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Magnetic Properties of Transition Metal Monolayers on Ta(001) Surfaces

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The magnetic and structural properties of transition metal (Mn, Fe, Co) monolayers on Ta(001) surfaces are investigated theoretically by using the first principles full-potential linearized augmented plane wave method. Mn and Fe monolayers become ferromagnetic on Ta(001) surfaces while Co monolayers becomes non-magnetic. The paramagnetism of Co monolayers is explained by the Stoner theory of magnetism. The magnetic coupling of a transition metal overlayer with a substrate is ascribed to the orbital hybridization between the *s* and *d* orbitals of the transition metal.

Keywords : transition metal, magnetism, surface magnetism, Ta

1. Introduction

When transition metals are put on a surface, they exhibit different forms of magnetic behavior depending on the conditions of the substrate. For example, an Mn monolayer becomes ferromagnetic on a W(001) surface while it is an antiferromagnet on noble metal surfaces. However, bulk Mn is a complex antiferromagnet. An Fe monolayer becomes an antiferromagnet on a W(001) surface while it is a ferromagnet on noble metal surfaces. Fe is a typical ferromagnet in bulk state. Co is also predicted to be an antiferromagnet on a W(001) surface and a ferromagnet on noble metal surfaces [2]. The atomic number of the transition metal plays an important role in determining the magnetic behavior, since the magnetic moment is determined on how the d-orbital is filled with electrons. For example, early transition metals (Cr, Mn) are antiferromagnetic and late transition metals (Fe, Co, Ni) are ferromagnetic on Pd(001). This trend is reversed on a W substrate [3].

The magnetic behavior of transition metals on W(001) surfaces has been studied in detail by the Blügel group [2, 3]. It will be interesting to compare the magnetic properties when the substrates are changed to materials other than W. Ta is a good candidate as a substitute of W. Ta has the same bcc crystal structure as W with an enlarged lattice constant. Furthermore, Ta belongs to the same 5*d* series as W with an atomic number that is smaller by one. We have investigated theoretically the magnetic properties of transition metal (Mn, Fe, and Co) monolayers grown on Ta(001) surfaces by using the first principle full-potential linearized augmented plane wave (FP-LAPW) method.

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2. Method

The electronic structure was calculated using the FP-LAPW [4, 5] method within a generalized gradient approximation [6]. The FP-LAPW method is an all electron calculation with no shape approximation for charge densities, potentials, and wave functions. The charge densities and potentials inside the muffin-tins are expanded by the lattice harmonics up to $l_{max}=8$. Core electrons are treated fully relativistically and valence electrons are treated semirelativistically. Transition metal monolayers on Ta(001) surfaces were modeled by single slabs with 9 symmetrical layers in total. The experimental value of the lattice constant of 3.30 Å for bulk Ta was adopted. The muffin-tin radii are 2.4 a.u. for Ta and 2.2 a.u. for 3d-transition metals. The interlayer distances between the Ta(001) surfaces and the transition metal monolayers were determined by minimizing the total energies. The magnetic behavior of paramagnetic (PM), antiferromagnetic (AFM) and ferromagnetic (FM) states was considered. For magnetic calculations, a p(1×1) structure was

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used for PM and FM and a c(2×2) structure was used for AFM. We used k_{max}=3.0 for Fe/Ta(001) and Co/Ta(001), which corresponds to about 1400 basis functions. For Mn/Ta(001), k_{max}=2.9 was used, which corresponds to about 1300 basis functions. 21 $k_{||}$ points in the irreducible wedge of the two dimensional Brillouin zone were used for integration in k-space in all the calculations.

3. Results

The total energies were calculated to obtain an equilibrium distance between the overlayer and the top layer in the PM, AFM, and FM states. Fig. 1 shows the total energy curves for Fe/Ta(001) as a typical example. The PM states have the highest total energy while the FM states have the lowest total energy. The total energy of Mn/Ta(001) shows a similar behavior as that of Fe/Ta (001). Co atoms are expected to have a magnetic moment of about 1 μ_B on Ta(001) due to the tendency that the magnitude of the magnetic moment decreases as the atomic number increases from Mn to Co. However, Co/Ta(001) is nonmagnetic at its equilibrium distance. The paramagnetism of Co/Ta(001) is explained below by the Stoner theory of magnetism. The results regarding structure and magnetic behavior are summarized in Table 1. The FM states are stable for the Mn and Fe overlayers on a Ta(001) surface while the PM state is most stable for the Co overlayer. Kubetzka *et al.* also reported an FM ground state for Fe/Ta(001) [1]. The magnetic moment of Fe, 2.1 μ_B, is similar to the bulk value of 2.2 μ_B. This stands in contrast to the usual tendency that the magnetic moment increases on a surface [7]. It has been reported that the magnetic moment of Fe is reduced to 1.72 μ_B due to relaxation on a Cr (001) surface [8]. Among the overlayers, both the interlayer distance and the magnetic moment are largest for Mn. The equilibrium PM overlayer distances are 2.18 Å, 2.08 Å, and 2.15 Å for Mn, Fe, and Co, respectively. When magnetism is turned on, the overlayers distances are increased, which means that magnetic interactions are important in determining the overlayer distances.

