Journal of Magnetism and Magnetic Materials 324 (2012) 2131–2135

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Exchange-induced phase separation in Ni–Cu films

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ARTICLE INFO

Article history:
Received 12 December 2011
Available online 23 February 2012

Keywords:
Dilute ferromagnet
Thin film
Phase diagram
Ferromagnetic resonance
Curie temperature

ABSTRACT

Magneto-structural properties of films of diluted ferromagnetic alloys Ni_xCu_{1-x} in the concentration range $0.7 < x < 1.0$ are studied experimentally. Films deposited by magnetron sputtering show partial phase separation, as evidenced by structural analysis and ferromagnetic resonance measurements. The phase diagram of the Ni_xCu_{1-x} bulk system is obtained using numerical theoretical analysis of the electronic structure, taking into account the interatomic exchange interactions. The results confirm the experimentally found partial phase separation, explain it as magnetic in origin, and indicate an additional metastable region connected with the ferromagnetic transition in the system.

1. Introduction

Magnetic multilayered films have been receiving much attention from the research community in the field of spintronics. Recently, a device has been proposed [1] and demonstrated [2,3], in which the magnetic and spin-transport properties are controlled thermo-electrically. At the core of the new design is a tri-layer (*F/f/F*) of two strong ferromagnets (*F*) exchange-coupled by a weakly ferromagnetic spacer (*f*) having the Curie point at/or near room temperature (300–500 K). The spacer *f*, controlling the exchange coupling between the outer layers, can be made of a diluted ferromagnetic alloy, such as Ni_xCu_{1-x} [2,3]. It is desirable that the spacer is compositionally uniform and does not contain multiple phases, which can form as a result of atomic segregation during film deposition or subsequent heat treatment.

Alloys Ni_xCu_{1-x} in their bulk form are *fcc* binary substitution alloys (α-phase), with Ni and Cu mutually solvable in any proportion up to the temperature of 627 K, where at $x=0.67$, the α phase separates into two, α_1 and α_2 [4]. According to [4], the phase diagram contains a metastable region with phase separation at the nickel concentration of $0.86 < x < 0.91$, which can lead to additional magneto-structural inhomogeneities in the system. The results, obtained in this work, however, are obtained using an empirical model, based on the experimental data. It is desirable to independently simulate the phase equilibria in the system Ni_xCu_{1-x}, first without using experimental information and then compare the results with the experimental findings. Of particular interest is the influence of the magnetic interactions in the system on the potential structural and/or compositional phase separation. The Curie temperature of bulk Ni_xCu_{1-x} alloys depends linearly on the Ni concentration [4]. By varying the Ni content from 0.5 to 1 the Curie temperature is changed from 0 K to 627 K. However, based on neutron scattering data, it was shown that both bulk samples [5,6] and sputtered films of Ni–Cu alloys [7] exhibit partial phase separation and form Ni rich clusters of typical size 5–10 Å and magnetic moment $8-12\mu_B$. The formation of such Ni clusters and the resulting magnetic inhomogeneity in the material can lead to anomalous magnetic [8] and transport [9] properties.

In this work we investigate the mechanisms behind phase formations in diluted ferromagnetic alloy films of Ni_xCu_{1-x} and explain using first-principles calculations the enhancing effect of the exchange interaction on phase separation in the ferromagnetic composition range of $0.7 < x < 1.0$. We discuss optimum ways to prepare films with enhanced magneto-structural homogeneity.

