Journal of Magnetics 6(3), 80-82 (2001)

A First-principles Study on Magnetism of $Fe_2/Ir_4(001)$ Superlattice

In Gee Kim and Jae Il Lee*

Department of Physics, Inha University, Incheon 402-751, Korea

(Received 9 April 2001)

We have investigated magnetism of $Fe_2/Ir_4(001)$ superlattice in terms of a first-principles calculation by using an all-electron full-potential linearized augmented plane-wave (FLAPW) method within the generalized gradient approximation (GGA). We considered two magnetic states, the ferromagnetic (FM) and antiferromagnetic (AFM) coupled states between the Fe layers. It was found that the FM state was energetically more stable than the AFM one by 0.166 eV. Calculated magnetic moments of the Fe layers were, in absolute values, 2.45 μ_B and 2.30 μ_B for the FM and AFM states, respectively. We also found that the Ir layers had very small magnetic moments less than 0.1 μ_B for both magnetic states. In all the magnetic states, the subinterface Ir layers were coupled antiferromagnetically to the interface Ir layers, while the interface Ir layers were always coupled ferromagnetically to the interface Fe layers. These results contradicted to recent experimental reports of magnetically "dead" Fe layers in Fe/Ir superlattices for which the Fe layer thickness was less than two atomic layers. We attributed that the experimentally observed "dead" Fe layers were due to possible interdiffusion between Ir and Fe layers.

Key words : first-principles calculation, transition-metal magnetism, electronic structure, superlattice, Fe, Ir

1. Introduction

Correlation between magnetism and crystal structures has drawn great attentions [1]. Progress in fabrication techniques, such as molecular beam epitaxy (MBE), enables us in realizing artificial structures of multilayers, superlattices, sandwiches, overlayers, etc. Artificial system displays noble properties, for example, revealing ferromagnetism in nonmagnetic materials, enhanced magnetic moments at surfaces and interfaces, perpendicular magnetic anisotropy, etc. [2] Recently, experimental observations on magnetically "dead" Fe layers in Fe/Ir(001) superlattices have been reported [3-5]. Since antiferromagnetically coupled layers in two-dimensional systems could lead no net magnetization [6], the antiferromagnetic state has been considered to explain the observed magnetically "dead" layers in extremely thin Fe layers (~2 atomic monolayers) on Ir(001) surface and Fe/Ir superlattices, in terms of *ab initio* density functional calculations [7-9]. Another possibility to explain the observed magnetically "dead" layers in ferromagnetic Fe layers is interdiffusion process at Fe and Ir interface. If Fe and Ir ions were intermixed at an interface, the wave function overlap, or hybridization, between Fe-3*d* and Ir-4*d* states is essentially increased for reducing the magnetic moment of Fe ion or possible antiferromagnetic coupling under intermixed environment. Unfortunately, no such considerations are known to our knowledge for the Fe/Ir systems yet.

In this paper, we present our first-principles calculational results on magnetism of thin Fe layers in Fe/Ir(001) superlattice system, based on the density functional total energy calculation in terms of the all-electron full-potential linearized augmented plane-wave (FLAPW) method [10]. We have considered two magnetic states, the ferromagnetic (FM) and antiferromagnetic (AFM) coupled states between two Fe layers. However, the paramagnetic (PM) state exhibits much higher energy (one order higher than the other magnetic states) so that we excluded possible PM state in the Fe layers. It was found that the FM state was, energetically, more stable than the AFM state by 0.166 eV. Calculated magnetic moments, in absolute values, were 2.45 μ_B and 2.30 μ_B for the FM and AFM states, respectively. These results contradict to the experimental observations on magnetically "dead" Fe layers for which the thickness of the Fe layer is less than five atomic monolayers (ML). Since the experimental interface roughness was controlled within two ML [3-5], a possible interdiffusion between Fe and Ir layers at interface was responsible for observed magnetically "dead" layers. We briefly described calculational model and method in Sec. II. Results and discussions are presented in Sec. III and a brief summary is given in Sec. IV.

