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Correlation between spin reorientation transition and Curie temperature of Ni_xPd_{1-x} alloy on Cu(001)

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The correlation between spin reorientation transition (SRT) and Curie temperature of Ni_xPd_{1-x} alloy on Cu(001) was investigated by ac susceptibility and magneto-optic Kerr-effect measurements. The result shows that Ni_xPd_{1-x} alloy exhibits a SRT at Ni concentration of 87% for 9 ML and of 96% for 7 ML. No reduction in Curie temperature was observed in the region of SRT as declared by F. Matthes *et al.* [J. Appl. Phys **91**, 8144 (2002)]. In addition it was found that Curie temperature increased with Ni concentration in different trends before and after SRT, which are attributed to the anisotropy variation.

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The ultrathin ferromagnetic films can reveal novel properties by virtue of their low dimensionalities. It is well known that the magnetic long-range order does not exist in an isotropic two-dimensional (2D) Heisenberg system at any finite temperature.[2] But this issue might be unrealistic since it is hard to physically realize the ultrathin ferromagnetic film without any anisotropy. In experiment, this topic has been addressed in the vicinity of the spin reorientation transition (SRT) in ultrathin magnetic films. Because at the SRT point, the magnetocrystalline anisotropy, surface anisotropy, and dipolar anisotropy could compensate in some cases. So it is very interesting to study the correlation between SRT and long-range magnetic order. In the nineties a suppression of magnetization within the SRT region was reported in Fe/Cu(001) and Fe/Ag(001) systems.[3,4] Recently a reduction in the Curie temperature was observed within the narrow gap of the SRT region in Fe/Ni/Cu(001) system.[5]

However SRT is not necessarily connected with a loss or weakening of long-range magnetic order. Whether the anisotropy within the SRT region is vanished or not depends on the SRT type.[6] If it is a continuous SRT, its anisotropy never disappears within the SRT region, where only the magnetization rotates between in plane and out of plane, e.g., Ni on Cu(001) system.[7,8] Interestingly a recent experiment on nine-monolayer Ni_xPd_{1-x} alloy demonstrated that there exist a SRT at 87% Ni concentration for Cu(001) substrate; meanwhile, a dramatic reduction in Curie temperature was observed within its SRT region.[1] Due to this striking experimental result we strive to investigate the correlation between SRT and Curie temperature of Ni_xPd_{1-x} alloy on Cu(001) substrate with a better-defined approach.

Instead of preparing one sample for one composition we adopt the composition wedge technique,[9,10] which can bridge the whole SRT region in one sample with a uniform thickness. This technique can help us get a more detailed picture about the variation in magnetic property versus the Ni concentration. Indeed we confirmed that there existed a SRT in the system as a function of Ni composition. But unlike the result declared in Ref. 1, no reduction in T_c was observed in the region of SRT in our experiment. And it was found that the increasing behavior of T_c with Ni concentration has different trends before and after SRT.

The experiment was carried out in an ultrahigh vacuum system equipped with reflection high-energy electron diffraction (RHEED), low-energy electron diffraction (LEED), cylindrical-mirror-analyzer-based auger electron spectroscopy (AES), magneto-optical Kerr effect (MOKE), and MOKE-based ac susceptibility. Before Ni_xPd_{1-x} alloy deposition the Cu(001) substrate was cleaned by cycles of Ar ion sputtering at 1 keV and subsequently annealed at 850 k for 15 min until a sharp (1×1) pattern appeared in LEED.[11] The growth temperature was kept at 300 K and the pressure was better than 8×10^{-10} mbar during the codeposition of Ni and Pd sources. The concentrations of the composition wedge were determined by AES. Two representative LEED patterns of Ni_xPd_{1-x} composition wedge sample shown in Figs. 1(a) and 1(b) demonstrated that the film was in a high-quality single crystalline fcc structure. The film thickness was monitored by RHEED intensity oscillation during the growth which was shown in Fig. 1(c). A detailed description about the growth method of composition wedge sample can be found elsewhere.[9]

The magnetic properties of Ni_xPd_{1-x} composition wedge sample were investigated through both MOKE and ac susceptibility measurements.[12–14] The ac susceptibility measurements were performed by using the longitudinal Kerr effect upon the application of an external magnetic field modulation in the film plane.[14–16] This modulation magnetic field had a frequency of 314 Hz and an amplitude of 2 Oe. The laser beam was focused to a 0.2-mm-size spot on the composition wedge sample. Thus the composition variation probed due to the wedge shape is only 1.2–1.3 % within the laser spot, so the composition within the laser spot can be considered uniform. To have a comparison with the result in our composition wedge sample was intentionally capped with 3 ML Cu because Ni_xPd_{1-x} alloy in the work by Matthes *et al.*[1] has a thinner critical thickness for SRT. And according to the previous experimental research on Ni/Cu(001) system the critical thickness for uncovered Ni film is about 10 ML while adsorption of CO or H_2 or capping with Cu could reduce the critical thickness by about 3–4 ML.[17] Thus, another benefit of capping sample with Cu is that it could avoid the complication of residual gas in the chamber.

