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Magnetic anisotropies of iron on the Pt(111) surface

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Abstract

We have studied magnetic anisotropies of Fe atoms on the platinum (111) surface, employing a fully relativistic pseudopotential and plane wave method with the local spin density approximation. We investigated three surface structures with different Fe monolayer coverages: full coverage, half-coverage and quarter-coverage. The effect of surface relaxation has been included. It was found that the magnetic easy axis of the system is within the surface plane for all systems investigated. In the system of an Fe chain on Pt(111), having an anisotropic local structure, the magnetic anisotropy energy is much enhanced after surface relaxation. This absolute value is larger compared with the value for bulk alloy and the magnetic easy axis is directed parallel to the alignment of Fe atoms.

1. Introduction

Stable high density magnetic recording media whose magnetic moment is rigid against thermal fluctuations are required. Magnetic anisotropy is the important property for stability of the media. FePt and CoPt alloys are considered good candidate of materials for developing new media in the next generation due to their large perpendicular magnetic anisotropy energy (MAE) [1–4].

Gambardella and co-workers fabricated a Co nanowire on Pt surface by using a self-assembly epitaxial technique and characterized magnetic properties of the system [5]. Subsequently, his group also investigated magnetic properties of Co nanoparticles containing several Co atoms on Pt surface both experimentally and theoretically [6]. A similar system, a thin Fe layer on the Pt surfaces, has been studied experimentally by several groups [7–9]. The result of Repetto *et al* indicated that the direction of the magnetic easy axis was changed depending on the coverage of the iron layers. Thus the magnetic anisotropy is sensitive to the local structure in the system consisting of magnetic atoms and transition metal substrate. The bottom-up approaches which have been performed theoretically could be important for understanding magnetic anisotropy deeply [6, 10, 11]. For example, Gambardella *et al*

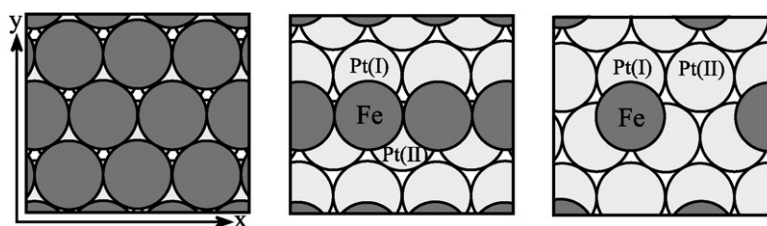


Figure 1. The schematic view of the surfaces; Fe full layer (left), Fe chain (middle), Fe quarter-layer (right). Fe and Pt atoms are specified by dark and light grey balls, respectively. The indications of Pt(I) and Pt(II) in the middle and right panels specify two kinds of Pt atoms in the top layer of the Pt substrate.

predicted a larger magnetic anisotropy energy of single cobalt atoms deposited onto Pt(111) surfaces. The works on the surface magnetism for Fe monolayers on W(110) have used an optimized atomic structure to calculate the magnetic anisotropy [12].

In the present work, by means of a fully relativistic pseudopotential approach, we have studied magnetic anisotropies and presented MAE of the assumed Fe monolayers on Pt(111) surface. These systems were found to have in-plane magnetic anisotropy. Although the preliminary results have been partially published elsewhere [13], in this work we found a larger MAE as a consequence of geometry optimizations and added results for a new surface having a low coverage.

2. System and method

The system considered is a repeated slab model with Fe atoms on Pt(111) surface. The surface structures of Fe and Pt atoms have never been determined. To estimate MAEs, however, we investigated the prototypes of three coverages for Fe monolayers: full coverage, half-coverage and quarter-coverage. In this work, we assumed that the Fe atoms occupied the fcc hollow sites on Pt(111) surface in all cases. For half-coverage, Fe atoms were assumed to form chains along the x -direction. The surface normal was taken as the z -direction. In the case of the quarter-coverage, the Fe atom does not touch the other Fe atoms. The schematic views from the surface normal are shown in figure 1. In the case of half- and quarter-coverages, there are two kinds of Pt atoms in the top layer. They are represented by Pt(I) and Pt(II) in figure 1. In the Fe chain system, the Fe atom has three Pt neighbours; one Pt(I) and two Pt(II) atoms, while in the Fe quarter-layer system, Fe has three Pt(I) neighbours and Pt(II) does not touch the Fe atom.