---

*Tel. +82-32-860-7654, e-mail: jilee@inha.ac.kr

2. Calculational Method

We constructed $Fe_2/Ir_4(001)$ superlattice with $p(1 \times 1)$ planar lattice constant of 5.13 a.u., which is the fcc Ir lattice constant [11]. Interlayer distance between Ir layers are taken to be 3.63 a.u., the half of fcc Ir lattice constant. We chose the Fe interlayer distance to be 3.03 a.u., half value of bct Fe obtained by atomic volume constant conversion of bcc Fe. Fe-Ir interlayer distance was taken to be 3.33 a.u., the average of the interlayer distances of fcc Ir and bct Fe. Our choice of these interlayer distances leads the length of c-axis of the $Fe_2/Ir_4(001)$ superlattice to be 20.57 a.u. In order to investigate antiferromagnetic couplings between Fe layers, we did not involve inversion symmetry and reflection symmetry about perpendicular plane of z-axis (z-reflection symmetry).

Kohn-Sham equation [12] was solved to obtain energy eigenvalues within the generalized gradient approximation [13] in terms of the FLAPW method. Charge densities and potential inside each muffin-tin sphere with radii of 2.20 a.u. for Fe and 2.55 a.u. for Ir were expanded with $l \leq 8$ lattice harmonics. About 500 LAPW basis functions per each **k**-point were used as variational set. Integration inside Brillouin zone was replaced by summation over 60 special **k**-points inside 1/8 irreducible wedge of the Brillouin zone. All core electrons were treated fully relativistically, while valence electrons were treated scalar relativistically, without considering spin-orbit coupling. Self-consistency was assumed when the root-mean-square distance between input and output charge (spin) density is less than 1×10^{-4} electrons/a.u.^{3}

3. Results and Discussions

Spin density contours on (110) plane are shown in Fig. 1 for the (a) FM and (b) AFM states. Solid lines and broken lines represent spin-up and spin-down, respectively. Contour starts from 5×10^{-4} electrons/a.u.^{3} and subsequent lines are increased by a factor of $\sqrt{2}$. It is easily found that the spin densities along c-axis are symmetric and antisymmetric for the FM and AFM states, respectively. Symmetricity and antisymmetricity along the c-axis of the spin density force the $Fe_2/Ir_4(001)$ superlattice system to be FM and AFM states, respectively, to display different layer-by-layer magnetization sequences. As we follow the line along Fe(Ia)-Ir(Ia)-Ir(I-1a)-Ir(I-1b)-Ir(Ib)-Fe(Ib) layer sequence, the layer-by-layer magnetization sequence of the FM state is UUD-DUU, while that of the AFM state is DDU-DUU, where U and D represent spin-up and spin-down states, respectively. These features indicate that relaxation of constraints of inversion and z-reflection symmetry during the self-consistent calculation does not change final results. Since the spin densities on Fe atomic sites are highly localized, one can expect that the magnetic moments of Fe layers are large.