We first explored the magnetic properties of 9 ML Ni_xPd_{1-x} composition wedge sample. Two representative MOKE hysteresis loops at RT were shown in Fig. 2(a), one

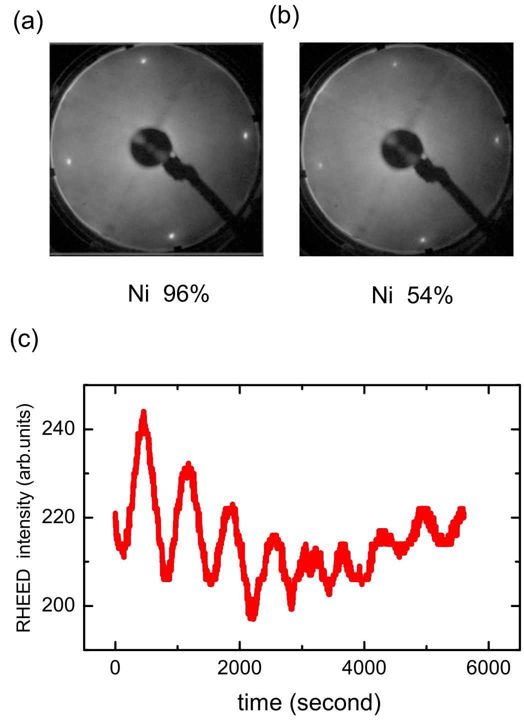

FIG. 1. (Color online) (a),(b) LEED patterns of 96% and 54% Ni concentrations in 9 ML Ni_xPd_{1-x} composition wedge sample (E=109 eV). (c) RHEED intensity oscillation during the growth in Ni_xPd_{1-x} composition wedge sample on Cu(001).

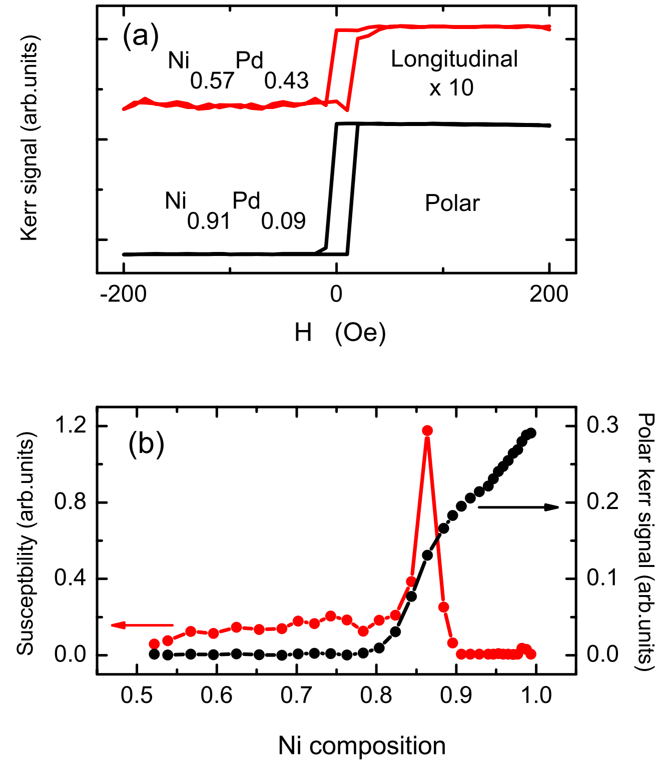

FIG. 2. (Color online) (a) The longitudinal MOKE hysteresis loop of 57% Ni and polar MOKE hysteresis loop of 91% Ni of 9 ML composition wedge sample. (b)The imaginary part of the ac susceptibility Im(χ) and remanent magnetization determined from polar MOKE of 9 ML composition wedge sample as a function of Ni composition.

for $Ni_{0.57}Pd_{0.43}$ with easy axis of magnetization in plane and the other for $Ni_{0.91}Pd_{0.19}$ with perpendicular easy axis. The remanent magnetization (M_r) determined from the polar Kerr signal as a function of Ni composition is plotted in Fig. 2(b). It shows that the remanent magnetization of polar MOKE increased gradually, which could suggest that the magnetization rotated continuously from in plane to out of plane. The imaginary part of the ac susceptibility [Im(χ)] as a function of Ni composition was also shown in Fig. 2(b). The intensity of Im(χ) showed a pronounced peak at the SRT position. It clearly demonstrated that SRT occurred at Ni concentration of 86% for 9 ML Ni_xPd_{1-x} alloy. Such SRT position was the same as the early experimental result in Ref. 1.

