L1₀-ordered FeNi film grown on Cu-Ni binary buffer layer

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Abstract. An FeNi film with a metastable L₁₀-ordered phase was fabricated by an alternate monatomic layer deposition. An Fe monolayer and a Ni monolayer were alternately grown on a Cu₀.₆Ni₀.₄ binary buffer layer. A (001) L₁₀-FeNi superlattice peak, which indicates the formation of a L₁₀ phase, was clearly observed by an X-ray diffraction. A long-range order parameter \( S \) of L₁₀-FeNi was estimated to be 0.5 ± 0.1. It was found that the film has a uniaxial magnetic anisotropy along \( c \)-axis by magnetization measurement. The uniaxial magnetic anisotropy energy \( K_u \) was evaluated to be \((5.0 ± 0.1) \times 10^6 \) erg/cc.

1. Introduction
Perpendicularly magnetized films (PMF) have been investigated as materials for magnetic recording media. By using PMF, magnetic recording media with higher density can be developed. However, even devices with PMF have a limit of integration, because magnetizations easily fluctuate by external heats when a cell size of magnetic materials is too small. A uniaxial magnetic anisotropy enhances a thermal stability of a magnetization. Therefore, development of PMF with high uniaxial magnetic anisotropy energy \( K_u \) is required. L₁₀-type ordered alloys have attracted as such materials. Particularly, L₁₀-type alloys with noble metals such as FePt, FePd and CoPt have high \( K_u \) of the order of \( 10^7 \) er/sg/cc [1, 2, 3, 4]. On the other hand, it was reported that L₁₀-FeNi also has high \( K_u \) of \( 1.3 \times 10^7 \) erg/cc in a bulk sample [5]. Therefore, it is expected to realize a “noble metal free perpendicularly magnetized film” by the development of L₁₀-FeNi films.

Existence of natural L₁₀-FeNi has been only confirmed in meteorites [6, 7]. An order-disorder transition temperature of L₁₀-FeNi phase is 320°C [8]. The ordering to L₁₀ phase never occurs over this temperature, but diffusion rates of atoms are extremely slow below this temperature. It takes more than \( 10^5 \) years to finish one atomic jump at 300°C [9]. Thus, the formation of L₁₀-FeNi is not easy by simple deposition processes. An Fe monolayer (ML) and a Ni ML are alternately stacked along the \( c \)-axis in L₁₀-FeNi. Thus, L₁₀-FeNi films may be fabricated by the alternately atomic deposition of Fe (1 ML) and Ni (1 ML). Actually, naturally non-existent materials such as L₁₀-FeAu and hcp-CoRu were artificially created by the alternately monatomic layer deposition [10, 11].

Previously, T. Shima et al. fabricated L₁₀-FeNi films [12]. The long-range order parameter \( S \) and the \( K_u \) were estimated to be 0.6 ± 0.2 and \( 6.3 \times 10^6 \) erg/cc, respectively. However, the \( S \) estimated by X-ray diffraction (XRD) had large margin of errors, because L₁₀-NiPt diffraction peaks overlapped L₁₀-FeNi superlattice peaks.

