Magnetic stripe melting at the spin reorientation transition in Fe/Ni/Cu(001)

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(Received 8 December 2004; revised manuscript received 17 February 2005; published 29 June 2005)

Magnetic stripe domains in Fe/Ni/Cu(001) were imaged and studied using photoemission electron microscopy. The stripe domain width decreases exponentially as the system approaches the spin reorientation transition (SRT) point. A reduction of the Curie temperature (T_C) is observed within a narrow gap of the SRT region. For film with fixed thickness, the stripe domains below T_C represent only part of the stripe phase due to a higher SRT temperature than T_C.

DOI: 10.1103/PhysRevB.71.224429 PACS number(s): 75.70.Ak

Magnetic long-range order in a two-dimensional (2D) system has been an intensively studied topic in condensed-matter physics. It is well known that magnetic long-range order does not exist in an isotropic 2D Heisenberg system at any finite temperature,[1] but could be established by adding a uniaxial magnetic anisotropy to the system.[2] In experiment, this topic has been addressed by investigating the so-called spin reorientation transition (SRT) in magnetic ultrathin films in which the perpendicular magnetocrystalline anisotropy and the in-plane dipolar magnetic anisotropy (shape anisotropy) can be adjusted by varying either the film temperature or the film thickness. At the SRT point, where the spin direction switches from perpendicular to in-plane directions of the film, the perpendicular magnetocrystalline anisotropy cancels the in-plane magnetic shape anisotropy. Therefore, an investigation of the magnetic long-range order at the SRT is expected to reveal the magnetic origin of the 2D Heisenberg system. Although macroscopic magnetic measurement shows a suppression of the magnetization within a pseudogap of the SRT,[3,4] microscopic domain imaging shows that the reduced macroscopic magnetization is not due to a loss of the magnetic long-range order but due to the formation of magnetic stripe domains.[5,6] Theoretically, it is found that the stripe domain phase has a lower energy than a single domain phase and that the stripe domain width should shrink rapidly towards a minimum value as the total effective magnetic anisotropy approaches zero.[7,8] The discovery of the magnetic stripe phase stimulated many research activities to investigate the stripe domain dynamics,[9] phase transitions,[10] and ground-state spin structures,[11] etc. For example, it is found that the stripe phase could evolve into a tetragonal liquid phase under certain conditions.[12,13] It is also shown that an in-plane exchange field could line up the stripe domain direction as well as to reduce the stripe domain width.[14] Despite many achievements, less attention has been paid to the stripe domain melting at the Curie temperature, especially near the SRT point. Obviously, more investigation is needed to better understand the magnetic origin of 2D magnetic systems. We report here our study of the Fe/Ni/Cu(001) system. Using photoemission electron microscopy (PEEM), we investigated the behavior of the magnetic stripe domains as a function of the Fe film thickness and temperature. We found that the underlying mechanism of the stripe domain formation and a reduction of the Curie temperature in the SRT region is a result of a crossover from the anisotropy length scale to the dipolar length scale.

