# Giant perpendicular magnetic anisotropy of an Ir monolayer on a NiAl(001) surface

Dongyoo Kim, Jeonghwa Yang, and Jisang Hong
*Department of Physics, Pukyong National University, Busan 608-737, Korea*
(Received 13 May 2009; revised manuscript received 4 July 2009; published 21 August 2009)

Using the state-of-the-art full potential linearized augmented plane-wave method, we have investigated the magnetic properties of Os and Ir monolayer (ML) film on NiAl(001) surface. It has been found that the one ML of Os and Ir film can have ferromagnetic ground state with magnetic moment of 0.35 and $0.64\mu_B$ on Ni terminated surface, whereas both films display no sign of magnetic state on Al terminated surface. In addition, the surface Ni atom has an induced magnetic moment of $0.26\mu_B$ in Ir/NiAl(001), while only $0.09\mu_B$ is observed in Os/NiAl(001). We attribute the existence of magnetism to the interaction between 5*d* of adlayer and 3*d* of surface Ni. Moreover, we have obtained that the Os/NiAl(001) and Ir/NiAl(001) films show a perpendicular magnetic anisotropy (PMA). Surprisingly, it appears that the Ir/NiAl(001) has a giant PMA energy of 7.18 meV.

DOI: 10.1103/PhysRevB.80.052404 PACS number(s): 75.30.Gw, 75.75.+a

In most of studies for magnetic properties of materials, the conventional 3*d* transition-metal elements such as Mn, Fe, Co, and Ni have been focused because it is believed that the itinerant magnetism is originated from the 3*d* electrons. However, on theoretical side the possibility of magnetism in 4*d* or 5*d* materials has also long been extensively investigated. According to many previous studies, it is found that even the 4*d* and 5*d* film can display magnetic state on nonmagnetic metal surface such as Ag(001).[1–6] Along with these theoretical studies, one can find experimental evidence that the 4*d* monolayer and cluster systems may display magnetic state.[7,8] In these studies, the major effort has been focused on the magnetic ground state and the magnitude of magnetic moment.

Magnetic anisotropy is one of the most fundamental and important physical quantities for both fundamental interests and novel magnetic device application purposes. Indeed, the perpendicular magnetic anisotropy (PMA) with large saturation magnetization is of interest for high-density magnetic recording media. In general, the strong spin-orbit coupling (SOC) is anticipated in late transition-metal series and it is expected to observe large magnetic anisotropy energy (MAE). In this respect, many different types of 3*d*–5*d* or 3*d*–4*d* hybridized systems are mostly explored.[9–13] Of course, one should note that the MAE is not only influenced by the absolute value of SOC, but also dependent on the wave-function character. Despite extensive studies on the magnetic ground state in 4*d* or 5*d* films, it is rare to find the investigation for magnetic anisotropy with realistic structure although the free standing system has been calculated.[14,15] In our previous experiences, we have observed that the MAE in free standing state can be substantially altered in the presence of supporting material. Thus, the inclusion of hybridization with substrate is essential for realistic material. Furthermore, no one has ever studied the possibility of giant PMA in pure 5*d* transition-metal film grown on specific surface. In this view, we will explore whether one can find 5*d* thin-film systems which show both ferromagnetic (FM) and giant PMA feature. To this aim, we choose Os and Ir thin films on NiAl(001) surface.

The full potential linearized augmented plane (FLAPW) method for electronic and magnetic properties is used. Therefore, no shape approximation is assumed in charge, potential, and wave-function expansions.[16–18] We treat the core electrons fully relativistically, and the spin-orbit interaction among valence electrons are dealt with second variationally.[19] The generalized gradient approximation is employed to describe exchange and correlation potentials.[20] Spherical harmonics with $l_{max}=8$ are used to expand the charge, potential, and wave functions in the muffin-tin region. Energy cutoffs of 225 Ry and 13.7 Ry are implemented for the plane-wave star function and basis expansions in the interstitial region. The lattice constant of 2.88 Å in lateral direction is employed and the NiAl(001) substrate is simulated by nine layers of slab. The employed muffin-tin radius of 5*d* and 3*d* elements is 2.5 atomic unit and 2.2 atomic unit, respectively. For Al atom, the muffin-tin radius of 1.6 atomic unit is used. It is assumed that the Os and Ir films are pseudomorphically grown on NiAl(001) surface. Nonetheless, the atomic position in vertical direction is relaxed and the optimized atomic structure is obtained via force and total-energy minimization procedure. We use 210 *k*-mesh points with Monkhorst-Pack method[21] during the course of entire calculations discussed in this report.

