



Effect of magnetic alloying on magnetic anisotropy in ultrathin fcc Ni-like films

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Abstract

For identifying the magnetic alloying effect on the magnetic anisotropy in the magnetic ultrathin films, the spin-reorientation transition was studied by preparing the ultrathin $\text{Fe}_x\text{Ni}_{1-x}$ and $\text{Co}_x\text{Ni}_{1-x}$ alloy films on Cu(100) with variations of coverage and alloy concentration x . The comparison between $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ and $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$ shows that the modification of the critical thickness for the spin-reorientation transition by the alloy concentration x for Fe is 1.35 times larger than modification by Co. The evolution of spin-reorientation transition in this Ni-dominant films could be mainly traced back into the local behavior of d-electrons in impurities and host element, corrected with the charge transfer of d-electrons between them.

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1. Introduction

The magnetic properties of the 3d-transition metal ultrathin films are strongly dependent on the d-band characteristics of the films, such as

band filling, band dispersion, and exchange splitting. This is not only because of the complicated 3d-band structures around Fermi energy with substantial density of states, but also because of the critical evolution of the band filling difference between the majority (n_\uparrow) and minority (n_\downarrow) bands. In alloy films, there exists an interplay between local behavior and itinerant properties of d-electrons, such as charge transfer between various elements due to different electronegativities. Thus, it is more difficult to clarify how the band shape

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and band filling influence the magnetic properties in alloy films. Theoretically, the band filling was often treated as a parameter with continuous variation of electron numbers. Many efforts were made on the study of the correlation between band structures and magnetic properties, such as the magnetic moment and anisotropy, as a function of d-electron number (n_d) by the first-principle calculation [1–7]. Different from the magnetic moment which follows the Slater–Pauling curve with nearly linear dependence on n_d , the magnetic anisotropy, however, exhibits more complicated behavior as a function of n_d . Either rapid oscillations [1,3] or slow variation [2,7] showed a non-monotonic n_d dependence of anisotropy.

In an alloy film, the added magnetic element, however, should not only result in change of the d-band filling, but also affect the band width or dispersion. The question arises herein is how the magnetic alloying manipulates the electronic structure in the alloy system. One could either consider it as an artificial system with a band structure overall determined by the averaged electron numbers from the alloy composition, or must go into the details of the alloying effect on the electronic states of each element involved, such as local density state, band shape, and exchange splitting. A strong effect, which is relevant to magnetotransport properties, of magnetic impurities from Fe and Cr on valence band of the Ni host has been observed by means of angle-resolved photoemission [8]. The strong variation of these effects suggest the concept of the magnetic alloying rather than an averaging band behavior or band filling determined by the corresponding electron number.

In this article, we report on a detailed measurement of the alloying effect on spin-reorientation transition (SRT) as well as magnetic anisotropy in fcc Ni-like films. Comparing the results from different magnetic impurities (Fe and Co), the evolution of the magnetic anisotropy at variation of alloy concentration can be well understood by merely considering a Ni-like band shape and a local exchange splitting determined by the magnetic moment of added element, entangled with the interplay of the local density of states and charge transfer from added and host elements.

2. Experiment

The magnetic alloy ultrathin films were prepared and investigated in situ in an ultrahigh vacuum (UHV) chamber [9]. To deposit the films with desired and subtle-varied alloy composition is relevant to the study on the drastic variation of critical thickness for SRT. The coverage and alloy composition of the alloy films in our experiments can be precisely controlled in the co-deposition technique to an accuracy of 0.05 ML and $\pm 0.5\%$, respectively [10,11]. Furthermore, Auger electron spectroscopy (AES) was employed for studying the chemical structure and for double check of alloy composition of the alloy films [10,12]. The medium energy electron diffraction (MEED) was taken for calibration of the film coverage as well as for providing an information of the morphology of the film surface [13,14]. To identify the structural properties of the films, the crystalline structure and interlayer distance of the alloy films were performed via the low energy electron diffraction (LEED) and LEED $I(E)$ in the kinematic approximation [15–18]. In addition, the study of the magnetic hysteresis loops was carried out by means of magneto-optical Kerr effect (MOKE) in polar and longitudinal configurations *quasi-simultaneously* with benefit of the lock-in technique to investigate the evolution of SRT with variation of alloy concentration of the films.

