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FCC/BCC Competition and Enhancement of Saturation Magnetization in Nanocrystalline Co–Ni–Fe films[¶]

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The structure, chemical composition, and magnetic properties of electrochemically deposited nanocrystalline Co–Ni–Fe films were investigated using a number of techniques. A high saturation magnetic induction up to $B_s = 21$ kG was attained. An enhancement of the saturation magnetization compared to the ideal anticipated one was revealed, which correlated with the nonlinear behavior of the structural phase composition and lattice parameters with the change of the composition.

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Soft magnetic thin films with a high magnetic moment are used in various applications, such as magnetic recording systems, high frequency planar inductors, and modern nonvolatile magnetic memory. Currently, such a widespread ferromagnetic substance is permalloy. However, ternary Co–Ni–Fe alloy possesses much higher saturation magnetization [1]. Moreover, competition among fcc and bcc crystalline phases decreases the coercive force and magnetostriction. At the same time, the complicated correlation between the deposition condition, microstructure, and magnetic properties is a basic constraint of the industrial application of this material. In this paper, we analyze the saturation magnetization, structure, and lattice parameters of electrodeposited thin Co–Fe–Ni films as a function of their chemical composition.

Nanocrystalline CoNiFe films were prepared by the electrochemical method on Cu(250 nm)/Cr(25 nm)/SiO₂(300 nm)/Si substrate. Uniaxial magnetic anisotropy was obtained by applying a permanent magnetic field around 800 Oe oriented along the substrate surface during the film deposition. Details of the film deposition can be found in [2].

The chemical composition of the films was determined by the energy dispersive analysis of x rays (SEM–EDS and TEM–EDS). The concentration of Co, Fe, and Ni varied between 0.30 to 0.54, 0.12 to 0.31, and 0.15 to 0.58, respectively. From x-ray diffraction (XRD) study, we have learned that the CoNiFe films are normally composed of the fcc and bcc competing structural phases, as illustrated in Fig. 1. More experimental details of investigation of the microstructure and composition were reported previously in our publications

[2–4] as well as in earlier works [1, 5]. In particular, it has been established that the films were highly textured with the (111) XRD line dominating for the fcc crystallites and (110) XRD line dominating for the bcc crystallites, and this kind of texture was kept even after the annealing [3]. Due to this fact, the two lines were normally used for estimations of the fcc-to-bcc fractions in the films. In the present work, the fcc fraction and bcc fraction present in the alloy were obtained from areas under the $(111)_{fcc}$, A_{fcc} and $(110)_{bcc}$, A_{bcc} XRD lines, respectively. From the XRD line widths, we estimated that the grain size for the fcc phase was about 30 nm for most of the samples, while it was about 10 to 20 nm for the bcc grains. The saturation magnetization I_s , as well as the anisotropy field H_k , was obtained from ferromag-

Fig. 1. XRD spectra of Co–Fe–Ni films. The smooth lines are fitted curves.

[¶] The text was submitted by the authors in English.

Fig. 2. Ferromagnetic resonance spectra for the EA and HA directions of the applied magnetic field.

Fig. 3. Ratio of the fcc and bcc fractions vs. the number of electrons per atom n_e .

netic resonance (FMR) measurements at the frequency $f = 9.40$ GHz in an external magnetic field range up to 5 kOe aligned along the film surface by comparing the resonance fields along the easy (EA) and hard (HA) axes and applying the Kittel relationships:

$$\omega^2 = \gamma^2 (H_R^{EA} + H_k)(H_R^{EA} + H_k + 4\pi I_s), \quad (1a)$$

$$\omega^2 = \gamma^2 (H_R^{HA} - H_k)(H_R^{HA} - H_k + 4\pi I_s), \quad (1b)$$

where $\omega = 2\pi f$ is the frequency of the applied RF field, γ is the gyromagnetic ratio, and H_R^{EA} and H_R^{HA} are the resonance values of the DC field applied in the EA and HA directions, respectively. Typical FMR spectra are demonstrated in Fig. 2 with more experimental details. Ferromagnetic resonance spectra for the EA and HA directions of the applied magnetic field of the FMR investigation were reported in [2]. Depending on the composition, the saturation magnetic induction varied between $4\pi I_s = 16.7$ and 21 kG, which is a high value compared to that of permalloy.

The variation of the chemical composition has a strong influence on the structure as well as on the magnetic properties [1–5]. To study the effect of the composition in a generalized way, we combine here the two independent parameters of the composition of the ternary alloy into a single number of electrons per atom:

$$n_e = 27x + 28y + 26z,$$

where x , y , and z are the concentrations of Co, Ni, and Fe in the alloy, respectively, with $x + y + z = 1$.

Figure 3 shows the natural logarithm of the A_{fcc}/A_{bcc} ratio as a function of the number of electrons per atom n_e in the Co–Ni–Fe alloy. The dashed line is the line of the equal population of the fcc and bcc phases. A quite pronounced peak of $\ln(A_{fcc}/A_{bcc})$ is seen at around $n_e = 27.0$, thus, evidencing a dominance of the fcc phase.

An increase in n_e above this value, i.e., a decrease in the Fe content, leads to a surprising decrease in the fcc

fraction and to a parity of the two phases. A reduction of n_e below 27.0, i.e., an increase in the Fe content, leads to a single bcc phase structure, which is the natural structure for the α -Fe.