An eletropolished Cu(001) single-crystal disk (10-mm-diam and 2-mm-thick) was cleaned in an ultrahigh-vacuum (UHV) system by cycles of Ar ion sputtering at ~2 keV and annealing at ~600 °C. fcc Fe film on Cu(001) has been a model system for the study of stripe domains, but the ferromagnetic (F) to antiferromagnetic (AF) transition at ~4 ML of Fe in Fe/Cu(001) makes it unclear whether the stripe domains observed in this system reveal the whole stripe phase.[15] To ensure an observation of the complete stripe phase, the Fe SRT thickness should be thinner than the F/AF transition thickness.[16] It is well known that Ni film grows epitaxially on Cu(001) and has an in-plane magnetization below ~7 ML thickness.[17] Thus to shift the Fe SRT thickness below the F/AF transition thickness, we weaken the perpendicular magnetic anisotropy by adding ~5 monolayer(ML) Ni to the Fe film so that the Fe/Ni(5 ML) film on Cu(001) undergoes the SRT within the Fe ferromagnetic phase. Fe/Ni(5.4 ML) film was grown epitaxially onto the Cu(001) by evaporating Ni and Fe from heated alumina crucibles. The evaporation rate was monitored by a quartz thickness monitor which was calibrated by reflection high-energy electron diffraction (RHEED) intensity oscillations. The Fe film was grown into a wedge shape by moving the substrate behind a shutter to facilitate a continuous change of the Fe film thickness. The wedge slope (~3 ML/mm) was determined by the Fe evaporation rate and the substrate moving speed. The Fe/Ni(5.4 ML)/Cu(001) was covered with ~10 ML Cu protection layer, and transferred immediately into the PEEM-II chamber at beamline 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 surface normal direction. The magnetic domain images were obtained at room temperature by taking the ratio of L_3 and L_2 edges, utilizing the effect of x-ray magnetic circular di-

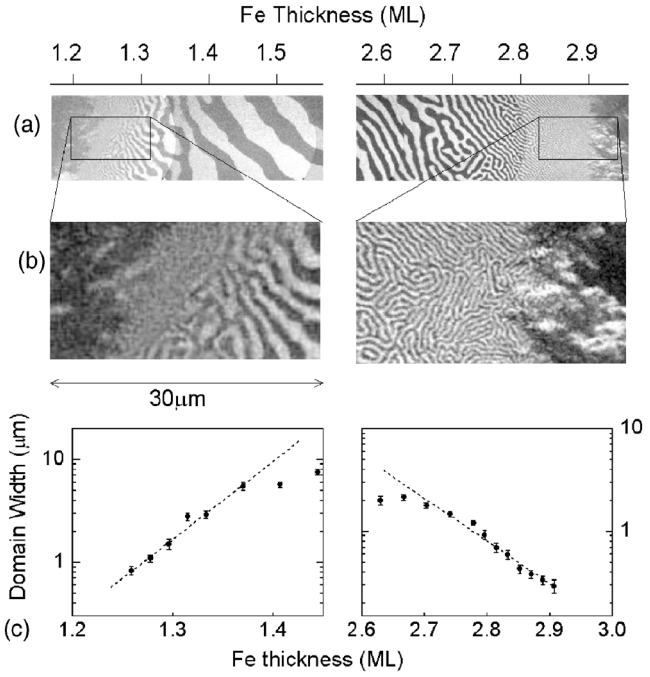

FIG. 1. (a) Magnetic domains of Fe(wedge)/Ni(5.4 ML)/Cu(001). (b) Zoom-in images near the SRT of 1.2 ML and 2.9 ML of Fe. (c) Stripe domain width decreases exponentially as the Fe film thickness changes towards the SRT point.

chroism (XMCD).[14] Since the Fe and Ni magnetizations are strongly coupled to behave as a single magnetic layer, we show only Fe PEEM images to represent the Fe/Ni magnetic domains in this paper.

Figure 1(a) shows the magnetic domain images of the Fe/Ni(5.4 ML) at different Fe thicknesses. As the Fe film thickness increases, we observed parallel-perpendicular-parallel SRT with the perpendicular magnetization in the range of 1.2 ML$<d_{\mathrm{Fe}}<$2.9 ML. The SRT at 1.2 ML Fe is caused by the increased perpendicular magnetic anisotropy with increasing Fe film thickness. The SRT at 2.9 ML Fe is due to the increased in-plane magnetic shape anisotropy with increasing Fe film thickness. This observation is not surprising because both Ni(5.4 ML) and thick Fe films have an in-plane magnetization so that the perpendicular magnetocrystalline anisotropy of the Fe film dominates only in a finite thickness range to generate a perpendicular magnetization. Nevertheless, magnetic stripe domains were observed in both SRT regions with the stripe domain width decreasing rapidly towards the SRT point [Fig. 1(a)].

