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Temperature-driven spin reorientation transition of magnetron sputtered nickel thin film*

Song Xiao-Hui(宋小会)† and Zhang Dian-Lin(张殿琳)

Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, *China*

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The temperature-driven spin reorientation transition of magnetron sputtered Ni/Si (111) systems has been studied. The relationship between ac initial susceptibility and temperature of nickel films with different thicknesses shows that the magnetization orientation changes from in-plane to out-of-plane with the increase of temperature. The temperature dependence of magnetoelastic, magneto-crystalline, and magnetostatic anisotropies determines the direction of the reorientation transition. The temperature-driven spin reorientation transition is supported by Hall coefficient measurements which show that its temperature dependence is similar to that of susceptibility.

Keywords: magnetic anisotropy, spin reorientation transition, ac initial susceptibility
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1. Introduction

In recent years there has been the considerable interest in the phenomenon of spin-reorientation transitions (SRTs) in ultrathin magnetic films due to its potential advantages in magneto-optical recording.[1−6] Such a transition is induced by the competition among the magnetostatic, magnetocrystalline, and magnetoelastic energies in which their relative importance may change with thickness or temperature of the films. The temperature-driven SRT is inherently related to the thermodynamics in a two-dimensional magnetic system, and it is in a sense more fundamental and more complex in the investigation of the SRT of magnetic thin films. However, not many experimental results were reported about the temperature-driven SRT behaviour.[7−9] As far as we know, no general theoretical functional dependence of magnetic anisotropy energy on temperature is established. More experiments would be beneficial to the further investigation in this respect.

Ni film is one of the most interesting candidates for the above purpose. A large number of reports are devoted to the SRTs in Ni/Cu systems. Ni films on Cu(100) substrate show a perpendicular magnetic anisotropy over a wide thickness range from 2 to 13 nm.[10−13] Previous investigations show that the magnetizations of Ni/Cu(001) film are oriented parallel to the plane for film thicknesses below 7 monolayer (ML). The films change their spin alignment to out-of-plane in a thickness range from 7[10] to 37 ML. Then a gradual transition back to in-plane magnetization occurs again with the further increase of film thickness.[12] Comparatively, results of Ni film on Si substrate appearing in the literature are much less than those of Ni/ Cu. The growth process of Ni film on Si substrate has been investigated systematically.[14−16] In this paper we report the temperature-driven SRT phenomenon in the Ni/Si(111) systems in thicknesses ranging from 5 to 20 nm.

2. Experiment

Ni thin films were deposited on Si(111) substrates by using dc magnetron sputtering method at an ambient temperature. A pure Ni (99.99%) target was used. The vacuum in the sputtering chamber was 1×10^{-4} Pa and the working Ar gas pressure was 0.25 Pa in the depositing process. The magnetizations of the films were measured by using a DC/AC measurement system with a sensitivity of about 2×10^{-8} emu. The film thicknesses were measured by using a Dektak 8

*Project supported by the National Natural Science Foundation of China (Grant No 10334080).
†E-mail: xhsong@aphy.iphy.ac.cn

system (Veeco Instruments, Inc) with a sensitivity of about several angstroms (The thicknesses of the films at the thin ends were calculated from the deposition time).

The ac susceptibilities as a function of temperature were measured for different nickel films in an ac field (633 H_2 and 10 Oe, 1 Oe $\approx$ 80 A/m) perpendicular to the film plane. The background DC field was eliminated to less than 10^4 T. The results for three Ni films of different thicknesses are shown in Fig.1. We see that for each sample, with the increase of temperature, the susceptibility first increases to a maximum and then decreases. The peak on the susceptibility curve results from the change of magnetic orientation with temperature just as discussed in Refs.[17,18]. The curves for different thicknesses indicate that the SRTs of the Ni films shift toward low temperatures with the decrease of film thickness. The transition temperatures of SRT are about 100, 180 and 220 K for the film thicknesses of 5, 10 and 20 nm, respectively, estimated from Fig.1. We argue that the shift is caused by the change of relative importance of the shape anisotropy and the magnetocrystalline anisotropy (in favour of the in-plane magnetization) with respect to the magnetoelastic anisotropy (causing the out-of–plane magnetization).

