

Anomalous Nernst Effect of Ni-Al Alloys and Application to Spin Seebeck Devices

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Abstract

We have investigated anomalous Nernst effect of Ni-Al alloys and it is applied to spin Seebeck devices. The metal thickness and composition dependence have been investigated. It was found that the electromotive force of the spin Seebeck devices is larger as nickel film is thinner and it is highest for Ni_{0.7}Al_{0.3}.

1. Introduction

Studies of thermoelectric conversion technique based on spin Seebeck effect are actively carried out [1, 2]. However, the efficiency is too small to make practical devices. In order to increase the power conversion efficiency, spin Seebeck devices combined with anomalous Nernst effect of Ni were proposed [3]. The electromotive force is increased for Ni (2.66 $\mu\text{V/K}$) compared with Pt electrode (1.65 $\mu\text{V/K}$). On the other hand, Mr. Sachi reported that anomalous Nernst effect of Fe is increased by Al addition [4]. However, effect of Al addition to Ni has not yet been reported. Then, we have investigated anomalous Nernst effect of Ni-Al alloys.

Schematic illustrations of (a) spin Seebeck effect, (b) anomalous Nernst effect, and (c) spin Seebeck device using anomalous Nernst effect are shown in Fig. 1. The electric field E_{ISHE} by spin Seebeck effect and E_{ANE} by anomalous Nernst effect are expressed by the following equations,

$$E_{ISHE} \propto J_s \times \sigma \quad (1), \quad E_{ANE} \propto M \times \Delta T \quad (2)$$

where J_s , σ , M , and ΔT , respectively, represent spin current vector, spin polarization vector, magnetization vector, and temperature gradient within the ferromagnetic metal [2, 3]. The electromotive force of anomalous Nernst effect occurs in ferromagnetic metal. Then, it is possible to increase electromotive force by replacing paramagnetic metal layer of spin Seebeck devices.

2. Spin Seebeck devices combined with anomalous Nernst effect of Ni

At first we used Ni as one of a ferromagnetic material. Recipe of fabricating thermoelectric transducer based on spin Seebeck and anomalous Nernst effects is shown in Table 1. Ce₁Y₂Fe₅O₁₂ (Ce:YIG) ferrimagnetic film was deposited by Metal Organic Decomposition (MOD) method, which is carried out in atmospheric pressure and the expensive vacuum apparatus is not necessary, on Gd₃Ga₅O₁₂ (GGG) (111) substrate. Ce:YIG was spin-coated on GGG (111) substrate, followed by decomposition and crystallization. The MOD process of Ce:YIG film was repeated 10 times to produce 400 nm thick film and finally annealed at 1050 °C for 3 h in pseudo air ambient (N₂:O₂=4:1). The surface roughness of Ce:YIG films was evaluated by atomic force microscope (AFM) and it was revealed that the surface is very rough. Then the sample surface was mechanically polished using diamond slurry with a diameter of 0.25 μm for 1 min. After cleaning the polished surface in acetone and pure water Ni of 10 ~ 40 nm thick was deposited on Ce:YIG film by vacuum deposition. Thereafter, the electromotive force was measured. Here temperature difference between substrate and Ni electrode is 50 °C and the applied magnetic field is 500 G. Electromotive force is

changed as a function of Ni film thickness as shown in Fig. 2. The electromotive force of Ni/Ce:YIG samples is higher than that of Ni/SiO₂/Si samples because the electromotive force by both inverse spin hall effect and anomalous Nernst effect exist. As Ni film is thinner the electromotive force is larger. This reason is that the resistance of Ni film is high when it is thin, resulting in the high electromotive force which is given by the current times resistance. We also confirmed that the electromotive force of Ni/Ce:YIG samples is higher than that of Pt/Ce:YIG. Next, we investigated the influence of the deposition method of Ni film. Ni was deposited on Bi₁Y₂Fe₅O₁₂ (Bi:YIG) by vacuum deposition or RF sputtering. Bi:YIG (300 nm) film was formed by MOD process on GGG (111) using the recipe shown in Table 1. The electromotive force for the sample with Ni film deposited by sputtering is higher than that deposited by vacuum deposition as shown in Fig. 3.