The distance between Fe and Pt was set to the length of the nearest Fe–Pt bond in the FePt alloy (5.11 au) as the initial geometry for the systems of full coverage and half-coverage. For the Fe–Pt length in quarter-coverage, we used the optimized counterpart in the system of half-coverage. We used the Pt substrate having a (111) surface and four atomic monolayers, which is extracted from the Pt crystal structure. A vacuum layer about 14 au thick was assumed.

We employed the local spin density approximation [14] in the Kohn–Sham theory [15]. We used the pseudopotential plane wave method [16, 17], the fully relativistic version [18, 19] in which the wavefunction has the two-component spinor form [20]. This method self-consistently contains the spin–orbit coupling (SOC), which is the main origin of magnetic anisotropy. The energy cut-offs of 30 and 300 Ryd were taken for wavefunctions and densities, respectively [17]. With this approach, we have relaxed only the atomic positions of Fe and Pt atoms in the first and second layers. The MAE is estimated from $E_x - E_z$ and $E_y - E_z$, where E_α is the total energy when the system is magnetized in the α -direction.

Table 1. The coordinates of surface Pt atoms (in au) with respect to the Fe atom. The ideal case means that Fe atoms are located at the fcc hollow sites extracted from Pt crystal structure and no surface relaxation is introduced. The third columns specify the number of nearest neighbours of Fe atoms around the Pt atom. The fourth column indicates the distance between Fe and Pt atom (in au).

		Number	Fe–Pt(1)	x	y	z
Fe full layer (ideal)	Pt	3	5.23	0.00	3.02	−4.27
Fe full layer	Pt	3	4.89	0.00	3.02	−3.85
Fe chain	Pt(I)	1	4.56	0.00	2.94	−3.50
	Pt(II)	2	4.69	2.62	−1.69	−3.51
Fe quarter-layer	Pt(I)	1	4.49	0.00	3.11	−3.25
	Pt(II)	0	6.90	5.23	3.02	−3.34

We have obtained the MAE in FePt alloy as a reasonable value previously [19], in which we used a dense mesh for \mathbf{k} -point sampling. In the present work we used the 12×12 mesh for the system of Fe quarter-layer and the 24×24 mesh for all the other cases [21]. The MAEs would be fairly well converged with respect to \mathbf{k} -points [13]. The spin and orbital magnetizations on atoms are estimated in the sphere with the radius of 2.5 au for Fe and Pt atoms.

3. Results and discussion

The Fe full layer of the ideal has the distance of 4.27 au ($=d_0$) between layers. The second layer (top layer of Pt) imposes an attractive force on the first (iron) layer and a repulsive one on the third layer. The distances between the second and third layers are 4.39, 4.43, and 4.39 au for the systems of Fe full layer, Fe chain and Fe quarter-layer, respectively. The two latter values are averaged ones due to slight bucklings. These distances are larger by a few % of d_0 [22]. The coordinates of surface Pt atoms with respect to the Fe atom are presented in table 1. As shown in table 1, the distance between the first and second layers ($=d_{12}$) is 3.85 au for the system of Fe full layer, decreased by 20% from the value of d_0 . As the coverage of Fe atoms decreases to the Fe chain and the Fe quarter-layer, the values of d_{12} are clearly decreased. In the system of Fe quarter-layer, the Pt(I) is pushed up to the Fe atom and the Pt(II) sinks down in the substrate, due to a strong attractive interaction between the Fe and Pt atoms and a slight repulsive one between Pt layers. The distortion of Pt atoms within the plane makes a relaxation of the attractive forces between neighbouring Fe and Pt atoms. In the system of Fe chain, the geometry of Fe atoms is more anisotropic within the xy -plane than the other systems.

Table 2 presents MAEs for systems investigated. For the Fe full layer and the Fe chain, the MAEs of similar systems have been reported in previous work, in which the Pt atoms in the top layer of substrate were not relaxed. The values of MAE have a similar order to the value for bulk alloy (2.61 meV/Fe atom) which was estimated by the same method [19]. In table 2, all the values are smaller than the bulk value except the MAE of $E_x - E_z$ in the system of Fe chain. The ideal case for Fe full layer provides a different value of about 40–50%, compared with the values from the optimized structure. The effect of geometry optimizations appears in MAE within factors of about twice. For the full monolayer and quarter-monolayer, optimizations reduce the modulus of MAE, while for the chain they enhance it. The reversal of sign is not observed in our calculation after optimizations.

