Epitaxial contact Andreev reflection spectroscopy of NbN/Co$_2$FeSi layered devices

Iduru Shigeta, Takahide Kubota, Yuya Sakuraba, Cor G. Molenaar, Joost N. Beukers, Shojiro Kimura, Alexander A. Golubov, Alexander Brinkman, Satoshi Awaji, Koki Takanashi, and Masahiko Hiroi

Citation: Appl. Phys. Lett. 112, 072402 (2018); doi: 10.1063/1.5007287
View online: https://doi.org/10.1063/1.5007287
View Table of Contents: http://aip.scitation.org/toc/apl/112/7
Published by the American Institute of Physics

Articles you may be interested in

Magnetic tunnel junctions with an equiatomic quaternary CoFeMnSi Heusler alloy electrode

Large moments in bcc Fe$_x$Co$_y$Mn$_z$ ternary alloy thin films

Perpendicular magnetic tunnel junctions with Mn-modified ultrathin MnGa layer

Mobility spectrum analytical approach for the type-II Weyl semimetal Td-MoTe$_2$

Precision measurement of the quantized anomalous Hall resistance at zero magnetic field

Temperature dependence of interlayer coupling in perpendicular magnetic tunnel junctions with GdO$_X$ barriers
Epitaxial contact Andreev reflection spectroscopy of NbN/Co$_2$FeSi layered devices

Iduru Shigeta,$^{1,a)}$ Takahide Kubota,$^{2,3}$ Yuya Sakuraba,$^4$ Cor G. Molenaar,$^5$ Joost N. Beukers,$^5$ Shojiro Kimura,$^6$ Alexander A. Golubov,$^{5,7}$ Alexander Brinkman,$^5$ Satoshi Awaji,$^6$ Koki Takanashi,$^{2,3}$ and Masahiko Hiroi$^1$

$^1$Department of Physics and Astronomy, Graduate School of Science and Engineering, Kagoshima University, Korimoto 1-21-35, Kagoshima 890-0065, Japan
$^2$Institute for Materials Research, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan
$^3$Center for Spintronics Research Network (CSRN), Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan
$^4$National Institute for Materials Science (NIMS), Sengen 1-2-1, Tsukuba 305-0047, Japan
$^5$Faculty of Science and Technology and MESA+ Institute for Nanotechnology, University of Twente, 7500 AE Enschede, The Netherlands
$^6$High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan
$^7$Moscow Institute of Physics and Technology, Dolgoprudny, Moscow Region 141700, Russia

(Received 2 October 2017; accepted 29 January 2018; published online 12 February 2018)

We investigated the spin polarization $P$ of Co-based Heusler alloy Co$_2$FeSi by epitaxial contact Andreev reflection (ECAR) spectroscopy using epitaxially grown superconductor NbN and Heusler alloy Co$_2$FeSi layered devices. Ferromagnetic Co$_2$FeSi possesses the highest Curie temperature ($T_C \approx 1100$ K) and the largest spontaneous magnetic moment ($p_s \approx 6 \mu_B$) in the class of Heusler alloys. The ECAR measurements revealed that the $P$ value of Co$_2$FeSi was $54 \pm 2\%$ with a finite barrier parameter $Z$, indicating that an intrinsic $P$ value in ECAR spectroscopy would exceed reported values in point-contact Andreev reflection spectroscopy. We therefore established not only the epitaxial integration of ferromagnetic Co$_2$FeSi with superconductor NbN on an MgO substrate but also the fabrication and evaluation techniques of their ECAR devices. This highly versatile superconducting spintronic system enables fundamental superconducting spintronic studies, and it is also a candidate for practical superconducting spintronic devices. Published by AIP Publishing, https://doi.org/10.1063/1.5007287

The study of superconducting and ferromagnetic bilayers, trilayers, and multilayers has great importance due to their potential applications ranging from fundamental superconducting spintronic studies$^{1,2}$ to practical superconducting spintronic devices, such as the spin Josephson junction$^{4,6,7}$ and superconductor-NbN should yield high injection-efficiency spin-selective ohmic contacts, thereby enabling the development of practical superconducting spintronic devices, such as the spin Josephson junction$^{4,6,7}$ and superconducting spin transistors$^{8,9}$ devices, with both low power consumptions and high speeds.