In this paper, we focused on a Cu₀.₆Ni₀.₄ binary buffer layer. This material has no diffraction peak around L₁₀-FeNi superlattice peaks in the XRD measurement, and it is expected this buffer layer should be non-magnetic. We have attempted to estimate the order parameter of L₁₀-FeNi more precisely by using a Cu₀.₆Ni₀.₄ buffer layer.
2. Experimental procedure
Films were grown by molecular beam epitaxy using two e-guns (Fe and Ni) and two Knudsen-Cells (Au and Cu) in ultrahigh vacuum (UHV) chamber (base pressure: $1 \times 10^{-8}$ Pa). Polished MgO (001) substrate was annealed at $570^\circ C$ for 30 min in the UHV chamber. Fe (1 nm) and Au (20 nm) layers were grown on the MgO substrate at 80°C. Then a Cu$_{0.6}$Ni$_{0.4}$ (93 nm) buffer layer was grown by co-deposition of Cu and Ni at 500°C. Finally, Fe (1 ML) and Ni (1 ML) were alternately grown at 187°C. The repetition time of Fe and Ni was 50 times. The deposition rate of Fe and Ni was 0.01 nm/s, which was controlled by a thickness monitor with quartz oscillators. Surface morphology and flatnesses of each layer were monitored by a reflection high-energy electron diffraction (RHEED). X-ray diffraction (XRD) with Cu-Kα radiation was performed to characterize crystal structures and confirm the formation of L1₀-FeNi. Magnetic properties were investigated by magnetization measurement using the superconducting quantum interference device (SQUID) magnetometer at room temperature. A same stacking structure without only an [Fe (1 ML) / Ni (1 ML)]$_{50}$ layer was also fabricated as a reference sample.

3. Results and discussion
Figure 1 shows RHEED images of (a) Cu$_{0.6}$Ni$_{0.4}$ buffer layer and (b) [Fe (1 ML) / Ni (1 ML)]$_{50}$ surfaces. Sharp streak patterns were observed for all the surfaces. This confirms the formation of flat surface and an epitaxial growth of Cu$_{0.6}$Ni$_{0.4}$ buffer layer, Fe, and Ni layers. Streaks between fundamental streaks in Fig. 1 (a) indicate the formation of an ordered alloy or any surface reconstruction.

Figure 1. RHEED images of surfaces of (a) Cu$_{0.6}$Ni$_{0.4}$ buffer layer and (b) [Fe (1 ML) / Ni (1 ML)]$_{50}$. The incident beams were directed along the azimuth of MgO $<100>$ and $<110>$.

Figure 2 shows XRD patterns of samples with and without an FeNi layer. (002) and (004) fundamental peaks of FeNi were observed around $2\theta = 51^\circ$ and $2\theta = 119^\circ$, respectively. In the pattern of sample without FeNi, there are three kinds of peaks from buffer layers. A peak around $2\theta = 49.5^\circ$ is not (002) Au peak, because (002) Au peak should appear around $2\theta = 44.4^\circ$. Thus, it is considered that the Cu$_{0.6}$Ni$_{0.4}$ layer and the Au layer were alloyed because the Cu$_{0.6}$Ni$_{0.4}$ was grown at high temperature (500°C). However, Au-Ni system is immiscible, therefore, there is a possibility that at least three phases were formed in the alloyed Au-Cu-Ni layer. C-axis lattice parameters ($c$) of the three phases in
the buffer were estimated from their peak positions. They were 3.69 (lower angle), 3.57 (middle angle) and 3.51 (higher angle) Å. It is considered that the phase of \( c = 3.69 \) Å includes a large amount of Au compared with other phases, because the lattice parameters for Au, Cu and Ni are 4.08 Å, 3.62 Å and 3.52 Å, respectively. The phase of \( c = 3.51 \) Å might mainly consist of Ni. The phase of \( c = 3.57 \) Å is a phase with an intermediate composition, although it is not definitely identified.

Figure 2 (b) shows a magnified image of Fig. 2 (a) around \( 2\theta = 25^\circ \). (001) \( L1_0 \)-FeNi superlattice peak was clearly observed only for the pattern with FeNi layer. A long-range order parameter \( S \) of \( L1_0 \)-FeNi was estimated to be \( 0.5 \pm 0.1 \) from the ratio of integrated intensities of the (001) superlattice peak and the (002) fundamental peak.