This composition-driven SRT can be understood in a similar way as Ni/Cu(001) system.[8,18,19] The SRT takes place when the magnetocrystalline anisotropy $K=K_V+K_S/d$ (d is the thickness of the film) gets balanced with the shape anisotropy $2\pi M^2$. The volume anisotropy K_V comes from the magnetoelastic anisotropy, which is due to the tetragonal distortion of the lattice structure on the Cu(001) substrate. The positive K_V is the driving force of the perpendicular easy axis, while the negative value of K_S favored the in-plane magnetization as well as the shape anisotropy. The incorporation of Pd in the Ni lattice could lead to a reduction in the lattice mismatch and thus a strain relaxation in the film. As a consequence the magnetoelastic contribution will be reduced. So at a fixed thickness the driving force of the perpendicular easy direction (K_V) gets larger from Pd-rich side to Ni-rich side. When K_V is large enough to balance other anisotropies at a proper concentration, the SRT will happen.

Now we concentrate on the correlation between SRT and Curie temperature of Ni_xPd_{1-x} alloy on Cu(001). The Curie temperature was determined by the ac susceptibility measurement as shown in the inset of Fig. 3(a). In Fig. 3(a) the Curie temperature of 9 ML Ni_xPd_{1-x} composition wedge sample versus Ni composition was plotted. A sharp peak of the susceptibility demonstrated the SRT position where no Curie temperature (T_c) reduction was found around the SRT region. And T_c was increasing from Pd-rich side to Ni-rich side. This trend can be simply understood by the fact that Ni is ferromagnetic while Pd is not. But at the same time we noticed that this increasing behavior was not simply like the bulk system of Ni_xPd_{1-x} alloy.[20] In our case, SRT actually makes a significant difference to the increasing behavior of T_c. The Curie temperature of Ni_xPd_{1-x} alloy increased very slowly as a function of Ni composition before SRT, while it raised more sharply after SRT happened. So the SRT position behaved like a turning point in the curve of T_c versus Ni composition. Thus this phenomenon could indicate that it is the variation in anisotropy that influences the increasing behavior of T_c.

To make sure the correlation between SRT and Curie temperature we tried to alter the SRT position. So we prepared a 7 ML Ni_xPd_{1-x} composition wedge sample capped with 3 ML Cu to have a comparison. And its magnetic properties were investigated in the same way. The sharp peak of Im(χ) shown in Fig. 3(b) demonstrated that its SRT occurred at Ni

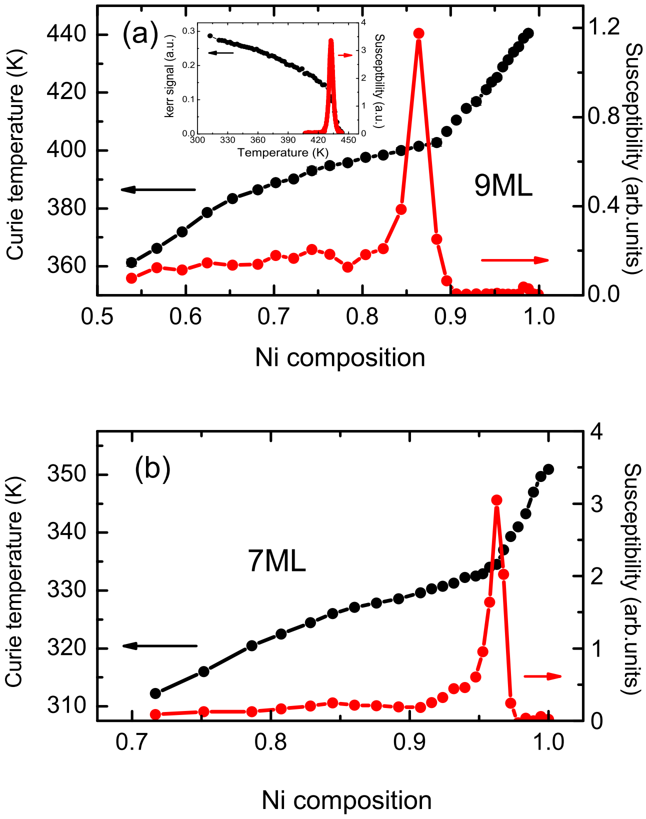

FIG. 3. (Color online) (a) Im(χ) and Curie temperatures versus Ni composition of 9 ML Ni_xPd_{1-x} composition wedge sample. An example of Curie temperature measurement by ac susceptibility is given in the inset, where the remanent magnetization determined by MOKE as a function of temperature is also shown. (b) Im(χ) and Curie temperatures versus Ni composition for 7 ML Ni_xPd_{1-x} composition wedge sample.

concentration of 96%. The Curie temperature of 7 ML Ni_xPd_{1-x} alloy versus Ni concentration was also shown in Fig. 3(b). The increasing behavior of Curie temperature was as similar as that of 9 ML. T_c increased monotonically versus Ni composition but in different trends. And the turning point was still the SRT position.

