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Electronic and magnetic structures in spin-spiral structures of free-standing Fe(110) monolayers with lattice constants, a, matching those of bulk bcc Fe (2.87 Å) and W (3.16 Å), were investigated by means of first-principles film full-potential linearized augmented-plane-wave calculations including intra-atomic noncollinear magnetism. For $a=2.87$ Å, the spin-spiral structures with wavelength around $7a$ are energetically favored over the collinear ferromagnetic state while those for $a=3.16$ Å turn out to be less favorable. The formation of the spin-spiral structures are found to result from a Fermi-surface nesting that leads to an instability of the ferromagnetic state. In addition, the spin-orbit coupling is found to play an important role to determine the magnetization rotation. These results offer an important step in understanding complex noncollinear spin-spiral magnetism in thin films. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2151822]

Noncollinear magnetism in helical spin-density-wave structures or spin-spiral structures, in which the magnetization is rotated along a certain direction in a crystal, has received much attention in fundamental and applied physics. So far, theoretical progress within spin-density-functional theory has succeeded in describing spin-density-wave or spin-spiral structures in itinerant materials such as bulk Cr, γ-Fe, and U compounds.[1–4] Moreover, even in well-known ferromagnetic materials, e.g., α-Fe, Co, and Ni, the spin-spiral order has been predicted to become stabilized under conditions such as high pressure.[5] The noncollinear magnetism may be governed by the exchange interactions with neighbors and more distant neighbors, the RKKY interaction, the spin-orbit coupling (SOC), as well as a Fermi-surface nesting that leads to a ferromagnetic instability.

Now, interest in the noncollinear magnetism at surfaces and interfaces has increased because of their potential technological importance, in which a breaking of symmetry and an enhanced SOC arising from a reduced dimensionality would give rise to noncollinear magnetism[6] with new and exotic features differing from those of bulk. Little is known quantitatively, however, about the detailed complexity of the spin-spiral structures in thin films. Therefore, it is strongly desirable to understand it from highly precise first-principles calculations.

Here, as a first important step, we have investigated spin-spiral structures in thin-film Fe(110) monolayers by means of the thin-film full-potential linearized augmented-plane-wave (FLAPW) method[7,8] that now also incorporates intra-atomic noncollinear magnetism.[9,10] The results obtained predict, for the Fe(110) monolayer with the lattice constant of bulk Fe, that the spin-spiral structures with wavelength around $7a$ are energetically favored over the collinear ferromagnetic state, while those for the W lattice constant turn out to be less favorable. In addition, we find that the SOC plays an important role in determining the rotation of the magnetization.

Calculations were carried out based on the local-spin-density approximation using the von Barth–Hedin exchange correlation[11] in the scalar relativistic approximation (SRA)[12,13] for the conduction electrons, i.e., without the SOC, and fully relativistically for the core electrons. To treat the intra-atomic noncollinear magnetism that allows its direction to vary continuously all over space,[9,10] the electron density and the effective potential are determined with a 2×2 density matrix and the basis functions are specified with the spin-independent LAPW basis in order to avoid discontinuity in augmentation of the basis functions at the muffin-tin (MT) radius. The LAPW basis with a cutoff of $|\mathbf{k}+\mathbf{G}|<3.6$ a.u.$^{-1}$ and MT sphere radii$=2.3$ a.u. are used; lattice harmonics with angular momenta up to $\ell=8$ are employed to expand the charge and magnetization density, the vector potential, and eigenvectors.

We employed a free Fe(110) monolayer slab with wave vector, $\mathbf{q}$, in the [001] direction in a unit cell and assumed lattice constants, a, matching those of (i) bulk bcc Fe (2.87 Å) and (ii) bulk bcc W (3.16 Å), and performed self-consistent total-energy calculations for changes of the wave vector. As a reference, calculations were performed without the spin-spiral structures, i.e., a collinear ferromagnetic monolayer state with the same lattice and computational parameters.

The calculated energies of the spin-spiral structures as a function of the wave vector, $\mathbf{q}=(00q)$, for the Fe(110) monolayers are shown in Fig. 1, where $q=0$ and 1 indicate the collinear ferromagnetic and antiferromagnetic states, respectively. The self-consistent results demonstrate that for $a=2.87$ Å (bcc Fe) the spin-spiral structures are energetically favored over the collinear ferromagnetic state when q is less than about 0.22. The energy minimum appears to be around $q=0.14$, corresponding to a wavelength of about $7a$, and is

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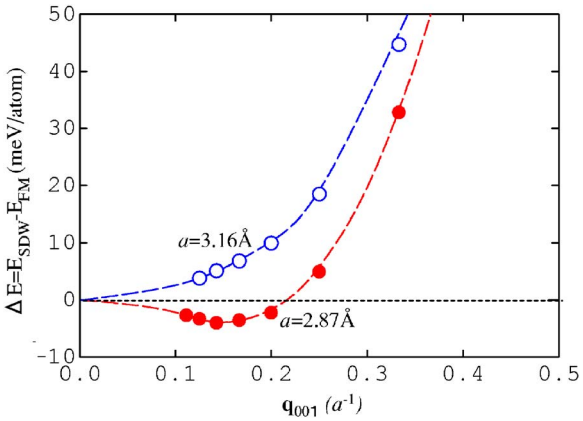

FIG. 1. Calculated energies of spin-spiral structures as a function of wave vector, $\mathbf{q}=(00q)$, for Fe(110) monolayers with lattice constants of 2.87 Å (closed circles) and 3.16 Å (open circles).

about 4 meV/atom lower in energy than the ferromagnetic state. Interestingly, the half-wavelength, $3.5a$ (10 Å), almost corresponds to the domain-wall width, 8 Å, in the free-standing Fe(110) monolayer predicted previously.[14] The spin moments integrated in the MT spheres in the spin-spiral structures are also found to depend on the q values as seen in Fig. 2, where the moments decrease monotonically from the 2.85 μ_B at $q=0$ to 2.61 μ_B at $q=1$ as q increases. The moments in the lowest energy state at $q=0.14$ result in 2.82 μ_B. In contrast, for $a=3.16$ Å (=bcc W) the spin-spiral structures turn out to be less favorable and the spin magnetic moments do not depend on the q values.