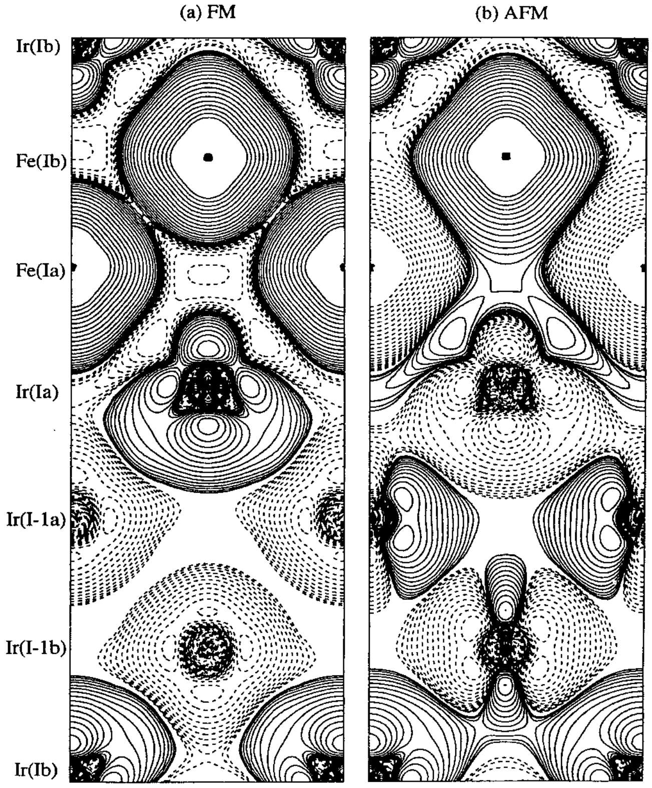

Fig. 1. Spin density contours on (110) plane of the (a) FM and (b) AFM states. Solid lines and broken lines represent spin-up and spin-down, respectively. Contour line starts from 5×10^{-4} electrons/a.u.^{3} and subsequent lines are increased by a factor of $\sqrt{2}$.

Table 1. Calculated magnetic moments (in units of μ_B) of $Fe_2/Ir_4(001)$ superlattice for ferromagnetically (FM) and antiferromagnetically (AFM) coupled Fe layers

Layer	FM	AFM
Fe(Ib)	+2.45	+2.30
Fe(Ia)	+2.45	−2.30
Ir(Ia)	+0.08	−0.08
Ir(I-1a)	−0.06	+0.07
Ie(I-1b)	−0.06	−0.07
Ir(Ia)	+0.08	+0.08

Calculated magnetic moments inside each muffin-tin sphere of $Fe_2/Ir_4(001)$ are summarized in Table 1, both for the FM and AFM states. As expected from the spin density contours in Fig. 1, absolute values of the magnetic moments of Fe layers are large to be 2.45 μ_B and 2.30 μ_B for the FM and AFM states, respectively. These are rather enhanced one with respect to the bulk bcc Fe moment of 2.2 μ_B [14]. Those large magnetic moments contradict to the experimental observations of magnetically "dead" Fe layers [3-5] in Fe/Ir(001) systems. In addition, such large magnetic moments are also calculated in Fe overlayers on Ir(001) surfaces [8]. Consequently, a natural question on origin of the observed magnetically "dead" layers from the experiments is arisen. There are four possibilities to frustrate or kill the magnetic moments at Fe layers: (i) two-dimensional structure of Fe layers is disordered if the thickness of Fe layers is small, (ii) there is segregation of Fe ions into the Ir layers for thin layers, (iii) the Fe layers are antiferromagnetically ordered when the Fe layer thickness is very small, (iv) and there are interface roughness or interdiffusion between Fe and Ir layers, which cannot be detected within the experimental resolution. For a disordered layer, coherence length should be much reduced to increase randomness of magnetic moments at Fe sites so that the total magnetization would appear to be zero. This fact explains the observed large Fe L_3 branching ratio in the X-ray magnetic circular dichroism (XMCD) [5]. Sharp (1×1) low energy electron diffraction (LEED) [5] and reflection high energy electron diffraction (RHEED) [3,4] signals, however, rule out this hypothesis. The RHEED results also eleminate the possibility of Fe segregation [3,4].

A hypothesis of antiferromagnetically ordered Fe layers in Fe/Ir(001) system is regarded as a prominent candidate for explaining the observations of magnetically "dead" Fe layers. With ignoring the different atomic basis, *i.e.*, Fe and Ir ions in the tetragonal unit cell, average Wigner-Seitz radius r_{WS} is calculated to be ~2.78 a.u. According to a fixed-moment calculation [15], fcc Fe could have high-spin (HS) state in ferromagnetic case and antiferromagnetic state. Corresponding magnetic moments are ~3.0 μ_B and ~2.5 μ_B for the HS and AFM states, respectively. In such case, the total energy difference between HS and AFM is ~100 meV, where the FM state is more stable than AFM state. In our case, total energy difference, $\Delta E = E_{FM} - E_{AFM}$, is calculated to be –0.166 eV, *i.e.*, the FM state is more stable than the AFM one. This energy difference is considerably large (~100 times larger than the fcc Fe) so that the possibility of AFM state as a metastable state of Fe layers in the $Fe_2/Ir_4(001)$ superlattice is hardly possible.

