Atomically Sharp Magnetic Domain Wall in Thin Film Fe(110): A First Principles Noncollinear Magnetism Study

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Magnetic domain wall structures in an Fe (110) monolayer are determined by the highly precise first principles full-potential linearized augmented plane-wave method including intra-atomic noncollinear magnetism. The self-consistent results demonstrate that the magnetization changes from one orientation to the opposite (180°) orientation within an 8 Å width without any abrupt rotation. This narrow domain wall is found to arise from band effects. Our results are consistent with and support domain walls having a 6 Å width recently observed in spin-polarized scanning tunneling microscopy experiments.

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Interest in magnetic domain walls (DW) has been greatly increased in both basic and applied research—including the constraint DW with a few nm width in quantum spin interfaces [1,2] and the role of the DW in the exchange bias [3,4] and nanoscale geometrical structures [5]. The DW, which changes the magnetization orientation from one easy axis to another, is known to be determined by a competition between the exchange energy and the anisotropy energy—as revealed in phenomenological continuum micromagnetic calculations [6,7]: The exchange energy tends to produce a slow variation of the magnetization while the anisotropy energy favors a rapid change from one easy axis to another, which leads to a stable DW width of the order of 10 nm in the bulk. To date, first-principles investigations of the DW have also greatly advanced [8,9] in which the magnetization rotation as seen in Bloch walls was successfully demonstrated and reliable exchange stiffness values were obtained.

Most recently, however, a narrow DW with a width of approximately two lattice constants (about 6 Å) was observed in pseudomorphically grown Fe monolayers on a W(110) substrate by spin-polarized scanning tunneling microscopy (SP-STM) experiments [10]. Adjusting the phenomenological calculations to this experimental observation, the very narrow DW has been explained by a large magnetic anisotropy (4.2 meV/atom) that favors a very narrow wall width. Although these phenomenological calculations are well justified in a large-scale system such as in bulk, it is not clear whether they can be applied to itinerant ultrathin film systems: The electronic and magnetic structures of ultrathin films are known to be strongly different from those in bulk [11,12] and the exchange stiffness (or integral) of the DW has a strong dependence on the DW width when the width becomes on the order of the atomic size [8]. Also, discontinuous magnetization changes in the DW cannot be ruled out, as seen in ultrathin Fe structures on Cu(001) where (i) a competition between ferromagnetic (FM) and antiferromagnetic (AFM) coupling and (ii) their long-range interactions beyond the second neighbor play a crucial role [13]. Hence, in ultrathin films the ground state of the DW appears to be governed by atomic-scale itinerant electron considerations and needs to be treated with first principles quantum mechanics.

Here, we present results of electronic and magnetic structures of the DW in an Fe(110) monolayer as obtained with the thin film full-potential linearized augmented plane-wave (FLAPW) method [14,15] that now also incorporates intra-atomic noncollinear magnetism [16,17]. These self-consistent calculations predict the DW width to be only 8 Å. While surprising at first, this result appears to support and confirm the SP-STM experimental findings (about 6 Å). Further, we find that this narrow DW originates from band effects. These predictions offer possible improvements in the quantitative understanding of DW properties in ultrathin films and invite further experimental confirmation.

Calculations were carried out based on the local spin density approximation (LSDA) using the von Barth-Hedin exchange correlation [18] in the scalar relativistic approximation [19,20] for the conduction electrons, i.e., without the spin-orbit coupling (SOC), and fully relativistically for the core electrons. To treat the intra-atomic noncollinear magnetism so as to allow its direction to vary continuously all over space [16,17,21], 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

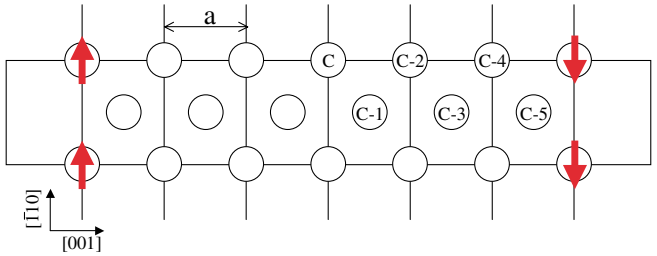

FIG. 1 (color online). Model and notation for a magnetic 180° domain wall structure in an Fe(110) monolayer, where C and C-n denote the center atom and n the atomic index for atoms away from the center. Arrows (red) indicate initial magnetic orientations at both sides of the domain wall; a is the bcc lattice constant of bulk Fe.

