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TEMPERATURE DEPENDENCE OF ANISOTROPY IN Co/X(Pt, Au, Ag, Ni) MULTILAYER FILMS*

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Temperature dependences of effective perpendicular anisotropy (PA) K_u and interface energy K_s for Co/X(Pt, Au, Ag, Ni) multilayer films (MFs) as well as of spontaneous magnetization M_s for Co/Pt and Co/Au MFs are reported. The dependence of K_s on T distinguishes between Co/Au, Co/Ni and Co/Pt, Co/Ag MFs, i.e., K_s of the former decreases with the increase of T, but it increases for the latter. Co/Pt MFs with the perpendicular anisotropy possess a large enhancement of M_s at low temperature, but there is not any enhancement for Co/Au MFs. We have discussed the source of PA in Co/Au and Co/Pt MFs and speculated that the strain due to the lattice mismatch may be the main cause of PA in Co/Au MFs, but it is not for Co/Pt MFs.

PACC: 7570; 7530G

I. INTRODUCTION

The origination of perpendicular anisotropy (PA) has, continuously, been a very interesting problem since the Pd/Co multilayer film was reported in 1985.[1] For a decade a lot of papers have been published to discuss the source of PA in the Co-based MFs such as Co/X(Pt, Pd, Au, Ag, Ir, Ni)[2-11] that were usually prepared by deposition(sputtering) or epitaxy. The aim of such researches is apparent because these multilayers are promising materials for magneto-optical (MO) recording[12,13] and their properties differ from the bulk.

In general, the PA is attributed to the reduced symmetry at interface[14], the crystal anisotropy or the magnetoelastic effects.[15-21] Theoretical calculations of the surface anisotropy energy on monolayer and of the crystal anisotropy on MFs based on band structure have also been done for several systems. The surface and crystal anisotropy could result in a favored spin orientation in monolayer and MFs, respectively.[22-26]

Besides the PA behavior, the theoretical calculation indicated that there is an enhancement of the atomic moment for the monolayer of some transition metals.[27] Moreover, in the form of MFs the spin polarization of Pd, Pt, Ir and a very large enhancement of cobalt orbital moment in Pd/Co MFs have been reported as well.[28-29]

Although a lot of information on Co/X MFs has been presented, the temperature dependences of the perpendicular anisotropy and the interface energy as well as the spontaneous magnetization have been little concerned with.

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According to the measurements of the variations of K_u and K_s with temperature from 80–290 K for Co/X(Pt, Au, Ag, Ni) MFs as well as the temperature dependence of magnetization from 4.2 to 300 K for Co/Pt and Co/Au MFs, the origin of the perpendicular anisotropy of these MFs is discussed in this paper qualitatively.

II. EXPERIMENTAL

The samples Co/X(Pt, Au, Ag and Ni) MFs were prepared by sputtering at room temperature on silicon (111) substrate in Ar atmosphere at a pressure of about 3 mbar. The background pressure was better than 10^{-8} mbar. The thickness of each sublayer in MFs was controlled by a predetermined sputtering time via computer. In Co/X(Pt, Au, Ag) MFs, the Co thickness varied from 0.3 to 2.0 nm, while Pt, Au, Ag thicknesses were fiixed at 1.5, 5.0, 5.0 nm, respectively. For the Co/Ni MFs the Ni thickness varied from 0.4 to 2.5 nm and the Co thickness was fixed at 0.2 nm.

Small and large angle XRD with Cu-K_α radiation were used to analyze the crystal structure. By means of a magnetic torquemeter and a superconducting quantum interference device (SQUID) magnetometer, the effective magnetic anisotropy constant K_u and the saturation magnetization M_s were determined as functions of temperature.

III. RESULTS

Small and large angle X-ray diffractions (XRD) indicate that Co/X(Au, Pt, Ag, Ni) MFs possess a common composition-modulated character and have a fcc structure with a pronounced (111) texture orientated along the vertical direction, but their modulated characters are quite different. In Co/Pt MFs two small angle peaks for Co$\leq$0.7 nm or three for Co$\geq$0.9 nm can be seen, but in Co/Au MFs, eight small angle peaks can be seen althouth the Co thickness is just 0.5 nm. It means that the interface of Co/Au MFs is very sharp and immiscible. For the Co/Ni MFs, two small angle peaks just appear for 0.4 nm Ni/0.2 nm Co, while only one peak appears for larger Ni thickness. It specifies that the interface is relatively sharp only for the smaller Ni thickness. In Co/Ag MFs, two small angle peaks could be seen for all the samples. It means that the miscible interface of Co/Ag MFs is independent of the Co thickness.