3. Results

Several characteristics could be altered while changing alloy composition of the films: crystalline structure, morphology, and electronic structure. Each of them will influence the magnetic behavior. It becomes crucial to confirm the structural invariance on the variation of the alloy composition. In the previous study, we found that the crystalline structure, lattice constant, as well as morphology of $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$ and $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ are invariant with alloy composition [10,11,9]. In addition, all these alloy films behave like the Ni/Cu(100) film. It implies that these alloy films are the Ni/Cu(100)-like systems at least within the composition range of $x \leq 8\%$, indicating that the

structure of these alloy films are Ni-dominant and varied insignificantly within the alloy composition in our study. These structural evidences allow us to attribute the change in the magnetic properties to the effect of electronic structures mentioned above.

To observe how SRT evolves with alloy concentration is crucial to characterize the alloy-induced magnetic behavior of the film. The critical thickness (d_c) of SRT, which was identified as the thickness where the magnetic easy axis transfers from in-plane to out-of-plane orientation, is a good characteristic to describe the evolution of SRT. The magnetic easy axis was identified by taking the hysteresis loops at 110 K with the applied field perpendicular and parallel to the film surface. As illustrated in Fig. 1 (open and solid circles), d_c of SRT for $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ varies from 7.5 ML for $x = 0\%$ [pure $\text{Ni}/\text{Cu}(100)$] to 16 ML for $x = 5\%$. No SRT was found for $x > 6\%$ with the coverage up to 20 ML. In comparison with $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$ [10] (open and solid triangles in Fig. 1), d_c of the $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ alloy film is more sen-

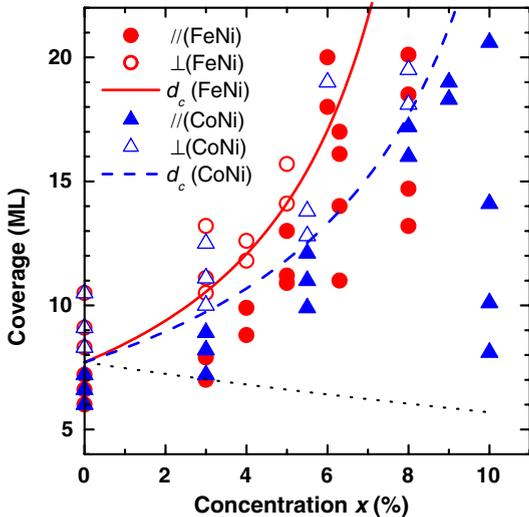


Fig. 1. Comparison of the magnetic phase diagrams of magnetic easy axis for $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ and $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$. The dotted curve represents the evaluation of d_c for $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ similar to Eq. (1) by linear superposition of magnetic moments and anisotropies of $\text{Fe}/\text{Cu}(100)$ and $\text{Ni}/\text{Cu}(100)$. The solid and dashed curves show the evaluation of d_c for $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ and $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$, respectively, in terms of Eq. (3) by considering the magnetostriction coefficient as a function of alloy concentration x in Ni.

sitive to the content of Fe. That is, the less Fe is needed to achieve the same d_c when alloys with Ni, as shown in Fig. 1.

The critical thickness in $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$ can be well evaluated by fitting magnetic moments and anisotropies of $\text{Co}/\text{Cu}(100)$ and $\text{Ni}/\text{Cu}(100)$ as [10]

$$d_c = \frac{-2[xK_s^{\text{Co}} + (1-x)K_s^{\text{Ni}}]}{[xK_v^{\text{Co}} + (1-x)K_v^{\text{Ni}}] - 2\pi[xM^{\text{Co}} + (1-x)M^{\text{Ni}}]^2} \quad (1)$$

