Structural, magnetic, and electronic properties of Fe/Au monatomic multilayers

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An Fe/Au monatomic multilayer, consisting of alternating single Fe and Au layers, has been studied by means of the self-consistent full-potential linearized augmented plane wave method. We show by total energy minimization that this artificial thin film is in the tetragonal $L1_0$ ordered structure with the ratio of the interlayer spacing to the intralayer lattice constant at 0.865. In this configuration, the magnetic moment in each monolayer, the spin-polarized electronic density of states, and the corresponding band structure are calculated. The results are discussed in connection with recent experiments. [S0163-1829(96)04030-1]

Magnetic multilayers have attracted considerable attention over the past few years.[1] A magnetic *monatomic* multilayer, consisting of alternating single atomic layers of magnetic and nonmagnetic elements, is the low thickness limit of a magnetic multilayer. One particular structure out of the stacking of alternating single atomic layers is the tetragonal $L1_0$ ordered structure shown in Fig. 1. Some ordered alloys are known to have this phase as their naturally occurring structure. For example, an FePt alloy can spontaneously order into the tetragonal $L1_0$ structure by a traditional heat treatment.[2,3] More importantly, the recent development of sophisticated growth techniques has made it possible to easily grow quality tetragonal $L1_0$ ordered structures that exist naturally,[4,5] and to fabricate such materials that otherwise do not exist in nature. One such example is the Fe/Au $L1_0$ structure. This structure does not exist naturally in the Fe-Au phase diagram near the equiatomic composition,[7] but can be fabricated layer-by-layer by molecular beam epitaxy.[8] This tetragonal $L1_0$ ordered FeAu material is of great technological interest because it adds a new member to a family of ferromagnets that may have significant application potential in magnetic recording.[9,10]

The magnetic properties of the artificial Fe/Au $L1_0$ structure have been characterized experimentally, but to date a complete theoretical study of the system is still lacking. In one previous study, the magnetic properties of the Fe/Au monatomic multilayer were investigated by the layer Korringa-Kohn-Rostoker (LKKR) method,[11] but not in the tetragonal $L1_0$ ordered structure. In this paper, an Fe/Au in $L1_0$ structure was investigated by means of the self-consistent full-potential linearized augmented plane wave method (FLAPW).[12] We show, by total energy minimization, that this artificial thin film is ferromagnetic and is in the tetragonal $L1_0$ ordered structure. The ratio of the interlayer to the intralayer lattice constant is 0.865, and in this structure the magnetic moment of Fe is $2.75\mu_B$. The spin-polarized electronic density of states (DOS) and the corresponding band structure of the $L1_0$ ordered FeAu are also obtained. We discuss these magnetic and electronic properties in connection with the experimental results.

The first-principles FLAPW total energy method has been shown to be highly accurate in predicting ground state structural properties of solids ranging from insulators to metals.[14] We apply the method here to find the optimum ratio c/a of the tetragonal $L1_0$ ordered FeAu. For the Fe/Au monatomic multilayer grown on the Au(100) buffer layer,[8] we set the in-plane lattice constant $a = a_{\mathrm{Au}} = 4.08$ Å in our calculations. The Brillouin zone sampling was performed using a 12^3 special k-point mesh, which yielded 126 points in the irreducible Brillouin zone. The muffin-tin (MT) radii of 2.35 a.u.

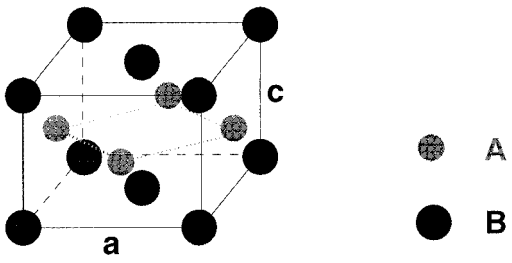

FIG. 1. The tetragonal $L1_0$ ordered structure.

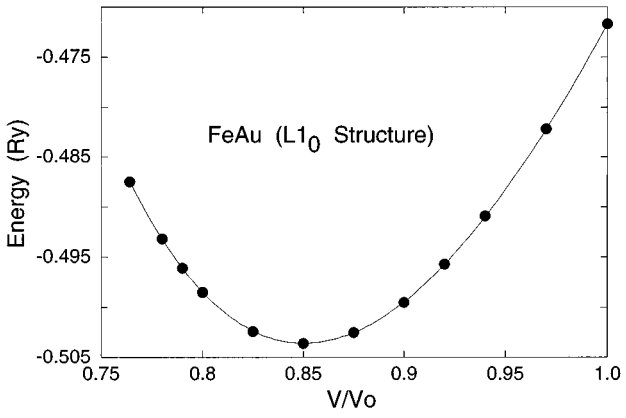

FIG. 2. Relative total energy as a function of volume for tetragonal $L1_0$ ordered FeAu. V_0 is the equilibrium volume of fcc Au corresponding to a lattice constant of $a_{\mathrm{Au}} = 4.08$ Å. The solid circles are the FLAPW calculated data and the solid line is a least squares fit to the Murnahan equation of state (Ref. 16).