A number of studies using Co-based Heusler alloys have recently achieved large tunneling magnetoresistance (TMR) and giant magnetoresistance (GMR) effects originating from the half-metallic nature of Co-based Heusler alloys. Giant TMR ratios over 2600% at low temperature have been reported in magnetic tunnel junctions (MTJs) with epitaxial Co$_2$Mn$_{1.24}$Fe$_{0.16}$Si$_{0.84}$ electrodes with the crystalline MgO barriers.$^{10}$ Large MR ratios over 30% at room temperature (RT) have also been observed in current-perpendicular-to-plane giant magnetoresistive (CPP-GMR) devices using epitaxial Co$_2$Fe$_{0.4}$Mn$_{0.6}$Si ferromagnetic electrodes,$^{11,12}$ which are one order of magnitude higher than those reported in the CPP-GMR devices with conventional ferromagnetic layers such as CoFe or NiFe electrodes. In contrast, only few reports exist for epitaxial NbN/Heusler alloy layered films and their devices so far; for example, giant coercivity enhancement in NbN/Co$_2$FeSi bilayers,$^{13}$ efficient injection of pure spin currents in NbN/Co$_2$MnSi devices,$^{14}$ superconductivity in NbN/Co$_2$Fe$_{0.4}$Mn$_{0.6}$Si/Au trilayers,$^{15}$ and investigation of odd frequency pairs generated in highly spin polarized ferromagnetic Josephson devices.$^{16}$

Point-contact Andreev reflection (PCAR) spectroscopy is an alternative powerful technique to evaluate the spin polarization of half-metallic ferromagnets as it is simple to fabricate junctions and to apply model calculations.$^{17-19}$ The large values of the spin polarization in half-metallic Heusler alloys were reported to be $74 \pm 2\%$ for Co$_2$MnGe$_{0.75}$Ga$_{0.25}$ and $72 \pm 2\%$ for Co$_2$MnGa$_{0.5}$Ge$_{0.5}$ in PCAR spectroscopy. In principle, the contact interface in PCAR spectroscopy is
Heusler alloy Co$_2$FeSi possesses the highest Curie temperature at 450 K. The NbN/Co$_2$FeSi layered ECAR devices was measured by 4-electrode fitting was carried out using the spin polarization spectroscopy. The crystal structure of NbN/Co$_2$FeSi/Au trilayer films but also the spin polarization of their ECAR devices. NbN is a type-II superconductor of the high critical temperature ($T_C$ ≈ 16 K), the long coherence length $\xi(0) \approx 5$ nm, and the long penetration depth $\lambda(0) \approx 200$ nm. The NbN/Co$_2$FeSi layered devices including ultra-clean interfaces and evaluated the spin polarization of Co$_2$FeSi by ECAR spectroscopy.

In this letter, we investigated not only the structural, magnetic, and transport properties of epitaxial NbN/Co$_2$FeSi/Au trilayer films but also the spin polarization of their ECAR devices. NbN is a type-II superconductor of the high critical temperature ($T_C$ ≈ 16 K), the long coherence length $\xi(0) \approx 5$ nm, and the long penetration depth $\lambda(0) \approx 200$ nm. The NbN/Co$_2$FeSi layered devices including ultra-clean interfaces and evaluated the spin polarization of Co$_2$FeSi by ECAR spectroscopy.

Epitaxially grown layered samples including the NbN/Co$_2$FeSi interface were prepared using an ultra-high vacuum (UHV)-compatible magnetron sputtering system with a base pressure below $1 \times 10^{-7}$ Pa. The surface of an MgO substrate was thermally cleaned in the UHV chamber before depositing NbN and Co$_2$FeSi layers. First, a superconducting NbN layer was directly grown at RT on an MgO single-crystal substrate using a niobium target by reactive sputtering in nitrogen and argon mixture gas. A Heusler alloy Co$_2$FeSi layer was also deposited at RT followed by in situ annealing at 450 °C to promote chemical ordering. The thicknesses of the NbN and Co$_2$FeSi layers were controlled to be 100 nm and 5 nm, respectively. After the film was cooled to RT, the film was finally capped by a 3-nm-thick Au protective layer. All the procedures were continuously carried out inside the UHV chambers without breaking the vacuum to maintain the clean interfaces.