by assuming linear variation of the magnetic moments and anisotropies between $\text{Co}/\text{Cu}(100)$ and $\text{Ni}/\text{Cu}(100)$. However, the estimation of d_c similar to Eq. (1) by linearly superposing the magnetic moments and anisotropies of $\text{Fe}/\text{Cu}(100)$ and $\text{Ni}/\text{Cu}(100)$ (listed in Table 1) makes large discrepancy from our experiments, as depicted by dotted line in Fig. 1. The failure of the linear model may result from the fact that $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ reveals “compressed” distortion along the surface normal while $\text{Fe}/\text{Cu}(100)$ is tetragonal “expanded”. It may result in different signs between them. Therefore, K_v adopted for $\text{Fe}/\text{Cu}(100)$ could fail to describe Fe in $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$. On the other hand, it is possible that K_v of Fe in $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ is still positive but the linear model does not work any more. Since K_v of Ni and Fe is positive a linear model will also yield a positive value for all concentrations. But due to the different structure (expanded versus compressed) of the Fe and Ni this linear model is flawed. However, many problems may be encountered for the argument of K_v for $\text{Fe}/\text{Cu}(100)$. This debate remains unsolved till now. Moreover, it would be also a good point to include further data of $\text{Fe}/\text{Cu}(100)$ [19]. Adopting $K_v = 77.7 \mu\text{eV}/\text{atom}$

Table 1
Magnetic moments and anisotropies of Fe and Ni

Element	M (μ_B)	K_s ($\mu\text{eV}/\text{atom}$)	K_v ($\mu\text{eV}/\text{atom}$)
Fe	2.5 ^a	115.8 ^b	103.6 ^b
Ni	0.57 ^c	-77 ^d	29 ^d

^a Ref. [6].

^b Ref. [26].

^c Ref. [25].

^d Ref. [21].

and $K_s = 120 \mu\text{eV}/\text{atom}$ for Fe/Cu(100) by Platow et al. [19], we got another descending curve similar to the dotted line in Fig. 3 but with different slope. For example, d_c varies from 6.6 ML to 7.0 ML for $x = 5\%$. It still fails for the description of Fe–Ni alloy.

One may doubt whether the magnetic anisotropy for Fe/Cu(100) is temperature-dependent variable and affect the tricky fitting of phase diagram for FeNi alloy. Another detailed study [20] on more reliable temperature-dependent anisotropy for Fe/Cu(100) provides a good opportunity to clarify this point. For the anisotropy of Fe/Cu(100) taken at about 110 K, the measurement temperature in our experiments, the fitting of phase diagram is a little closer to the experimental one than the other fitting. However, the value of critical thickness is still decreasing with variation of Fe composition x . This descending tendency will be even enhanced if one adopts the anisotropy of Fe/Cu(100) at higher temperature. Obviously, although taking the temperature-dependent anisotropy constants for fcc Fe/Cu(100) from the previous study (Ref. [20]) can give a temperature-dependence of the critical thickness, it still failed to describe the experimental results of evolution of the critical thickness at variation of the alloy composition. Thus, the FeNi alloy ultrathin films in our experiments can not be just simply treated as a linear combination of fcc Fe and Ni films on Cu(100) substrate even taking the temperature-dependence of the anisotropy for fcc Fe into account.

The previous study indicated that the SRT for Ni/Cu(100) is mainly attributed to the volume-type magnetoelastic anisotropy (K_v^{Ni}) [21]. It can be expressed as

$$K_v^{\text{Ni}} = -\frac{3}{2}(c_{11} - c_{12})\lambda_{100}(\varepsilon_1 - \varepsilon_2), \quad (2)$$

where c_{11} and c_{12} are elastic stiffness constants, ε_1 and ε_2 represent the strain and the tetragonal distortion, respectively; λ_{100} denotes the magnetostriction coefficient along the [100] direction. Apparently K_v^{Ni} depends on the strain of the film and magnetostriction coefficient λ_{100} . As mentioned in our previous study [9], the strain of $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ keeps almost invariant for dif-

ferent compositions x . By contrast, the magnetostriction coefficient (λ_{100}) is greatly sensitive to the alloy composition x . It has been shown, by first principle calculation [7], to be strongly dependent on the minority band filling n_d , which is proportional to the alloy concentration x in Ni. Therefore the magnetoelastic anisotropy varies dramatically upon variation of alloy concentration. The evaluation of d_c can thus be implemented as

