Transport and magnetic properties of fully-epitaxial superconducting NbN/half-metallic Heusler alloy Co$_2$MnSi bilayer films

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Fully-epitaxial NbN/Co$_2$MnSi bilayer films were grown on MgO(001) single crystalline substrates by a sputter technique. Structural, electric transport and magnetic properties were investigated for the fabricated films. The resulting NbN/Co$_2$MnSi bilayer films show superconducting critical temperature $T_c$ was of 16.0 K and the saturation magnetization $M_s$ at room temperature was of 790 emu/cm$^3$. The electric resistivity of the fabricated films was also measured under magnetic fields and the upper critical field $B_{c2}$ was estimated to be $B_{c2} > 30$ T in the case of the applied magnetic field parallel to the MgO(001) single crystalline substrate. In addition, NbN/Co$_2$MnSi junctions were made by an electron-beam lithography technique. The enhancement of a zero-bias conductance peak (ZBCP) was observed in differential conductance in the NbN/Co$_2$MnSi junctions. The ZBCP disappeared above $T_c = 16.0$ K of superconducting NbN films.

Key words: half-metal, superconductor/ferromagnet junctions, Andreev reflection, Heusler alloy, NbN

1. Introduction

The field of spintronics has attracted increased attention because half-metallic ferromagnets that have an energy gap in only majority spin channel can generate completely spin polarized conduction electrons. Half-metals are expected to lead to great advancement in spintronic devices, such as magnetic random access memories and magnetic field sensors. In order to search for half-metals, there are several determination techniques of the spin polarization $P$ of materials: for example, tunneling magnetoresistance (TMR) effect$^3$, Andreev reflection$^2$ and Zeeman splitting$^3$. Actually the $P$ value of full-Heusler alloy Co$_2$MnSi was reported as $P = 89\%$ at $T = 2$ K from the TMR ratio of Co$_2$MnSi/Al-O/Co$_2$MnSi devices$^4$. Their TMR ratio drastically reduced with the increase of temperature up to room temperature. This is however the evidence that full-Heusler alloy Co$_2$MnSi is half-metal at least at low temperatures.

The spin polarization among different determination techniques does not coincide each other; for example, $P = 89\%$ in the case of the TMR device$^4$ but $P = 56\%$ in the case of the Andreev reflection method$^5$ at liquid helium temperatures for full-Heusler alloy Co$_2$MnSi. One of the possible reasons for different $P$ values results in realization of the different crystalline states at each junction interface. This is because the TMR devices are controlled in the atomic scale at an interface, whereas the roughness at a junction interface could not be essentially avoid for point-contact Andreev reflection (PCAR) methods$^6$, due to adjusting contact resistance between a superconducting needle and a ferromagnetic Heusler alloy. The features of superconducting NbN films are chemically stable, high superconducting critical temperature $T_c$ and high upper critical magnetic field $B_{c2}$ in the conventional metallic superconductors. Furthermore its fully-epitaxial growth is possible on single crystalline MgO and Co-based Heusler alloys$^7$. Superconducting Josephson devices are actually realized using NbN multilayer films, such as magnetic sensors, filters and voltage standards.

In this paper, we focus on properties of the fully-epitaxial superconducting NbN/half-metallic Heusler alloy Co$_2$MnSi bilayer films and their junctions. The fully-epitaxial films can prevent roughness effects at a junction interface, which are possible to reduce the spin polarization due to the spin flip scattering or inelastic scattering for incident electrons. We report (i) the fabrication process of fully-epitaxial NbN/Co$_2$MnSi bilayer films, (ii) the structural, electric transport and magnetic properties of their fully-epitaxial bilayer films and (iii) their junction properties for Andreev reflection measurements.

2. Experimental procedure

An epitaxially grown superconducting NbN film was fabricated directly on a MgO(001) single crystalline substrate, and then a Co$_2$MnSi Heusler film was grown on the top of the NbN film without breaking vacuum using an ultrahigh vacuum magnetron sputtering system. Here NbN films are deposited using an Nb target by reactive sputtering in nitrogen and argon mixture gas. At this point, the NbN/Co$_2$MnSi bilayer film was annealed at 450 °C because of helping to form the L2$_1$-ordered structure for the Co$_2$MnSi Heusler layer. The thicknesses of NbN and Co$_2$MnSi films were controlled to 100 nm and 5 nm, respectively. An Au film with a thickness of 3 nm was finally deposited on the top surface as a protective layer. Structural properties...
for fabricated films were confirmed by x-ray diffraction (XRD) and reflection high-energy electron diffraction (RHEED). The saturation magnetization and in-plane magnetic anisotropy were also investigated using a vibrating sample magnetometer (VSM) at room temperature. The electric resistivity was measured by a standard four-point probe method under magnetic fields up to 14.5 T in the temperature range between 4.2 K and 300 K. Magnetic fields were applied to two directions, which were parallel and perpendicular to MgO(001) substrates. Using the fully-epitaxial NbN/Co2MnSi bilayer films, junctions for Andreev reflection measurements were made with the circular pillars in the range between 40 nm and 4 μm in diameter by an electron-beam lithography technique. The differential conductance of NbN/Co2MnSi junctions was measured using a physical property measurement system (PPMS) by a conventional modulation technique in the temperature range between 2K and 20 K.