$\ell = 8$ are employed to expand the charge and magnetization density, the vector potential, and eigenvectors. The DW in the Fe(110) monolayer is modeled by a monolayer slab containing 13 atomic rows in the [001] direction, as sketched in Fig. 1, separated by vacuum regions of length $2a$, which demonstrates a singular DW with two semi-finite domains. The experimental lattice parameter, a, of bulk bcc Fe is assumed. With an initial magnetic configuration of a 180° DW as indicated by arrows in the figure, we determined self-consistently the direction as well as the magnitude of the magnetization density without any constraints. As a reference, self-consistent calculations were performed without the DW (i.e., a collinear FM monolayer state) with the same lattice and computational parameters.

The relative angle ϕ (shown as circles) of the integrated spin moments in the MT spheres and the difference in the magnitude of the magnetization ΔM (shown as triangles) between systems with and without the DW for the Fe(110) monolayer are presented in Fig. 2. For comparison, the calculated ϕ and ΔM for the DW in the bulk [8] are also shown in the figure (open points). In the bulk case, the moments in the DW rotate linearly from one orientation to the opposite (180°) orientation with almost no change of magnitude, since the exchange energy favors the slower variation of the magnetization—as expected from phenomenological calculations .

In contrast, in the Fe(110) monolayer the moments rotate rapidly within approximately three lattice constants about the center without any discontinuous changes. Thus, the self-consistent calculations demonstrate the narrow DW structure and its DW width is 8 Å, when estimated from the tangent of the angle at the center. These results are qualitatively consistent with and support the SP-STM observations of the Fe/W(110) monolayer [10].

The behavior of the magnitude of the moments for the DW in the monolayer differs from the bulk case. On moving from one side of the DW, the moments decrease by $0.04\mu_B$ at the center. The effect of intra-atomic non-

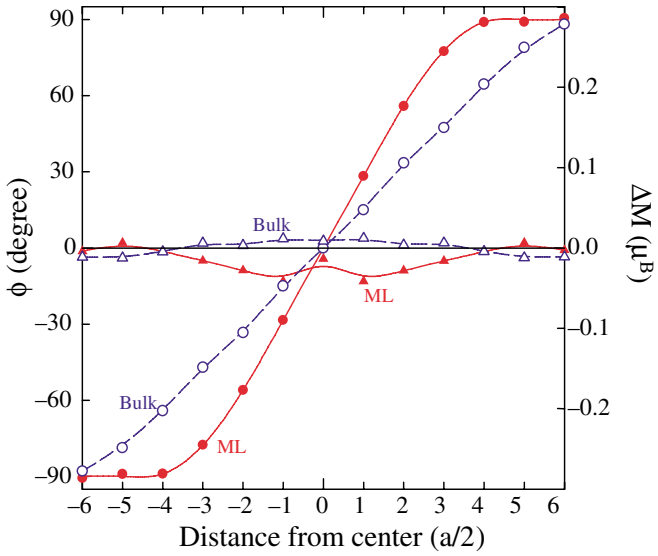

FIG. 2 (color online). Relative angle ϕ of the spin moment direction (circles) in the muffin-tin spheres and the difference in the magnitude, ΔM (triangles), between systems with and without the domain wall. Solid (red) and open (blue) points represent the results for Fe(110) monolayer and bulk, respectively; a is the bcc lattice constant of bulk Fe.

collinear magnetism appears mainly near the center: the noncollinearity at the C, C-1, and C-2 atoms in Fig. 1, calculated as the moment away from the average direction by $\Delta M_{\mathrm{NCM}} = \int_{\mathrm{MT}} |m_\perp(\vec{r})| d\vec{r}$, where $m_\perp(\vec{r})$ is the perpendicular component of the average orientation, is 0.06–0.07μ_B, which corresponds to about 2%–3% of their total moments; ΔM_{NCM} at both ends of the DW is negligibly small—less than 0.01 μ_B.