In order to discuss the stability of the spin-spiral structures, we calculated the Fermi surfaces of the ferromagnetic Fe(110) monolayers; shown in Fig. 3. For $a=2.87$ Å, one closed Fermi surface in the majority-spin state clearly possesses a nesting feature that creates an electron pocket centered at the Γ point. The majority-spin d bands are almost fully occupied, and so the bands crossing E_F arise mainly from electrons in largely dispersive sp-like or vacuum states. Importantly, one of the nesting lines is perpendicular to the [001] ($\Gamma-H$) direction. In the minority-spin states, some nesting feature can be also assigned perpendicular to the [001] direction, which now arise mainly from the d electrons.

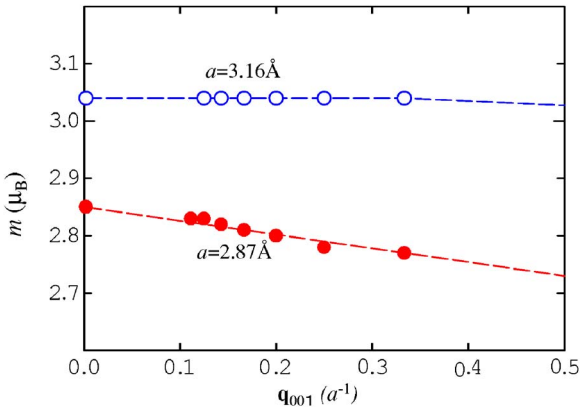

FIG. 2. Spin moments integrated in muffin-tin spheres in spin-spiral structures for Fe(110) monolayers with lattice constants of 2.87 and 3.16 Å. The notations are the same as in Fig. 1

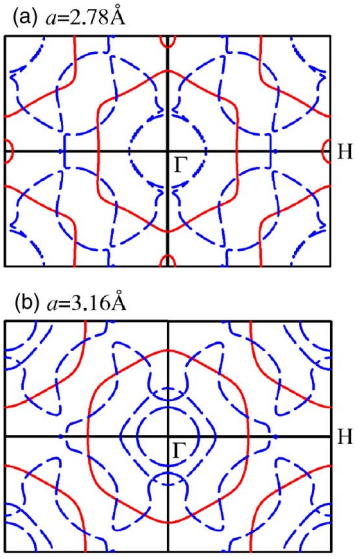

FIG. 3. Calculated Fermi surfaces of ferromagnetic Fe(110) monolayers with lattice constants of (a) bulk Fe (2.87 Å) and (b) bulk W (3.16 Å). Solid and dashed lines represent majority-spin and minority-spin states, respectively.

Thus, the nesting vector connecting majority-spin and minority-spin states in the [001] direction will give rise to singularities in the generalized susceptibility that leads to an instability of the ferromagnetic state against the formation of the spin-spiral structures obtained. The nesting vector was estimated to be about 0.2, which roughly corresponds to the energy minimum around $q=0.14$ in Fig. 1. In contrast, the Fermi surfaces for $a=3.16$ Å tend to lose the nesting feature, and therefore the ferromagnetic instability is no longer excited. Of course, further investigations will be necessary for a discussion including the effects of the substrate, since substrates significantly change the electronic and magnetic structures of the thin films, as demonstrated previously.[15]

Finally, it is interesting to consider the effect of the SOC, since the spin symmetry in the spin-spiral structures breaks the crystal symmetry when the spin and orbital moments are coupled through the SOC. Furthermore, changes of the electronic structures near E_F due to the spin-spiral order would significantly affect SOC-induced magnetic phenomena, as demonstrated in calculations of the magnetocrystalline anisotropy in thin films.[16]

We consider here the SOC effects on the magnetization rotations in the spin-spiral structures, which may be typically characterized by two modes where the rotational axis of the magnetization is parallel ([001]) or normal ([$\bar{1}$10]) to the spin-spiral axis of [001]. They correspond to Bloch-type and Neel-type rotations, respectively, as seen in the domain-wall structures. Since both rotations are related by a 90° rotation about the easy [110] axis of Fe(110) monolayer in spin space,[17] we apply the second variation SOC method based on the force theorem[18]—as successfully demonstrated in magnetocrystalline anisotropy energy calculations[16,19]—to obtain the energy difference, $\Delta E=E_{\mathrm{Neel}}-E_{\mathrm{Bloch}}$. By use of the SRA eigenvectors, we obtained ΔE to be −0.14 meV/atom for the Fe(110) monolayer with $a=2.87$ Å at $q=0.14$. Thus, the Neel-type rotation is favored more than the Bloch-type one.

The results are again consistent with the magnetization rotation in the domain wall in the free-standing Fe(110) monolayer, as demonstrated previously.[17]

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