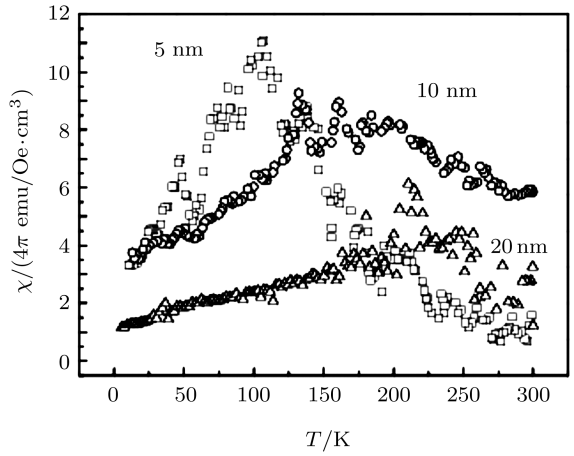


Fig.1. The low field susceptibility as a function of temperature for nickel thin films, with an ac magnetic field being perpendicular to the film plane.

3. Result and discussion

The magnetization orientation in a thin film is determined by the total anisotropic energy density that can be represented as $K^{\rm eff}\sin^2\theta$, where θ is the angle between the magnetization and the film normal and $K^{\rm eff}$ includes the contributions of all relevant anisotropic energies and is expressed as

$$K^{\rm eff} = -2\pi M_{\rm s}^2 + K_{\rm MC}^{\rm b} + K_{\rm ME}^{\rm b} + (K_{\rm I} + K_{\rm S})/d. \quad (1)$$

Here, $-2\pi M_{\rm s}^2$ is the shape anisotropy with $M_{\rm s}$, the saturation magnetization; $K_{\rm MC}^{\rm b}$ is the bulk magnetocrystalline anisotropy; $K_{\rm ME}$ is the magnetoelastic anisotropy; d is the film thickness; $K_{\rm S}$ and $K_{\rm I}$ are the surface anisotropy and the interface anisotropy, respectively. An earlier discussion[11] ascribed the thickness-induced spin reorientation of Ni films to the competition between $K_{\rm ME}^{\rm b}$ and $-2\pi M_{\rm s}^2$ plus $(K_{\rm I} + K_{\rm S})/d$. The discussion ignored the magnetocrystalline anisotropy, which could be reasonable because it had a small value at room temperature. However, the case is quite different when we look at the temperature-driven SRT. Since both the shape and magnetocrystalline anisotropic energies increase with the decrease of temperature, and the magnetoelastic anisotropic energy decreases with the increase of thickness, the competition between them plays an important role in the phenomenon.

In the following we will give a rough estimation to see how the SRT occurs. We notice that, while $K_{\rm ME}^{\rm b}$ is in favour of out-of-plane alignment, the other three terms in expression (1) all support the in-plane orientation. When $K_{\rm ME}^{\rm b}$ dominates, $K^{\rm eff}$ is positive, and the spin will be aligned out-of-plane. If the sum of the remaining terms exceeds $K_{\rm ME}^{\rm b}$, $K^{\rm eff}$ is negative, and we will have spin aligned in the plane. When the film thickness is larger than a critical value $d_{\rm c}$ (around 13ML[11]), $K_{\rm ME}^{\rm b}$ changes with T through $K_{\rm ME} = B\varepsilon = -B\eta(d_{\rm c}/d)^{2/3}$,[5] where B is the magnetoelastic constant; ε is the strain; η is the lattice mismatch between the film and the substrate and η increases with the decrease of temperature because of the big difference in thermal expansion between Ni and Si. This difference in expansion leads to a lattice mismatch as large as 2.73% at low temperatures. The temperature dependence of magnetocrystalline anisotropy is given by $K_{\rm MC} = K_0{\rm e}^{-aT^2}$.[19] The shape anisotropy $-2\pi M_{\rm s}(T) = -2\pi M_{\rm s}(0)(1 - T/T_{\rm c})^{3/2}$. Since $T_{\rm c}$ is dependent on d, the shape anisotropy depends on both T and d. The surface anisotropy and the interface anisotropy do not change with temperature in the range we discussed.[13] The temperature-driven SRT

occurs when K^{eff} changes its sign. Both the dependences of K_{ME}^{b} and $-2\pi M_{\text{s}}^2$ on d lead to a shift of the transition toward higher temperatures with the increase of d. This accords with what we observed (Fig.1). Figure 2 shows the change of K^{eff} with temperature for a sample with a thickness of 20 nm estimated from the above expressions. We see that the crossover from positive to negative occurs around 220 K, consistent with the experimental observation.