Fabricated trilayer films were patterned into a pillar shape for a CPP-type four-terminal device structure using electron beam lithography and Ar ion milling. The designed diameter of circular pillars changed from 60 nm to 4 μm on one substrate. The differential conductance $G(V)$ of the NbN/Co$_2$FeSi layered ECAR devices was measured by standard ac lock-in detection at a frequency of 1 kHz. The normalized conductance curve $G(V)/G_N$ was fitted to the modified Blonder-Tinkham-Klapwijk (BTK) model in order to evaluate the spin polarization $P$ of the conductive $sp$-electrons in the Heusler alloy Co$_2$FeSi. A multiple parameter fitting was carried out using the spin polarization $P$, the superconducting energy gap $\Delta$, the dimensionless barrier parameter $Z$, and the broadening parameter $\omega$ as the variable parameters.

The crystal structure of NbN/Co$_2$FeSi/Au trilayer films was confirmed at RT by X-ray diffraction (XRD) using monochromatic Cu-Kα radiation and reflection high-energy electron diffraction (RHEED). Figure 1(a) shows the 2θ profile for a NbN/Co$_2$FeSi/Au trilayer film. The result of the 2θ profile exhibits that the NbN layer is single phase within the resolution limits of the XRD. Figure 1(b) shows the rocking curve of the NbN(002) peak in Fig. 1(a). A full-width at half-maximum (FWHM) of 1.55° in the rocking curve suggests that the strain of the epitaxially grown NbN(001) layer on the MgO substrate is relatively small when compared to NbN films of different nitrogen concentrations with lower $T_C$. Here, we note that there are no Bragg reflections for the Co$_2$FeSi and Au layers in Fig. 1(a) because of the thin layer thickness of the order of a few nanometers which might be under the detection limit of the XRD facility. The epitaxial growth and the chemical ordering of the Co$_2$FeSi layer were therefore evaluated by using RHEED images. Figures 2(a) and 2(b) show the RHEED images of a Co$_2$FeSi layer surface along Co$_2$FeSi[100] and Co$_2$FeSi[110] directions. The RHEED images denote that the superlattice diffraction lines [arrows in Fig. 2(b)] from the L$_{2,1}$-ordered phase of Co$_2$FeSi were confirmed when an electron-beam was incident to the...
The XRD and RHEED measurements demonstrate the epitaxial growth of both NbN and Co\(_2\)FeSi layers. From the XRD patterns and RHEED images, the epitaxial relationships of the layered sample were confirmed as follows, from the substrate to top: MgO(001)[100] \(\rightarrow\) NbN(001)[100] \(\rightarrow\) Co\(_2\)FeSi(001)[110].

The magnetization \(M(H)\) curves of the NbN/Co\(_2\)FeSi/Au trilayer film were investigated at RT by using a vibrating sample magnetometer (VSM). The magnetic properties of the trilayer film arise from the Co\(_2\)FeSi layer, which is ferromagnetic at RT while the NbN and Au layers are paramagnetic. Figure 3 shows the \(M(H)\) curves measured for an unpatterned trilayer film with a 5-nm-thick Co\(_2\)FeSi layer. The blue and red lines correspond to the \(M(H)\) curves along the Co\(_2\)FeSi[100] and the Co\(_2\)FeSi[110] directions, respectively. The in-plane magnetic easy-axis is the Co\(_2\)FeSi[100] direction.