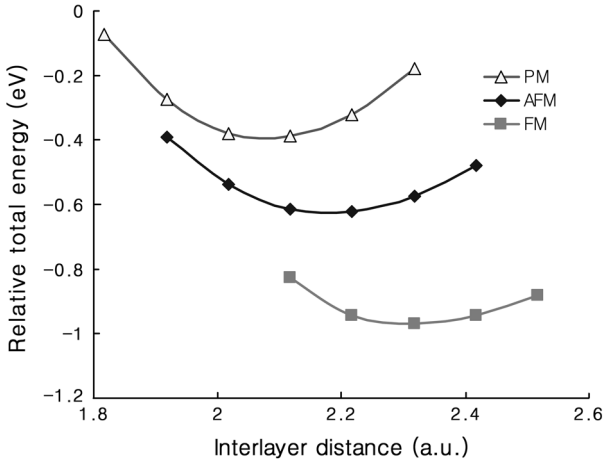

Fig. 1. Relative total energies of Fe/Ta(001) in PM, AFM, and FM states as a function of the interlayer distance. Zero energy has no significant meaning here but is the total energy for a paramagnetic state.

Table 1. Equilibrium properties.

	Magnetic state	Magnetic moment (μ_B)	Overlayer distance (Å)
Mn	FM	3.1	2.66
Fe	FM	2.1	2.31
Co	PM	0.0	2.15

For a better understanding of the paramagnetic behavior of Co/Ta(001), the *d*-orbital local DOS (LDOS) of the overlayer Co atom as well as of Fe and Mn are plotted in Fig. 2 for the equilibrium overlayer distances. The change of overall shape of the LDOS as the atomic number changes can be explained with the rigid band model. As the atomic number increases from Mn to Co, the energy band moves towards lower energy as the band is filled. There is a peak at the Fermi level in the LDOS of Mn while the Fermi level is located at the trough of the LDOS for Co. According to the Stoner theory of magnetism, a paramagnetic state is not stable if $IN(0) > 1$ where $N(0)$ is the LDOS at the Fermi level and I is the Stoner parameter of the transition metal overlayer atom. The Stoner parameters are 0.41 eV, 0.46 eV, and 0.49 eV for metallic Mn, Fe, and Co, respectively [2]. Hence, the Stoner factors [$IN(0)$] are 1.62, 1.32, and 0.38 for the Mn, Fe, and Co overlayers, resepectively, which means that the PM state is stable for Co/Ta(001) and unstable for Mn/Ta(001) and Fe/Ta(001). Although we have used the Stoner parameters of the bulk transition metals, the Stoner theory explains the paramagnetism of Co/Ta(001) quali-

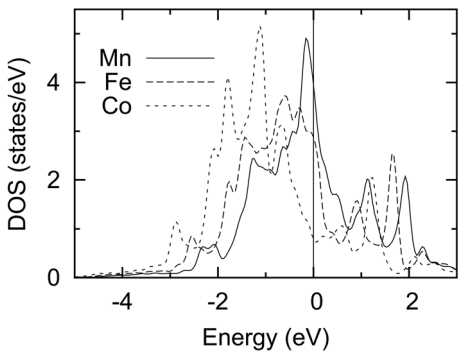

Fig. 2. *d*-orbital LDOS of the overlayers (Co, Mn, and Fe) at the equilibrium overlayer distance.