Anomalous Hall effect is sensitive to the magnetic property of metals[20] and can be used to investigate the magnetic behaviour of metals. It is interesting to note that the temperature-driven SRT in our Ni films is supported by the synchronous changes in magnetic susceptibility and extraordinary Hall signal (Fig.3).

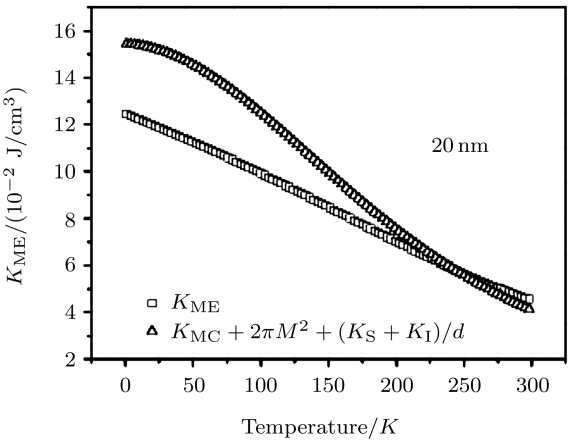


Fig.2. The magnetic anisotropy versus temperature for a 20 nm thick nickel film, where the values of K_{ME}, $K_{\text{S}} + K_{\text{I}}$ and K_{MC} are cited from Refs.[13] and [19] respectively.

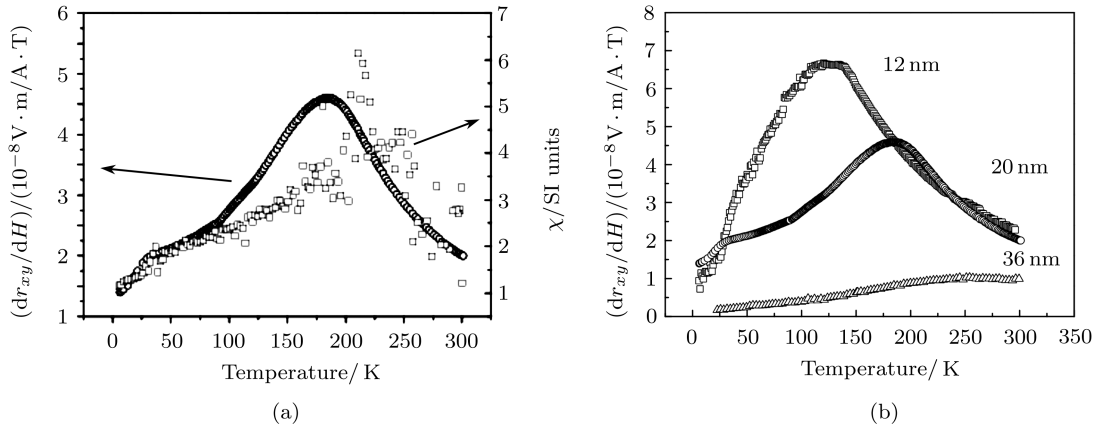


Fig.3. (a) The differential Hall resistivity and susceptibility versus temperature for a 20 nm thick Ni film, and (b) the differential Hall resistivity versus temperature for three Ni films with different thicknesses.

4. Conclusion

The temperature-driven spin reorientation transition has been found in the magnetron sputtered Ni films of thicknesses below 20 nm. The transition shifts toward lower temperatures with the decrease of thickness. The results accord with the plots where the competition between different magnetic anisotropic energies gives rise to this phenomenon. The transition is further supported by the synchronous changes in magnetic susceptibility and extraordinary Hall signal.

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