3. Anomalous Nernst effect of Ni-Al alloys

Ni was deposited on SiO₂ (1.6 μm)/Si. Then Al was deposited on Ni in succession. In mass ratio, Ni:Al = 8:2. The total thickness of Ni and Al is about 20 nm. The samples were annealed at 600 °C ~ 900 °C for 30 min in nitrogen atmosphere. X-ray diffraction (XRD) spectra of NiAl films annealed at various temperatures are shown in Fig. 4. XRD peak of NiAl (110) appears in $2\theta \approx 44^\circ$. The peak height of NiAl annealed at 800 °C is the highest. The electromotive force was observed for only NiAl annealed at 900 °C. We considered that for 900 °C annealed sample, the crystal phase may be different from other samples because the XRD peak position is slightly different. We measured electromotive force of NiAl alloy with various Al contents. NiAl alloys were deposited on SiO₂. In mass ratio, Ni:Al = 9:1, 8:2, 7:3, 6:4, 5:5. These were annealed at 900 °C for 30 min in nitrogen atmosphere. XRD spectra of NiAl alloy of various Al contents are shown in Fig. 5. As Al content is higher the XRD intensity of NiAl (110) is decreased. The electromotive force of NiAl alloys depending on Al content is shown in Fig. 6. It is found that the electromotive force of Ni_{0.7}Al_{0.3} is the highest. However, the result was inferior to that of Fe-Al alloys [4]. There are several reason of this. We fabricated Ni-Al alloys from layer structure, whereas Fe-Al alloys was sputtered from the alloy target [4]. Another reason it that the saturation magnetization of Ni is less than Fe.

4. Conclusions

We have fabricated spin Seebeck devices using anomalous Nernst effect of Ni-Al alloys. For pure Ni it is confirmed that as Ni film is thinner the electromotive force is increased. The electromotive force of Ni film deposited by sputtering is higher than that deposited by vacuum deposition. For Ni-Al alloys it was found that the appropriate anneal condition is 900 °C, and the composition of Ni_{0.7}Al_{0.3} is the best for the electromotive force.

References

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- [3] A. Kirihara, *et al.*, Scientific Reports **6**, 17 (2016).
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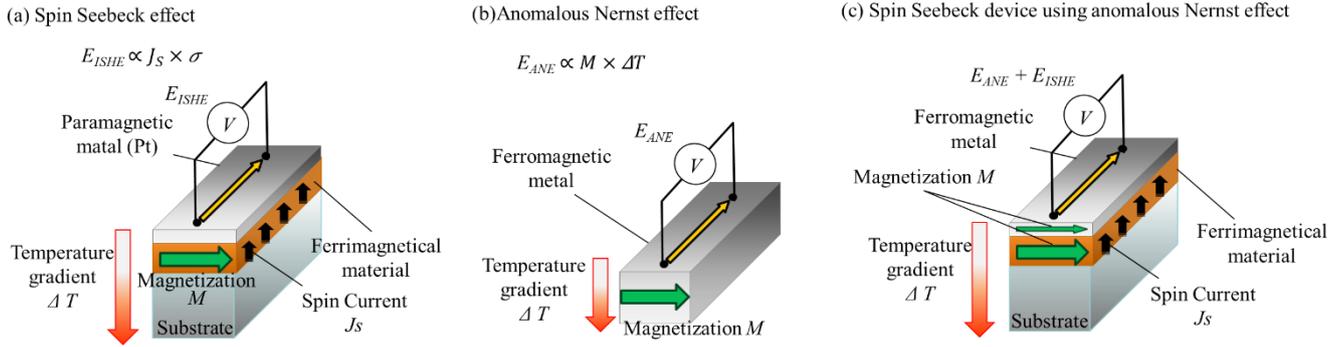


Fig. 1 Schematic illustrations of (a) spin Seebeck effect, (b) anomalous Nernst effect and (c) spin Seebeck device using anomalous Nernst effect.

Table 1 Recipe of fabricating thermoelectric transducer based on spin Seebeck and anomalous Nernst effects.