Before patterning into the device structure, the resistivity \(\rho(T)\) of the NbN/Co\(_2\)FeSi/Au trilayer film was measured by the dc four-terminal method under magnetic fields up to 17 T in the temperature range between 4.2 K and 300 K. Figure 4 shows temperature-dependent \(\rho(T)\) for magnetic fields applied parallel to the surface of a NbN/Co\(_2\)FeSi/Au trilayer film, varying in 1 T steps from 0 to 17 T. As shown in Fig. 4, the superconducting transition temperature \(T_c\) decreased monotonically with the increase in the magnetic fields, while the superconducting transition width \(\Delta T_c\) was almost unchanged. In Fig. 4, we obtained that the normal resistivity \(\rho_{20K}\) at 20 K was 69.2 \(\mu\Omega\)-cm and the residual resistivity ratio (RRR) was 1.12. \(T_c\) was defined as the temperature at which the resistivity dropped to half its normal state value and it was determined to be 16.3 K in the absence of a magnetic field.
of magnetic fields, which coincided with that of our 100-nm-thick NbN monolayer films. In addition, $T_c$ is comparable to that of the maximum value for NbN films deposited at RT.$^{24}$

The upper critical field $\mu_0H_{c2}(T)$ was also evaluated from the $\rho(T)$ measurements in Fig. 4. The inset of Fig. 4 presents the temperature dependence of the upper critical fields $\mu_0H_{c2}(T)$ and $\mu_0H_{c2}(0)$ of a NbN/Co$_2$FeSi/Au trilayer film in two magnetic field directions applied parallel and perpendicular to the surface of the trilayer film. For the out-of-plane magnetic fields, $T_c$ is rapidly suppressed with a broadening of the superconducting transition, which is in contrast to those observed in the in-plane magnetic fields. Such an anisotropy is ascribed to 2-dimensional (2D) geometry of the film, leading to contribution of the vortex motion in the out-of-plane magnetic field.

The upper critical field $\mu_0H_{c2}(0)$ at 0 K was determined from the Bardeen-Cooper-Schrieffer (BCS)-type function $\mu_0H_{c2}(T) = \mu_0H_{c2}(0)[1 - (T/T_c)^2]^{\beta}$, using the trilayer film’s $T_c$ with $\mu_0H_{c2}(0)$ and $\alpha$ and $\beta$ as fitting parameters.$^{30}$ We found $\mu_0H_{c2}(0) = 23.2$ T with $\alpha = 1.26$ and $\beta = 1.09$ in the parallel magnetic field (blue line) and $\mu_0H_{c2}(0) = 16.2$ T with $\alpha = 1.26$ and $\beta = 1.00$ in the perpendicular magnetic field (red line). Considering that the thickness $d_{NbN}$ of the NbN layer is 100 nm and its coherence length $\xi_{NbN}(0)$ at 0 K is about 5 nm, the superconductivity of the trilayer film should be in the three-dimensional (3D) regime, due to satisfying $d_{NbN} \gg \xi_{NbN}(0)$. Indeed, $\mu_0H_{c2}(T) \propto 1 - T/T_c$ was substantially fulfilled in both magnetic field directions, corresponding to $\alpha = 1$ and $\beta = 1$ in the 3D regime.$^{31}$

The spin polarization of ferromagnetic Heusler alloy Co$_2$FeSi was evaluated using NbN/Co$_2$FeSi layered ECAR devices, which were patterned from the trilayer films excellent in magnetic and superconducting properties. The differential conductance $G(V)$ of the ECAR devices, whose interface was well-defined at the atomic scale, was measured by ECAR spectroscopy. In this study, at least 20 different devices ranging in diameter from 60 nm to 1 mm were measured, whereas $G(V)$ was obtained for less than 100-nmdiameter devices. It was difficult for large size devices to apply bias voltage beyond the superconducting energy gap $\Delta$ because their device resistance decreased with the increase in the pillar diameter.

