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Minority spin transport in epitaxially grown nickel-iron nitride films

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Abstract

20-60 nm-thick epitaxial $Ni_xFe_{4-x}N$ (x = 0, 1, 3, and 4) films were successfully fabricated on $SrTiO_3$ (001) single-crystal substrates by alternating substrate temperature (T_{sub}). Crystal orientation and degree of order of N site were improved with the increase of T_{sub} for x = 1 and 3. The lattice constant and saturation magnetization decreased as the Ni content increased. Curie temperature of the Ni_3FeN film was estimated to be 266 K from the temperature dependence of magnetization. Negative anisotropic magnetoresistance effects were observed for x = 1 and 3, and this result indicates that the minority spin conduction is dominant in $Ni_xFe_{4-x}N$(x = 1 and 3).

1. Introduction

Anti-perovskite type 3d transition metal ferromagnetic nitrides are a promising candidate as a new spintronics material. Fe_4N, one of these compounds, is theoretically predicted to have a large negative spin-polarization of electrical conductivity [1], and negative anisotropic magnetoresistance (AMR) effects originating from the negative spin-polarization of Fe_4N were reported [2]. $Ni_xFe_{4-x}N$, where Fe atoms are partially replaced with Ni atoms, is theoretically expected to exhibit a larger negative spin-polarization of density of states at Fermi level (P_D = –0.86) than Fe_4N, and can be a candidate material for spintronics with a high spin-polarization. According to previous studies, the saturation magnetization (M_s), lattice constant, and Curie temperature (T_c) of powdered $Ni_xFe_{4-x}N$ ($0 \leq x \leq 3.6$) monotonically decreased with increasing Ni content [3], and T_c is lower than room temperature (RT) for $x \geq 3$. However, this is not consistent with recent report that Ni_3FeN epitaxial films exhibited a large positive AMR ratio of approximately 6% at RT [4]. Therefore, it is not made clear whether the T_c is above RT or not for Ni_3FeN. In this work, we fabricated 20-60 nm-thick $Ni_xFe_{4-x}N$ (x = 0, 1, 3, and 4) epitaxial films, and examined their magnetic and magnetotransport properties by measuring magnetization curve and AMR effect.

2. Experiment

We grew 20-60 nm thick $Ni_xFe_{4-x}N$(x = 0, 1, 3, and 4) films on $SrTiO_3$ (STO) (001) single-crystal substrates by molecular beam epitaxy using solid sources of Ni and/or Fe and radio-frequency (RF) N plasma. Substrate temperature (T_{sub}) was varied from 150 to 550 °C and the Ni/Fe ratio was controlled by their crucible temperatures. The crystalline quality of grown films was evaluated by reflection high-energy electron diffraction (RHEED), ω-2θ x-ray diffraction (XRD), and ϕ-2$\theta\chi$ XRD measurements with Cu-$K\alpha$ radiation. Occupation probability of N atom at the body center of $Ni_xFe_{4-x}N$ and the degree of order (S) were evaluated from the integrated intensities of the fundamental $Ni_xFe_{4-x}N$ (200) and super lattice $Ni_xFe_{4-x}N$ (100) peaks from x-ray diffraction patterns. Magnetic measurements were carried out for approximately 60 nm-thick $Ni_xFe_{4-x}N$ films utilizing the vibrating sample magnetometer and superconducting quantum interference device magnetometer. A photolithographic process and ion milling were used to pattern the Al-capped 50 nm-thick $Ni_xFe_{4-x}N$(x = 1 and 3) films with stripes 0.2 mm wide and 6 mm long for resistance measurements. The stripes were patterned along the $Ni_xFe_{4-x}N$ (x = 1 and 3) [100] and [110] direction. The AMR measurements were performed in the temperature range of 5-300 K, by a DC four-probe method using the external magnetic field of 30 kOe, and the electric current of 0.2 mA.

3. Result and discussion

Fig. 1 shows the ω-2θ XRD and RHEED patterns of 20 nm-thick $Ni_xFe_{4-x}N$(x = 0, 1, 3, and 4) films fabricated at T_{sub} = 450, 550, 350, and 250 °C, respectively. Only (001) oriented diffraction peak and streaky RHEED patterns were observed for x = 0, 1, and 3, which is indicative of epitaxial growth of single-phase nitrides. In the case of Ni_4N, however, other phases such as Ni_8N were confirmed, suggesting the decomposition due to release of N atoms. The full width at half maximums (FWHMs) for $NiFe_3N$(002) and Ni_3FeN(002) diffraction lines, measured by x-ray ω-scan rocking curves decreased with increasing T_{sub} and showed the smallest values of FWHM = 0.280 and 1.248° at T_{sub} = 550 and 400 °C, respectively. In addition, the S of $NiFe_3N$ and Ni_3FeN reached the largest values of S = 0.71 and 0.75 at the respective T_{sub}. The S value was calculated to be 0.75 for Fe_4N fabricated at T_{sub} = 450 °C, whereas we were not able to calculate it for Ni_4N because of its supper lattice diffraction peak being very weak. The lattice constants of $Ni_xFe_{4-x}N$(x = 0, 1, 3, and 4) films were determined to be 3.798, 3.790, 3.756, and 3.745 Å, respectively. The decrease of lattice constants with increasing x in $Ni_xFe_{4-x}N$ is well explained by the replacement of Fe atoms with smaller radius Ni atoms.