Stripe domains result from the competition between the short-range exchange interaction and the long-range dipole interaction. For a 2D Heisenberg square lattice with lattice constant a, nearest-neighbor exchange interaction J, and uniaxial magnetic anisotropy K, the Hamiltonian is

$$H=-J\sum_{\langle i,j\rangle}\vec{S}_i\cdot\vec{S}_j-K\sum_i S_{iz}^2 + \frac{\Omega}{4\pi}\sum_{i,j}\frac{\vec{S}_i\cdot\vec{S}_j-3(\vec{S}_i\cdot\hat{r}_{ij})(\vec{S}_j\cdot\hat{r}_{ij})}{[(n_i-n_j)^2+(m_i-m_j)^2]^{3/2}}. \quad (1)$$

Here $\vec{S}_i$ is the spin at site i, $\Omega=2\pi\mu^2/a^3$ is the dipolar interaction strength, and μ is the magnitude of the magnetic moment at each lattice site. By minimizing the total energy, the stripe domain width (in unite of lattice constant a) can be derived,[7,14]

$$L=\frac{5J\pi^2}{6\Omega}\frac{\exp(\sqrt{\pi^4JK_e/\Omega^2}+1)}{\sqrt{\pi^4JK_e/\Omega^2}+1}, \quad (2)$$

where $K_e=K-\Omega$ is the effective magnetic anisotropy. For an ultrathin film, the values of J, K, and Ω need to be normalized by the film thickness. If the ultrathin film consists of d atomic layers, one can view that the d spins in the vertical direction are rigidly tightened together so that the exchange interaction J of the 2D lattice in the model should be magnified by a factor of d. The magnetic anisotropy K remains unchanged if it is surface type or scales linearly with d if it is volume type. For the dipolar interaction, however, one has to consider the short- and long-range parts separately. The short-range part forms the conventional shape anisotropy, thus it scales linearly with the film thickness. The long-range part comes from the dipolar interaction between two blocks of d spins at a distance much greater than the film thickness, thus it should scale with the square of the film thickness.[7] For the case of Fe/Ni bilayers, J and K should be normalized as $J=J_{\mathrm{Fe}}d_{\mathrm{Fe}}+J_{\mathrm{Ni}}d_{\mathrm{Ni}}$ and $K=K_{\mathrm{Fe}}^V d_{\mathrm{Fe}}+K_{\mathrm{Ni}}^V d_{\mathrm{Ni}}+K_{\mathrm{Ni\text{-}Cu}}^S+K_{\mathrm{Fe\text{-}Ni}}^S+K_{\mathrm{Fe\text{-}Cu}}^S$ (V and S denote the volume and surface contributions), where the film thickness d is measured in units of layer spacing ($a_\perp=1.8$ Å). The short- and long-range dipole interactions are $\Omega_S=2\pi(d_{\mathrm{Fe}}\mu_{\mathrm{Fe}}^2+d_{\mathrm{Ni}}\mu_{\mathrm{Ni}}^2)/(a_{//}^2a_\perp)$ and $\Omega_L=2\pi(d_{\mathrm{Fe}}\mu_{\mathrm{Fe}}+d_{\mathrm{Ni}}\mu_{\mathrm{Ni}})^2/a_{//}^3$, where $a_{//}$ is the in-plane lattice constant ($a_{//}=2.55$ Å). Then the K_e and Ω in Eq. (2) should be replaced by $K_e=K-\Omega_S$ and Ω_L, respectively, and L should be measured in units of the in-plane lattice constant. At the SRT point ($K_e=0$), the stripe domain width reaches a minimum value of $L_{\min}=5eJ\pi^2/6\Omega_{\mathrm{L}}$. Away from the SRT point, K_e can be expressed as $K_e(d_{\mathrm{Fe}})\approx[\partial K_e(d_{\mathrm{Fe}}^0)/\partial d_{\mathrm{Fe}}]\delta d_{\mathrm{Fe}}$, where $\delta d_{\mathrm{Fe}}=d_{\mathrm{Fe}}-d_{\mathrm{Fe}}^0$ is the thickness deviation away from the SRT point. Then it is easy to realize from Eq. (2) that the stripe domain width depends exponentially on the Fe film thickness near the SRT point. This property was reported in Ref. 14 and can also be seen clearly in Fig. 1(c), where the logarithm of the stripe domain width depends linearly on the Fe film thickness in both SRT regions near 1.2 and 2.9 ML of Fe.

