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# Fabrication of $L1_0$-FeNi phase by sputtering with rapid thermal annealing

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ABSTRACT

FeNi films were directly deposited on MgO(001) substrates by co-sputtering and rapid thermal annealing (RTA). The formation of the $L1_0$ phase was investigated for films with different thicknesses and different annealing conditions by grazing incidence X-ray diffraction. For the FeNi films with a thickness of 5 nm, superlattice 001 and 110 peaks were observed after annealing at a heating rate of 50 °C/s, which indicates that three variants of $L1_0$ grains were formed in the films. The maximum long-range order parameter, which corresponded to a volume-averaged parameter, was approximately 0.11. For the FeNi films with a thickness of 30 nm, a superlattice 110 peak was only observed after annealing at a heating rate of 50 °C/s, and the formation of 001 textured grains was clarified. Magnetic properties also changed depending on the FeNi film structures. The formation mechanism of $L1_0$-FeNi is discussed based on the strain caused by RTA in the FeNi films.



## 1. Introduction

For a perpendicular recording media and motor magnets, materials with large uniaxial magnetic anisotropy ($K_u$) are necessary and investigated for several materials [1–4]. $L1_0$-ordered alloys, such as $L1_0$-FePt and $L1_0$-FePd, are known to reveal large $K_u$ [5–7]. Therefore, these alloys are promising for high-density perpendicular recording media or high-maximum-energy magnets. Although many $L1_0$-ordered alloys include a noble element, leading to a high cost for industrial products, $L1_0$-FeNi includes only low-cost elements and exhibits large saturation magnetization ($M_s$; 1200–1300 emu/cc) and $K_u$ ($1.3 \times 10^7$ erg/cc), excellent corrosion resistance, and high Curie temperature [8–11]. In other words, this material is a "noble-element-free and large $K_u$ material". From previous studies [12–14] involving $L1_0$-FeNi film experiments, it was found that $K_u$ increased with the order parameter ($S$) and was induced by the spin-orbit coupling of Fe 3*d* electrons in the $L1_0$ structure. However, the formation of the $L1_0$-FeNi phase is quite difficult with conventional methods such as annealing. Because the order-disorder transition temperature (~320 °C) is too low compared to the melting point (>1400 °C), the formation-energy difference between the $L1_0$ and $A1$ phases is also small [15–18], therefore, the ordering process hardly occurs. In nature, $L1_0$-FeNi is found in iron-meteorites [10,11,18–20]; experimentally it was obtained using special techniques such as neutron irradiation in the applied magnetic field [8,9], and the films were fabricated by molecular beam epitaxy (MBE) on a buffer layer with good lattice match to $L1_0$-FeNi [12,14,21–24]. Recently, the formation of an $L1_0$-FeNi alloy was also reported when using layer-by-layer sputtering deposition [25] or various alloying methods [26–29]; however, the volume fraction of $L1_0$-FeNi was small.

In this paper, we fabricated $L1_0$-FeNi films on MgO(001) substrates by conventional sputtering and rapid thermal annealing (RTA). In certain cases, the RTA method is effective in enhancing the $L1_0$ ordering process; [001] textured $L1_0$-FePt and CoPt films were easily obtained on polycrystalline substrates [30–33]. Here, the relation between annealing conditions and structural and magnetic properties for the FeNi films including $L1_0$-FeNi phase were systematically investigated.

## 2. Experimental procedures

All FeNi films were prepared by co-sputtering of Fe (RF) and Ni (DC) directly onto clean MgO(001) substrates at room temperature

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using an ultra-high vacuum (base pressure: $< 3 \times 10^{-6}$ Pa) magnetron sputtering system [34]. Sputtering powers of the Fe and Ni targets were 31 and 11 W, respectively. The Ar gas pressure during sputtering was 0.2 Pa, and the co-deposition rate was 0.015 nm/s. The total thicknesses of the co-deposited FeNi films were 5, 30, 50, and 100 nm. The composition of FeNi was estimated to be $Fe_{47}Ni_{53}$ (at.%) from electron probe microscopy analysis. After deposition, the FeNi films were annealed by using an RTA system consisting of an infrared light source and a vacuum chamber (starting pressure: $< 5 \times 10^{-4}$ Pa). The annealing conditions, such as an annealing temperature (250–400 °C) and annealing time (5–20 h), were used. To study the effect of heating rate on the formation of $L1_0$-FeNi phase, it was changed from 0.1 °C/s to 50 °C/s. The RTA method involves a rapid heating rate (50 °C/s) followed by holding the temperature constant. The typical heating rate was 0.1 °C/s and followed by holding the temperature constant for thermal annealing (TA). For structural characterization, grazing incidence X-ray diffraction (GI-XRD) measurements were carried out with an in-plane scan ($\varphi$-$2\theta$, where $\varphi$ is the rotation angle of the sample, and $2\theta$ is the detector position) using synchrotron radiation at the synchrotron radiation facility of SPring-8 (BL46XU). The incident angle of the X-ray beam was 0.28° with the film surface, and samples were enclosed in a He-gas-exchange chamber to reduce the background due to scattering from air. To use anomalous dispersion, X-ray energy was chosen near the Fe-*K* absorption edge (about 7.110 keV) because the superlattice peak intensity should be enhanced at the Fe-*K* absorption edge compared to that at off-resonance energies. The *S* was evaluated with the following Equation:

$$S = \sqrt{\frac{\left(I_{sup}/I_{fund}\right)^{exp}}{\left(I_{sup}/I_{fund}\right)^{cal}_{S=1}}}, \quad (1)$$

where $I_{sup}$ and $I_{fund}$ are integrated intensities in the GI-XRD measurement for the superlattice and fundamental peaks, respectively. The superscript of "*exp*" means the experimentally observed value estimated by peak-fitting using a pseudo-Voigt function, and "*cal*" means the theoretical value with the perfect order ($S = 1$) calculated with the following Equation:

$$I = |F|^2 \frac{1}{sin2\theta}, \quad (2)$$

where *F* is the structure factor, and 1/sin2θ is the Lorentz factor. $|F|^2$ is described with the following Equation:

$$|F|^2 = 4\left\{\left\{(f_{0Fe} + f'_{Fe}(\varepsilon))e^{-M_{Fe}} \pm (f_{0Ni} + f'_{Ni}(\varepsilon))e^{-M_{Ni}}\right\}^2 + \left\{f''_{Fe}(\varepsilon)e^{-M_{Fe}} \pm f''_{Ni}(\varepsilon)e^{-M_{Ni}}\right\}^2\right\}, \quad (3)$$

where $f_{0Fe}$ and $f_{0Ni}$ are the normal atomic scattering factors of Fe and Ni [35], and $e^{-M_{Fe}}$ and $e^{-M_{Ni}}$ are the Debye-Waller factors [36] of Fe and Ni, respectively. In this study, X-ray energy was adjusted to 7.11 keV (Fe-*K* adsorption edge); therefore, the anomalous dispersion effect was taking place, and *f'* and *f"* are the real and imaginary parts of anomalous scattering factors, respectively. Our measurements required neither adsorption correction nor polarization correction, because in-plane geometry and incident X-rays with linearly polarized light were used. In addition, high-resolution transmission electron microscopy (HRTEM) observation was used for the microstructural analysis of FeNi films. Cross-sectional images were observed with electron beam//MgO[100] direction. Magnetization curves were measured by a superconducting quantum interference device (SQUID) at room temperature. Magnetic fields up to ±20 kOe were applied along the parallel and perpendicular directions to the plane.

## 3. Results

### 3.1. Annealing time dependence for FeNi films (5 nm)

Fig. 1(a) shows GI-XRD profiles for an FeNi film (5 nm) annealed at 350 °C at a heating rate of 50 °C/s and held for 20 h. The incident directions of the X-ray beam are related to the scattering vector (***K***) along the MgO [100] or [110], the results of which are shown with broken and solid lines, respectively. In the profile for the ***K***//MgO [110] direction, superlattice 110 and fundamental 220 peaks were observed. This orientation relationship indicates that the [001] axis (i.e., *c*-axis) is perpendicular to the MgO(001) substrate. On the other hand, in the profile with the ***K***//MgO[100] direction, superlattice 001 and fundamental 200 peaks were observed. With in-plane XRD measurement, observation of diffraction from (100) planes is possible; however, for the $L1_0$ structure, a 100 peak should not be observed due to its structural symmetry [5]. Thus, this peak was indicated as 001 in the profile instead of the 100 peak. This orientation relationship indicates that the [001] axis (i.e., *c*-axis) for $L1_0$-FeNi is parallel to the MgO(001) substrate. Therefore, both superlattice diffraction peaks confirm the formation of two variants of the $L1_0$-FeNi films, that is, the *c*-axis is perpendicular and parallel to the film plane. In general, an $L1_0$-ordered alloy with high tetragonality is easily formed because RTA induces tensile strain to the

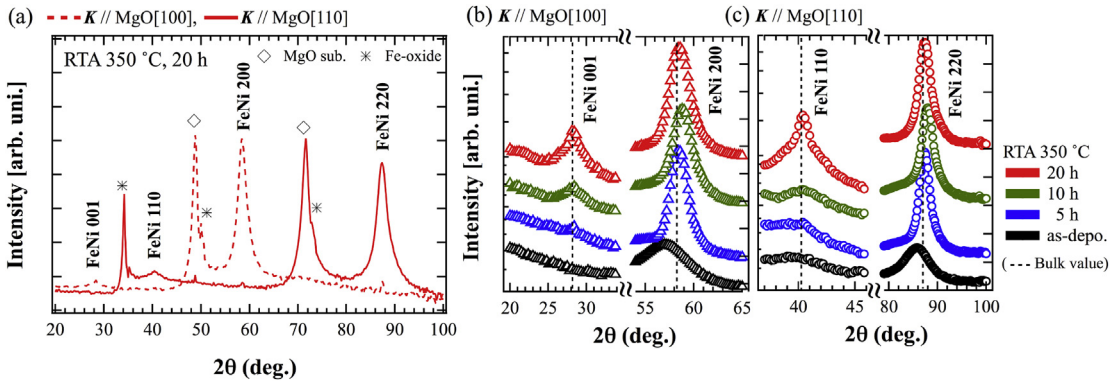


**Fig. 1.** (a) GI-XRD profiles for FeNi film (5 nm) on MgO(001) substrates annealed at 350 °C at a heating rate of 50 °C/s for 5–20 h. (b) Profiles in a narrow range around FeNi(001) and (200) peaks. (c) Profiles in a narrow range around FeNi(110) and (220) peaks.