The effective anisotropy constants K_u vs either the Co thickness for Co/X(Pt, Au, Ag) or the thickness of Co+Ni for Co/Ni MFs are given in Fig. 1, in which the data of Co/Pt and Co/Au MFs are quoted from our previous paper[30] for comparison with Co/Ag, Co/Ni MFs. We can see that Pt/Co MFs possess the largest perpendicular anisotropy, $K_u = 1.2 \times 10^7$ erg·cm^{-3}, and it appears at about one monlolayer of cobalt in period. For Co/Au MFs, K_{umax} is located at about 1.5 monolayer of cobalt, and its value, of about

5×10^6 erg·cm^{-3}, is just one half that of Co/Pt. $K_{umax} = 1 \times 10^6$ erg·cm^{-3} for Co/Ni MFs is the smallest among Co/X(Pt, Au, Ni) MFs and it is located at 0.7 nm thickness of Ni. Co/Ag MFs have negative anisotropy constant in the whole range of Co thickness.

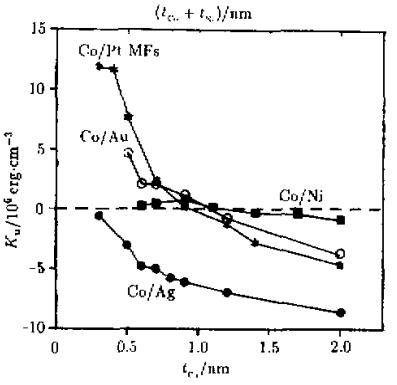

Fig. 1. The curves of K_u vs thicknesses of Co and Co+Ni for Co/pt, Co/Au, Co/Ag and Co/Ni MFs.

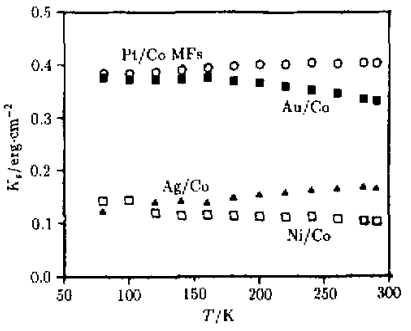

Fig. 2. The Variations of K_s as a function of temperature for Pt/Co, Au/Co, Ni/Co and Ag/Co MFs.

According to our measurements, the variations of K_u with temperature for Co/X(Pt, Au, Ag, Ni) MFs are quite different. K_u increases with the decrease of temperature for all thicknesses of cobalt or nickel in Co/Au and Co/Ni MFs, but $K_u(T)$ is dependent on the Co thickness in Co/Pt and Co/Ag MFs. With Co thickness less than 0.4 nm $K_u(T)$ of Co/Pt MFs is similar to that of Co/Au MFs, that is, K_u increases with the decrease of temperature, but it is smaller at low temperature than at high temperature when Co thickness is over 0.4 nm. In Co/Ag MFs, the minus K_u value is nearly independent of T when the thickness of Co is larger than 0.8 nm.

Sugimoto *et al.*[31] reported previously that Co/Au and Co/Pt MFs show an opposite dependence of K_u on temperature. However, they did not observe the dependence of K_u on T, which, we found, is closely related to the Co thickness in Co/Pt MFs.

Below, we can see that these different temperature dependences of K_u in Co/X(Pt, Au, Ag, Ni) MFs will affect the temperature dependence of the interface energy K_s.

In general, the K_u of the magnetic MFs can be expressed as follows:

$$K_u = 2K_s/t_{Co} + (K_v - 2\pi M_s^2) , \tag{1}$$

where t_{Co} is the thickness of the Co layer in Co/X(Pt, Au, Ag) MFs and it should be replaced by the thickness of Co plus Ni layers, i.e., the period thickness, for Co/Ni MFs; K_v and $2\pi M_s^2$ are the bulk anisotropy energy and demagnetization energy for the magnetic layer in MFs, respectively. By means of plots of K_u t_{Co} vs. t_{Co} or t_{Co+Ni}, K_s and $K_v - 2\pi M_s^2$

can be obtained. If the data of $K_u(T)$ are known, then the temperature dependence of the values of K_s and $(K_v - 2\pi M_s^2)$ can also be deduced.