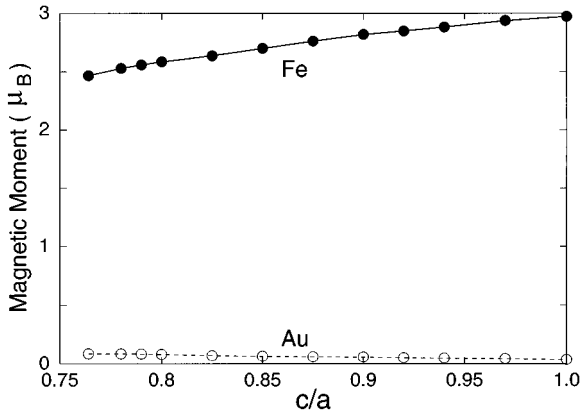

FIG. 3. Magnetic moments of Fe and Au in the tetragonal $L1_0$ ordered FeAu as a function of the c/a ratio of interlayer-to-intralayer lattice constants. The solid circles are the FLAPW calculated data for Fe with the solid line as a guide for the eye, and open circles are calculated data for Au along with the dashed line as a guide for the eye.

and 2.50 a.u. for Fe and Au, respectively, as well as the energy cutoff of $R_{\mathrm{MT}}K_{\max}=10$ were kept fixed in the calculations. The Ceperley-Alder local-density expression[13] for the exchange-correlation potential was used. Self-consistency was achieved when the total energy was stable to within 0.01 mRy/unit cell.

The FLAPW total energy as a function of volume for the tetragonal $L1_0$ ordered FeAu is plotted in Fig. 2, along with fits to the Murnaghan equation of state[16] from which one determines the equilibrium lattice constant and bulk modulus. The ratio of interlayer to intralayer lattice constant in this quasiequilibrium[15] phase was found to be $c/a=0.865$, which corresponds to a 15% volume contraction with respect to the equilibrium Au volume. This optimum ratio ($c/a=0.865$) is smaller than the experimental value of 0.94.[8] The equilibrium interplanar spacing may be near the value expected for the hard sphere radii of the elements. For example, the equilibrium ratio of 0.957 for the tetragonal $L1_0$ ordered FePt (Ref. 4) is close to the value of 0.922 (Ref. 17) estimated from the hard sphere radii of fcc Pt ($r_{\mathrm{Pt}}=1.386$ Å) and bcc Fe ($r_{\mathrm{Fe}}=1.243$ Å). The same argument gives rise to $c/a=0.857$ for FeAu ($r_{\mathrm{Au}}=1.443$ Å from fcc Au), which is very close to our calculated ratio of 0.865. The quality of the film as measured by the long range order parameter S is only about 0.3, compared to $S=1$ for perfectly ordered alloys. The experimental value of c/a was determined from the x-ray diffraction which makes direct measurements of *correlations* between atomic positions. Local disorder may displace some Fe and Au atoms from their equilibrium positions, and may change some Fe-Au stacking to Au-Au or Fe-Fe stacking faults. One sees that the value of c could be affected by the local disorder, and may become larger if more Au-Au stacking faults occur during the growth. This might explain the discrepancy between the experimental and calculated values.

Magnetism of transition metals is generally enhanced by decreasing the coordination number or by reducing the symmetry of the system.[18,19] A single monolayer of Fe has lower symmetry and a smaller coordination number than that of bulk bcc Fe. It has been shown that a single monolayer of Fe has a large enhanced magnetic moment of $3.4\mu_B$,[19] while the magnetic moment of bulk bcc Fe is only $2.2\mu_B$. The degree of symmetry and coordination number of Fe layers in the Fe/Au $L1_0$ structure are between bulk bcc Fe and single monolayer Fe. One expects that the magnetic moment of Fe

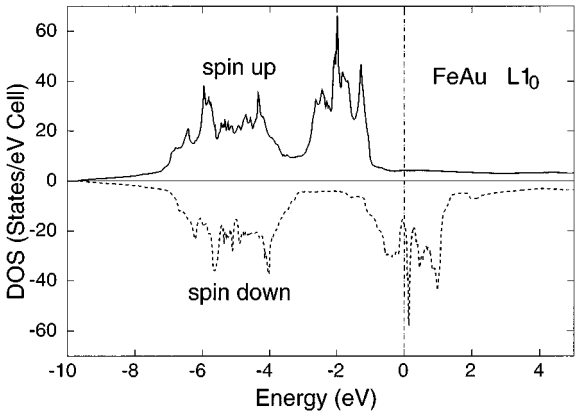

FIG. 4. The total electronic DOS of spin up and spin down for the tetragonal $L1_0$ ordered FeAu.