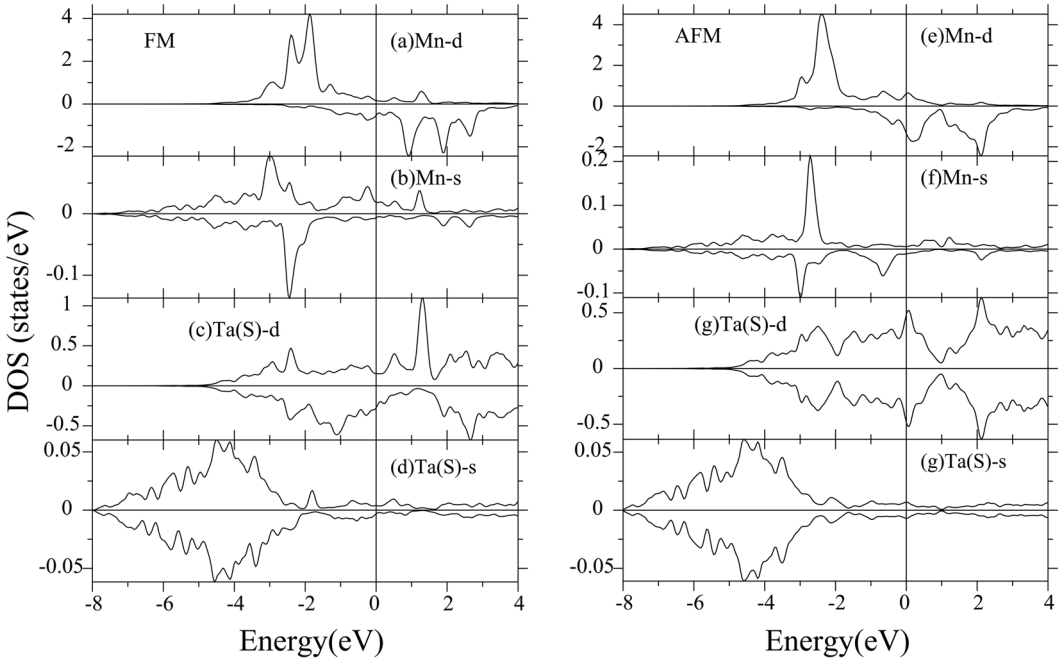

Fig. 3. LDOS of the overlayer Mn and top layer Ta at the equilibrium interlayer distance. Positive or negative sign of LDOS represents spin direction.

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In order to explain the ferromagnetic properties of Mn and Fe monolayers on Ta(001), we investigated the LDOS of the transition metal overlayers and the top layer Ta. For example, Fig. 3 shows the local DOSs of the Mn-*s*, Mn-*d*, Ta-*s*, and Ta-*d* orbitals for the FM and AFM Mn/Ta(001). The Mn-$d\uparrow$ band is almost fully occupied, while the Mn-$d\downarrow$ is partially occupied, which gives Mn a magnetic moment of 3.1 μ_B. The magnetic moment in Mn atoms becomes stable through coupling with substrate atoms as well as other Mn atoms in the overlayer. Since the Mn-*d* electrons are localized in character, coupling occurs via