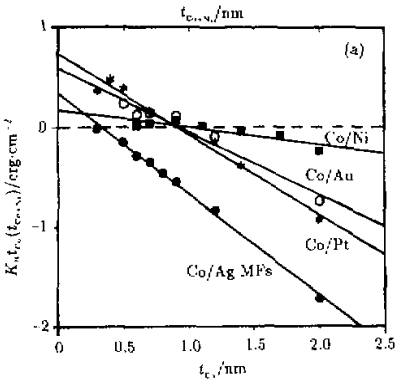

Fig. 3 (a) $K_u\, t_{Co}$ vs. t_{Co} or t_{Co+Ni} for Co/Pt, Co/Au, Co/Ag and Co/Ni MFs.

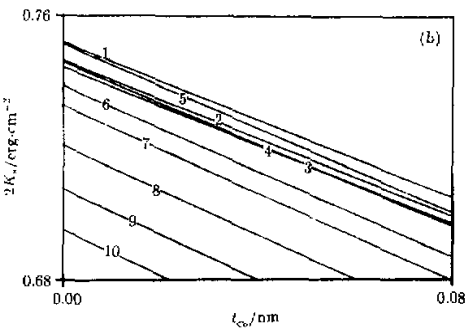

Fig. 3(b) $2K_s$ vs. t_{Co} of Co/Au MFs in the temperature range of 80 to 260 K. Curves 1 to 10 correspond to 80 to 260 K, respectively.

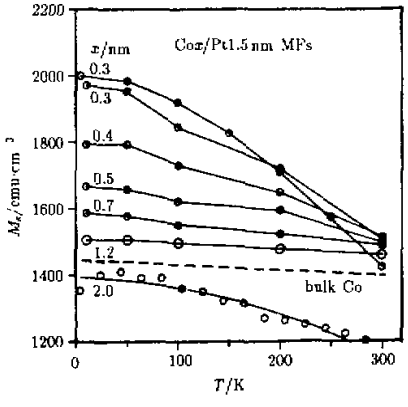

Fig. 4. $M_s(T)$ of Co/Pt MFs as the function of the variant Co layer thickness in Co/Pt MFs and for bulk cobalt.

Figure 2 shows the curves of K_s vs. T for the measured samples. At room temperature, $K_s = 0.42, 0.33, 0.16$ and $0.10\,\mathrm{erg\cdot cm^{-2}}$ for Co/Pt, Co/Au, Co/Ag and Co/Ni MFs, respectively. As indicated in Fig. 2, K_s decreases gradually with the decrease of T for Co/Pt and Co/Ag MFs, while it has an opposite temperature dependence for Co/Au and Co/Ni MFs. Moreover, the K_s for the Co/Pt and Co/Au MFs approaches to the same value when T decreases; Co/Ag and Co/Ni MFs have the same behavior.

In order to obtain the curves of K_s vs. T, as shown in Fig. 2, the plots of $K_u(T)\, t_{Co}$ vs. t_{Co} for Co/X(Pt, Au, Ag) MFs and $K_u(T)$ t_{Co+Ni} vs. t_{Co} for Co/Ni MFs were made. Figure 3 (a) gives such a plot at room temperature for Co/X(Pt, Au, Ag, Ni) MFs and Fig. 3 (b) shows, as an example, the plot of K_s vs. t_{Co} in the temperature range of 80–260 K for Co/Au MFs. It is noticed that the intercept on the vertical axis, as shown in Fig. 3 (a), is a measure of the quantity of $2K_s$.Figure. 4 shows the variation of the spontaneous magnetization M_s as a function of T from 4.2 to 300 K for Co/Pt MFs with different Co thicknesses (0.3, 0.4, 0.5, 0.7, 1.2 and 2.0 nm). We can see that M_s has not any apparent enhancement at room temperture for all Co thickness. The enhancement appears obviously only at low

temperature and is closely related to the Co thicknesses. The thinner the Co thickness, the larger the enhancement of M_s. Moreover, $M_s(T)$ of the Co/Pt MFs with Co thickness $=1.2$ nm approaches to that of the bulk. These behaviors are in good agreement with those previously reported on Ag/Co/Ag(100) epitaxial sandwich films.[32] However, we have not observed any M_s enhancement on Co/Au MFs sputtered on Al foil at low temperature.

