Thickness-dependent magnetic anisotropy in ultrathin Fe/Co/Cu(001) films

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Using the full potential linearized augmented plane wave method, we have explored the thickness-dependent magnetic anisotropy of ultrathin Fe films with 1 ML Co underlayer on Cu(001) as well as other magnetic properties. It has been found that the magnetic moment of Co underlay is almost intact with increasing Fe film thickness. A similar trend has been observed in interior Fe layers, whereas the surface Fe magnetic moment is greatly enhanced compared to those in other layers. Through magnetic anisotropy calculations, we have found that the 1 ML Fe film on Co/Cu(001) has a perpendicular magnetic anisotropy, but the direction of magnetization changes to in-plane with increasing Fe coverage. It has been found that the spin-orbit interactions around the middle of Brillouin zone play an essential role in the spin reorientation transition.

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I. INTRODUCTION

In the last decade, magnetism in low dimensions has been the focus of a great amount of research because the magnetic properties found in low dimensions can be utilized for magnetic nanodevice applications such as magnetic fields or spin filtering sensors and high density magnetic recording media. Recently, studies for various magnetic properties of ultrasmall nanoscale or atomic scale magnetic materials are particularly interesting since many peculiar physical properties are observed which were not found in bulk or macroscopic states.

Among various magnetic properties found in magnetic materials, the magnetic anisotropy is one of the most important quantities since this physical phenomenon is closely related to the utilization of direction of magnetization for application purposes. In the studies of magnetic anisotropy in surface magnetism, the change of magnetization direction in ultrathin films, the so-called spin reorientation transition (SRT), has long been an intriguing issue. Typically, Ni/Cu(001) has been well known as a prototype for the thickness-dependent SRT system.[1] Indeed, this SRT phenomenon has been found in various other systems, for example, Ni/Fe/Ni on W(110),[2] Fe/Cu(001).[3] In addition, there are many experimental studies for the magnetic properties of Fe/Co/Cu films[4–9] and in these systems the thickness of Co underlayer is mostly in the range of several monolayers. In addition, one may find theoretical studies for the SRT phenomenon in Fe-Co alloy and Fe films on Co(001) surface.[10,11] Very recently, interesting SRT in ultrathin Fe/Co/Cu(001) films has been experimentally observed[12] with an ultrathin Co underlayer. It has been proposed that the SRT from out-of-plane to in-plane occurs about 2 ML Fe coverage if the Co underlayer coverage is approximately 1 ML and the different SRT behaviors take place for different Co underlayer coverage.

The physical origins of magnetic anisotropy are magnetic dipolar interaction (shape anisotropy) and the magnetocrystalline anisotropy due to spin-orbit interaction. The magnetic dipolar interaction always prefers in-plane magnetization in thin film structure, whereas the spin-orbit interaction manifests very complicated behaviors. It is well understood that the magnetic anisotropy of materials is substantially dependent on subtle changes in underlying electronic structures, so the interpretation of magnetic anisotropy cannot be made in terms of simple physical quantities. From a theoretical view point, one may note the second order perturbation theory for the relation between the orbital anisotropy and the direction of magnetization.[13] However, there is one requirement that should be satisfied to apply this result, i.e., the spin-orbit coupling via minority spin channel dominates all other interaction processes. Regarding this issue, we have found that the spin flip interaction is substantially important for the determination of magnetization direction.[14,15] On the other hand, phenomenologically the experimental data for magnetic anisotropies are usually interpreted in terms of competition between surface and volume contributions to the magnetic anisotropy. Nonetheless, there are difficulties for understanding the direction of magnetization in such a way since the separation of these two effects is not physically clear in ultrathin films, and it has been found from our previous calculations that the spin orbit coupling through spin flip channel cannot be completely ignored.

Therefore, one needs to fully consider quantitative methods in order to understand the magnetic anisotropy occurring in nanoscale ultrathin magnetic films. In this Brief Report, we will theoretically explore the changes of magnetic moments of Fe and Co atoms with increasing film thickness and mainly focus on the thickness dependent SRT in ultrathin Fe/Co/Cu(001) films to reveal the physical origin. To compare the results observed in experimental data,[12] we have considered 1 ML of Co underlayer on Cu(001) and changed the Fe coverage from 1 to 4 ML thickness.