To explore the physical meaning of Eq. (2), we discuss here the minimum value and the exponential change of the stripe domain width at the SRT. To understand the result of $L_{\min}=2.27J\pi^2/\Omega_{\mathrm{L}}$ given by Eq. (2), note that the energy competition at the SRT point ($K_e=0$) takes place between the short-range exchange interaction and the long-range dipole interaction. Since stripe domains can be viewed as a static spinwave whose excitation energy is $E\sim J\pi^2/\lambda^2$ (λ is the spinwave wavelength), the formation of the stripe domains of stripe width L should cost energy of $\delta E\sim+J\pi^2/L^2$. On the other hand, dipole interaction favors opposite alignment of perpendicular spins. Then the dipole interaction between a stripe of L up-spins and a stripe of L down-spins should lower the energy by $\delta E\sim\Omega_{\mathrm{L}}(LL/L^3)=\Omega_{\mathrm{L}}/L$. Therefore, at

the SRT point, a balance of the short-range exchange interaction and the long-range dipole interaction leads to a minimum stripe domain width that satisfies $J\pi^2/L_{\min}^2 \sim \Omega_L/L_{\min}$ or $L_{\min} \sim J\pi^2/\Omega_L$, agreeing with the exact result of $L_{\min} = 2.27J\pi^2/\Omega_L$. Taking the values of $d_{Fe} \approx 2.9$ ML, $\mu_{Fe} \approx 2.6\mu_B$, $J_{Fe} \approx 36$ meV for fcc Fe,[18] and $d_{Ni} \approx 5.4$ ML, $\mu_{Ni} \approx 0.6\mu_B$, $J_{Ni} \approx 4.6$ meV for fcc Ni,[19] we estimate $L_{\min} = 2.27J\pi^2/\Omega_L \approx 0.31$ μm, which agrees reasonably well with the experimental value of $L_{\min} \approx 0.29 \pm 0.04$ μm.

After the discussion of $L_{\min}$, we now discuss the exponential dependence of the stripe domain width on the magnetic anisotropy. Note that the uniaxial magnetic anisotropy favors a ferromagnetic single domain phase and the dipolar interaction favors a stripe domain phase. The exponential increase of the stripe domain width with increasing magnetic anisotropy actually reflects a crossover from the dipole-dominated regime to the anisotropy-dominated regime. This crossover can be better understood with the expression of the magnetic anisotropy length of $L_{ani} = \sqrt{J/K_e}$ and the dipole length of $L_{dip} = J/\Omega_L$. As is well known, L_{ani} (or L_{dip}) defines a length scales beyond which the magnetic anisotropy (or the dipole interaction) dominates the exchange interaction J. Thus the behavior of the system should be dominated by the dipole interaction in the regime of $L_{dip} \ll L_{ani}$ and by the anisotropy in the regime of $L_{ani} \ll L_{dip}$, respectively. By rewriting Eq. (2) in terms of L_{ani} and L_{dip}, we have

$$L = \frac{5\pi^2 L_{dip}}{6} \frac{\exp(\pi^2 L_{dip}/L_{ani} + 1)}{\pi^2 L_{dip}/L_{ani} + 1}. \quad (3)$$

Then Eq. (3) expresses merely that the transition from the stripe phase ($L \sim L_{dip}$) to the single domain phase ($L \sim \infty$) is a result of the crossover from the dipole-dominated regime ($L_{dip} \ll L_{ani}$) to the anisotropy-dominated regime ($L_{ani} \ll L_{dip}$). It is this length-scale crossover that results in the exponential change of the stripe domain width in the SRT region.