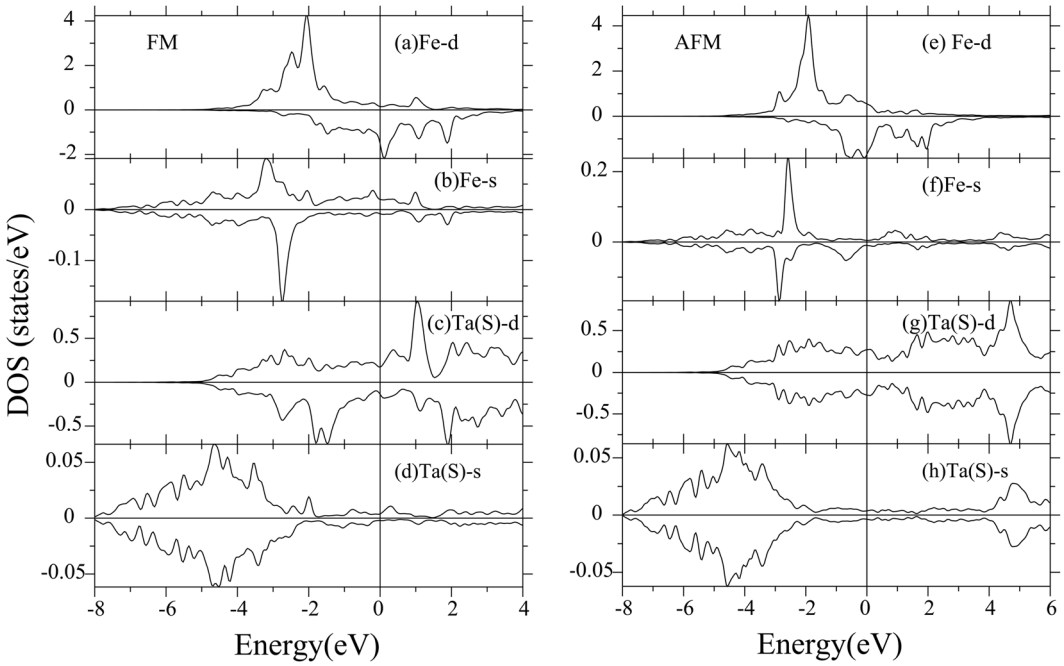

Fig. 4. LDOS of the overlayer Fe and top layer Ta at the equilibrium interlayer distance. Positive or negative sign of LDOS represents spin direction.

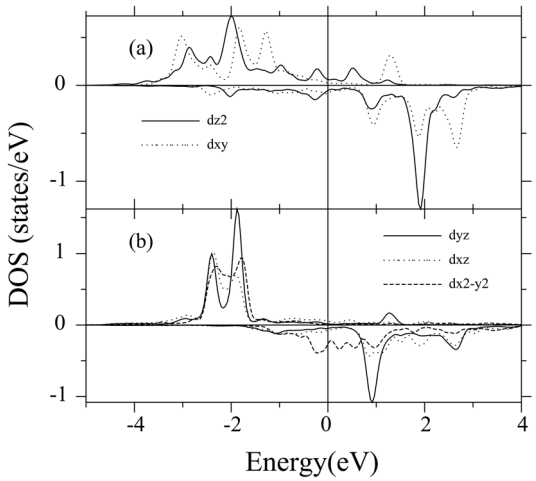

Fig. 5. LDOS split by the crystal field of overlayer *d*-orbitals at the equilibrium interlayer distance. (a) d_{z^2} (solid) and d_{xy} (dotted) (b) d_{yz} (solid), d_{xz} (dotted), and $d_{x^2-y^2}$ (dashed).

the overlayer *s*-orbitals. Considering that magnetic moment is determined by the Mn-$d\uparrow$ orbital, the ↑ electrons in the Mn-*s* orbital, that is the Mn-$s\uparrow$ electrons, play an important role. The Mn-$s\uparrow$ electrons hybridize with Mn-$d\uparrow$ electrons at –3.0 eV below the Fermi level for FM as can be seen in Fig. 3(b), while this occurs at –2.7 eV for AFM as can be seen in Fig. 3(f). The lower total energy of the FM state than that of the AFM state can be ascribed to the location of Mn-$s\uparrow$ orbitals in the DOS. The Ta-*d* electrons of the top layer are more important than the Ta-*s* electrons in coupling with Mn atoms as can be seen in Figs. 3(c) and (d). Fig. 4 depicts the local DOSs of the Fe-*s*, Fe-*d*, Ta-*s*, and Ta-*d* orbitals for the FM and AFM Fe/Ta(001). The Fe-$s\uparrow$ electronic states are located at –3.2 eV for FM and –2.6 eV for AFM, which reflects the lower total energy of FM than that of AFM.

In order to observe in detail the role of the Mn-*d* orbital in hybridization, the LDOSs of Mn-*d* orbitals were split according to the crystal field in Fig. 5 as a typical example. Note the DOS at –3 eV, where the magnetic coupling is made, in which electronic states consist mainly of Mn-d_{z^2} and Mn-d_{xy} orbitals. From the shape of the orbitals, we can expect that Mn-d_{z^2} electrons are magnetically coupled with the substrate and that Mn-d_{xy} electrons couple with adjacent Mn atoms in the overlayer. Electrons from d_{yz}, d_{xz}, and $d_{x^2-y^2}$ orbitals are most important in the forming of the magnetic moments in the overlayer.

4. Conclusion

We have investigated the structural and magnetic properties of transition metal monolayers on Ta(001) surfaces. Fe and Mn monolayers have ferromagnetic ground states while a Co monolayer has a paramagnetic ground state. The paramagnetic behavior of Co layers can be explained with the Stoner theory since the paramagnetic state has a low DOS at the Fermi level while those of Mn and Fe are high enough to give rise to magnetic orderings. Magnetic coupling of the transition metal overlayer occurs when the *s* electrons of the transition metal hybridize with d_{z^2} and d_{xy} orbitals in the same atom. From the shape of the orbitals, we see that the d_{z^2} electrons are to couple with the substrate and that the d_{xy} electrons are to couple with other adjacent transition metals in the overlayer.

Acknowledgments

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