties. In this article, we report the calculated structural, electronic, and magnetic properties of a Mn monolayer on an Fe(111) substrate. With the aid of our atomic force approach, a large surface multilayer relaxation is found which, as expected, affects the interfacial magnetic ordering significantly.

The Mn/Fe(111) system is simulated by a 13 layer slab consisting of 11 Fe layers in a bcc lattice and a pseudomor-

phic Mn layer on each side. The lattice constant in the lateral plane is chosen from experiment for bulk bcc Fe ( $a_{\text{Fe-Fe}}=7.66$  a.u.), while the vertical positions of all the atoms (including Mn and Fe) are optimized according to the calculated atomic forces.<sup>7</sup> Within the muffin-tin (MT) spheres ( $r_{\text{Mn}}=2.3$  a.u.,  $r_{\text{Fe}}=2.1$  a.u.), lattice harmonics with angular-momentum  $l$  up to 8 are employed to expand the charge density, potential, and wave functions. More than 60 a.u. augmented plane waves per atom are used as a variational basis set. The core states are treated fully relativistically and the valence states are treated semirelativistically (i.e., without spin-orbit coupling). Integrations over  $k$  space are substituted by summations over 18 special  $k$  points in the irreducible 2D Brillouin zone.<sup>8</sup> Convergence is assumed when the average root means squared distance between the input and output charge and spin densities is less than  $2.0\times 10^{-4}$  e/(a.u.).<sup>3</sup> This criterion ensures the reliability of the total energy and force up to 0.5 mRy and 1 mRy/a.u., respectively.

As a crucial reference system, it is very important to obtain highly precise results for the atomic structure and bonding properties of the clean Fe(111) surface. Experimentally, the Fe(111) surface is known from (LEED) to undergo larger multilayer relaxations due to its roughness:  $\Delta d_{12}=-16.9\%$ ,  $\Delta d_{23}=-9.8\%$ ,  $\Delta d_{34}=4.2\%$ , and  $\Delta d_{45}=-2.2\%$ .<sup>9</sup> The optimized structure obtained through the atomic force approach is shown in Table I. The multilayer relaxations (compared to the distance between two adjacent (111) planes in bulk bcc Fe) was found to be  $\Delta d_{12}=-15.4\%$ ,  $\Delta d_{23}=-18.6\%$ ,  $\Delta d_{34}=1.9\%$ , and  $\Delta d_{45}=-5.1\%$ . In contrast with the experimental measurement, the Fe(2) plane appears to undergo the largest downward relaxation since  $d_{23}$  (1.27 a.u.) is even smaller than  $d_{12}$  (1.32 a.u.). This unusual relaxation deserves experimental scrutiny, since it is found that the optimized structure is about 0.1 eV lower in total energy than obtained using the measured geometry.<sup>10</sup>

TABLE I. Calculated equilibrium position  $z$  (in a.u.) measured from the center plane and magnetic moment  $M$  (in  $\mu_B$ ) for the clean Fe(111) surface and Mn/Fe(111) in its FM and AFM states.

Atom	Mn	Fe(1)	Fe(2)	Fe(3)	Fe(4)	Fe(5)	Fe(6)
Fe(111)							
$z$	...	7.18	5.86	4.59	3.00	1.52	0.00
$M$	...	2.62	2.23	2.39	2.09	2.14	2.26
FM Mn/Fe(111)							
$z$	8.97	7.22	6.05	4.50	2.99	1.51	0.00
$M$	3.47	1.88	2.20	2.03	2.05	2.08	2.01
AFM Mn/Fe(111)							
$z$	8.84	7.22	6.05	4.53	2.99	1.53	0.00
$M$	-3.43	2.01	2.10	2.01	2.32	2.26	2.36

Following the deposition of Mn, the large surface relaxation in the Fe(111) substrate is removed for most of the Fe layers except the interfacial one. Quantitatively, as listed in Table I, the calculated multilayer relaxations in the Fe(111) substrate for parallel (antiparallel) Mn-Fe alignment become  $\Delta d_{12} = -25\%$  ( $-25\%$ ),  $\Delta d_{23} = -0.6\%$  ( $-2.6\%$ ),  $\Delta d_{34} = -0.3\%$  ( $-1.3\%$ ), and  $\Delta d_{45} = -5.1\%$  ( $-6.4\%$ ). Substantial Mn-induced changes can be found for the surface Fe(1) layer ( $\Delta z = +0.04$  a.u.) and especially for subsurface Fe(2) ( $\Delta z = +0.19$  a.u.) layers. Interestingly, there is no significant difference between atomic structures of the Fe(111) substrate for the Mn-Fe FM and AFM magnetic ordering, which indicates that the different Mn-Fe magnetic orderings play only a minor role for the atomic structure for the Fe(111) substrate in this system.

