Magnetic Bubble Domain Phase at the Spin Reorientation Transition of Ultrathin Fe/Ni/Cu(001) Film

J. Choi,[1] J. Wu,[1] C. Won,[1,2] Y. Z. Wu,[1,3] A. Scholl,[4] A. Doran,[4] T. Owens,[1] and Z. Q. Qiu[1]

[1]*Department of Physics, University of California at Berkeley, Berkeley, California 94720, USA*

[2]*Department of Physics, Kyung Hee University, Seoul 130-701, Korea*

[3]*Surface Physics Laboratory (National Key Laboratory) and Advanced Material Laboratory, Fudan University, Shanghai 200433, China*

[4]*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

(Received 8 June 2006; revised manuscript received 1 March 2007; published 18 May 2007)

Magnetic domain phases of ultrathin Fe/Ni/Cu(001) are studied using photoemission electron microscopy at the spin reorientation transition (SRT). We observe a new magnetic phase of bubble domains within a narrow SRT region after applying a nearly in-plane magnetic field pulse to the sample. By applying the magnetic field pulse along different directions, we find that the bubble domain phase exists only if the magnetic field direction is less than ~10 degrees relative to the sample surface. A temperature dependent measurement shows that the bubble domain phase becomes unstable above 370 K.

DOI: 10.1103/PhysRevLett.98.207205 PACS numbers: 75.70.Ak

The interplay between the long-range magnetic dipolar interaction, the short-range magnetic exchange interaction, and the on-site magnetocrystalline anisotropy leads to many interesting phenomena of magnetic domain formation [1]. In particular, the formation of magnetic stripe domains at the so-called spin reorientation transition (SRT) of an ultrathin magnetic film [2,3] is shown to be associated with the two-dimensional (2D) magnetic long-range order [4–6]. Thus understanding various magnetic domain phases and their competition at the SRT of a magnetic ultrathin film becomes fundamentally important to the understanding of the 2D magnetic nature. Although the magnetic stripe phase was recently observed in experiment [7–9] and has been applied to explain the reduction of the macroscopic magnetization at the SRT, it is not answered if the magnetic stripe phase is the only ordered magnetic phase in a 2D magnetic system; i.e., it is not clear if other domain phases could also exist to compete with the stripe domain phase in a 2D magnetic system. The difficulty in answering this question lies in the long-range nature of the magnetic dipole interaction that any theoretical model has to presume a domain structure before calculating its total energy [6,10]. Therefore, the theory can only compare the energy difference of given domain structures rather than predict the ground state domain structure. On the other hand, it is well known that there exist magnetic domain phases other than the stripe phase in thick ferromagnetic films [11]. For example, micron-sized magnetic bubble domains have been observed in thick garnet films [12] and layer-structured ferromagnet [13]. The discovery of the bubble domain phase in thick garnet films broadens significantly the phase diagram of the magnetic domains and has forced people to reexamine the mechanism of the magnetic domain formation. However, it is unclear if the bubble domain phase observed in thick films retains as the film thickness is reduced to the nanometer regime. A more general question is if the stripe domain phase is the only ordered magnetic phase at the SRT of an ultrathin film or there could exist other ordered magnetic phases to compete with the stripe phase. To explore other possible 2D magnetic domain phases, theoretical effort has been made to study the stability of the stripe domain phase as well as the possible phase diagram of a 2D magnetic system [14]. Computer simulations were also performed to search for other domain structures [15,16]. The result suggests that the bubble domain phase could have a comparable energy to compete with the stripe domain phase in a 2D magnetic system [17]. Despite the great theoretical effort, no experimental evidence has been found to confirm the existence of new magnetic domain phases at the SRT although more symmetrical and dynamical stripe phases were reported recently [18–20]. In this Letter, we show unambiguously that there exists a magnetic bubble domain phase at the SRT of Fe/Ni/Cu(001) ultrathin films.