II. NUMERICAL METHOD

The thin film version of full potential linearized augmented plane (FLAPW) method was employed in our calculations. 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 was employed to describe exchange correlation.[20] Spherical har-

TABLE I. The calculated spin magnetic moments (in μ_B) of Co and Fe atoms for various Fe thickness.

Atom	1 ML	2 ML	3 ML	4 ML
Co	1.65	1.60	1.57	1.58
Fe_1	2.84	2.52	2.52	2.52
Fe_2		2.84	2.62	2.62
Fe_3			2.86	2.63
Fe_4				2.86

monics with $l_{max}=8$ were used to expand the charge, potential, and wave functions in the muffin tin region. Energy cutoffs of 225 and 13.7 Ry were implemented for the plane wave star function and basis expansions in the interstitial region. We have used 210 k-mesh points during the course of entire calculations discussed in this report. The convergence for all physical quantities investigated in the present work has been carefully checked. The Fe and Co films are assumed to grow pseudomorphically, hence the Cu(001) lattice constants are employed in lateral directions and the vertical position of all constituents are optimized via force and total energy minimization procedures. The Cu(001) substrate is simulated by seven layers of Cu(001) film

III. RESULTS AND DISCUSSIONS

In Table I, we first present calculated magnetic moments of Co in underlayer and Fe adatoms for various structures. As shown in Table I, it has been found that the Co underlayer has rather stable magnetic moment although one can find little change for 1 ML Fe coverage. In surface Fe atom, the magnetic moment is greatly enhanced compared to that of bulk state which is about $2.26\mu_B$ and this is a typical phenomenon observed in most of thin film structures. Likewise the finding in Co underlayer, the surface Fe magnetic moment is insensitive to the existence of neighboring Fe sublayers. Even in other Fe layers, no significant modifications in magnetic moments with increasing Fe thickness have been found. All these features indicate that the essential magnetic interaction is rather limited by nearest interlayer interaction, not strongly affected by other layers.

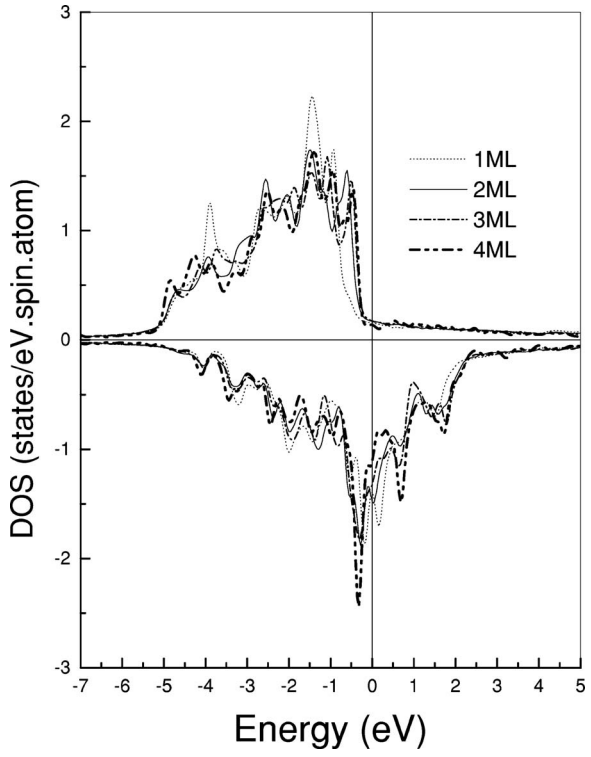

FIG. 1. Fe thickness-dependent DOS of the Co underlayer.