Process	Condition
1. Spin-coating	500 rpm 5 s 3000 rpm 30 s
2. Drying (in air)	150 °C 3 min
3. Temperature rising	35 °C/min
4. Pre-baking (in air)	500 °C 5 min
5. Annealing (N ₂ :O ₂ = 4:1)	• 1050 °C 3 h (Ce:YIG) • 720 °C 14 h (Bi:YIG)
6. Nickel Deposition	10 ~ 40 nm

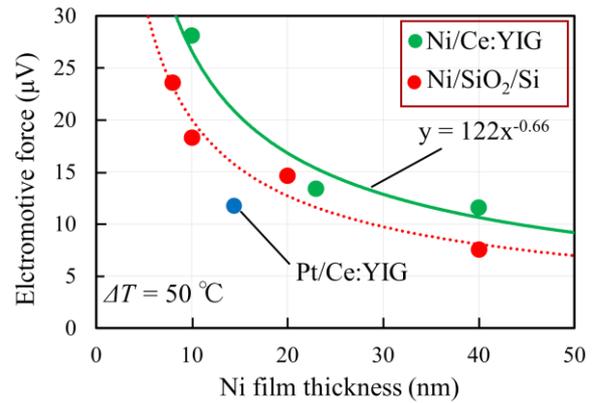


Fig. 2 Electromotive force of Ni/Ce:YIG and Ni/SiO₂/Si as a function of Ni film thickness.

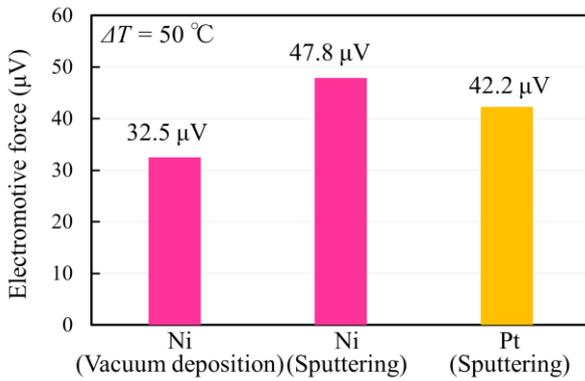


Fig. 3 Comparison of electromotive force for various electrodes.

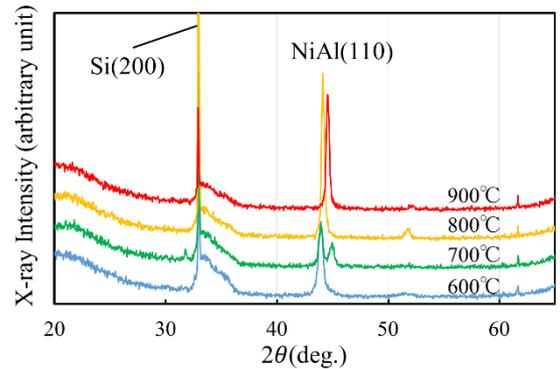


Fig. 4 XRD spectra of NiAl film annealed at various temperatures.

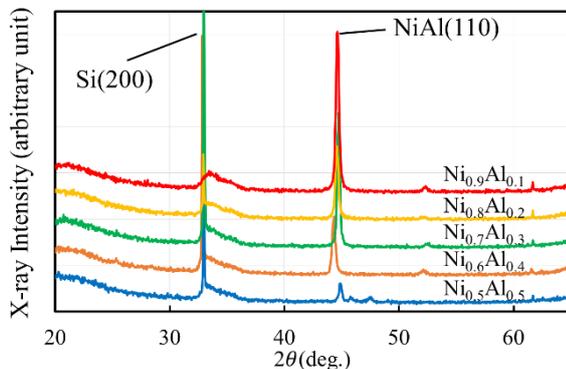


Fig. 5 XRD spectra of NiAl alloys with various Al content.

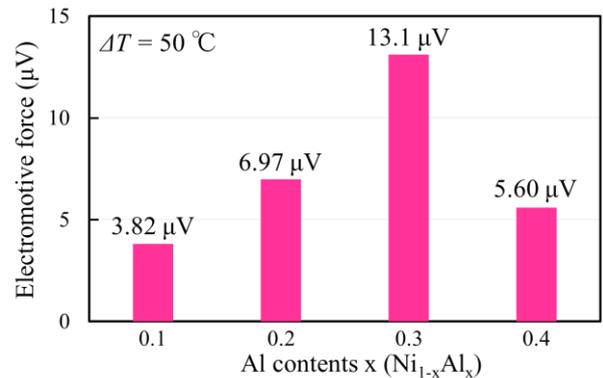


Fig. 6 Electromotive force for NiAl alloys depending on Al content