3. Results and discussion

3.1 Crystal structures

The crystal structures of NbN films were investigated by XRD patterns. Figure 1(a) shows the 2θ profiles for NbN films with 100 nm in thickness at various nitrogen partial pressures. The resulting lattice constants for a-axis of the NbN films are given in Table 1. The mismatch of lattice constants between a MgO(001) substrate and a NbN(001) film was improved with the decrease of nitrogen partial pressures. The lattice mismatch was estimated as 5.2% in the case of the nitrogen partial pressure of 5.9%. Figure 1(b) shows rocking curves at various nitrogen partial pressures, corresponding to the NbN films in Fig. 1(a). As shown in Fig. 1(c), RHEED images denote that Co2MnSi(110) superlattice diffraction lines [arrows in Fig. 1(c)] from the L21-ordered structure were confirmed when an electron-beam was incident to the (110)-direction of Co2MnSi surface. The epitaxial growth of NbN(001) films and Co2MnSi(001) films on MgO(001) single crystalline substrates was confirmed for all samples using 2θ profiles, rocking curves and RHEED images.

3.2 Magnetization

The magnetization of NbN/Co2MnSi bilayer films was measured as a function of magnetic fields at room temperature. The magnetization is selectively sensitive only to the Co2MnSi layer because the Co2MnSi film is the ferromagnetic state but the NbN film is the normal state at room temperature. Figure 2 shows the magnetization curves under magnetic fields applied to the Co2MnSi(100)- and Co2MnSi(110)-directions. The experimental results indicate that there was the in-plane magnetic anisotropy for the Co2MnSi films and the magnetic easy-axis was the Co2MnSi(110)-direction. The magnetization was almost saturated at 100 G and the saturation magnetization $M_s$ was 790 emu/cm³, which was slightly smaller value than 1050 emu/cm³ of the bulk Co2MnSi samples.

3.3 Electric transport

The electric resistivity $\rho(T)$ was observed as a function of temperature for NbN films and NbN/Co2MnSi bilayer films. Figure 3 illustrates nitrogen partial pressure dependence of electric resistivity $\rho(T)$ of superconducting NbN films with 100 nm in thickness. As shown in Fig. 3, the $T_c$ increases with the decrease of
MgO-sub./NbN/Co$_2$MnSi

Fig. 2 Magnetization as a function of applied magnetic fields to the Co$_2$MnSi(100)- and Co$_2$MnSi(110)-directions for the NbN/Co$_2$MnSi bilayer film at room temperature. The saturation magnetization $M_s$ is slightly smaller value than that of the bulk Co$_2$MnSi sample. The magnetic easy-axis is the Co$_2$MnSi(110)-direction.

Fig. 3 Electric resistivity $\rho(T)$ as a function of temperature for a NbN films at various nitrogen gas pressures. The superconducting critical temperature $T_c$ increases with the decrease of nitrogen gas pressures. The maximum $T_c$ is 16.0 K at the nitrogen partial pressure of 5.9%.

nitrogen partial pressures and $T_c$ was optimized at $N_2 = 5.9 \%$ during sputtering at ambient substrate temperature. Using NbN films optimized $T_c$, as $T_c = 16.0$ K, we have measured the electric resistivity $\rho(T)$ of both NbN and NbN/Co$_2$MnSi films under magnetic fields up to 14.5 T. Figures 4(a) and 4(b) show magnetic field dependence of the electric resistivities $\rho(T)$'s for NbN and NbN/Co$_2$MnSi films, respectively. As shown in Figs. 4(a) and 4(b), $T_c$'s of both films are equal to 16.0 K at $B = 0$ T, and $T_c$ decreases with increasing magnetic fields. However, the resistivity $\rho(T)$ behaviors under magnetic fields are different: (i) $T_c$ of the NbN/Co$_2$MnSi bilayer film decreases rapidly compared to that of the NbN film. (ii) The superconducting transition width $\Delta T$ of the NbN/Co$_2$MnSi bilayer film is narrower than that of the NbN film. The differences are obviously caused by

Fig. 4 Electric resistivity $\rho(T)$ under magnetic field $B$ applied perpendicular to MgO(001) substrate for (a) the NbN film and (b) the NbN/Co$_2$MnSi bilayer film.