The magnetic easy axis is directed in the x -direction for the full coverage and half-coverage and the y -direction for the quarter-coverage. As in the previous work [13], it was found that the magnetic easy axis is within the surface plane for all cases calculated. In the case of full coverage and quarter-coverage, the MAEs obtained from $E_x - E_z$ and $E_y - E_z$ are almost the same. This is due to the small anisotropy of geometries within the surface plane. In the Fe

Table 2. The magnetic anisotropy energies (in meV/Fe atom) for Fe full layer, Fe chain (Fe half-layer) and Fe quarter-layer on Pt(111) surface. The values in parentheses specify the data before introducing atomic relaxations.

	$E_x - E_z$		$E_y - E_z$	
Fe full layer (ideal)	-0.66		-0.61	
Fe full layer	-0.47	(-0.72)	-0.41	(-0.60)
Fe chain	-4.13	(-2.31)	-1.88	(-0.79)
Fe quarter-layer	-0.77	(-0.89)	-0.98	(-0.99)

chain case, the magnetic easy axis is directed with the alignment of Fe atoms. The absolute of the MAE for $E_x - E_z$ is the largest in the present work, which seems to be related to an anisotropic geometry of surface.

In the experiment, the Fe chain at the step on the Pt(997) surface showed that the easy axis was normal to the chain direction and was canted from the surface plane by 10° , having an out-of-plane component of magnetic anisotropy [9]. This experimental direction is different from that of our result. This could be ascribed to the existence of edge Pt atoms at the step in the experiment. As we will refer to in the later paragraphs, our result for Fe chain on Pt(111) would imply that the detail of the local structure changes the magnetic anisotropy sensitively. The out-of-plane component of the experiment was qualitatively reproduced in the previous calculations which have taken into account the step in their models without surface relaxation [10, 11]. However, the magnetic anisotropy for the Fe chain at the step has been slightly controversial. The inclusion of surface relaxation at the step might resolve the controversy.

The in-plane magnetization anisotropy may correspond to the recent experiment on the molecular beam epitaxy-grown superlattices with (111) orientation [23]. Repetto *et al* in their experiment showed that the direction of the easy axis was out of the plane below three-monolayer coverage for the systems of Fe layers on Pt(111) surface [9], which seems to be contrary to our results. This difference may be attributed to an intrinsic difference of the local atomic structures between our models and their structures in the experiment. In the latter they observed the weak tendency of Fe incorporation into the Pt top layer and three-dimensional islands which have Fe second and third layers even at coverages lower than one monolayer, while our models could be too simple to include an essential local network of atoms at the surfaces.

The spin and orbital magnetic moments are presented for Fe and Pt atoms in surface layers in table 3. The plus sign for orbital moments means that the moment is parallel to the spin moment. The magnetic moments on Fe atoms gradually increase almost overall as the coordination numbers of Fe atoms decrease. The values presented are comparable to the values for the results for the bulk alloy [19]. The relationship between the MAE and the orbital moment anisotropy was previously derived from the second perturbational formula [24]. This relation requires a large orbital moment and a strong SOC to obtain a large MAE. However, it is difficult to see a property of magnetic anisotropy by observing a table of orbital magnetic moments, like table 3.

Assuming a large exchange splitting, the MAE of $E_x - E_z$ may be well understood using the second perturbational formula [25];

$$E_x - E_z = (\xi)^2 \sum_{o,u} \frac{|\langle o|\ell_z|u\rangle|^2 - |\langle o|\ell_x|u\rangle|^2}{\epsilon_u - \epsilon_o}, \quad (1)$$

where o and u specify occupied and unoccupied minority spin states and the ℓ_x and ℓ_z are angular momentum operators. The formula for $E_y - E_z$ is obtained by replacing x with y in

Table 3. The spin and orbital magnetic moments (in μ_B) for Fe and Pt atoms in surface layers. The labels of x , y , z indicate the magnetization direction of the systems. The third columns indicate spin magnetic moments for the systems magnetized along the z -direction. The spin magnetic moments for x and y polarizations are not shown due to a small difference (less than $0.002 \mu_B$) from the value for z polarization.