$$d_c(n_d) = \frac{-2K_s^{\text{Ni}}}{K_v^{\text{Ni}}(n_d) - 2\pi M_{\text{Ni}}^2(n_d)} \quad (3)$$

by approximation of linear dependence of magnetoelastic anisotropy (K_v^{Ni}) as well as magnetic moment (M^{Ni}) on 3d-band filling n_d (Ref. [7]) and, in turn, on the alloy concentration x in $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$. Noted that the surface magnetic anisotropy K_s^{Ni} in Eq. (3) was assumed to be invariant with alloy concentration, which is compatible with the previous study [22]. The solid curve illustrated in Fig. 1 represents the evaluated d_c as a function of alloy concentration x from Eq. (3), where $M^{\text{Ni}}(n_d)$ is taken by superposition of magnetic moments for Fe and Ni listed in Table 1. Apparently, the trend of d_c evaluated by this method agrees very well with the experimental results. It should be emphasized that within the low Fe content limit ($x \leq 8\%$), the magnetic anisotropies of $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ can not be oversimplified as a linear combination of those of Fe/Cu(100) and Ni/Cu(100). Iron in the $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ films behaves like an electron regulator to modify the band characteristics of the Ni/Cu(100) films. Besides, our simulation of phase boundary is very similar to that by Thamankar et al. [22] where the magneto-elastic properties of bulk Fe–Ni was taken into account to derive the value of d_c . This is mainly because that both simulations was based on the same tendency for magnetostriction coefficient λ_{100} of Ni while alloying with Fe.

4. Discussion

It is reasonable to expect the stronger modification of SRT by Fe than that by cobalt when these two elements are alloyed into Ni films since the

difference of d-electron numbers between Fe and Ni is twice of that between Co and Ni. It agrees with our result, as shown in Fig. 1. Furthermore, for the same critical thickness d_c , the concentration of Co (x_{Co}) is about 1.35 times of the concentration of Fe (x_{Fe}), as shown in Fig. 1. It means that modification of d_c for SRT by the alloy concentration x for Fe is 1.35 times larger than modification by Co. On the other hand, according to Eq. (3) there exists a pole in the $d_c(x)$ curve for $K_v - 2\pi M^2 = 0$. This pole describes the transition from Ni-type SRT to Fe-type SRT or Co-type films (no SRT). By comparing the pole positions between $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ and $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$, a factor of 1.32 between the concentration of Co and Fe in alloy can be obtained. This is very close to 1.35 in the above evaluation. Not that magnetic moment could only affect the pole position a minor shift and change the factor from 1.32 to 1.37 if we take the magnetic moment of Fe from $2.5\mu_B$ to $3.0\mu_B$. The factor of about 1.35 can mainly be attributed to the facts as follows: First, the distributions of electrons in metals iron, cobalt, and nickel are different from those in free atoms. For example, there are five spin-up and one spin-down d-electrons for a free iron atom such that there is no spin-up hole in the d-orbit of iron atom. However, there exists an incompletely filled majority-spin band for the metal iron. Second, what affects d_c of SRT in $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$ and $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ is likely to be the difference of filling numbers Δn_d between the majority and minority bands, rather than the total number of d-electrons n_d . The magnetic anisotropies which influence d_c of SRT originate from the interplay between magnetic moment and spin-orbit interaction. Each of them is correlated to the Δn_d in the films. The average Δn_d of nickel with iron composition will then be suppressed due to the vacancy of majority band in iron [23]. A simple estimation of d_c modification by alloy concentration of iron (x_{Fe}) and cobalt (x_{Co}) can therefore be made by considering the differences of the filling numbers for majority and minority bands in iron, cobalt, and nickel. For the same d_c , the relation of the concentration x_{Fe} in Fe–Ni alloy and x_{Co} in Co–Ni alloy was expressed, by considering the averaged d-band asymmetry $\Delta n_d^{\text{alloy}}$, as

