Fabrication of L1₀-FeNi films by nitrogen topotactic extraction from FeNiN films

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Ferromagnetic materials possessing large uniaxial magnetic anisotropy energy ($K_u$) and composed of earth abundant elements are required in order to realize the next-generation permanent magnets. We have focused on the L1₀-ordered FeNi alloy as a rare-earth free high $K_u$ ferromagnetic material [1]. Recently, the synthesis of L1₀-FeNi powder by nitrogen topotactic extraction from FeNiN powder was reported, and a high degree of order ($S$) was achieved [2]. However, the mechanism of nitrogen extraction and the exact $K_u$ value of L1₀-FeNi formed by nitrogen extraction are still unclear. Thereby, the experiment using the single-crystal FeNiN and L1₀-FeNi film is required. In this study, we grew epitaxial FeNiN films by molecular beam epitaxy (MBE), and fabricated L1₀-FeNi films by the nitrogen topotactic extraction [3].

20 nm-thick FeNiN films were grown on SrTiO₃(STO)(001) substrates by MBE supplying Fe, Ni, and radio-frequency (RF) N₂ plasma, simultaneously [4]. Supply rates of Fe and Ni were controlled so that Fe/Ni composition was 1. The growth temperature, N₂ gas flow rate, and RF input power were fixed to be 300 °C, 1.0 sccm, and 250 W, respectively. Denitriding was performed by ex-situ furnace annealing at 300 °C for 4 h under H₂ gas flow rate of 1 L/min at ambient pressure. Structure of the samples was characterized by out-of-plane and in-plane x-ray diffraction using Cu-Kα radiation. Magnetization curves were measured by vibrating sample magnetometer at room temperature, and the $K_u$ value was estimated by magnetic torque measurements.

From the XRD measurements, we confirm the multi-domain epitaxial growth of FeNiN films on STO(001) with epitaxial relationships of FeNiN[001](100)/STO[100](001) and FeNiN[010](100)/STO[100](001). These epitaxial relationships are maintained after the denitriding, which means that L1₀-FeNi films textured with the $a$-axis perpendicular to the film plane with two variants have been fabricated. The uniaxial magnetic anisotropy along two in-plane $c$-axis directions of the $a$-axis textured L1₀-FeNi films is confirmed by the magnetization curve measurements. The $K_u$ value is calculated to be $4.4 \times 10^6$ erg/cm³ by magnetic torque measurements. From the relationship between $K_u$ and $S$ reported for L1₀-FeNi films prepared by MBE [5], $K_u$ of $4.4 \times 10^6$ erg/cm³ corresponds to $S$ of 0.4. As a next step, we try to grow $c$-axis textured single-crystal FeNiN films, and form $c$-axis textured single-crystal L1₀-FeNi films with the high $K_u$ and $S$ values by the in-situ nitrogen topotactic extraction method.

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