An electrochemically polished Cu(001) single crystal was cleaned in an ultrahigh vacuum (UHV) chamber by cycles of Ar^+ sputtering at ~2 keV and annealing at ~600 °C. Fe and Ni cross wedges were grown epitaxially onto the Cu(001) to permit Fe and Ni thickness variations. Fe/Cu(001) has been a prototype model system for the study of stripe domains. However, the ferromagnetic to antiferromagnetic transition at ~5 ML of Fe [21,22] makes it unclear whether the stripe domains observed in this system reveal the complete stripe domain phase. It is shown that adding a thin Ni layer to the Fe film could shift the SRT of the Fe/Ni/Cu(001) to the ferromagnetic region of the fcc Fe [23] so that a complete SRT can be studied in Fe/Ni system. The Fe/Ni/Cu(001) film was covered with a 20 Å Cu protection layer before being transferred to the Photoemission Electron Microscopy (PEEM) chamber at beam line 7.3.1.1 of the Advanced Light Source (ALS) at the Lawrence Berkeley National Laboratory. The x-ray beam was circularly polarized and incident at an angle of 60° to the sample surface normal direction. The magnetic domain images were obtained at room temperature by taking the ratio of the Fe peak intensities at the L_3 and L_2 edges using the right circular polarized x-ray beam, utilizing the effect of x-ray magnetic circular dichroism (XMCD). Since the Fe and Ni magnetizations are strongly coupled to behave as a single magnetic layer, we show only Fe PEEM images in this Letter to represent the Fe/Ni magnetic domains.

Figure 1(a) shows the magnetic domain image of Fe/Ni(10.6 ML) film. We observe a dramatic change of the domain pattern at 3.75 ML of Fe: the magnetic domains change from regular stripe domains below 3.75 ML of Fe to irregular domains above 3.75 ML. After rotating the sample by 90 degrees around the surface normal direction, we found that the domain contrast below 3.75 ML remains unchanged while the domain contrast above 3.75 ML changes. Since the magnetic contrast depends on the angle between the incident x-ray beam direction and the magnetization, the above result shows that the Fe/Ni(10.6 ML) film undergoes a SRT from a perpendicular magnetization below 3.75 ML Fe to an in-plane magnetization above 3.75 ML Fe. We found that the Fe SRT thickness increases with increasing Ni thickness from $d_{\mathrm{Fe}} \sim 2.9$ ML at $d_{\mathrm{Ni}} \sim 5.5$ ML to $d_{\mathrm{Fe}} \sim 4.3$ ML at $d_{\mathrm{Ni}} \sim 13.8$ ML [Fig. 2(a)]. This result is not surprising since Ni has a SRT at ~7 ML from in-plane magnetization to out-of-plane magnetization with increasing thickness [24]. Thus increasing the Ni thickness strengthens the perpendicular magnetic anisotropy of the Fe/Ni film to result in an increase of the Fe SRT thickness.

To explore new magnetic domain phases, we magnetized the sample with an in-plane magnetic field (~1 kOe) for a few seconds before imaging the sample in its remanent state. After applying the in-plane magnetic field, the stripe domain phase changes into bubble domain phase in a narrow thickness range of the SRT while remaining in the stripe phase elsewhere [Fig. 1(b)]. Although the conversion of the stripe domains to bubble domains occurs only within a finite thickness range, the existence of the bubble domains at the SRT is unambiguous as we observed the bubble domains throughout the studied Ni thickness range [Fig. 2(b)–2(e)]. We also applied the magnetic field pulse along both the in-plane [110] and [100] directions, and found that the appearance of the bubble domain phase is independent of the in-plane magnetic field direction. Using the diameter as the bubble domain width, we found that the bubble domain width decreases exponentially with Fe thickness in the same manner as the stripe domains [Fig. 1(d)]. This result shows

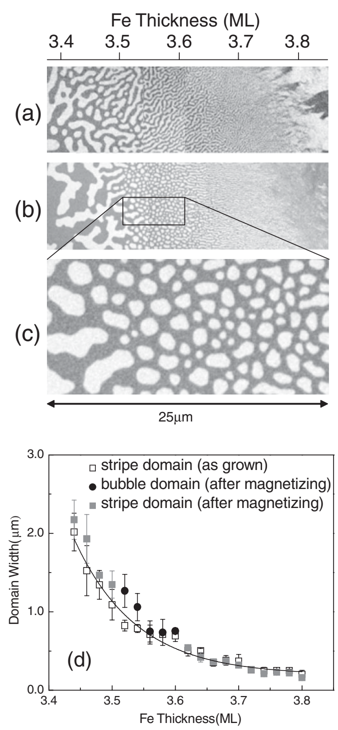

FIG. 1. Magnetic domain images of Fe(wedge)/Ni(10.6 ML)/Cu(001) for (a) as grown sample and (b) after applying a nearly in-plane magnetic field pulse. (c) Zoom-in image of the bubble domains in the SRT region. (d) Domain width as a function of the Fe film thickness, before and after in-plane magnetization. The solid line is a guide to the eye.