Since the experimental interface roughness was controlled within two monolayers [3-5], we, thus, attributed that a possible roughness and interdiffusion between Fe and Ir layers at interface is responsible for observed magnetically "dead" layers. When intermixing, due to interface roughness and interdiffusion, occurs at Fe/Ir interface, wavefunction overlap, or hybridization between Fe-3*d* and Ir-4*d* states, might be increased, because coordination number of other atomic basis is possibly increased whereas interatomic distances between Fe and Ir ions is possibly reduced. Such hybridization effect between the Fe-3*d* and Ir-4*d* states causes increment of diffuseness of Fe-3*d* states to reduce, to frustrate, or to kill absolute value of magnetic moment of Fe ions. A possibility of AFM coupling between Fe ions under such intermixed environments cannot be ignored also, because the interatomic distances between Fe ions should be much extended for inducing the oscillatory exchange coupling between Fe ions through Ir ions. Magnetism of Fe/Ir superlattice under such intermixed situation will be discussed in a paper published elsewhere [16].

4. Summary

To summarize, we have investigated the magnetism of $Fe_2/Ir_4(001)$ superlattice in terms of a first-principles calculation by using the FLAPW method within the GGA. We have considered two magnetic states, FM and AFM states. Calculated magnetic moments are in absolute value to be 2.45 μ_B and 2.30 μ_B for the FM and AFM states, respectively. These result contradicts to recent experimental observations of magnetically "dead" Fe layers in Fe/Ir systems if thickness of Fe layers are less than 5 monolayers. We thought that these observations of "dead" magnetic layers are resulted from interdiffusion between Fe and Ir layers at interface, in viewpoint of total energy. This work requires a more detailed study for interdiffusion effects on the magnetism in Fe/Ir superlattice.

Acknowledgments

This work is supported by 2000 Inha University Specialized Research Project.

References

[1] J. Häglund, Phys. Rev. B **47**, 566 (1993) and references therein.

[2] A. J. Freeman and R. Wu, J. Magn. Magn. Mater. **100**, 497 (1991).

[3] S. Andrieu, E. Snoeck, P. Arcade, and M. Piecuch, J. Appl. Phys. **77**, 1308 (1995).

[4] S. Andrieu, F. L. Razafindramisa, E. Snoeck, and M. Piecuch, Phys. Rev. B **52**, 9938 (1995).

[5] W. L. O'Brien and B. P. Tonner, J. Vac. Sci. Technol. A **13**, 1544 (1995).

[6] R. Wu and A. J. Freeman, Phys. Rev. B **44**, 4449 (1991).

[7] K. Louzazna and A. Haroun, Solid State Commun. **112**, 649 (1999).

[8] I. G. Kim, J. I. Lee, W. Mannstadt, A. J. Freeman, unpublished.

[9] D. C. A. Stoeffler, J. Magn. Magn. Mater. **198-199**, 306 (1999).

[10] E. Wimmer, H. Krakauer, M. Weinert, and A. J. Freeman, Phys. Rev. B **24**, 864 (1981); M. Weinert, E. Wimmer, and A. J. Freeman, Phys. Rev. B **26**, 4571 (1982).

[11] C. Kittel, *Introduction to Solid State Physics* 7th Ed. (Wiley, New York, 1996) p. 23.

[12] W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).

[13] J. P. Perdew, J. A. Chevary, S. H. Vosko, K. A. Jackson, D. J. Singh, and C. Fiolhais, Phys. Rev. B **46**, 6671 (1992).

[14] J. Callaway and C. S. Wang, Phys. Rev. B **16**, 2095 (1977).

[15] V. L. Moruzzi, P. M. Marcus, and J. Kübler, Phys. Rev. B **39**, 6957 (1989).

[16] I. G. Kim, J. I. Lee, and S. C. Hong, unpublished.