Fig. 5 Upper critical field $B_c2(T)$ as a function of temperature for (a) NbN and (b) NbN/Co$_2$MnSi films. The magnetic field was applied to parallel and perpendicular to the MgO(001) substrate.
the additional ferromagnetic CoMnSi layer. Here we estimated the upper critical field $B_{c2}$ of the fabricated films from Figs. 4(a) and 4(b). Figure 5 represents the resulting $B_{c2}(T)$ at finite temperatures for (a) NbN and (b) NbN/CoMnSi films, where the magnetic field $B$ was applied in the direction parallel or perpendicular to the MgO(001) substrate. The upper critical field $B_{c2}(0)$ at 0 K was obtained by the following equation:

$$B_{c2}(T) = B_{c2}(0) \left(1 - \frac{T}{T_c}\right)^{\alpha},$$

(1)

where $B_{c2}(T)$ is the upper critical field at finite temperature, $B_{c2}(0)$ is the upper critical field at 0 K, $T_c$ is temperature and $\alpha$ is the number obtained from fitting analysis. The broken lines and dotted lines in Fig. 5 are the fitting results of Eq. (1) to the experimentally obtained $B_{c2}(T)$. The resulting parameter values of $B_{c2}(0)$ and $\alpha$ are given in Table 2. In this analysis, $T_c$ is fixed to 16.0 K, which was obtained from the electric resistivity measurements. The maximum $B_{c2}(0)$ was estimated to 31.8 T in the case of the applied magnetic field parallel to the MgO(001) substrate for the NbN film.

### 3.4 Differential conductance

Andreev reflection measurements using superconducting junctions are one of powerful methods used for investigating ferromagnetic half-metallic materials. Differential conductance $\sigma(V)$ was therefore measured for NbN/CoMnSi junctions. Figure 6 represents the resulting $\sigma(V)$ of a NbN/CoMnSi junction with the circular pillar of 100 nm in diameter for various temperatures. As shown in Fig. 6, the enhancement of zero-bias conductance peak (ZBCP) was observed. The dip structure approximately at 4 mV was caused by the superconducting energy gap $\Delta$ of the NbN layer, but this was larger than $\Delta = 2.9$ mV of the NbN films in the previous report. This is because the spectral structure in the differential conductance was smeared by additional broadening effects except for the thermal broadening; for example, finite lifetime of quasiparticles in superconducting NbN films. The inset of Fig. 6 illustrates temperature dependence of zero-bias conductance $\sigma_0(T)$ of the same junction in Fig. 6. The $\sigma_0(T)$ value decreased quickly just below $T_c$ and the $\sigma_0(T)$ behavior was similar to temperature dependence of the superconducting energy gap $\Delta(T)$ of the Bardeen-Cooper-Schrieffer (BCS) theory. The experimental result proves that the ZBCP originates in the superconductivity at the NbN/CoMnSi junction interface.

We here discuss our ZBCP data in the differential conductance, compared with the extended Blonder-Tinkham-Klapwijk (BTK) model. In the extended BTK model, if the counter-electrode is a half-metal ($P = 100\%$), normalized differential conductance $\sigma(V)\sigma_N = 0$ within the superconducting energy gap $\Delta$, due to the reduction of Andreev reflection. Here $\sigma_N$ is the differential conductance in the normal state. On the other hand, $\sigma(V)\sigma_N = 2$ within $\Delta$ even if the counter-electrode is a normal metal ($P = 0\%$). Therefore, such an enhancement of the ZBCP in Fig. 6 cannot be explained in the framework of the extended BTK theory. Hence, we were not unfortunately able to estimate the spin polarization of our epitaxial CoMnSi films by the Andreev reflection technique. Meanwhile, there are several theoretical expectations for the appearance of the ZBCP in half-metal/superconductor junctions: for example, the odd frequency pairs or impurities at a spin-active interface. To clarify this issue, further experimental and theoretical works are needed.

### 4. Conclusions

We have succeeded to fabricate fully-epitaxial superconducting NbN/half-metallic Heusler alloy CoMnSi bilayer films and their junctions for Andreev reflection measurements. The fabrication condition of the NbN/CoMnSi bilayer films was optimized, such as $T_c = 16.0$ K for NbN films and $M_s = 790$ emu/cm$^3$ for CoMnSi films, respectively. The enhancement of the ZBCP was observed in the differential conductance of NbN/CoMnSi junctions and the ZBCP disappeared above $T_c$. 

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**Table 2** Fitting results of the upper critical field $B_{c2}(T)$ for NbN and NbN/CoMnSi films using Eq. (1).

<table>
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<tr>
<th>Materials</th>
<th>$B_{c2}(0)$</th>
<th>$\alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NbN (B $\parallel$ MgO-sub.)</td>
<td>31.8</td>
<td>0.976</td>
</tr>
<tr>
<td>NbN (B $\perp$ MgO-sub.)</td>
<td>18.3</td>
<td>0.886</td>
</tr>
<tr>
<td>NbN/CoMnSi (B $\parallel$ MgO-sub.)</td>
<td>31.3</td>
<td>1.090</td>
</tr>
<tr>
<td>NbN/CoMnSi (B $\perp$ MgO-sub.)</td>
<td>17.2</td>
<td>0.855</td>
</tr>
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</table>

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![Fig. 6](image-url)

**Fig. 6** Differential conductance of the NbN/CoMnSi junction with the circular pillar of 100 nm in diameter. The enhancement of the ZBCP was observed. The inset shows the temperature dependence of the zero-bias conductance $\sigma_0(T)$. 

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