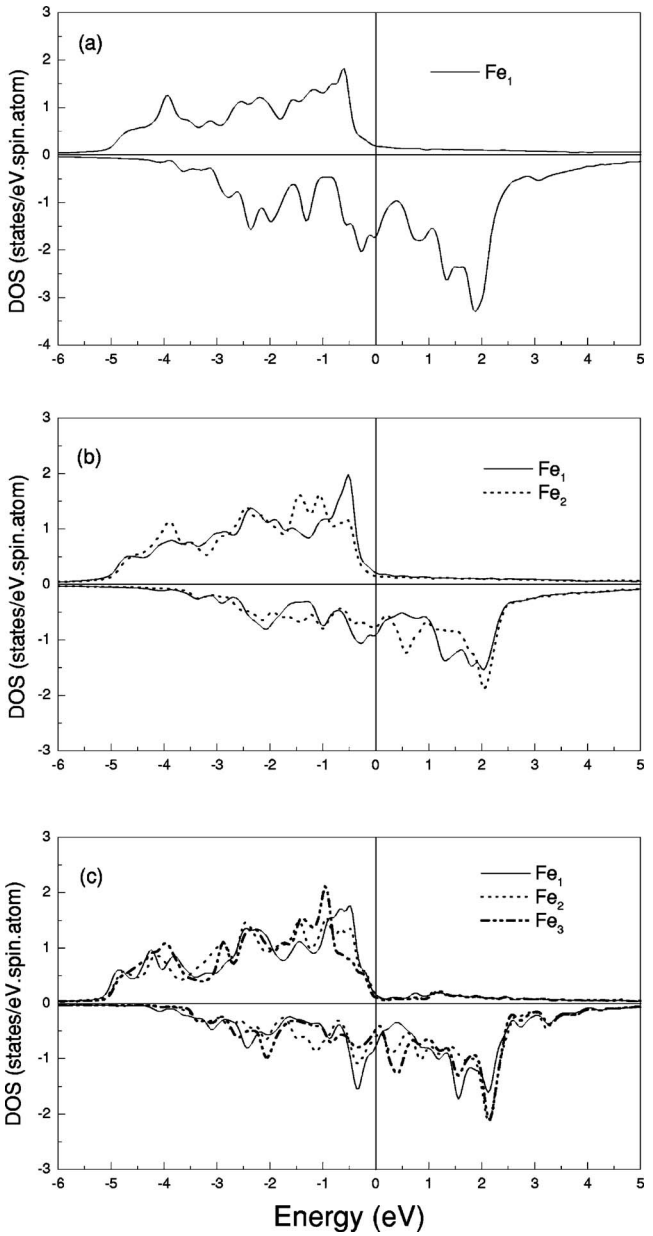

FIG. 2. DOS of interior Fe layers in (a) 2 ML, (b) 3 ML, (c) 4 ML Fe thicknesses.

We now explore the density of states (DOS) characters. The positive part is for majority spin states whereas the minority spin states are described in negative part. In Fig. 1, we show the DOS of the Co underlayer for different Fe thick-