We have first calculated the magnetic state of free standing Os and Ir films of 1 ML thickness. Three magnetic states such as FM, antiferromagnetic (AFM), and paramagnetic (PM) spin configurations are considered and the total energy of FM state is set to zero for reference. In Table I, the calculated total energies, spin, and orbital magnetic moments are presented. It is clearly shown that both Os and Ir films have FM ground state with large magnetic moment. One may note that the spin and orbital magnetic moments of Ir film in AFM state are almost negligible; however the energy difference compared with PM state is noticeable. This is due to the inclusion of SOC in AFM calculation. The next issue is to find the indication of magnetic state in Os and Ir films on NiAl(001) surface. The NiAl(001) alloy substrate can have two different surface geometries such as Ni terminated and Al terminated structures. We have considered both systems and found that the Os and Ir films manifest nonmagnetic state on Al terminated surface. Therefore, the Ni terminated surface has been chosen in the following calculations. The optimized atomic structure of Os/NiAl(001) and Ir/

 

TABLE I. Calculated total energy ($E$), spin magnetic moment ($\mu_S$), and orbital magnetic moment ($\mu_L$) of free standing Os and Ir atoms.

| | FM | AFM | PM |
|---|---|---|---|
| Os film | | | |
| E (meV) | 0 | 25 | 278 |
| $\mu_S$ | 1.30 | 1.42 | |
| $\mu_L$ | 0.14 | 0.28 | |
| Ir film | | | |
| E (meV) | 0 | 96 | 284 |
| $\mu_S$ | 1.55 | 0.002 | |
| $\mu_L$ | 0.45 | 0.001 | |

TABLE III. Calculated total energy ($E$), spin magnetic moment ($\mu_S$), and orbital magnetic moment ($\mu_L$) of 1 ML thickness Os, Ir, and surface Ni atoms.

| | FM | AFM | PM | PM(SOC) |
|---|---|---|---|---|
| Os film | | | | |
| E(meV) | 0 | 5 | 174 | 4 |
| $\mu_S$(Os, Ni) | 0.35,0.09 | 0.04,0.0 | | |
| $\mu_L$(Os, Ni) | 0.007,0.004 | 0.003,0.0 | | |
| Ir film | | | | |
| E(meV) | 0 | Not appear | 178 | 8 |
| $\mu_S$(Ir, Ni) | 0.64,0.26 | | | |
| $\mu_L$(Ir, Ni) | 0.21,−0.02 | | | |

NiAl(001) is presented in Table II. The relative distance between Os or Ir adlayer and Ni surface is denoted by $d(s\text{-}1)$ and other $d(i\text{-}j)$ stands for the relative distance of adjacent two layers in substrate material. We find that the adsorption of 1 ML of Os and Ir films does not significantly affect the geometric feature of substrate material although the Ir adlayer induces small inward relaxation. With this optimized structure, the calculated total energy and magnetic moments are shown in Table III. Here, we have also presented the total energy of PM state with inclusion of SOC for comparison with the energy of AFM state. The Os 1 ML film still maintains the FM ground state, but both spin and orbital magnetic moments are significantly suppressed on NiAl(001). This feature is definitely due to the hybridization with supporting material. In Ir/NiAl(001), the FM ground state is achieved as well and the spin magnetic moment is about $0.64\mu_B$. The induced spin magnetic moment of $0.26\mu_B$ in surface Ni is observed too. Besides, it is found that the spin alignment of Ir atoms is always parallel after self-consistent calculation even if we initially set up antiparallel spin configuration. Through the total-energy calculations, we have obtained that the Os/NiAl(001) and Ir/NiAl(001) films have adsorption energies of 2.885 and 2.95 eV, respectively. Note that the calculated adsorption energy of Os and Ir films on Al terminated surface is 3.36 and 3.73 eV, respectively. We have further analyzed the reason why the Ir and Os films have FM state only on Ni terminated surface. In Ir/NiAl(001), the charge transfer about 0.11 electrons from Ir $5d$ to Ni $3d$ in majority-spin state is found, while the reverse transfer about 0.15 electrons from Ni $3d$ to Ir $5d$ in minority-spin state takes place. In Os/NiAl(001), the similar hybridization between Os $5d$ and Ni $3d$ electron is observed, but the charge transfer is very weak compared with that in Ir/NiAl(001) system. Due to this feature, the surface Ni has very small induced magnetic moment in Os/NiAl(001) film. On Al terminated surface, there is no such $d$ state in Al atom and this results in nonmagnetic state. Thus, we believe that the ferromagnetism stems from the hybridization between $5d$ of adlayer and $3d$ of surface Ni.

