Comparative study of the *d*-band filling effect on the magnetic behavior of Co_xNi_{1-x} and Fe_xNi_{1-x} ultrathin films on Cu(100)

C. C. Kuo, S. F. Chuang, W. Pan, W. C. Lin, and Minn-Tsong Lin[a)]
Department of Physics, National Taiwan University, Taipei 106, Taiwan

The ultrathin Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) alloy films with $x \leqslant 10\%$ were prepared for the study of spin-reorientation transition with precise variation of composition and coverage of the films. The spin-reorientation transition with the variation of the film coverage was observed in Co_xNi_{1-x}/Cu(100) with $x < 10\%$ as well as Fe_xNi_{1-x}/Cu(100) with $x < 6\%$. This transition was found to be strongly affected by the alloy composition of the alloy films. The critical thickness for the spin-reorientation transition changes from 7.5 ML with $x = 0$ to 17.5 ML with $x = 8\%$ for Co_xNi_{1-x}/Cu(100). A more sensitive composition-driven effect has been found in Fe_xNi_{1-x}/Cu(100) by varying the Fe concentration for the critical thickness to 16 ML with $x = 5\%$. The more drastic influence of the concentration of Fe than Co on the critical thickness of the spin-reorientation transition can be attributed to the greater deviation of the difference in $3d$ electron number of majority and minority bands between Fe and Ni than that between Co and Ni. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1456401]

Numerous attempts have been made to reconfigure the easy axis, or so-called the spin-reorientation transition (SRT), of the magnetic ultrathin films. It has been shown that the SRT can be manipulated not only by the coverage,[1–3] but also by the temperature[4–6] of the films. In addition, the composition of alloy ultrathin films is another factor determining the SRT if two composite elements prefer different easy axes. The alloy composition-driven SRT has been successfully demonstrated for the Fe_xCo_{1-x}/Cu(100)[7] and Co_xNi_{1-x}/Cu(100)[8] systems. In both of these two alloy ultrathin films, the morphology, crystalline structure, and interlayer distance keep invariant in the range of the alloy composition where the SRT is observed. The alloy composition can thus be treated as a variable that modifies the $3d$-band filling for the films. As a consequence, the spin-reorientation transition found in the Fe–Co and Co–Ni alloy ultrathin films is attributed to the evolution of the $3d$-band filling in these systems.[7,8] It provides a venue to investigate the effect of $3d$-band filling on the magnetic characteristics such as the magnetic anisotropy and, in turn, the spin-reorientation transition of the films.

In particular, there is good evidence to show a critical evolution of *inverse* SRT similar to Ni/Cu(100) for Co_xNi_{1-x}/Cu(100) alloy films driven by the subtle variation of alloy composition x.[8] The critical thickness of the SRT was changed drastically from 7.5 to 17.5 ML for x varying from 0% to 8%. This system can be identified as a Ni/Cu(100)-like system since the deviation of the crystalline structure between Ni/Cu(100) and these alloy films is insignificant. The alloy composition in this system can therefore be treated as a variation of the effective filling of the $3d$-band electrons for Ni/Cu(100) films since Co is the element adjoining Ni with the $3d$-electron number decreasing by one. In addition, the difference in number of $3d$-electrons between Fe and Ni is twice as large as that between Co and Ni. An interesting question then arises: Will a more drastic variation of critical thickness for SRT be found than Co_xNi_{1-x}/Cu(100) films if Fe is substituted for Co? In addition, the complicated transitions of the structural and magnetic anisotropy in Fe/Cu(100) films[9] may be involved in the alloy films during the range of the composition in which the SRT is observed. Therefore, it is imperative to conduct a comparative study of the correlation between structure and magnetic properties for Co_xNi_{1-x}/Cu(100) and Fe_xNi_{1-x}/Cu(100) films.

In this work, a comparative study, including the structural and magnetic properties, of the magnetic ultrathin Co_xNi_{1-x}/Cu(100) and Fe_xNi_{1-x}/Cu(100) alloy films were performed. The alloy composition-driven SRT were found in both of these two alloy films. In addition, the contribution of the concentration of Fe and Co on the critical thickness of the SRT was also estimated and discussed.