2. Experimental methods

Films of diluted ferromagnetic Ni_xCu_{1-x} alloys, with $0.7 < x < 0.9$ in Ni concentration, 100 nm thick, were deposited at room temperature on thermally oxidized Si substrates using DC magnetron co-sputtering from Cu and Ni targets. Substrates of 150×10 mm in size, placed above the Cu and Ni targets, along the line connecting the centers of the targets, were used to produce a continuous and essentially linear compositional gradient along the substrate strip. The base pressure in the deposition chamber was $\sim 5 \times 10^{-8}$ Torr and the Ar pressure used during

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doi:10.1016/j.jmmm.2012.02.027

deposition was 5 mTorr. The deposition rate for Ni and Cu in the center of the substrate was ~ 0.5 Å/s. Samples for magneto-structural measurements were cut from the substrate strip into short sections with dimensions of 5×10 mm. In total, 30 samples of various composition Ni_xCu_{1-x} were produced in the same fabrication cycle under the same conditions. The thickness of the films was determined using a surface profilometer. The composition of the films was determined using x-ray dispersion spectroscopy analysis.

Ferromagnetic resonance (FMR) was used to determine the effective magnetization of the Ni_xCu_{1-x} films (M_{eff}) and their Curie temperature (T_c). FMR measurements were performed at 9.45 GHz using a Bruker ELEXSYS-E500 spectrometer equipped with a goniometer for angle-dependent measurements and a variable temperature cryostat. The effective magnetization was obtained from the resonance fields using the Kittel formulae [10]:

$$\frac{\omega}{\gamma} = H_{\perp} - 4\pi M_{eff},$$

$$\frac{\omega}{\gamma} = \sqrt{H_{\parallel}(H_{\parallel} + 4\pi M_{eff})}, \qquad (1)$$

where $H_{\perp}$ and $H_{\parallel}$ are the measured perpendicular and in-plane FMR resonance fields, respectively, ω—frequency and γ—the gyromagnetic ratio.

The Curie temperature of the films was determined from the FMR data as temperature at which either M_{eff} or the FMR signal vanished. The degree of magneto-structural non-uniformity in the films was estimated from the width of the FMR peaks (magnetic non-uniformity leads to broader FMR peaks).

Mechanical stress arising from mismatches in the lattice parameters of the substrate and the films can lead to perpendicular to the plane magnetic anisotropy in the films through magnetostriction. This magnetostrictive contribution to M_{eff} can be estimated using [11]:

$$4\pi M_{eff} = 4\pi M_s - H^*_{\perp}. \qquad (2)$$

Here M_s is the saturation magnetization of the film, $H^*_{\perp} = -2\lambda\sigma/3$—magnetostriction-induced perpendicular-anisotropy field, λ—magnetostrictive constant, σ—mechanical stress in the film. It follows from (2) that for a strong magnetostrictive contribution and/or small saturation magnetization, the effective magnetization can become negative, which corresponds to its perpendicular-to-the-plane orientation in the absence of external fields [10].

3. Experimental results and discussion

Fig. 1 shows the temperature dependence of the FMR resonance fields for Ni_xCu_{1-x} films with different Ni content. For $Ni_{0.71}Cu_{0.29}$ and $Ni_{0.77}Cu_{0.23}$ films, $H_{\perp} < H_{\parallel}$ in a broad temperature range, which indicates an out-of-plane magnetization orientation for these compositions. For films with higher concentrations of Ni (x=0.82–0.92), $H_{\perp} > H_{\parallel}$ and the magnetization is in-plane at zero field.

The temperature dependence of the effective magnetization obtained using (1) and the measured resonance fields (see Fig. 1) are shown in Fig. 2. For all compositions, the effective magnetization first increases with increasing temperature from 150 K to $\sim$320 K and subsequently decreases above $T > 320$ K. This indicates a significant magnetostrictive contribution to the anisotropy. For low Ni concentrations, $Ni_{0.71}Cu_{0.29}$ and $Ni_{0.77}Cu_{0.23}$, M_{eff} is negative, while for $Ni_{0.82}Cu_{0.18}$, $Ni_{0.85}Cu_{0.15}$, $Ni_{0.87}Cu_{0.13}$, $Ni_{0.89}Cu_{0.11}$ and $Ni_{0.92}Cu_{0.08}$ $M_{eff} > 0$. Thus, in films with low Ni concentrations and therefore low saturation magnetization, magnetostriction results in perpendicular magnetic anisotropy.