It was shown that the spin-spin correlation length in a 2D Heisenberg system is $\xi \approx \exp(2\pi J/T)$.[20] This correlation length grows to infinity as the temperature is lowered toward zero, reflecting the Mermin-Wagner theorem that there is no magnetic long-range order at any finite temperature. However, a length scale L introduced by a physical quantity could stop the growth of ξ, thus in principle trigger a possible phase transition.[21] Although it is difficult to obtain the exact scaling function, it is intuitive to estimate the transition temperature (if it exists) by setting $L \approx \xi = \exp(2\pi J/T_C)$, which leads to $T_C \approx 2\pi J/\ln L$. For example, the presence of a uniaxial magnetic anisotropy K introduces an anisotropy length of $L_{ani} = \sqrt{J/K_e}$. Thus the magnetic anisotropy could trigger a magnetic phase transition at $T_C \approx 2\pi J/\ln L_{ani} = 4\pi J/\ln(J/K_e)$, which is the result derived by Bander and Mills using renormalization-group theory.[2] With the presence of both the magnetic anisotropy and the dipole interaction, the Curie temperature T_C should be determined by the lower value of L_{ani} and L_{dip}. Far below the SRT thickness, $L_{ani} \ll L_{dip}$ so that the Curie temperature of the film is determined by $T_C \sim 2\pi J/\ln L_{ani}$, which increases with the film thickness ($J \propto d$). As the film thickness increases further to approach the SRT point, the L_{ani} diverges while the L_{dip} remains a smooth function. Therefore, the Curie temperature of $T_C \sim 2\pi J/\ln L_{ani}$ will decrease as $L_{ani} \to \infty$ until it reaches the L_{dip}-determined Curie temperature of $T_C \sim 2\pi J/\ln L_{dip}$. Thicker than the SRT thickness, the film has an in-plane magnetization with a higher Curie temperature determined by the in-plane magnetic anisotropy. Then a length scale crossover from $L_{ani} < L_{dip}$ to $L_{ani} > L_{dip}$ in the SRT region should also manifest as a reduction of the Curie temperature at the SRT. If the measurement temperature is lower than $T_C \sim 2\pi J/\ln L_{dip}$, a complete stripe domain phase should be observed. If the measurement temperature is higher than $T_C \sim 2\pi J/\ln L_{dip}$, there should exist a paramagnetic gap within a narrow thickness range in the SRT region. This situation can be observed in the SRT at 1.2 ML of Fe [Fig. 1(b)] in which the stripe domains disappear in a narrow region between the perpendicular magnetic stripe phase and the in-plane ferromagnetic phase. However, such a paramagnetic phase does not exist in the SRT region at 2.9 ML of Fe at room temperature. This is because the T_C at 2.9 ML Fe is higher than room temperature. To verify that the paramagnetic phase also exists in the SRT region at 2.9 ML of Fe, we increased the sample temperature and took magnetic domain images around 2.9 ML of Fe (Fig. 2). At room temperature, the stripe domains exist in the entire SRT region. At $T = 365$ K, the stripe domains starts to melt in some regions. At $T = 370$ K, it can be clearly seen that a paramagnetic gap develops at the SRT point. At $T = 380$ K, the paramagnetic gap is further widened. The result of Fig. 2 verifies that the T_C of a thin film is reduced at the SRT. It should be mentioned that the magnetic spatial resolution of our PEEM instrument is ~ 100 nm so that the absence of the stripe domains at high temperature is not due to the instrumental resolution. However, we cannot rule out the possibility of a fast-moving stripe domain phase (the acquisition time for each PEEM image is ~ 10–100 s), i.e., we cannot distinguish between a conventional paramagnetic phase and a dynamical stripe phase.