From the positions of the $(111)_{fcc}$ and $(110)_{bcc}$ XRD lines, the lattice parameters a_{exp}^{fcc} and a_{exp}^{bcc} were obtained, which also varied with the chemical composition. In the simplest ideal case, the lattice parameter of a random binary alloy can follow the linear Vegard law [6] if the pure components have the same type of lattice. For a ternary alloy with a different structure of the pure components (pure Co and Ni crystals have the fcc structure, while pure Fe preferably has the bcc structure), the experimental lattice parameter can be compared with a concentration weighted combination based on atomic volumes per atom as suggested by Zen [7]. Assuming that, in both the fcc and the bcc phase, the volume per atom is the same and taking into account that there are two atoms in the bcc and four atoms in the fcc lattices, we write for ideal lattice parameters

$$a_{ideal}^{fcc} = a_{Co}^{fcc}x + a_{Ni}^{fcc}y + \sqrt[3]{2}a_{Fe}^{bcc}z, \quad (2a)$$

$$a_{ideal}^{bcc} = \frac{1}{\sqrt[3]{2}}(a_{Co}^{fcc}x + a_{Ni}^{fcc}y) + a_{Fe}^{bcc}z. \quad (2b)$$

The ratios $a_{exp}^{fcc}/a_{ideal}^{fcc}$ and $a_{exp}^{bcc}/a_{ideal}^{bcc}$ of the experimental lattice parameters to the ideal anticipated ones as a function of the composition in terms of the number of electrons per atom n_e are plotted in Fig. 4. The lattice parameters of fcc-Co, fcc-Ni, and bcc-Fe are equal to 3.544 Å, 3.515 Å, and 2.867 Å, respectively. The volume required per single atom in Fe is about 5.9% larger than that per Co atom and about 8.5% larger than that per Ni atom. Therefore, the largest change of the alloy lattice parameter occurs when the concentration of Fe varies. The largest values of a_{ideal}^{fcc} and a_{ideal}^{bcc} equal to

Fig. 4. Ratios of the experimental lattice parameters to the ideal anticipated ones versus those predicted by Eqs. (2) for the bcc (a) and fcc (b) structural phases. The solid line is a linear fit to the data.

Fig. 5. Enhancement of the saturation magnetization. The solid line is a linear fit to the data.

3.61 Å and 2.867 Å, respectively, correspond to a dominance of Fe in the alloy.

The deviation of the ratios from unity can be interpreted as a nonlinear deformation of the crystal lattice cells. When the Fe concentration decreases, i.e., n_e increases, the $a_{\text{exp}}^{\text{bcc}}$ lattice parameter decreases faster

(Fig. 4a) than predicted by Eq. (2b). In the fcc phase, the lattice parameter $a_{\text{exp}}^{\text{fcc}}$ is larger than what follows from Eq. (3a), so the ratio $a_{\text{exp}}^{\text{fcc}} / a_{\text{ideal}}^{\text{fcc}}$ is larger than unity at a small concentration of Fe (large value of n_e). However, it falls down below unity at a larger iron concentration (smaller value of n_e), see Fig. 4b. One can also note a sharp decrease in $a_{\text{exp}}^{\text{bcc}}$ around $n_e = 27.0$, i.e., in the region of the peak of $\ln(A_{\text{fcc}}/A_{\text{bcc}})$. It could be qualitatively interpreted as a strong contraction of the bcc nanosize grains due to an interface mismatch effect of dominating fcc grain boundaries. The fcc grains experience a size expansion in the same region due to a similar mismatch effect from the neighboring bcc grain boundaries, as can be seen in Fig. 4b.

Similar to the ideal lattice parameters, one can compose an ideal anticipated saturation magnetization I_S^a of the Co–Ni–Fe alloy as a liner combination of the partial magnetizations

$$I_S^a = I_S^{\text{Co}} x + I_S^{\text{Ni}} y + I_S^{\text{Fe}} z, \quad (3)$$

where the partial magnetic moments I_S^x of pure fcc-Co, fcc-Ni, and bcc-Fe are 1400, 480, and 1700 G, respectively. With such a representation, we can expect a variation of $4\pi I_S^a$ in the range of 11 to 17 kG for the variation of the composition in the investigated films. The observed $4\pi I_S^{\text{obs}}$ values diverge systematically from the anticipated values.

One can see from Fig. 5 that, while the ratio I_S^{obs} / I_S^a is close to one at low $n_e \approx 26.8$, it tends to increase at high n_e values. Most of the experimental points are positioned above the dashed line, which levels the observed and anticipated values. It is evident from the graph that the observed saturation magnetization shows a kind of enhancement compared with the ideal anticipated values, especially for lower iron concentrations or a higher number of electrons per atom. In other words, the data show a kind of tendency to slow down the decrease in the magnetic moment with a decrease in the iron concentration.

A deviation from the Vegard law or from the Zen law for a binary system is a common observation, and there is no reason why these deviations could diminish in a ternary alloy. Evidently, this kind of nonlinear deformation is connected with a complicated rearrangement of the outer shell electron configurations in a disordered alloy. For the case of competing nanocrystalline structural phases, the interface mismatch causes nonlinear deformations, which also induce an additional reconfiguration of the outer shell electrons. However, comparing the data, we can conclude that the internal (change in composition) and the external (interface effect) induced electron reconfigurations that have a different influence on the magnetic properties of

the alloy. While systematic nonlinearity, i.e., a general decrease in $a_{\text{exp}}^{\text{bcc}}/a_{\text{ideal}}^{\text{bcc}}$ and increase in $a_{\text{exp}}^{\text{fcc}}/a_{\text{ideal}}^{\text{fcc}}$ with increase in n_e correlate with the systematic behavior of I_S^{obs}/I_S^a , there is no clear evidence of a strong effect of the interface effect on I_S^{obs}/I_S^a , which definitely is present in the lattice parameter behavior. Moreover, from comparison of Figs. 3 and 5, we can conclude that the enhancement of I_S^{obs}/I_S^a is more evident when the parity of the competing phases stabilizes.

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