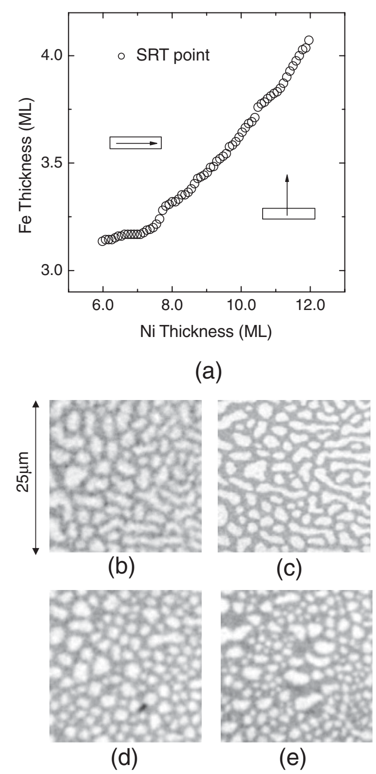

FIG. 2. (a) SRT position in the Fe-Ni thickness plane. Magnetic bubble domain images of (b) $d_{\mathrm{Fe}} = 2.9$ ML, $d_{\mathrm{Ni}} = 5.5$ ML; (c) $d_{\mathrm{Fe}} = 3.1$ ML, $d_{\mathrm{Ni}} = 8.0$ ML; (d) $d_{\mathrm{Fe}} = 3.6$ ML, $d_{\mathrm{Ni}} = 10.8$ ML; (e) $d_{\mathrm{Fe}} = 4.1$ ML, $d_{\mathrm{Ni}} = 13.6$ ML.

that the formation of bubble domain phase is likely originated from the same mechanism as the stripe domains, i.e., from the competition between the dipole interaction, the exchange interaction, and the magnetic anisotropy [9].

Noticing that the bubble domains have different domain patterns for spin-up and spin-down domains, the formation of the bubble domain phase actually breaks the up-down symmetry of the stripe domain phase. However, a perfect in-plane magnetic field should not break the up-down symmetry of a film. Then the formation of the bubble domains indicates that the applied magnetic field pulse was not perfectly in the film plane. This is conceivable because it is very difficult to align the magnetic field direction exactly in the film plane in experiment. Conservatively speaking, we could only guarantee that the ''in-plane'' magnetic field direction is within ~5 degrees to the sample surface. We did the following experiment to verify that a small off-normal magnetic field component indeed exists to trigger the bubble domains. After applying an ''in-plane'' magnetic field pulse and obtaining the bubble domains [Fig. 3(a)], we reversed the current direction of the electromagnet to apply a magnetic field pulse to the sample. Since the reversal of the electromagnet current reverses the magnetic field direction (both in-plane and out-of-plane direction), the up-down bubble domains should reverse their contrasts accordingly. The domain image after applying the reversed magnetic field [Fig. 3(b)] indeed shows that all bubbles reverse their contrast as compared to the image of Fig. 3(a), proving that there exists a small normal component of the magnetic field in generating the bubble domains. Then it is interesting to ask how much of the normal magnetic field component is needed to generate the bubble domain phase. We applied a magnetic field pulse at different angles relative to the sample surface and found that the bubble domain phase disappears for magnetic field direction greater than ~10° to the sample surface [Fig. 4].

To further isolate the effect of the in-plane and out-of-plane components of the magnetic field on the formation of the bubble domains, we applied the magnetic field pulse with 0.5, 1.5, and 1.8 kOe to the sample at different angles. We find that for magnetic field direction less than 5° relative to the sample surface, the bubble domain phase

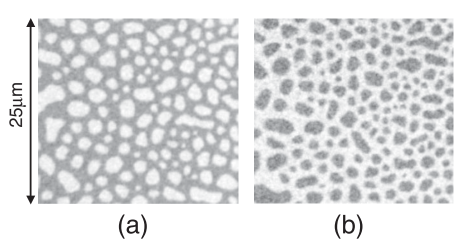

FIG. 3. Bubble domain phase of Fe(3.55 ML)/Ni(10.6 ML)/Cu(001) (a) after applying a nearly in-plane magnetic field pulse and (b) after applying a magnetic field pulse in the opposite direction. The reversal of the domain contrast shows that there exists a small normal component of the magnetic field pulse.