$$\begin{aligned}\Delta n_d^{\text{alloy}} &= \Delta n_d^{\text{Fe}} x_{\text{Fe}} + \Delta n_d^{\text{Ni}} (1 - x_{\text{Fe}}) \\ &= \Delta n_d^{\text{Co}} x_{\text{Co}} + \Delta n_d^{\text{Ni}} (1 - x_{\text{Co}}),\end{aligned}\quad (4)$$

where Δn_d^{Fe} , Δn_d^{Co} , and Δn_d^{Ni} represent the difference of majority and minority bands for iron, cobalt, and nickel, respectively. Note that Δn_d is similar to the “magnetic moment” if the orbital moment is not taken into consideration. For the transition metals, the orbital moment is insignificant although it should be enhanced at the surface. Thus, Δn_d could be analogous to the magnetic moment for a rough estimate. The ratio of the alloy concentration $x_{\text{Co}}/x_{\text{Fe}}$, which is equivalent to the relative contribution of the alloy concentration Fe on d_c in comparison with Co, can be simplified as

$$\frac{x_{\text{Co}}}{x_{\text{Fe}}} = \frac{\Delta n_d^{\text{Fe}} - \Delta n_d^{\text{Ni}}}{\Delta n_d^{\text{Co}} - \Delta n_d^{\text{Ni}}}.\quad (5)$$

In order to estimate the $x_{\text{Co}}/x_{\text{Fe}}$ ratio in terms of Δn_d^{Fe} , Δn_d^{Co} , and Δn_d^{Ni} , we adopted the filling numbers for majority and minority bands of Co and Fe by modifying the spin-dependent density of states for fcc Ni (Ref. [24]) with the spin splitting (equivalent to Δn_d) proportional to magnetic moment in accordance with the Stoner theory [23]. Meanwhile the band shapes for added Fe and Co were kept invariant with that for host element Ni, as illustrated in Fig. 2. This assumption is reasonable since the band shape with tiny alloy concentration is mainly determined by periodically neighboring

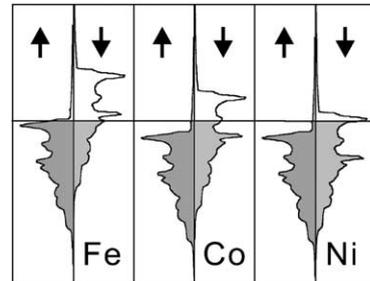


Fig. 2. Density of states for majority and minority bands in Fe, Co, and Ni. The densities of states for iron and cobalt were evaluated by taking the band shape of fcc nickel [24] modified with the spin splitting proportional to magnetic moment in accordance with the Stoner theory [23].

Table 2

3d-Band filling numbers (n_d) and densities of states at Fermi level [$N_d(E_F)$] for transition metal Fe, Co, and Ni. The n_d 's as well as $N_d(E_F)$'s of iron and cobalt were evaluated by modifying the spin-dependent density of states of fcc nickel [24] with the spin splitting proportional to magnetic moment in accordance with the Stoner theory [23], as illustrated in Fig. 2

	Majority (\uparrow)		Minority (\downarrow)		Asymmetry ($\uparrow - \downarrow$)	
	n_d	$N_d(E_F)$	n_d	$N_d(E_F)$	Δn_d	$\Delta N_d(E_F)$
Fe	4.614	1.020	2.109	1.290	2.505	-0.270
Co	4.756	0.131	2.951	1.110	1.805	-0.979
Ni	4.756	0.131	4.105	1.977	0.651	-1.846

host elements. Table 2 listed the spin-dependent band filling numbers (Δn_d 's) as well as the densities of states at Fermi level ($\Delta N_d(E_F)$'s) for Fe, Co, and Ni. Using the estimated values in Table 2, x_{Co}/x_{Fe} can be evaluated as 1.61 which is much less than two, the expected value from the d-electrons of free atoms. That is, 1.61 times of Co content is need as compared with Fe for achieving the same d_c under our d-band asymmetry consideration. It means that Δn_d rather than n_d should be the principal factor to modify the magnetic anisotropies and, in turn, to affect d_c in the films. Furthermore, if the charge transfer δ between nickel and iron or cobalt is taken into consideration [23], the effect of alloy content for iron and cobalt on d_c expressed in Eq. (5) can be modified as