Fig. 2 shows the M_s of $Ni_xFe_{4-x}N$ films as a function of x. Those obtained from first-principle calculation are also plotted. The M_s values were 1300 ± 70 emu/cm^3 and 1060 ± 50 emu/cm^3 for Fe_4N and $NiFe_3N$, respectively, at RT, and they increased to 1520 ± 80 emu/cm^3, 1150 ± 60 emu/cm^3 at 2 K. In contrast, the hysteresis curve was not obtained for Ni_3FeN at RT but obtained at 2 K with M_s = 480 ± 20 emu/cm^3. T_c of Ni_3FeN film was determined to be 266 K from the temperature dependence of magnetization. In our present experimental result, Ni_4N was paramagnetic, differently from our first-principle calculation.

Fig. 3 shows the temperature dependence of AMR ratio of $Ni_xFe_{4-x}N$(x = 1 and 3) films. The AMR ratios of $NiFe_3N$ (Ni_3FeN) at 5 K were −1.0% (−1.6%) and −0.5% (−4.0%) in I // [100] and I // [110] directions, respectively. Negative AMR ratio was obtained at almost all of the temperature. According to recent theory of Kokado *et al.* in the framework of the two-current model that takes into account the *s-d* scattering, negative AMR effect is explained by scattering process of $s_\uparrow \rightarrow d_\uparrow$ or $s_\downarrow \rightarrow d_\downarrow$ [5]. It is understood that P_D of $Ni_xFe_{4-x}N$(x = 1 and 3) are negative, therefore, these negative AMR effects are originated from scattering process of $s_\downarrow \rightarrow d_\downarrow$, and indicates that the minority spin transport is dominant in $Ni_xFe_{4-x}N$(x = 1 and 3).

4. Conclusion

$Ni_xFe_{4-x}N$(x = 0, 1, 3, and 4) epitaxial films were successfully prepared on STO(001) single-crystal substrates and their crystalline qualities, magnetic properties, and magneotransport properties were investigated. The crystal orientation and S values were evaluated for samples grown at various T_{sub}. With increasing T_{sub}, the FWHMs of $NiFe_3N$(002) and Ni_3FeN(002) diffraction lines decreased and they reached the smallest values of 0.28 and 1.248° at T_{sub} = 550 and 400 °C, respectively, and their S values reached the highest values of 0.71 and 0.75. The lattice parameters and M_s decreased for $Ni_xFe_{4-x}N$ as the Ni composition increased. Negative AMR effects were observed for x = 1 and 3, indicating that the minority spin electron dominate the electrical conductivity in $Ni_xFe_{4-x}N$(x = 1 and 3).

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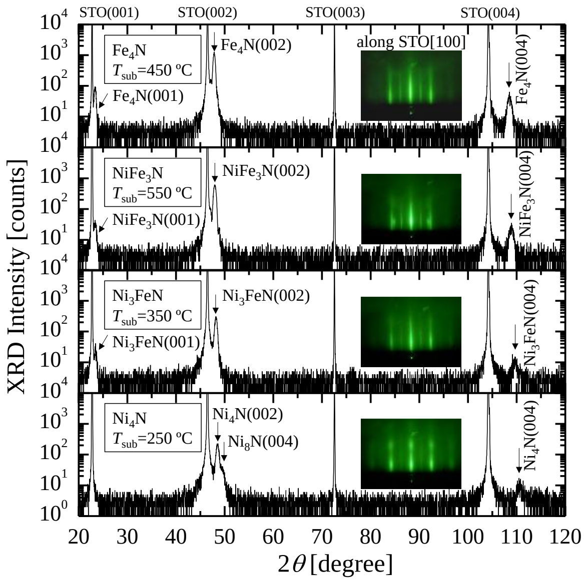

Fig. 1 ω-2θ XRD and RHEED patterns of 20 nm-thick $Ni_xFe_{4-x}N$ (x = 0, 1, 3, and 4) films.

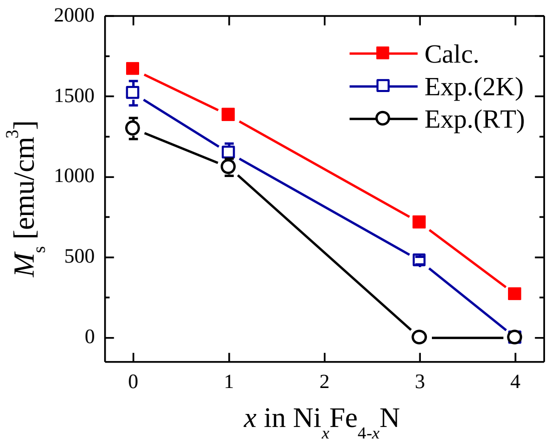

Fig. 2 Saturation magnetization of $Ni_xFe_{4-x}N$(x = 0, 1, 3, and 4).

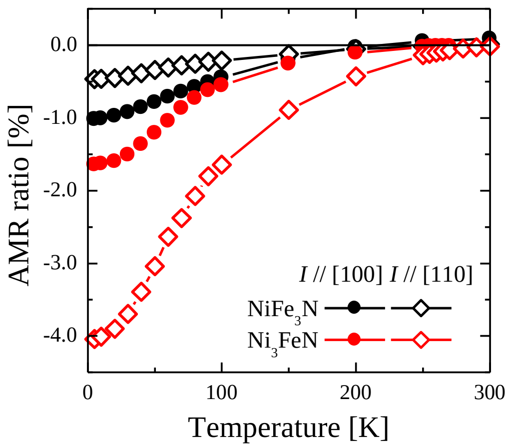

Fig. 3 Temperature dependence of AMR ratio of $Ni_xFe_{4-x}N$(x = 1 and 3).