![Figure 2](image_url)

Figure 2. (a) XRD patterns of samples with and without [Fe (1 ML) / Ni (1 ML)]\textsubscript{50} layer. (b) Magnified view around \( 2\theta = 25^\circ \).
Figure 3 shows magnetization curves of the L1₀-FeNi film when the magnetic field was applied in parallel and perpendicular to the film plane. Magnetization curves of the sample without FeNi layer were also measured. Unfortunately, it was found out that the sample without FeNi showed slight ferromagnetism (the saturation magnetization $M_s$ of alloyed buffer layer was about 60 emu/cc). Thus, magnetization curves of the L1₀-FeNi film (Fig. 3) were obtained by subtracting magnetization curves of the sample without FeNi. The reason for the formation of ferromagnetic buffer layer even though Cu₀.₆Ni₀.₄ usually does not show ferromagnetism is considered as follows. The formation of at least three phases of the alloyed buffer layer was confirmed from the XRD measurements. Therefore, it is considered that any ferromagnetic phase different from Cu₀.₆Ni₀.₄ was formed, and this phase showed ferromagnetic behavior in the magnetization curve of the sample without FeNi layer. A $K_u$ of the L1₀-FeNi film was evaluated by sum of an area enclosed between two magnetization curves (Fig.3) and the shape anisotropy $2\pi M_s^2$ (The value of area was minus because an easy magnetization axis was the in-plane direction). Uniaxial magnetic anisotropy of $K_u = (5.0 \pm 0.1) \times 10^6 \text{[erg/cc]}$ was obtained ($M_s = 1125 \pm 5 \text{[emu/cc]}$). It is considered this value will be increased by further optimization of growth conditions.

4. Summary
An FeNi film with a metastable L1₀-ordered phase was fabricated by an alternate monatomic layer deposition of Fe (1 ML) and Ni (1 ML). Buffer layers and the FeNi layer were epitaxially grown on the MgO (001) substrate. A clear (001) L1₀-FeNi superlattice peak was successfully observed by an XRD measurement. The $S$ of L1₀-FeNi was estimated to be $0.5 \pm 0.1$. It was revealed that the easy magnetization axis of FeNi film lay in parallel to the plane from magnetization curves obtained by subtracting magnetization of the buffer layer. The $M_s$ and the $K_u$ of FeNi film was estimated to be $1125 \pm 5 \text{emu/cc}$ and $(5.0 \pm 0.1) \times 10^6 \text{erg/cc}$, respectively. There is a possibility that $K_u$ will be further enhanced by an improvement of the long-range order parameter.

Acknowledgements
This work was partly supported by a Grant-in-Aid for Scientific Research (22246087) from Japan Society for the Promotion of Science.
References

[1] Lairson B M, Visokay M R, Sinclair R and Clemens B M 1993 Appl. Phys. Lett. 62 639
[2] Gehanno V, Samson Y, Marty A, Gilles B and Chamberod A 1997 J. Magn. Magn. Mater. 172 26
[3] Lairson B M, Visokay M R, Marinero E E, Sinclair R and Clemens B M 1993 J. Appl. Phys. 74 1922
[4] Barmak K, Kim J, Lewis L H, Coffey K R, Toney M F, Kellocc A J and Thiele J-U 2005 J. Appl. Phys. 98 033904
[5] Paulevé J, Chamberod A, Krebs K and Bourret A 1968 J. Appl. Phys. 39 989
[6] Albertsen J F, Jensen G B and Knudsen J M 1978 Nature 273 453
[7] Mehta S, Novotny P M, Williams D B and Goldstein J I 1980 Nature 284 151
[8] Paulevé J, Dautreppe D, Laugier J and Néel L 1962 J. Phys. Radium 23 841
[9] Scorzelli R B 1997 Hyp. Interact. 110 143
[10] Takanashi K, Mitani S, Sano M, Fujimori H, Nakajima H and Osawa A 1995 Appl. Phys. Lett. 67 1016
[11] Himi K, Takanashi K, Mitani S, Yamaguchi M, Ping D H, Hono K and Fujimori H 2001 Appl. Phys. Lett. 78 1436
[12] Shima T, Okamura M, Mitani S and Takanashi K 2007 J. Magn. Magn. Mater. 310 2213