IV. DISCUSSION

The strain model to explain the perpendicular anisotropy on Co/X(Au, Pd, Pt) MFs is based on the lattice mismatch between Co and X layers and a lattice mismatch of about 14%, 12% and 10% could occur in Co/Au, Co/Pd and Co/Pt MFs, respectively. If the magnetostriction coefficient of Co layer in MFs is minus, then the joint effect of a negative magnetostriction and a tensile force from the lattice mismatch on Co layer could produce a perpendicular anisotropy. However, PtCo alloy film[33] and Co/Ni MFs[26] with PA have been reported in recent years. The former has no lattice mismatch and the latter shows very small mismatch. Moreover, theoretical calculations[24,26] based on the band structure can also explain the source of PA in such Co/X MFs. Consequently, it is still a challenge whether the strain model is correct or a common model is needed to respond to the source of PA in those MFs.

As for the strain model, one can express the total energy in MFs as follows:

$$K_u = K_v + K_d + K_{me}(t_{Co}) , \tag{2}$$

where $K_v = K_1 + K_2$ is the crystalline energy term, K_d the demagnetization energy term, and K_{me} the term of magnetoelastic anisotropy obtained from the magnetoelastic energy $E_{me} = -(c\epsilon) \cdot \Lambda$, here c is the elasticity tensor. $\epsilon = (\epsilon_1, \epsilon_2, \epsilon_3)$ is the strain, and Λ is the magnetostriction coefficient of bulk cobalt. The resultant anisotropy for a (0001) hcp Co film is[18]

$$K_{me} = -[c_{11} + c_{12} - 2c_{13}^2/c_{33}](\Lambda_A + \Lambda_B)\epsilon(t_{Co}) . \tag{3}$$

In Eqs. (2) and (3) the interface energy K_s, which is usually taken into account for PA in MFs, does not appear. In order to correlate the third term in Eq. 2 with the interface energy K_s^{∞}, we rewrite Eq. (2) as

$$K_u = K_v + K_d + A_0\epsilon_0/t_{Co} , \tag{4}$$

$$A_0 = -(c_{11} + c_{12} - 2c_{13}^2/c_{33})(\Lambda_A + \Lambda_B) , \tag{5}$$

where ϵ_0 is the strain for unit cobalt thickness. Then, from the plot of $K_u t_{Co}$ vs t_{Co},

$$K_u t_{Co} = 2K_s + (K_v + K_d)t_{Co} , \tag{6}$$

$$K_s = (1/2)A_0\epsilon_0 \ , \tag{7}$$

we can obtain K_s originated from the magnetostrain energy and deduce ϵ_0 if the interface energy is known.

Assume that the elastic constants c_{ij} is independent of temperature, because its temperature coefficient is at least two orders of magnitude less than that of $\wedge_A$ or $\wedge_B$.[34,35] Inserting the values of c_{ij}, $\wedge_A(T)$ and $\wedge_B(T)$[34,35] into Eqs. (5) and (7) and adjusting $\epsilon_0(T)$ values in Eq. (7), and fitting the calculated values of $K_s(T)$ in Eq. (7) to the experimental values $K_s(T)$, as indicated in Fig. 2, for Co/Au and Co/Pt MFs, then we obtain the curves of ϵ_0 vs T, as shown in Fig. 5. From this figure we can see that ϵ_0 of Co/Au MFs increases with increasing T with $\delta\epsilon_0 = 5 \times 10^{-4}\,\mathrm{deg}^{-1}$; however, ϵ_0 of Co/Pt MFs increases more quickly than Co/Au MFs at a rate, $\delta\epsilon_0 = 2 \times 10^{-3}\,\mathrm{deg}^{-1}$, that is, four times as large as that of Co/Au MFs.

From any handbook of physical constants one can find the coefficients of the linear thermal expansion β of 13.7, 14.1 and 8.9 ($10^{-6}\,\mathrm{deg}^{-1}$) for Co, Au and Pt, respectively. The coefficients β for Co and Au are almost the same, but they are quite different for Co and Pt. One can express $\delta\epsilon_0 \approx (l/l_0)\delta l/\delta T = \beta$, here l_0 and l are the linear length before and after the thermal expansion. According to the β values given above, $\delta\epsilon_0\beta \approx 1.4 \times 10^{-5}\,\mathrm{deg}^{-1}$ for Co in Co/Au MFs, which could be compared to the fitting value $\delta\epsilon_0 \approx 5 \times 10^{-4}$. But $\delta\epsilon_0 \approx 2 \times 10^{-3}$ is much larger than the extimated value for Co in Co/Pt MFs.