To discuss the stability of the DW, we calculated the valence charge density differences between the density of the collinear FM state from that of the DW state in the Fe(110) monolayer; this is shown in Fig. 3 on the (110)

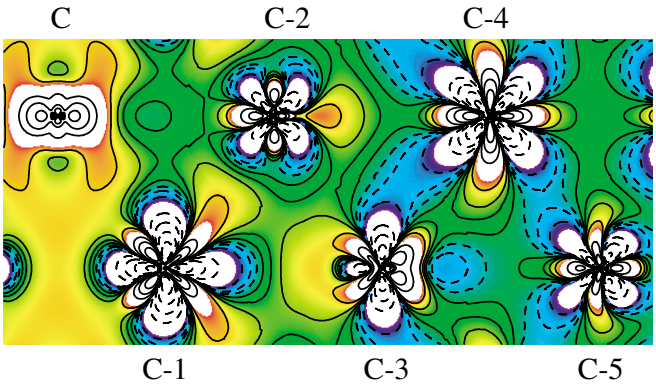

FIG. 3 (color online). Valence charge density differences between domain wall and ferromagnetic states on the (110) plane of an Fe(110) monolayer. Contours start from $1 \times 10^{-4} e/\mathrm{a.u.}^3$ and increase successively by a factor of 2. Solid (red) and dashed (blue) contours denote charge accumulation and depletion, respectively.

plane. The charge redistribution can be seen clearly at the center of the DW where charge accumulation in the interstitial region is observed. Although the lower (110) lattice symmetry causes a complicated charge redistribution at each atom site, the C and C-1 charge densities of the d_{xy} states (whose orbitals point to nearest neighbor atoms) increase, and those at C-2 decrease but again increase at C-3. These charge redistributions appear to lead to changes in the chemical bonding between atoms. We further find that the charge density in the d_{z^2} state at each atom (not shown in the figure) spreads out into the vacuum region compared to that in the collinear FM monolayer state.

The spin-projected density of states (DOS) along the average moment directions at the C and C-5 atoms for the DW and collinear FM states in the Fe(110) monolayer are shown in Fig. 4. The global features of the DOS both in the DW and collinear FM states are similar; however, small differences can be seen, indicating the breaking of the symmetry degeneracy by the presence of the DW. Further, the overall DOS energy-levels in the DW shift to lower energy while the shifts are no longer observable at both sides of the DW, as seen at the C-5 atom [Fig. 4(b)]. (The d bands at the C, C-1, and C-2 atoms are lowered by about 10 meV.) Thus, by introducing the DW, the bands prefer to energetically induce the narrow DW. In the case of the DW in the bulk, we confirmed that there are no such DOS shifts introduced by the DW, indicating no gain in band energy, which produces the linear magnetization rotation as seen in Fig. 2.

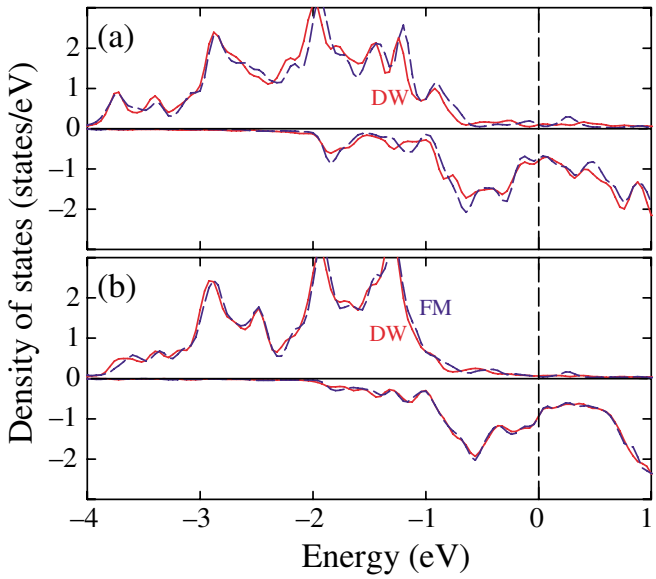

FIG. 4 (color online). Spin-projected density of states (DOS) along the average moment directions for the C (a) and C-5 (b) atoms in the Fe(110) monolayer. Solid (red) and dashed (blue) lines represent the DOS in the domain wall and collinear ferromagnetic states, respectively.

The DW formation energy, ΔE_{DW}, calculated as the difference in the total energies between the systems with and without the DW ($E_{\mathrm{DW}} - E_{\mathrm{FM}}$) within the scalar relativistic approximation (no SOC included) is found to be -26 meV/cell, corresponding to -3 meV/atom in the DW, i.e., the narrow DW is energetically favorable. As the DW width is constricted to a smaller length, the DW turns out to be unstable;, e.g., a similar calculation for the Fe(110) monolayer containing only five atomic rows (7 Å) in Fig. 1 yields a ΔE_{DW} that is 187 meV (37 meV/atom), and the magnetization orientation rotates linearly from one end to the other. Thus, the exchange contribution tends to produce a slower variation of the magnetization—as expected from phenomenological calculations—but the energy has a minimum at a DW width of 8 Å.