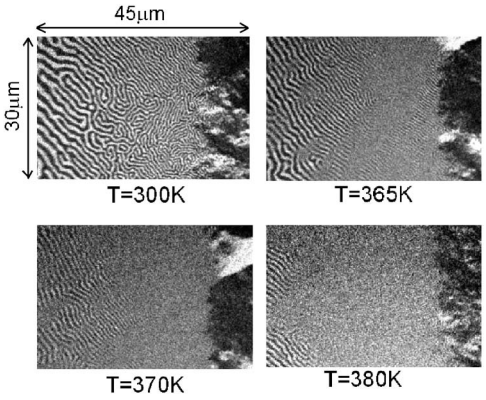

FIG. 2. Stripe domains of Fe/Ni(5.4 ML)/Cu(001) at the SRT near 2.9 ML of Fe. The Fe thickness difference between left and right edges is ~0.2 ML. Paramagnetic phase appears at high temperature within a narrow gap.

It is also worth noting that the stripe domain width at the boundary between the stripe phase and the paramagnetic phase is greater than $L_{\min}$ (see the images at $T = 300$ K and

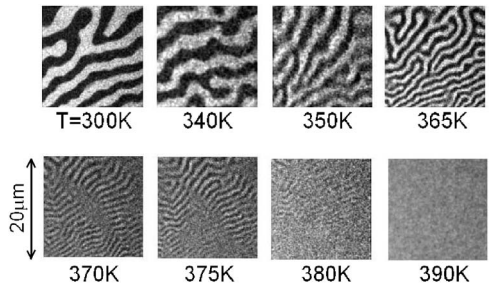

FIG. 3. PEEM images ($20\ \mu m \times 20\ \mu m$) of Fe(2.7 ML)/Ni(5.4 ML)/Cu(001) at different temperatures. The system undergoes a perpendicular-to-paramagnetic transition at $T=380$ K before the SRT occurs.

$T=380$ K in Fig. 2). This can be easily understood that the film becomes paramagnetic before K_e reaches zero. This result also implies that for a film of fixed thickness, an observation of the magnetic stripe domains does not ensure an observation of the SRT point. If the SRT occurs at a higher temperature than T_C ($T_{SRT} > T_C$), the observed stripe domains will be only part of the complete stripe phase and the stripe domain width at T_C should be greater than L_{min}. Only if the SRT occurs at a temperature lower than T_C ($T_{SRT} < T_C$) will the magnetization undergo perpendicular-parallel-paramagnetic transitions and the stripe domain width will reach its minimum value of L_{min} at the SRT point.[22] Then for a system with a perpendicular-paramagnetic transition, the stripe domain width at T_C could serve as an indicator of how high the T_{SRT} is above T_C. To see this effect, we took stripe domain images of Fe(2.7 ML)/Ni(5.4 ML)/Cu(001) film at different temperatures (Fig. 3). As the temperature increases, the stripe domain width decreases as well as the domain contrast. At $T=370$ K, we start to observe the melting of the stripe domains in some regions. Above $T=380$ K, the stripe domains disappear completely due to the perpendicular-paramagnetic transition. The width of the stripe domains at different temperatures is determined from the PEEM images and plotted in Fig. 4. The stripe domain width decreases exponentially with increasing temperature until the system reaches the Curie temperature of $T_C=380$ K. It should be mentioned that the stripe domain phase is completely recovered after cooling the sample back to room temperature. The decrease of the stripe domain width with increasing temperature is due to the decrease of K_e towards the SRT temperature, $K_e(T) \approx [dK_e(T_{SRT})/dT](T-T_{SRT})$, similar to the decrease of K_e with increasing Fe film thickness towards the 2.9 ML SRT thickness (Fig. 1). Note that both J and Ω should depend weakly on temperature; the value of $L_{min}=0.3\ \mu m$ should be reached if the complete stripe domain phase is present in the temperature-dependent result. The fact that there is a wider domain width of $\sim 0.55\ \mu m$ than $L_{min}=0.3\ \mu m$ at T_C shows that the system has a nonzero K_e at T_C, i.e., the system reaches T_C at a lower temperature before reaching T_{SRT}. Assuming a linear dependency of K_e on temperature around SRT, we extrapolate a SRT temperature of $T_{SRT}=395$ K to reach $L_{min}=0.3\ \mu m$, which is about 15 K higher than the Curie temperature. Using this method, we extract T_{SRT} in the range of 1.5 ML $< d_{Fe} <$ 2.8 ML within which T_{SRT} is greater than T_C. Together with the perpendicular-paramagnetic and parallel-paramagnetic transitions, we construct a phase diagram in the T-d_{Fe} plane for the sample of Fe/Ni(5.4 ML)/Cu(001) (Fig. 5). The Curie temperature increases with the Fe film thickness except for a sharp reduction within a narrow region of the SRT. The overall increase of T_C with film thickness is a common characteristic of magnetic thin films. The suppression of T_C at the SRT, however, is due to the reduced magnetic anisotropy.