		Spin		Orbital	
		z	x	y	z
Fe full layer (ideal)	Fe	2.943	0.082	0.082	0.087
	Pt	0.193	0.034	0.034	0.040
Fe full layer	Fe	2.883	0.074	0.074	0.082
	Pt	0.245	0.035	0.035	0.048
Fe chain	Fe	3.045	0.107	0.091	0.098
	Pt(I)	0.266	0.058	0.046	0.037
	Pt(II)	0.262	0.057	0.044	0.047
Fe quarter-layer	Fe	3.161	0.100	0.100	0.097
	Pt(I)	0.251	0.057	0.042	0.032
	Pt(II)	0.089	0.026	0.025	0.009

equation (1). The parameter ξ is an average of SOC coefficients. The large ξ value for Pt is one of reasons for the large MAE of CoPt and FePt alloys. The pair of occupied and unoccupied states around the Fermi level is important. In Fe–Pt systems, the Fermi level exists in minority spin states of Fe 3d, and sensitively depends on the hybridizations among Fe 3d orbitals and with Pt 5d orbitals. These hybridizations substantially affect the above matrix elements of ℓ_x and ℓ_z through changes of the orbitals on Fe atoms.

The partial density of states contributed from the individual atom is estimated using the formula

$$D_{LM}^I(\varepsilon) = \sum_i d_{LM,i}^I \delta(\varepsilon_i - \varepsilon) \quad (2)$$

$$d_{LM,i}^I = \int_0^{r_c} |R_{LM,i}^I(r_I)|^2 r_I^2 dr_I + \sum_{nm} \langle \Phi_i | \beta_n^I \rangle q_{nm}^I \langle \beta_n^I | \Phi_i \rangle \delta_{L,L_n} \delta_{L,L_m} \delta_{M,M_n} \delta_{M,M_m} \quad (3)$$

$$R_{LM,i}^I(r_I) = \int Y_{LM}^*(\hat{r}_I) \Phi_i(\mathbf{r}) d\hat{r}_I \quad (4)$$

$$\Phi_i(\mathbf{r}) = \sum_{LM} Y_{LM}(\hat{r}_I) R_{LM,i}^I(r_I) \quad (5)$$

where i and LM specify the complex indices of bands and \mathbf{k} and the angular quantum numbers, respectively. Φ_i is a pseudowavefunction and Y_{LM} is a cubic harmonic function. The q_{nm}^I and β_n^I are parts of the pseudopotential formalism [17].

Figure 2 shows densities of states projected to the components of $d_{3z^2-r^2}$, d_{xz} , d_{yz} , d_{xy} and $d_{x^2-y^2}$ for the Fe atom of systems. When we pay attention to the systems of Fe full layer and Fe chain, the components of d_{yz} , d_{xy} , and $d_{x^2-y^2}$ show large differences due to differences of surface geometry, namely because in the system of Fe chain there is no Fe atom at the nearest fcc hollow site in the y -direction.

First, we note that on the minority spin state, the density of states of the d_{yz} component for the minority spin around the Fermi level in the Fe chain system is decreased from the counterpart for the Fe full layer.

On the basis of the analysis of matrix elements (for example $\langle d_{yz} | \ell_z | d_{xz} \rangle = 1$, $\langle d_{yz} | \ell_x | d_{3z^2-r^2} \rangle = \sqrt{3}, \dots$ etc [25]), equation (1) implies that decreases of the SOC between