In Figs. 1(a) and 1(b), the density of states (DOS) of Os and Ir films is presented. It is clearly that the free standing Os and Ir films have a spin asymmetry and the Ir film has larger exchange splitting than that of Os film. On NiAl(001) surface, the spectral shapes of DOS of Os and Ir films have been changed and the broadening of band width is observed due to the hybridization with substrate. In particular, it has been found that the Ir layer is more strongly hybridized with surface Ni layer. This causes sizable induced spin polarization in surface Ni, whereas the spin asymmetry is quite weak in Os/NiAl(001). Overall, the DOS feature nicely account for the calculated magnetic moments.

The central issue of this report is to investigate the possibility of giant PMA in $5d$ transition-metal system. For the calculations of MAE arising from SOC, we employ torque method which provides very stable results.[22] The positive MAE stands for perpendicular magnetization to the film surface, whereas the negative one means in-plane magnetization. The calculated MAE is presented in Table IV. As shown, the free standing Os film has a large PMA energy of 23.26 meV, while the Ir free film has an in-plane magnetization. However, on NiAl(001) surface, the MAE of Os film is substantially suppressed although the PMA is still preserved. Very interestingly, the Ir film displays spin reorientation transition from in-plane to perpendicular to the film surface with giant PMA energy of 7.18 meV due to the interaction with substrate atoms. Note that the calculated shape anisotropy of Os and Ir films is 23 $\mu$eV and 31 $\mu$eV, respectively.

Through the decomposition of SOC channels, it is found that the MAE from spin-flip interactions, i.e., E(↑↓) plays an essential role for the direction of magnetization as shown in Table IV. Beside, the orbital anisotropy of supported systems is presented. One can see that the orbital anisotropy is quite

TABLE II. Calculated vertical distances (in Å) of Os and Ir films on Ni terminated surface.

| | $d(s-1)$ | $d(1-2)$ | $d(2-3)$ | $d(3-4)$ | $d(4-5)$ |
|---|---|---|---|---|---|
| Os | 1.51 | 1.43 | 1.44 | 1.45 | 1.45 |
| Ir | 1.53 | 1.39 | 1.47 | 1.44 | 1.45 |

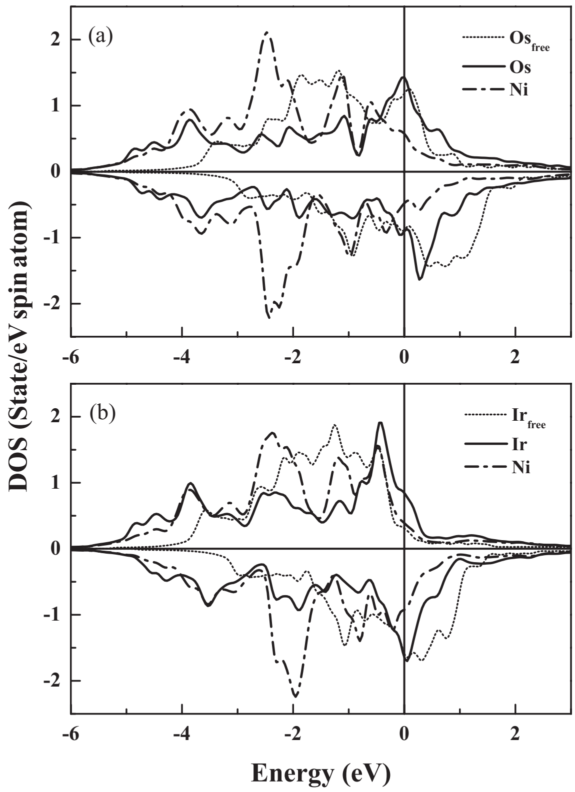


FIG. 1. Calculated DOS; (a) Os free standing and on NiAl(001), (b) Ir free standing and on NiAl(001). The dotted line is for DOS of free standing. The solid and dash-dotted lines are for adlayer and surface Ni, respectively.