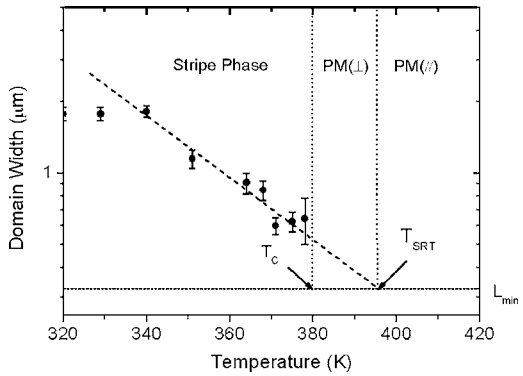

FIG. 4. Stripe domain width of Fe(2.7 ML)/Ni(5.4 ML)/Cu(001) vs temperature. Extrapolating the domain width to the minimum value of stripe domains ($L_{min}=0.31\ \mu m$) derives a SRT temperature of $T_{SRT}=395$ K.

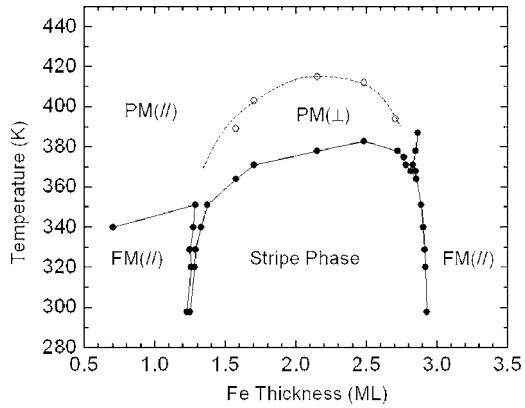

FIG. 5. Magnetic phase diagram of Fe/Ni(5.4 ML)/Cu(001). The dashed and solid lines are to guide the eye.

In summary, the Fe/Ni(5.4 ML)/Cu(001) system was investigated as a function of the Fe film thickness and temperature. As a result of a crossover from the anisotropy length scale to the dipolar length scale, we found that the stripe domain width decreases exponentially towards the SRT point, and that the Curie temperature of the film is reduced to result in a paramagnetic phase within a narrow thickness gap in the SRT region. In addition, the SRT temperature at a fixed Fe film thickness is higher than the Curie temperature. A magnetic phase diagram was constructed in the temperature-thickness plane to describe the magnetic phases of this system.

This work was supported by National Science Foundation Grant No. DMR-0405259, U.S. Department of Energy Contract No. DE-AC03-76SF00098, Chinese National Science Foundation, Chinese Science and Technology Ministry for 973 Project, and ICQS of CAS.

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