As is known, Mn atoms prefer AFM alignment in a free-standing monolayer with a large local magnetic moment of  $4.32 \mu_B$ <sup>11</sup> when calculated with the lattice constant of Ag.<sup>12</sup> AFM ordering was also found theoretically recently for Mn/Fe(001) and Mn/Co(001).<sup>5</sup> Here, the total energy of the AFM state is also well below the FM state (by as much as 0.8 eV/cell) for the Mn/Fe(111) system (here we assumed that Mn atoms form a FM sheet in the lateral plane). Such a large energy difference unambiguously indicates the stability of the AFM ground state for Mn/Fe(111).

As was also found in our previous calculations for bulk fcc Mn<sup>13</sup> and Mn/Fe(001),<sup>5</sup> the equilibrium Mn-Fe bond length,  $d_{\text{Mn-Fe}}$ , is longer for the FM state (4.47 a.u.) than that for the AFM state (4.31 a.u.). As discussed previously by Oguchi and Freeman,<sup>13</sup> this is due mainly to a shortened interatomic distance enhancing the hopping integrals which results in a band broadening in the FM case but a stronger exchange splitting in the AFM case. For systems with a half-filled band and strong magnetic moment, the AFM case is energetically favorable.

To provide a clearer insight into the bonding mechanism, the Mn induced charge redistributions for the Mn/Fe(111) systems, obtained by subtracting the superimposed charge density from a free Mn monolayer and a clean Fe(111) surface from the self-consistent charge density for the corresponding adsorption system, are presented in Fig. 1. A strong Mn-Fe(3) chemical bonding is obviously indicated by the  $d_{z^2}$  character of the contours around Fe(3). Note that electrons deplete from the Mn, Fe(1) and Fe(2) sites and accumulate in the interfacial region: this feature is surpris-

ingly close to the behavior of a  $P$  adsorbate on Fe(111).<sup>14</sup> Similarly, this can be understood as the relatively larger spatial extension of the Mn  $3d$  wave function, which is more easily affected by the surrounding Fe atoms. The Mn  $3d$  wave function feels the attraction from the Fe nuclei and strong Coulomb and Pa.u.li repulsions from the high electronic density region around the Fe atoms. As a result, the Mn  $3d$  electrons are squeezed into the intermediate region between Mn and Fe atoms.

The strong chemical interaction, as was also revealed in many other systems,<sup>15</sup> will frustrate the surrounding spin polarization. As also listed in Table I, the magnetic moment is enhanced by  $0.40 \mu_B$  in the clean Fe(111) surface layer from the bulk value,<sup>15</sup>  $2.22 \mu_B$ . For the FM state, the magnetic moment of Fe(1) is diminished by 0.74 to  $1.88 \mu_B$  due to the presence of the Mn adsorbate. As expected, the magnetic moment of Fe(3), the nearest Fe atom to the Mn adsorbate, is also markedly reduced by 0.36 to  $2.03 \mu_B$ . Similarly, the