When the SOC is introduced self-consistently and the magnetization orientations of both sides of the DW orient along the easy $\langle 110 \rangle$ axis, the spin density structure is not found to change significantly; now the ΔE_{DW} is found to be -29 meV/cell. Hence, the effect of the magnetic anisotropy from the SOC changes the DW formation energy by only -3 meV/cell, which corresponds to -0.3 meV/atom [22]. This anisotropy contribution is 1 order of magnitude smaller than the exchange one, so the narrow DW clearly arises from the changes in the electronic band structures presented above.

Finally, we comment on the magnetic ground state in an infinite Fe(110) monolayer. The lower DW energy in our semifinite 13-row atom lattice calculations implies some kind of a helical spin-density wave (SDW) state. In case of the DW in the Fe monolayer on the W(110) substrate, however, the large magnetocrystalline anisotropy (MCA), arising from the strong SOC at the W substrate interface, plays a key role in determining the ground state. The magnetization rotation in the DW, which changes the orientation from the easy axis, increases the total energy. Since the MCA energy in the Fe/W(110) case is estimated to be about 1–3 meV/Fe-atom [23,24], which corresponds to the magnitude of our DW energy within the scalar relativistic approximation, the ground state in the system having a large MCA would be the FM state. Of course, further investigations will be necessary for a quantitative discussion, including the effects of the W(110) substrate.

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[1] J. B. A. N. van Hoof, K. M. Schep, A. Brataas, G. E. W. Bauer, and P. J. Kelly, Phys. Rev. B **59**, 138 (1999).

[2] J. L. Prieto, M. G. Blamire, and J. E. Evetts, Phys. Rev. Lett. **90**, 27201 (2003).
[3] J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).
[4] A. E. Berkowitz and K. Takano, J. Magn. Magn. Mater. **200**, 552 (1999).
[5] P. Bruno, Phys. Rev. Lett. **83**, 2425 (1999).
[6] S. Chikazumi, *Physics of Ferromagnetism* (Wiley, New York, 1964).
[7] A. Hubert and R. Schäfer, *Magnetic Domains* (Springer-Verlag, Berlin, 1998).
[8] K. Nakamura, T. Ito, A. J. Freeman, L. Zhong, and J. Fernandez-de-Castro, J. Appl. Phys. **93**, 6879 (2003).
[9] J. Schwitalla, B. L. Györffy, and L. Szunyogh, Phys. Rev. B **63**, 104423 (2001).
[10] M. Pratzer, H. J. Elmers, M. Bode, O. Pietzsch, A. Kubetzka, and R. Wiesendanger, Phys. Rev. Lett. **87**, 127201 (2001).
[11] A. J. Freeman and R. Q. Wu, J. Magn. Magn. Mater. **100**, 497 (1991).
[12] R. Wu and A. J. Freeman, J. Magn. Magn. Mater. **200**, 498 (1999).
[13] T. Asada and S. Blügel, Phys. Rev. Lett. **79**, 507 (1997).
[14] E. Wimmer, H. Krakauer, M. Weinert, and A. J. Freeman, Phys. Rev. B **24**, 864 (1981).
[15] M. Weinert, E. Wimmer, and A. J. Freeman, Phys. Rev. B **26**, 4571 (1982).
[16] K. Nakamura, A. J. Freeman, D. S. Wang, L. Zhong, and J. Fernandez-de-Castro, Phys. Rev. B **65**, 12402 (2002).
[17] K. Nakamura, T. Ito, A. J. Freeman, L. Zhong, and J. Fernandez-de-Castro, Phys. Rev. B **67**, 14420 (2003).
[18] U. von Barth and L. Hedin, J. Phys. C **5**, 1629 (1972).
[19] D. D. Koelling and B. N. Harmon, J. Phys. C **10**, 3107 (1977).
[20] A. H. MacDonald, W. E. Pickett, and D. D. Koelling, J. Phys. C **13**, 2675 (1980).
[21] L. Nordström and D. J. Singh, Phys. Rev. Lett. **76**, 4420 (1996).
[22] Considering the computational accuracy, this value has too small a magnitude to conclude whether the anisotropy has positive or negative contribution to the DW stabilization due to the small SOC effect in the 3*d* metals.
[23] X. Qian and W. Hübner, Phys. Rev. B **64**, 92402 (2001).
[24] I. Galanakis, M. Alouani, and H. Dreyssé, Phys. Rev. B **62**, 3923 (2000).