The magnetic alloy ultrathin films were prepared and investigated *in situ* in an UHV chamber.[10] The coverage and alloy composition of the alloy films in our experiments can be precisely controlled to an accuracy of 0.05 ML ±0.5%, respectively.[11] The crystalline structure as well as interlayer distance of the alloy films were performed via the low energy electron diffraction (LEED) and LEED $I(E)$ in the kinematic approximation.[3] Auger electron spectroscopy was also employed as the tool for studying the chemical structure of the alloy films. In addition, the study of the magnetic hysteresis loops was carried out by means of magneto-optical Kerr effect in polar and longitudinal configurations *quasi-simultaneously* with benefit of the modulation and lock-in technique to investigate the evolution of magnetic structures, such as spin-reorientation transition.

The medium energy electron diffraction (MEED) intensities of Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) alloy ultrathin films for all the alloy compositions ($x \leqslant 10\%$) exhibit the regular oscillations up to four layers, as shown in

a)Author to whom correspondence should be addressed; electronic mail: mtlin@phys.ntu.edu.tw

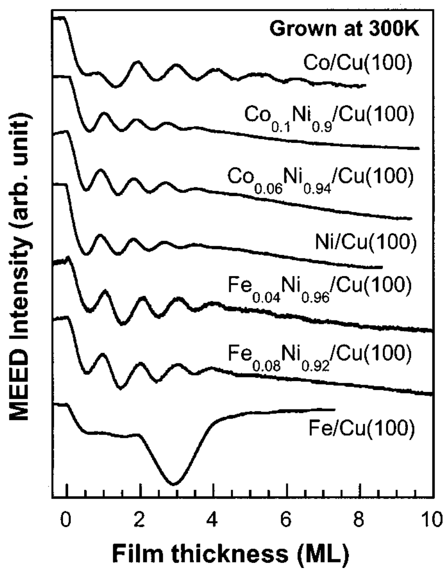

FIG. 1. MEED intensity oscillations of Co/Cu(100), Ni/Cu(100), Fe/Cu(100) and alloy ultrathin films with different compositions. All the films were prepared at 300 K.

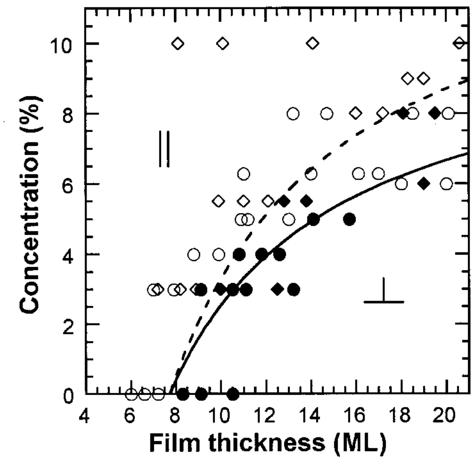

FIG. 3. Comparison of the magnetic phase diagrams for Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) alloy films. The solid circles (●), hollow circles (○), solid diamonds (♦), and hollow diamonds (◇) represent the out-of-plane and in-plane easy axes for the Fe_xNi_{1-x}/Cu(100) films as well as the out-of-plane and in-plane easy axes for the Co_xNi_{1-x}/Cu(100) films, respectively. The phase boundary of SRT for Fe–Ni and Co–Ni alloy are illustrated by solid and dashed curves, respectively.

Fig. 1. It is obvious that the characteristic in MEED oscillations of the alloy films look fairly similar to that of the pure Ni/Cu(100). Nevertheless, there are significant deviations of MEED characteristic between the alloy films and pure Fe/Cu(100) or Co/Cu(100) films. It implies both of these alloy films are the Ni/Cu(100)-like systems within the composition range of our experiments. Similar results can also be found in the LEED patterns for Ni/Cu(100), Fe_xNi_{1-x}/Cu(100), and Co_xNi_{1-x}/Cu(100) films. The LEED patterns in the Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) films both exhibit the crossing streaks on the LEED spots, just as the Ni/Cu(100) films do.[11] This result agrees well with the previous scanning tunnel microscope study in Ni/Cu(100).[12] The 3D platelike islands from the additional periodic structure in the [011] and $[0\bar{1}1]$ directions account for the crossing streaks along these two directions in the LEED patterns.

The average vertical interlayer spacing ($a_\perp$) for both the Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) alloy ultrathin films determined by means of LEED $I(E)$ measurement are compiled in Fig. 2. It is obvious that $a_\perp$ for all the alloy composition in both alloy films are almost the same within an accuracy of ± 0.02 Å. Different from the Fe/Cu(100) films, which exhibit a structure transition from fcc to bcc structure,[9] there is no structure variation or lattice relaxation for the alloy films for all the alloy compositions and thickness investigated. In addition, the vertical interlayer spacing for the alloy films are fairly close to the pure Ni/Cu(100) films, as shown in Fig. 2. It again confirms our assumption that both Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) films are Ni-dominant systems behaving like the Ni/Cu(100) films.