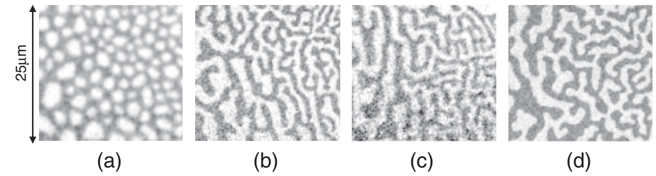

FIG. 4. Magnetic domain images of Fe(3.3 ML)/Ni(9 ML)/Cu(001) after applying a magnetic field pulse (a) less than ~5° from the sample surface, (b) ~10° from the sample surface, (c) ~20° from the sample surface, and (d) perpendicular to the sample surface.

always forms for all the magnetic fields of 0.5, 1.0, 1.5, and 1.8 kOe. For magnetic field direction greater than 10° relative to the sample surface, the bubble domain phase does not form regardless of the magnetic field strength. Therefore, the formation of the bubble domain phase depends only on the direction of the magnetic field pulse and is independent of the strength of the magnetic field. In particular, it is not the in-plane (or out-of-plane) component of the magnetic field alone that determines the formation of the bubble domains. For example, when the magnetic field is 10° (or 20°) relative to the sample surface, the magnetic bubble domain phase does not form even when the 1.8 kOe magnetic field generates an in-plane component of 1.77 kOe (or 1.69 kOe) whereas the bubble domain phase is already formed for a nearly in-plane magnetic field as low as 0.5 kOe. One possible mechanism of the bubble domain formation is the incomplete saturation of the sample where the in-plane component of the magnetic field, in the presence of a perpendicular magnetic component, causes the domain wall to move inhomogeneously [25,26]. However, this mechanism requires an incomplete saturation of the sample and a specific relation between $H_\perp$ and $H_{//}$. Our result that the appearance of the bubble domain phase is independent of the strength of the magnetic field and that $H_{//} > 1$ kOe will wipe out the stripe domains completely [9] show that this mechanism is unlikely to explain our observation.

To test the stability of the bubble domain phase, temperature dependent domain imaging was performed after creating the bubble domains. Figure 5 shows the magnetic domain images of Fe(3.5 ML)/Ni(10.6 ML) at different temperatures. Below 360 K, the film remains in the bubble domain phase. At $T = 360$ K, the bubble domains become elongated, signaling a transition from the bubble domain phase to the stripe domain phase. At $T = 370$ K, the stripe domains are fully developed. After cooling the sample back to room temperature, the film remains in the stripe domain phase rather than recovering the bubble domain phase. It should be mentioned that the smaller stripe domain width at high temperature is due to the decreased effective magnetic anisotropy [20]. The result of Fig. 5 suggests that the bubble domain phase may have a higher free energy than the stripe domain phase in the absence of a magnetic field. However, there should exist an energy

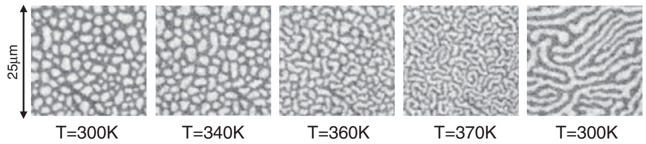

FIG. 5. Bubble domains near Fe(3.5 ML)/Ni(10.6 ML)/Cu(001). The bubble domains change to the stripe domains after increasing the temperature to 370 K. The stripe domains remain after cooling to room temperature.

barrier between the bubble and stripe phases that keeps the bubble domain phase metastable below 370 K. On the other hand, the conversion of the stripe phase to the bubble phase within a nearly in-plane magnetic field indicates that the bubble domain phase has a lower energy than the stripe domain phase within the nearly in-plane magnetic field in a narrow region of the effective magnetic anisotropy (K_e). The stability of stripe domain phase has been discussed in terms of periodic domain boundary fluctuations, and the result has been controversial. In particular, the instability of an isolated single stripe does not necessarily imply the instability of the stripe phase. In thick garnet films, the appearance of the bubble domain phase usually requests the presence of an external magnetic field and the micron-sized bubble phase exists in a wide range of the magnetic anisotropy [11]. This property seems to not explain our result that the submicron-sized bubble domains in Fe/Ni film appear only in a narrow region of the SRT and are metastable in the absence of an external magnetic field. For a 2D magnetic system, the total energy calculation predicts [17] that the bubble domain phase is energetically favorable over the stripe domain phase when the spin-up (or -down) area fraction is less than 0.28. However, this model cannot fully explain our annealing result and why a nearly in-plane magnetic field is needed to generate the bubble domain phase. In Ref. [14], the phase diagram of a ferromagnetic film in ($H_{//}$, $H_{\perp}$, T) space was discussed but without including the bubble domain phase. Obviously, in order to obtain a clear understanding of our experimental results, the stability of the stripe and the bubble domain phases needs to be understood as a function of K_e, $H_{//}$, and $H_{\perp}$ in the future studies. We hereby present our experimental observation to stimulate research on the bubble domain phase, and furthermore on the true ground state domain structure of a magnetic ultrathin film at the SRT.