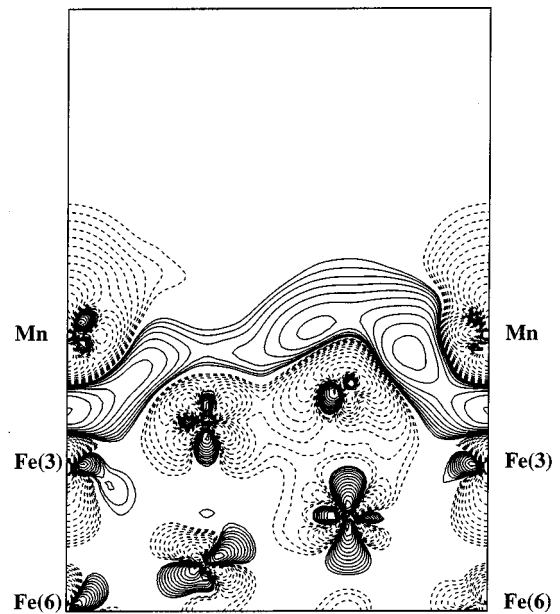


FIG. 1. Calculated charge density difference between the AFM Mn/Fe(111) system and the direct superposition of their reference constituents. Contours shown on a (110) plane starting from  $5 \times 10^{-4} e/a.u.^3$  and with successive contours increased by a factor of  $\sqrt{2}$ . Solid and dashed lines represent positive and negative differences, respectively.

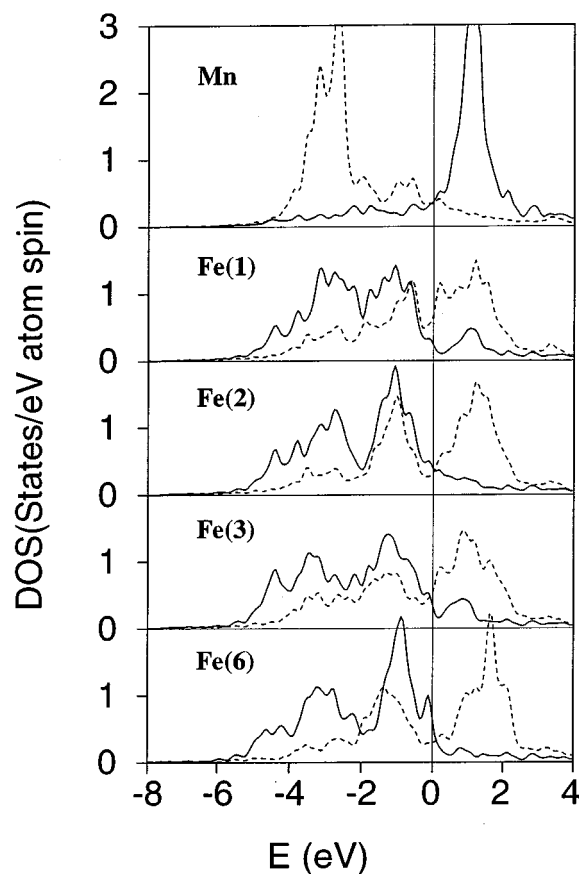


FIG. 2. The projected density of  $d$  states in the Mn and Fe muffin-tin spheres for the AFM Mn/Fe(111) system. Solid (dashed) lines represent majority (minority) spin.

magnetic moment of Fe(1) and Fe(3) drop to  $2.01 \mu_B$  in the AFM state of Mn/Fe(111)—indicating the detrimental effects of the Mn adatoms on the spin polarization of surrounding Fe atoms in both cases.

The projected density of states (DOS) for Mn and Fe muffin-tin spheres are plotted in Fig. 2. The DOS curves for the center Fe(6) layer show typical bulklike character for bcc Fe,<sup>10</sup> with  $E_F$  located near the top of the majority spin bands and in the valley of the minority spin bands. The DOS curves for Fe(1) and Fe(3) are substantially different from those for the center Fe(6) layer, indicating both a long-ranged surface

effect in the (111) oriented overlayer and a strong interaction between Mn adsorbate and Fe substrate. Compared to the clean Fe(111) surface, the strong hybridization between Mn and the Fe(111) substrate can be seen from the DOS of majority spin for Fe(1) and Fe(3). For the case of the clean Fe(111) surface,<sup>10</sup> the  $d$  band narrowing diminishes the majority spin holes in the outer layers [especially for the surface Fe(1) layer] and thus results in the surface magnetic moment enhancement. Here the presence of Mn causes a more pronounced DOS peak just about  $E_F$  for Fe(1) and Fe(3). This explains the detrimental effects of the Mn adatoms on the spin polarization of surrounding Fe atoms: the hybridization between Mn and Fe creates majority spin antibonding states above  $E_F$  and thus reduces the magnetic moments of the Fe atoms in the substrate, as shown in Table I.

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