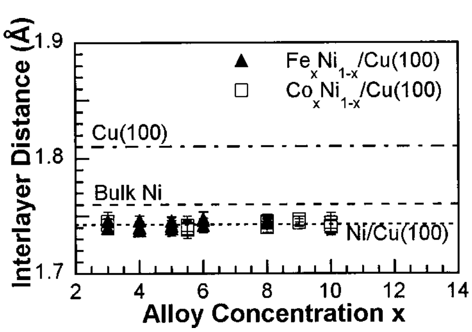

FIG. 2. Interlayer distance of Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) alloy ultrathin films with variation of alloy composition.

As mentioned, the magnetic properties of the alloy films were characterized by the hysteresis loops in terms of Kerr intensities. The magnetic hysteresis loops of the alloy films Fe_xNi_{1-x}/Cu(100) for $x=3\%$, 4%, 5%, 6%, and 8% as well as Co_xNi_{1-x}/Cu(100) for $x=3\%$, 5.5%, 8%, 9%, and 10% were investigated at 110 K with variation of the film thickness. The easy axis of these films is identified by both the polar and longitudinal geometries of hysteresis loops. For Co_xNi_{1-x}/Cu(100) films, the critical thickness of the SRT varies from 7.5 ML for $x=0$ [pure Ni/Cu(100)] to 17.5 ML for $x=8\%$. There is no SRT found for $x\geqslant 10\%$ with the coverage up to 20 ML. The similar behavior was also found in Fe_xNi_{1-x}/Cu(100), but it was more sensitive to the Fe concentration. The critical thickness of SRT varies more drastically to 16 ML for $x=5\%$, and no such transition can be observed for $x\geqslant 6\%$, as summarized in Fig. 3.

Comparing the electron configurations of the Fe, Co, and Ni atoms, the difference in number of $3d$ electrons between Fe and Ni is twice as large as that between Co and Ni. It is reasonable to expected that the influence of iron on the magnetic behavior is stronger than cobalt as these two elements are alloyed with Ni. However, the influence of Fe concentration in Fe_xNi_{1-x}/Cu(100) alloy films was found to be less than expected. The effect of the alloy composition x for Fe on the critical thickness for the SRT is about 1.4 times as large as that for Co, with the estimation of the electron number n_d in $3d$ minority band made by fitting the magnetostric-

TABLE I. Average number of electrons in 3d band for transition metals Fe, Co, and Ni.[a]

	$3d\uparrow$	$3d\downarrow$	Asymmetry ($3d\uparrow - 3d\downarrow$)
Fe	4.6	2.4	2.2
Co	5.0	3.3	1.7
Ni	5.0	4.4	0.6

[a]See Ref. 14.

tion coefficients λ_{001} as a function of n_d.[13] The deviation between the experimental result and expectation (two times larger) should be attributed to fact that the distribution of electrons in metal is somewhat different from that in a free atom. The 3d-band filling for the solid metals Fe, Co, and Ni are given in Table I.[14] With these modified values, the effect of 3d-band filling for Fe caused by the alloy concentration on the magnetic anisotropy is about $(2.2-0.6)/(1.7-0.6) = 1.45$ times as large as Co, which is very close to our experimental results.

In conclusion, the critical thickness of spin-reorientation transition for alloy ultrathin films Fe_xNi_{1-x}/Cu(100) and Co_xNi_{1-x}/Cu(100) were investigated and compared in this work. In comparison to the Co_xNi_{1-x}/Cu(100) films, Fe_xNi_{1-x}/Cu(100) exhibit more drastic influence of the alloy concentration x on the critical thickness of SRT. Based on the insignificant structure variation of the films in all the compositions, these composition-driven spin-reorientation transitions can be attributed to the effect of 3d-band filling on the magnetic anisotropy for the Ni/Cu(100)-like films. Based on our experimental results, the difference of the 3d electron number between the majority and minority bands for the Fe–Ni, and Co–Ni alloys was also successfully applied to estimate the effect of 3d-band filling of Fe_xNi_{1-x}/Cu(100) films as compared with Co_xNi_{1-x}/Cu(100) films.

This research was supported by the grant from the National Science Council at Taiwan through Contract No. NSC-90-2112-M-002-036.

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