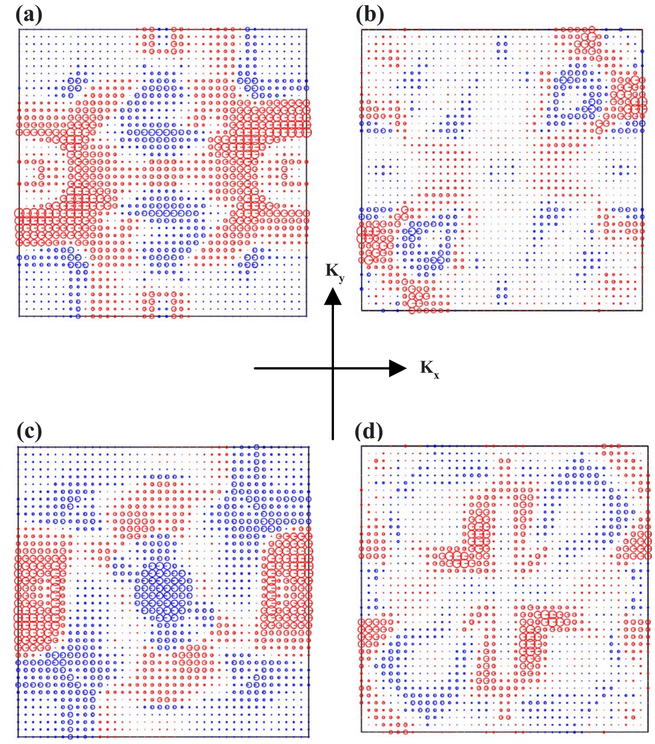


FIG. 2. (Color online) Distribution of magnetic anisotropy energy over two-dimensional BZ; (a) free standing Os, (b) Os/NiAl(001), (c) free standing Ir, and (d) Ir/NiAl(001).

small and even the direction is different from the spin magnetization. Thus, the simple interpretation in terms of orbital anisotropy cannot be applicable to this system. In this issue, one may argue that the more generalized approach has been proposed,[23] but the validity of such interpretation for magnetic anisotropy is not yet clear. It is also feasible to analyze the wave-function character provided that the magnetic anisotropy is mainly determined by one specific state among five possible $d$ wave functions. Nevertheless, it is unrealistic to assume that all the wave functions can be described by one particular state over the whole two-dimensional Brillouin zone (2D BZ).

In Fig. 2, we thus present the distribution of magnetic anisotropy over the 2D BZ. The red circle implies PMA energy and the blue circle means in-plane contribution at a given $k$ point. Note that the magnitude of MAE is proportional to the size of circle. For free standing Os in Fig. 2(a), one can clearly see that large portion of 2D BZ contains strong PMA and this results in huge PMA energy. On NiAl(001) surface in Fig. 2(b), relatively small area contributing to PMA is observed around the corner of BZ. This causes significant reduction in PMA energy. For Ir free standing film in Fig. 2(c), we have found very strong contributions to PMA near the BZ boundary, roughly along the $k_y$ direction. Nonetheless, the contribution to in-plane magnetization is found in large area. By adding all these counteracting effects, the free standing 1 ML of Ir film shows large in-plane MAE. Very interestingly, on NiAl(001) surface, the in-plane contributions are significantly suppressed. Of course, one can also observe that the area used to maintain strong PMA in free standing state disappears as well. But, the other part of 2D BZ displays large PMA and this compensates the absence of PMA. Overall, the giant PMA is realized in Ir/NiAl(001). As shown in Fig. 2, we find that various $k$ points in arbitrary direction provide similar magni-

TABLE IV. Calculated magnetic anisotropy energy (in meV) and orbital moment anisotropy (in $\mu_B$). The positive MAE corresponds to perpendicular magnetization, and the negative one is for in-plane magnetization.

| | Free standing | On surface | E(↑↑) | E(↓↓) | E(↑↓) | $\langle L_z \rangle$ | $\langle L_x \rangle$ |
|---|---|---|---|---|---|---|---|
| Os film | | | | | | | |
| E(meV) | 23.26 | 1.57 | 0.93 | −2.94 | 2.12 | 0.04 | 0.10 |
| Ir film | | | | | | | |
| E(meV) | −8.27 | 7.18 | −0.47 | −8.40 | 21.15 | 0.30 | 0.34 |

tude of MAE for both in-plane and PMA. This indicates that it is impossible to single out few dominant contributions to magnetic anisotropy. Instead, one has to take into account all the opposite contributions and the overall magnetic anisotropy is arisen from accumulative effect at each $k$ point. This complex behavior hinders us from simply interpreting the result in terms of correlation between magnetic anisotropy and wave-function change along particular direction.