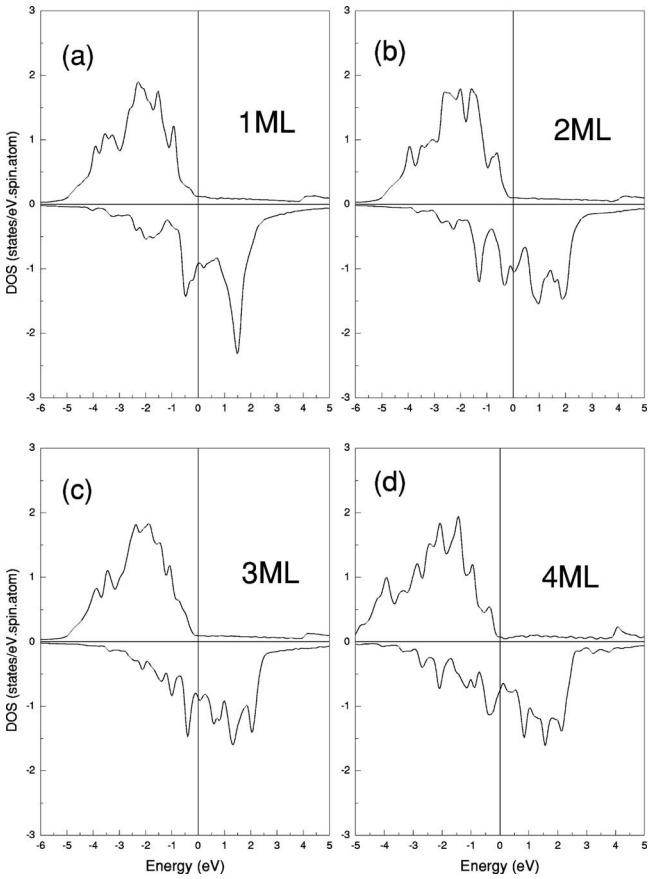

FIG. 3. DOS of surface Fe layer in (a) 1 ML, (b) 2 ML, (c) 3 ML, (d) 4ML thicknesses.

nesses. The overall trends of DOS spectral shape are unchanged although one can find small variation in the majority spin states below the Fermi level in 1 ML Fe thickness. Since the magnetic moment is simply the difference of number of electrons in filled states, the DOS feature can account for the stable magnetic moment of Co underlayer. In Fig. 2, we show the DOS of interior Fe layers. As in Co underlayer, one can see no physically meaningful modification in the DOS of Fe_1 which is directly adjacent to Co underlayer and this accounts for the calculated moments in Table I. In other Fe layers, one may note a small but sizable enhancement of the minority spin DOS around 0.5 eV above the Fermi level resulting in reduction of the number of electrons in filled states, e.g., Fe_2 in 3 ML thickness, Fe_2 and Fe_3 in 4 ML thickness, whereas the majority spin states remain almost empty. Therefore, as presented in Table I the magnetic moment in these atoms are slightly larger than that of Fe_1. In Fig. 3, we display the DOS of surface Fe layers. One can see that there is no observable changes in DOS for surface Fe layers with increasing Fe thickness. It seems that the magnetic moment is mostly determined by nearest interlayer interaction, i.e., the local exchange interaction is an essential factor. However, one should study the physical properties of interlayer exchange couplings in various layers to answer the question whether the local exchange interaction is really dominant in the determination of magnetic moments.[21–23]

We now focus on the calculations of the magnetic anisotropy of materials. The magnetic anisotropy is greatly dependent on subtle changes in the underlying electronic structure of material. Hence, it is required to employ very accurate numerical methods to deal with spin-orbit interaction resulting from a relativistic effect. In this Brief Report, we use the torque method.[24] It is known that the torque method provides very stable results even with fewer k points compared to methods that employe different schemes since the torque method calculates the magnetic anisotropy energy via expectation values of the angular derivative of the spin-orbit Hamiltonian.

TABLE II. The calculated magnetic anisotropy energies (in μeV/atom).

Thickness	E_{MAE}	$E_{\uparrow\uparrow}$	$E_{\uparrow\downarrow}$	$E_{\downarrow\downarrow}$
1 ML	29	28	70	−65
2 ML	−18	13	43	−79
3 ML	−50	12	43	−124
4 ML	−40	9	35	−88

The theoretically calculated magnetic anisotropy energy (MAE) per transition metal atom for various structures are presented in Table II. The positive E_{MAE} means that the direction of magnetization is perpendicular to the surface, while the negative one implies in-plane magnetization. As shown in Table II, we have obtained that the direction of magnetization is perpendicular to the surface for 1 ML Fe thickness, whereas the direction of magnetization is in-plane for thicker films.