In summary, the spin reorientation transition (SRT) of the Fe/Ni/Cu(001) system was investigated. After applying a nearly in-plane magnetic field, the stripe domain phase within a narrow thickness region undergoes a transition to the bubble domain phase. A small out-of-plane component of the "in-plane" magnetic field is crucial to generate the bubble domain phase. It is also shown that tilting the magnetic field out-of-plane more than 10° from the in-plane direction prevents the formation of the bubble domain phase. A temperature dependent measurement shows that the bubble domain phase changes back to the stripe domain phase above 370 K, indicating that the bubble domain phase is a metastable phase in the absence of a magnetic field.

This work was supported by National Science Foundation No. DMR-0405259, US Department of Energy No. DE-AC03-76SF00098, National Natural Science Foundation of China, and 973-Project under Grant No. 2006CB921300.

[1] C. Kittel, Rev. Mod. Phys. **21**, 541 (1949).
[2] D. P. Pappas, K. P. Kämper, and H. Hopster, Phys. Rev. Lett. **64**, 3179 (1990).
[3] Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. Lett. **70**, 1006 (1993).
[4] N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).
[5] Myron Bander and D. L. Mills, Phys. Rev. B **38**, 12 015 (1988).
[6] A. B. Kashuba and V. L. Pokrovsky, Phys. Rev. Lett. **70**, 3155 (1993).
[7] R. Allenspach and A. Bischof, Phys. Rev. Lett. **69**, 3385 (1992).
[8] A. Vaterlaus *et al.*, Phys. Rev. Lett. **84**, 2247 (2000).
[9] Y. Z. Wu *et al.*, Phys. Rev. Lett. **93**, 117205 (2004).
[10] Y. Yafet and E. M. Gyorgy, Phys. Rev. B **38**, 9145 (1988).
[11] A. Hubert and R. Schäfer, *Magnetic Domains* (Springer, New York, 1998).
[12] A. H. Bobeck and Hed Scovil, Sci. Am. **224**, 78 (1971); T. H. O'Dell, Rep. Prog. Phys. **49**, 589 (1986).
[13] T. Fukumura *et al.*, Science **284**, 1969 (1999).
[14] Ar. Abanov, V. Kalatsky, V. L. Pokrovsky, and W. M. Saslow, Phys. Rev. B **51**, 1023 (1995).
[15] A. B. MacIsaac, K. De'Bell, and J. P. Whitehead, Phys. Rev. Lett. **80**, 616 (1998).
[16] E. Y. Vedmedenko, H. P. Oepen, A. Ghazali, J.-C. S. Lévy, and J. Kirschner, Phys. Rev. Lett. **84**, 5884 (2000).
[17] K. Ng and D. Vanderbilt, Phys. Rev. B **52**, 2177 (1995).
[18] O. Portmann, A. Vaterlaus, and D. Pescia, Nature (London) **422**, 701 (2003).
[19] O. Portmann, A. Vaterlaus, and D. Pescia, Phys. Rev. Lett. **96**, 047212 (2006).
[20] C. Won *et al.*, Phys. Rev. B **71**, 224429 (2005).
[21] J. Thomassen, F. May, B. Feldmann, M. Wuttig, and H. Ibach, Phys. Rev. Lett. **69**, 3831 (1992).
[22] D. Li, M. Freitag, J. Pearson, Z. Q. Qiu, and S. D. Bader, Phys. Rev. Lett. **72**, 3112 (1994).
[23] Xiangdong Liu and Matthias Wuttig, Phys. Rev. B **64**, 104408 (2001).
[24] B. Schulz and K. Baberschke, Phys. Rev. B **50**, 13 467 (1994).
[25] A. Bauer, E. Mentz, and G. Kaindl, J. Magn. Magn. Mater. **198–199**, 489 (1999).
[26] A. Enders, D. Repetto, D. Peterka, and K. Kern, Phys. Rev. B **72**, 054446 (2005).