With regard to the ECAR devices analyzed by the modified BTK model, the typical device resistance is about 10 $\Omega$. Figure 5 shows the analytical result of the normalized $G(V)/G_N$ for a NbN/Co$_2$FeSi layered ECAR device at 4.2 K, indicating that the experimentally obtained $G(V)/G_N$ was in good agreement with that of the modified BTK model except for the region of the dip structure. As shown in Fig. 5, the peak and dip structures were observed at 2.33 mV and 4.13 mV, respectively. The temperature evolution of $G(V)$ was also measured to investigate the origin of the peak and dip structures. Figure 6 presents the intensity plot of $G(V)/G_N$ as a function of temperature and applied bias voltage for the same device in Fig. 5. We found that the peak and dip structures disappeared above $T_c$, indicating that these structures originate from the superconductivity. Hence, the peak corresponds to the superconducting energy gap $\Delta$, and the dip structure might be attributed to the superconducting critical current effect.$^{32}$

From the modified BTK analysis in Fig. 5, the spin polarization was determined to be $P = 55.0\%$ with $\Delta = 2.00$ meV, $\omega = 0.24$ meV, and $Z = 0.57$, as fitting parameters. In general, the intrinsic $P$ value of ferromagnets can be deduced by extrapolating $P$ down to $Z = 0$ in the $P(Z)$ plot.$^{29,33}$ However, since Figs. S1(a)–S1(c) (supplementary material) exhibit that the conductance spectral shapes were almost reproducible in the different devices on the same substrate and the $P$ values were practically independent of $Z$ ranging between 0.5 and 0.6, the $P$ values for individual ECAR devices were averaged out. We found the average value of the spin polarization $\langle P \rangle = 54\pm2\%$. Although it was reported that the intrinsic $P$ values of Co$_2$FeSi ranged from 49% to 55% in PCAR spectroscopy,$^{34-37}$ the intrinsic $P$ value of Co$_2$FeSi in ECAR spectroscopy would exceed the
reported values in PCAR spectroscopy by extrapolating down to $Z = 0$. This is because (i) $P$ increases monotonically with the decrease in $Z$; (ii) ECAR spectroscopy can determine an ideal intrinsic $P$ value in ferromagnets, due to the ultra-clean superconductor/ferromagnet interface, when compared with PCAR spectroscopy. Here, we note that the finite $Z$ value in the ECAR devices might suggest the possibility of little interdiffusion at the NbN/Co$_2$FeSi interface by the annealing process to promote chemical ordering of the Co$_2$FeSi layer. In addition, several reports exhibited that the half-metallicity can be realized for Co$_2$FeSi by the Fermi level tuning on the substitution for other elements with different valence electrons.\[38-40]\] Indeed, a high MR ratio was reported at RT for the Co$_2$Fe$_{0.4}$Mn$_{0.6}$Si based CPP-GMR devices.\[11,12\] Hence, the quaternary Heusler alloys based on Co$_2$FeSi are promising for half-metallic electrodes of superconducting spintronic devices.

In summary, we have demonstrated that Co-based Heusler alloy Co$_2$FeSi was epitaxially integrated with superconductor NbN on the MgO substrate while keeping high-quality magnetic and superconducting properties. The NbN/Co$_2$FeSi layered ECAR devices including ultra-clean interfaces were fabricated by patterning from the epitaxial trilayer films. The average value of the spin polarization $\langle P \rangle$ for Co$_2$FeSi was determined to be $54 \pm 2\%$ with the finite $Z$ by ECAR spectroscopy, indicating that the intrinsic $P$ value in ECAR spectroscopy would exceed the reported values in PCAR spectroscopy. The combination of the highly spin-polarized Heusler alloy with the superconductor enables fundamental superconducting spintronic studies. Since the $P$, $p_s$, and $T_c$ of Co-based Heusler alloys can be controlled by the substitution of their constituent elements, this highly versatile superconducting spintronic system is also a candidate for practical superconducting spintronic devices.

See supplementary material for the additional figures of the conductance spectral shapes.

We would like to thank K. Makise, K.-i. Matsuda, S. Kawabata, Y. Asano, and Y. Tanaka for helpful advice and discussions. I.S. acknowledges supports by JSPS KAKENHI Grant No. JP16K04933. A.A.G. acknowledges partial support by Ministry of Education and Science of the Russian Federation, Grant No. 14Y26.31.0007. This work was carried out under the Inter-University Cooperative Research Program of the Institute for Materials Research, Tohoku University (Proposal No. 16K0083). This work was performed at High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University (Project No. 16H0028).

---