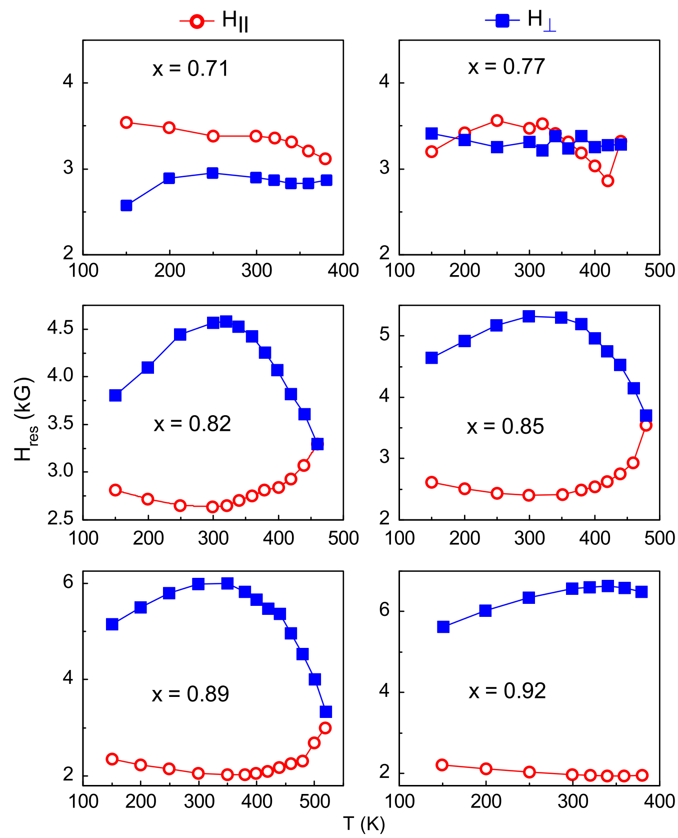

Fig. 1. FMR resonance fields for Ni_xCu_{1-x} films versus temperature. (–■–)—external field perpendicular to the film plane, (–○–)—field in the film plane.

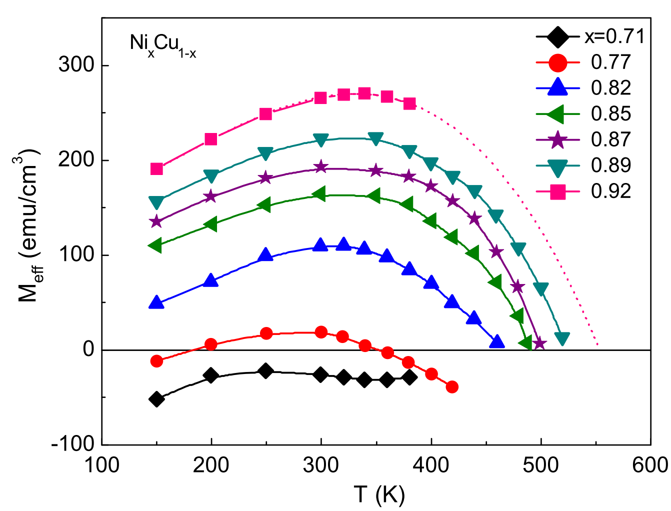

Fig. 2. Temperature dependence of the effective magnetization of Ni_xCu_{1-x} films with different Ni content x. Dashed line is the approximation for x=0.92.

The temperature of the Curie transition from the ferromagnetic to the paramagnetic state (T_c) for Ni_xCu_{1-x} films with x=0.82; 0.85; 0.87; 0.89 and 0.92 was determined as the temperature at which $M_{eff} \rightarrow 0$. For $Ni_{0.71}Cu_{0.29}$ and $Ni_{0.77}Cu_{0.23}$ films with strong magnetostriction and out-of-plane magnetization, it was more appropriate to determine the T_c directly from the temperature dependence of the respective amplitudes of the FMR signal (Fig. 3). T_c in this case was determined as the temperature at which the FMR signal amplitude approached zero (Fig. 3).