The SRT position of 7 ML shifted to the Ni-rich side compared to that of 9 ML. To understand this phenomenon we used the following phenomenological model. If the film thickness gets thinner K_S/d will become larger. Then the volume anisotropy K_V has to be larger in order to satisfy the condition for SRT happening. As illustrated above, K_V is proportional to the mismatch between Ni_xPd_{1-x} alloy and Cu(001) substrate. So for thinner thickness the SRT position should shift to the larger mismatch side which is the Ni-rich side. With this phenomenological model a simple magnetic phase diagram is given in Fig. 4(a), which showed SRT boundary dependent on Ni concentration and film thickness.

According to our experimental results no reduction in the Curie temperature was observed around the SRT region. One reason could be that the SRT of Ni_xPd_{1-x} is continuous like what happens in Ni/Cu(001) system. Its anisotropy never disappears, but the easy direction of the magnetization rotates continuously within the transition.[6]

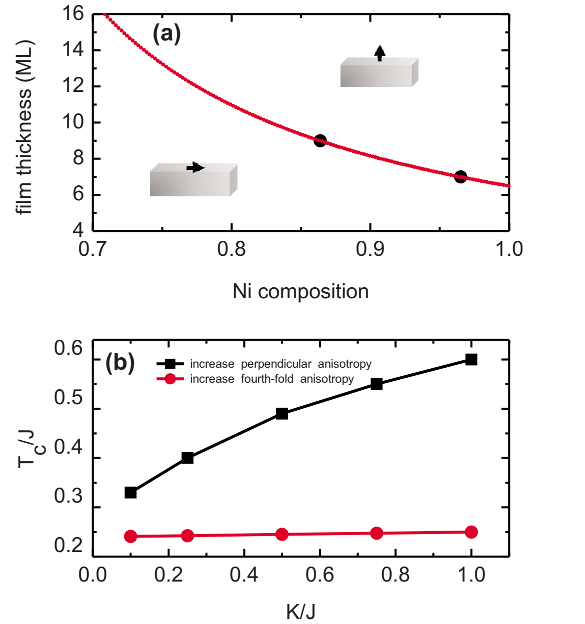

FIG. 4. (Color online) (a) Magnetic phase diagram of Ni_xPd_{1-x} alloy. The line separated the regimes of in-plane and perpendicular magnetization. (b) Curie temperatures versus increasing perpendicular anisotropy and fourfold in-plane anisotropy.

The other possibility could be that at the SRT point there still exists some residual perpendicular anisotropy which cants the magnetization 45° away from normal. This residual anisotropy is caused by variations in the perpendicular anisotropy across the film surface.[21] Another interesting phenomenon we found in our experiments is that the variation in magnetic anisotropy influenced the increasing behavior of Curie temperature. And the experimental results suggested that increasing the perpendicular anisotropy could enhance T_c much higher than the fourfold in-plane anisotropy. A simple Monte Carlo simulation was performed to demonstrate the impact of the anisotropy by studying the 2D Heisenberg model with perpendicular anisotropy $H=-J\Sigma_{ij}[S_i^xS_j^x+S_i^yS_j^y+(K+1)S_i^zS_j^z]$ and fourfold in-plane anisotropy $H=-J\Sigma_{ij}[(K+1)S_i^xS_j^x+(K+1)S_i^yS_j^y+S_i^zS_j^z]$, where K is the anisotropy parameter and J is the ferromagnetic exchange constant. The Curie temperature versus increasing perpendicular anisotropy and fourfold in-plane anisotropy were plotted in Fig. 4(b), respectively. The calculation result clearly showed that T_c can be enhanced much higher by raising the perpendicular anisotropy than the fourfold one. Simply speaking, it is because the fourfold in-plane anisotropy is a higher-order term compared to the perpendicular uniaxial anisotropy. So the perpendicular anisotropy breaks the symmetry of the system more seriously than the fourfold one. As a consequence the uniaxial anisotropy should be much easier in triggering the ferromagnetism than the fourfold anisotropy in low-dimensional system. This behavior is an interesting phenomenon which is a unique property of the ultrathin films.

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