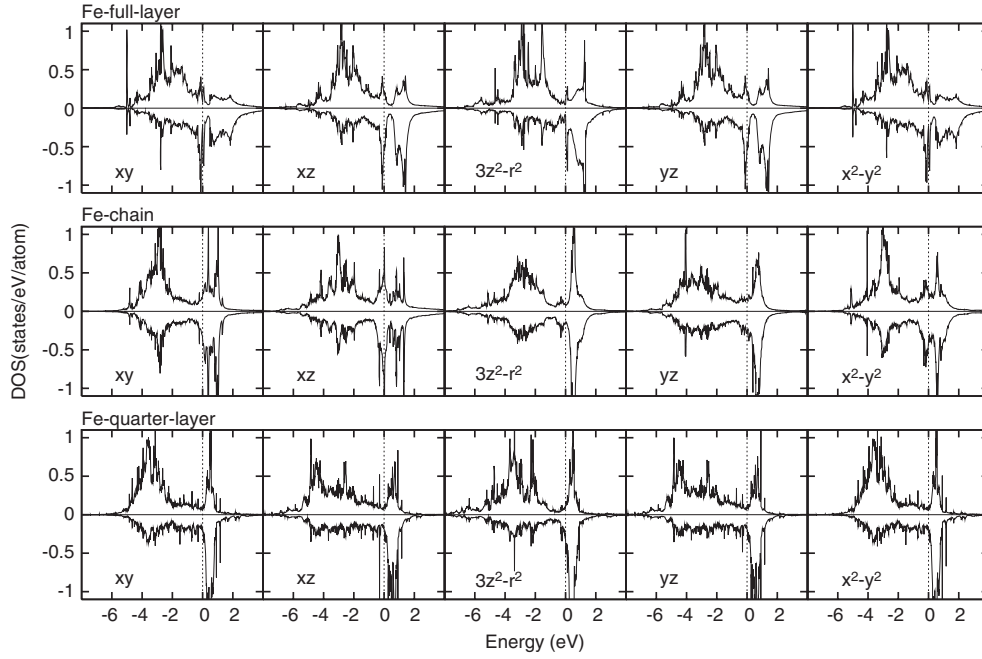


Figure 2. Partial densities of states projected to the components of d_{xy} , d_{xz} , $d_{3z^2-r^2}$, d_{yz} , $d_{x^2-y^2}$ in the Fe atom for the Fe full layer (top), Fe chain (middle) and Fe quarter-layer (down) on Pt(111) surface.

d_{yz} and d_{xz} give a negative contribution to the MAE of $E_x - E_z$. With this consideration, decreases of the SOC between d_{yz} and $d_{3z^2-r^2}$ and between d_{yz} and $d_{x^2-y^2}$ give a positive $E_x - E_z$. It is supposed that the increase of in-plane magnetic anisotropy in the Fe chain is mainly attributable to the decrease of SOC interactions between d_{xz} and d_{yz} states. By geometry optimizations for the Fe chain the Fe–Pt(I) bond length is shortened and, thus, the increase of hybridization between Fe $3d_{yz}$ and Pt(I) $5d_{yz}$ in the minority spin state lowers the density of minority spin states around the Fermi level. This may be a main reason for the large enhancement of in-plane anisotropy in the Fe chain (see table 2).

The SOC between d_{xy} and d_{xz} (d_{yz}) gives a negative contribution to the MAE of $E_x - E_z$ ($E_y - E_z$), implying that the anisotropy within the in-plane case would be explained for the Fe chain system. This consideration is consistent with the similar values for $E_x - E_z$ and $E_y - E_z$ in the systems of Fe full layer and Fe quarter-layer. Although the electronic structures would seem to benefit from a relatively large quantity of $|\langle o|\ell_x|u\rangle|^2$ for the systems of Fe and Pt(111) surface, which imply an in-plane easy axis, the estimation of MAE requires a realistic calculation.

For the system of the Fe quarter-layer, minority spin states are considerably pushed up to above the Fermi level and the densities of states for all d components in the vicinity of the Fermi level are much decreased, resulting in a decrease in MAE. The decreases at the Fermi level originate from the disappearing of the hybridization between the neighbouring Fe atoms. If we assume a rigid band picture, the large magnetic anisotropy energy, which was predicted for the single Co atom on Pt(111) surface [6], may be supported by a large peak in the minority spin state above the Fermi level.

4. Summary

We have studied three surface structures of different coverages for Fe atomic monolayers on Pt(111) surface, using the local spin density approximation. We have estimated the MAEs and investigated effects of surface relaxation. The results show that the MAEs are sensitive to the atomic local geometry of the surface through the hybridizations among Fe 3d and Pt 5d orbitals. Unlike the perpendicular magnetic anisotropy of bulk alloy, the magnetic easy axes were found to be the in-plane directions to the nearest neighbours within the Fe layer. The system of Fe chain, having an anisotropic local atomic structure, shows the largest MAE in the present work and the magnetic easy axis which is directed with the alignment of Fe atoms. The system of Fe quarter-layer, in which the disappearance of neighbouring Fe atoms lowers the density of states at the Fermi level, has a smaller MAE in a given surface area. To make a direct comparison with the experiment on Fe thin layers on Pt(111), we may use a more complex surface in which the local structure provides out-of-plane magnetic anisotropies.

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