In conclusion, we have investigated the magnetic properties of Os and Ir films on NiAl(001) surface. It has been achieved that the Os and Ir films have FM ground state on Ni terminated NiAl(001) surface with magnetic moment of 0.35 and $0.64\mu_B$, but the FM state is missing on Al terminated surface. It is found that the surface Ni has an induced magnetic moment of $0.26\mu_B$ in Ir/NiAl(001), whereas very weak induced moment is observed in Os/NiAl(001). We attribute the existence of FM state to the hybridization between $5d$ of adlayer and $3d$ of surface Ni. It has been obtained that both Os/NiAl(001) and Ir/NiAl(001) films have PMA. It is of interest to note that the Ir film manifests a giant PMA with MAE of 7.18 meV. This is the first theoretical prediction that the huge magnetic anisotropy in 2D film can be realized even in the presence of supporting surface and we hope that this result will stimulate further experimental verification.

This work was supported by the Korea Science and Engineering Foundation (KOSEF) grant funded by the Korea government (MEST) (Grant No. R01-2008-000-20014-0), and by the Korea Research Foundation Grant funded by the Korean Government (MOEHRD, Basic Research Promotion Fund) (Grant No. KRF-2007-331-C00102).

[1] S. Blügel, Phys. Rev. Lett. **68**, 851 (1992).
[2] M. J. Zhu, D. M. Bylander, and L. Kleinman, Phys. Rev. B **43**, 4007 (1991).
[3] R. Q. Wu and A. J. Freeman, Phys. Rev. B **45**, 7222 (1992).
[4] V. S. Stepanyuk, W. Hergert, K. Wildberger, R. Zeller, and P. H. Dederichs, Phys. Rev. B **53**, 2121 (1996).
[5] I. Turek, J. Kudrnovsky, M. Sob, V. Drchal, and P. Weinberger, Phys. Rev. Lett. **74**, 2551 (1995).
[6] Olle Eriksson, R. C. Albers, and A. M. Boring, Phys. Rev. Lett. **66**, 1350 (1991).
[7] R. Pfandzelter, G. Steierl, and C. Rau, Phys. Rev. Lett. **74**, 3467 (1995).
[8] A. J. Cox, J. G. Louderback, S. E. Apsel, and L. A. Bloomfield, Phys. Rev. B **49**, 12295 (1994).
[9] C. Moutafis, S. Komineas, C. A. F. Vaz, J. A. C. Bland, T. Shima, T. Seki, and K. Takanashi, Phys. Rev. B **76**, 104426 (2007).
[10] A. P. Mihai, J. P. Attané, A. Marty, P. Warin, and Y. Samson, Phys. Rev. B **77**, 060401(R) (2008).
[11] I. Galanakis, M. Alouani, and H. Dreyssé, Phys. Rev. B **62**, 6475 (2000).
[12] M. Abes, M. V. Rastei, J. Vénuat, L. D. Buda-Prejbeanu, A. Carvalho, G. Schmerber, J. Arabaski, E. Beaurepaire, J. P. Bucher, A. Dinia, and V. Pierron-Bohnes, Mater. Sci. Eng., B **126**, 207 (2006).
[13] T. Seki, T. Shima, K. Yakushiji, K. Takanashi, G. Q. Li, and S. Ishio, IEEE Trans. Magn. **41**, 3604 (2005).
[14] Y. Mokrousov, G. Bihlmayer, S. Heinze, and S. Blügel, Phys. Rev. Lett. **96**, 147201 (2006).
[15] Lucas Fernández-Seivane and Jamie Ferrer, Phys. Rev. Lett. **99**, 183401 (2007).
[16] E. Wimmer, H. Krakauer, M. Weinert, and A. J. Freeman, Phys. Rev. B **24**, 864 (1981).
[17] M. Weinert, E. Wimmer, and A. J. Freeman, Phys. Rev. B **26**, 4571 (1982).
[18] M. Weinert, J. Math. Phys. **22**, 2433 (1981).
[19] D. D. Koelling and B. N. Hamon, J. Phys. C **10**, 3107 (1977).
[20] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
[21] H. J. Monkhorst and J. D. Pack, Phys. Rev. B **13**, 5188 (1976).
[22] X. D. Wang, R. Q. Wu, D. S. Wang, and A. J. Freeman, Phys. Rev. B **54**, 61 (1996).
[23] G. van der Laan, J. Phys.: Condens. Matter **10**, 3239 (1998).