$$\frac{x_{Co}}{x_{Fe}} = \frac{[\Delta n_d^{Fe} - \Delta N_d^{Fe}(E_F)\delta] - [\Delta n_d^{Ni} + \Delta N_d^{Ni}(E_F)\delta]}{[\Delta n_d^{Co} - \Delta N_d^{Co}(E_F)\delta] - [\Delta n_d^{Ni} + \Delta N_d^{Ni}(E_F)\delta]}, \quad (6)$$

where $\Delta N_d^{Fe(Co,Ni)}(E_F)$ represents the difference of spin density of states at Fermi energy for Fe(Co, Ni), as listed in Table 2. Due to almost the same electronegativity of iron (1.83), cobalt (1.88), and nickel (1.89), the charge transfer between the two elements alloyed in films is insignificant for $Co_xNi_{1-x}/Cu(100)$ and $Fe_xNi_{1-x}/Cu(100)$. However, the densities of states at Fermi energy could be significantly different for iron, cobalt, and nickel. Therefore, even a little charge transfer will strongly alter the spin asymmetry Δn_d and result in significant modification of x_{Co}/x_{Fe} ratio. By adopting the values listed in Table 2, x_{Co}/x_{Fe} was approximated as $1.61 - 2.1\delta$. If there exists, for example, a charge transfer of about 0.1 electron [23], x_{Co}/x_{Fe} could be modified as 1.40 which is closer to our experimental result.

It is noted that the averaged d-band asymmetry Δn_d considered to evaluate d_c should be traced back to the effect of local behavior of d-electrons in each element of alloy. Nevertheless, the consideration of charge transfer in Eq. (6) reflects the itinerant behavior of d-electrons just like the overall d-band filling effect in alloy films. In addition, there could be no universal behavior for the d_c variation on d-band filling for overall alloy compositions since there could be more complicated evolution including not only electronic but also structural variations with x . Furthermore, the physical origin of the magnetic anisotropy should be the spin-orbit coupling in the materials. In the non-relativistic limit, the spin-orbit coupling is proportional to the spin moment \mathbf{S} and orbital moment \mathbf{L} , with some spin-orbit coefficient which is structural dependent. If the structures are kept invariant, the orbital moment as well as spin-orbit coefficient should be also unchange. Therefore, the evolution of magnetic anisotropy could be conducted in a good approximation with the magnetic moment under this situation, which is described by Δn_d in our analysis. This could be hold, in particular for the very low concentration x in our experiments. In stead of dealing with the anisotropy directly, the band filling asymmetry, Δn_d was evaluated in our analysis.

5. Summary

As well known, the magnetic anisotropy is really difficult to quantitatively study for both theoretically and experimentally approach due to its several order of magnitude less than magnetic moment. However, by observing the evolution of the critical

thickness for spin-reorientation transition, it is possible to analyze the evolution of magnetic anisotropy quantitatively. Also, by means of the successfully theoretical approach [7], the rigid band approximation could be checked in comparison with the experimental data. The spin-reorientation transition for ultrathin films $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ was investigated to study how the magnetic behaviors are modified by the alloy composition. The composition-driven spin-reorientation transitions for both $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ and $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$ are shown to be attributed to the effect of alloy concentration on the magnetostriction coefficient and, in turn, on the magnetoelastic anisotropy of the $\text{Ni}/\text{Cu}(100)$ -dominant films. Based on the experimental results, the asymmetry of the 3d electron numbers between the majority and minority bands was also successfully applied to evaluate the contribution of iron on the critical thickness of spin-reorientation transition in the $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ films as compared with that of cobalt in the $\text{Co}_x\text{Ni}_{1-x}/\text{Cu}(100)$ films. It reveals that the band asymmetry is the key factor to affect the magnetic anisotropies of the films. Additionally, a small charge transfer between the magnetic impurities and host element Ni was also taken into consideration as the further correction to evaluate the contribution of different impurities on the critical thickness of spin-reorientation transition in Ni-like films. These evaluations might help us to clarify the interplay between local and itinerant behaviors of d-electrons in alloy films under ultrathin limit.

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