According to the experimental data,[12] the critical Fe thickness for SRT from perpendicular magnetization to in-plane direction is about 2 ML since the perpendicular magnetization is found at Fe 1.7 ML when the Co underlayer coverage is 0.75 ML, and in-plane magnetization takes place at Fe 2.5 ML for 1.25 ML Co underlayer. Consequently, we have realized that the theoretical calculations are consistent with the experimental data. As noted, the second order perturbation theory predicts that the direction of magnetization is parallel to the direction of orbital magnetization provided the spin-orbit interaction through minority spin channel dominates all other interactions. To provide a more comprehensive understanding of the magnetic anisotropy we have analyzed the relative importance of spin-orbit interaction through each spin-orbit coupling channel. In Table II, the $E_{\uparrow\uparrow}$, $E_{\uparrow\downarrow}$, and $E_{\downarrow\downarrow}$ stand for the MAE resulting from majority-majority, majority-minority, and minority-minority spin-orbit channels, respectively. It is clearly presented that the $E_{\downarrow\downarrow}$ plays the most important role in determination of magnetic anisotropy, except for the 1 ML Fe thickness structure and in this case the spin-flip interaction has the largest contribution in magnitude to the magnetic anisotropy. From the calculated quantities, we have found that the $E_{\uparrow\downarrow}$, i.e., spin-flip interaction, cannot be negligible although the $E_{\downarrow\downarrow}$ is the largest in magnitude for most of the systems. These findings tell that the simple physical interpretation of MAE based on the orbital anisotropy is not applicable to these systems. As a result, it is necessary to investigate the characters of interaction in each k point in the two-dimensional Brillouin zone. In Fig. 4, we present the distribution of magnetic anisotropy energy

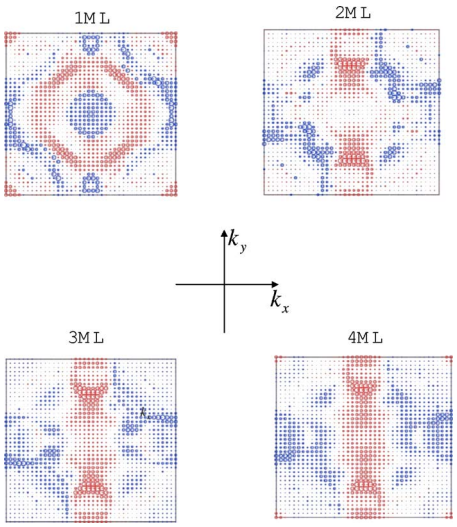

FIG. 4. (Color online) Thickness-dependent magnetic anisotropy energy distributions over the two-dimensional Brillouin zone.

the in two-dimensional Brillouin zone. The red circles mean that the spin-orbit coupling at the specific k point contributes to out-of-plane magnetization, while the blue circles are for in-plane magnetization. The size of a circle is proportional to the magnitude of the magnetic anisotropy. Note that for 1 ML Fe thickness, the contributions to perpendicular magnetization are originated mostly around the middle of Brillouin zone (i.e., along a circle with radius of $\frac{\pi}{2a}$, where a is a lattice constant). At the 2 ML coverage, substantial changes have been occurred. There are almost no significant contributions to the magnetic anisotropy near the Brillouin zone center (Γ point) used to have in-plane magnetization due to cancellation between in-plane and out-of-plane conctributions, thus this region is not important for SRT. The large part of the Brillouin zone maintained for perpendicular magnetization also vanishes and starts to have strong in-plane components in the same region. For thicker films the components for perpendicular magnetization arise again near the zone center, but at the same time in-plane components in other parts of the Brillouin zone have appeared. Overall, we have realized that the changes of spin-orbit interaction around the middle of Brillouin zone are decisive and attribute these variations in the spin-orbit coupling to the SRT at 2 ML Fe coverage.

In summary, we have investigated the thickness-dependent magnetic anisotropy of ultrathin Fe/Co/Cu(001) films. It has been found that the magnetic moment is mostly influenced by the nearest interlayer interaction and this feature results in the insensitivity of the magnetic moment to the film thickness. From the magnetic anisotropy calculations, we have shown that the calculated SRT is consistent with experimental data. Despite the insensitivity of magnetic moment with film thickness, the physical properties of spin-orbit interaction are substantially modified. The distributions of MAE shows that the most drastic changes in spin-orbit interactions occur in the middle parts of Brillouin zone and these features are the origin for SRT at 2 ML Fe thickness.

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