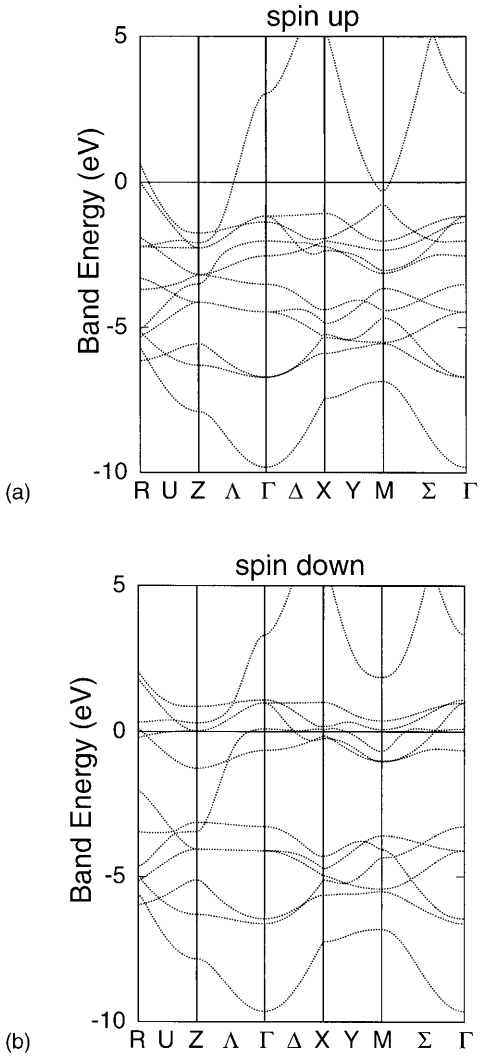

FIG. 5. Spin-up band (a) and spin-down band (b) for the tetragonal $L1_0$ ordered FeAu.

in the $L1_0$ ordered FeAu is between $2.2\mu_B$ and $3.4\mu_B$. This is confirmed by our calculations. The magnetic moments of Fe and Au in the Fe/Au $L1_0$ structure are plotted in Fig. 3 as a function of c/a. The solid circles for Fe are calculated values; the solid line is a guide for the eye. The open circles are calculated values for Au with the dashed line as a guide for the eye. One sees that magnetic moment of Fe increases slowly from $2.47\mu_B$ to $2.97\mu_B$ as the ratio c/a increases from 0.74 to 1. The trend of enhancing the magnetization in Fe layers results from weakening the correlation between Fe layers as their interlayer separation increases. The Au layers in $L1_0$ ordered FeAu are slightly spin polarized by the Fe monolayer between them. The magnetic moment of Au decreases from $0.088\mu_B$ to $0.037\mu_B$ as c/a increases from 0.74 to 1; the induced magnetization is reduced as a result of increasing the separation between Au layers and Fe layers. It is worth pointing out that the small induced magnetic moment of Au indicates weak coupling between the Fe monolayer and the Au monolayer.

In the structure of $L1_0$ ordered FeAu with $c/a=0.865$, the magnetic moment of Fe is found to be $2.75\mu_B$. This enhanced magnetization has been observed,[8] but the experimental value of $2.5\mu_B$ is less than what we calculated. If one chooses the experimental value of 0.94 for c/a, the calculated magnetic moment of Fe is $2.88\mu_B$, which is much larger than the experimental value $2.5\mu_B$. The existence of the local disorder in the film may reduce the measured magnetic moment of Fe.

The electronic density of states and band structure of the tetragonal $L1_0$ ordered FeAu with $c/a=0.865$ are presented in Figs. 4 and 5, respectively. The peaks in the spin-up DOS, as shown in Fig. 4, are located far below the Fermi level, and their corresponding bands are occupied as shown in Fig. 5(a). However, the spin-down band in Fig. 5(b), as expected, is partially filled. Notice that the Fermi energy is near a local minimum of the spin-down DOS in Fig. 4. This implies that the tetragonal $L1_0$ ordered FeAu is stable. The spin-up and spin-down DOS peaks around energy of -5 eV, shown in Fig. 4, are mainly from the d orbitals of Au atoms. One sees that they are slightly shifted from each other, which gives rise to a small magnetic moment of Au (order of $0.06\mu_B$). The spin-up DOS peaks around energy of -2 eV and spin-down DOS peaks around an energy of 0.05 eV are mainly d orbitals of Fe. The large split between spin-up and spin-down DOS peaks explains the large Fe magnetic moment (order of $2.75\mu_B$).

The MBE achieved Fe/Au monatomic multilayer[8] may become a new member of the tetragonal $L1_0$ family of ferromagnets which generally exhibit a strong uniaxial magnetocrystalline anisotropy. Current members are FePt-, FePd-, CoPt-, and MnAl-based alloys.[9,10] A consequence of the weak coupling between Fe layers and Au layers as discussed above is that the spins of the d electrons of the Fe layers are more confined to the c-axis direction, which leads to a large c-axis magnetic anisotropy. This strong magnetic anisotropy perpendicular to Fe and Au atomic planes has been observed.[8]

In conclusion, we have calculated structural, magnetic, and electronic properties of the Fe/Au monatomic multilayer system by the FLAPW method. This artificial thin film with the tetragonal $L1_0$ ordered structure has the value of 0.865 for the c/a ratio. It is found that this Fe/Au monatomic multilayer system with large magnetic moment of $2.75\mu_B$ for Fe may belong to the tetragonal $L1_0$ family of ferromagnets.

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