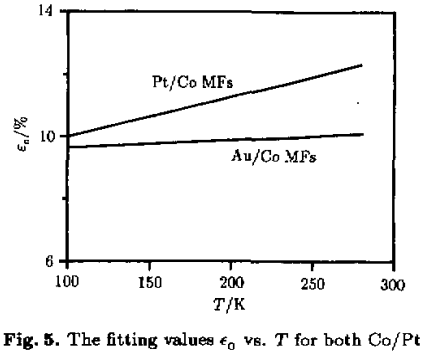

Fig. 5. The fitting values ϵ_0 vs. T for both Co/Pt and Co/Au MFs.

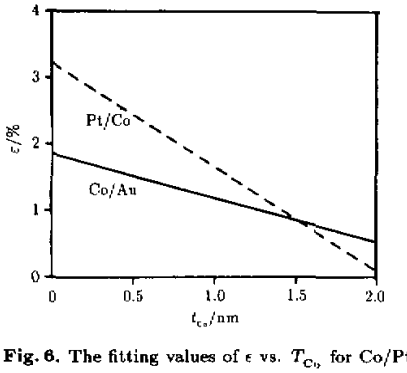

Fig. 6. The fitting values of ϵ vs. T_{Co} for Co/Pt, Co/Au MFs at room temperature.

Figure 6 shows the curves of ϵ vs. t_{Co} at room temperature for Co/Au and Co/Pt MFs, which are obtained from such a way that one should fit K_u obtained from Eq. (2) and Eq. (3) to the experimental curves of K_u vs. t_{Co} for both MFs, where the quantity $\epsilon(t_{Co})$ in Eq. (3) is a fitting parameter. It is apparent from this picture that at 1.5 monolayer Co in Co/Au MFs, $\epsilon \approx 1.6\%$ is in agreement with the recent result, measured by X-ray diffraction[17,18], but at one monolayer Co in Co/Pt MFs, $\epsilon \approx 3\%$ seems to be too large if we allow that its

lattice mismatch is smaller than that of Co/Au. Moreover, the interface of Co/Pt MFs is not as sharp as that of Co/Au, and the mixing interface of Co/Pt MFs will release strain. In view of the anisotropy magnitude, $K_{u\ max}$ of Co/Pt MFs is twice as large as $K_{u\ max}$ of Co/Au MFs, therefore the strain may only partially contribute to K_u for Co/Pt MFs, if it does.

By means of magnetic circular X-ray dichroism measurement[28], a great enhancement of Co orbital moment in Co/Pd MFs, compared with that in bulk Co, has been found. From our measurement of $M_s(T)$ for Co/Pt MFs shown in Fig. 4, a large enhancement of M_s of Co/Pt with thin Co layer at low temperature is visible. The cobalt atomic moment σ is about $2.35\mu_B$ for 0.3 nm Co/1.5 nm Pt MFs at 4.2 K, which is close to the theoretical value $2.4\mu_B$ on an unsupported monolayer of Co(001) with Ag lattice parameter.[32] But it is a little larger than the previous measurement ($\approx 2\mu_B$) on 1 to 2 monolayer Co at 5 K in Ag/Co/Ag(001) epitaxial sandwich structure.[32] If we assume that an enhancement of Co orbital moment in Co/Pt MFs appears, we should consider the contribution of the partially unquenched orbital moment to the anisotropy because the crystal field with D_{3d} point group coupled with the unquenched orbital moment would result in a preferential orientation of spin. However, it has not been considered in recent publications for the explanation of the perpendicular anisotropy in Co/Pd and Co/Pt MFs. For Co/Au MFs, we have not found any enhancement of M_s, as in the previous publication.[31] It may be the reason why the source of the anisotropy of Co/Au MFs differs from that of Co/X(Pd, Pt) MFs.

It is well known that the film with the perpendicular anisotropy must have the (111) crystal texture in both MFs and alloy films. In fact, the previous calculations[24,26] on the basis of the quadric perturbation theory pointed out that [111] axis perpendicular to the film plane is very important for the appearance of PA in MFs. Consequently, the coupling of spin with orbit originating from the partially unquenched Co orbital moment may be another source responding to the PA in Co/X(Pd, Pt) MFs. Recently, the in-situ measurement of stress for Co/Pd MFs indicates that Co layer is affected by compressive stress[16] at the beginning. Obviously, it does not meet the requirement of tensile force on Co layer. Therefore, the strain model is not a common one to explain the perpendicular anisotropy in Co/X MFs.

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