Greatly enhanced generation efficiency of pure spin currents in Ge using Heusler compound Co$_2$FeSi electrodes

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Received January 8, 2014; accepted January 19, 2014; published online February 6, 2014

We show nonlocal spin transport in n-Ge-based lateral spin-valve devices with highly ordered Co$_2$FeSi/n$^+$.Ge Schottky tunnel contacts. Clear spin-charge signals in Hanle effect curves are demonstrated at low temperatures, indicating the generation, manipulation, and detection of pure spin currents in n-Ge. The obtained spin generation efficiency of $\sim 0.52$ is about two orders of magnitude larger than that for a previously reported device with Fe/MgO tunnel barrier contacts. Considering the spin-related behavior with temperature evolution, we infer that it is necessary to simultaneously demonstrate a high spin generation efficiency and improve the quality of the transport channel to realize Ge-based spintronic devices.

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Solid-state spintronic devices in a scalable solid-state framework have been proposed.1–4) For compatibility with existing electronic devices on semiconductor platforms, all-electrical means of generation, transport, and detection of spin-polarized carriers through semiconductor channels such as GaAs,5) Si,6) and Ge7) are important technologies. To realize low power consumption for actual applications, it is more effective to use Schottky tunnel contacts than insulating tunnel barrier contacts because of the low parasitic resistance in nanoscale devices.3,8–10)

Because high electron and hole mobilities are required in next-generation channel materials, Ge-based spintronics compatible with Si large-scale integration (LSI) has also been explored recently by many groups.7,11–15) To date, by using nonlocal spin-valve (LSV) measurements in Ge-based laterally configured devices, pure spin currents were generated and detected up to $\sim 200$ K.7,16,17) Unfortunately, above 100 K, spin manipulation via Hanle-type spin precession has not been realized in nonlocal four-terminal geometries. Even though devices with Fe/MgO tunnel barrier contacts were used,7,16) Hanle effect curves in nonlocal four-terminal geometries were observed only at less than 50 K. In general, even at low temperatures, the spin lifetime of the n-Ge channel is frequently estimated to be less than 1 ns,7,16) which is significantly shorter than those of n-GaAs5) and n$^+$.Si.18) The short spin lifetime can lead to a short spin diffusion length in the channel region. This means that, in lateral device structures, the experimental demonstration of pure spin current transport is more difficult in n-Ge than in n-GaAs and n$^+$.Si.

To date, the use of highly ordered Co$_2$FeSi$_{1-x}$Al$_x$ Heusler compounds as the spin injector and detector has been explored to improve the efficiency of spin generation in the GaAs channels,19,20) although n-GaAs channels cannot be compatible with Si LSI, where Co$_2$FeSi$_{1-x}$Al$_x$ compounds have a relatively large spin polarization like that of a half-metallic material.21–24) Because we have established a technique for growing highly ordered Co$_2$FeSi ($x = 0$) on Ge using low-temperature molecular beam epitaxy (MBE),25) we can combine the Co$_2$FeSi electrodes with the reported spin injection/detection techniques.14,17) If a large spin accumulation is generated in the n-Ge channel by using highly ordered Co$_2$FeSi, lateral transport with spin manipulation, i.e., the Hanle effect curve, can be observed at higher temperatures.

In this paper, using L2$_1$-ordered Co$_2$FeSi/n$^+$.Ge Schottky tunnel contacts, we demonstrate lateral transport of pure spin currents detected by four-terminal nonlocal Hanle effects in n-Ge up to 225 K. The spin generation efficiency is markedly enhanced and is about two orders of magnitude larger than that previously reported by Zhou et al.7) Co-based Heusler compounds will be a candidate for the metallic source and drain material in Ge-based spintronic applications. We also discuss the spin-related behavior with temperature evolution.

As schematically shown in Fig. 1(a), we fabricated n-Ge-based lateral spin-valve devices (LSVs) with Co$_2$FeSi/n$^+$.Ge Schottky tunnel contacts. The fabrication process is as follows. First, we formed a phosphorous (P)-doped n-Ge(111) channel ($P^+ \approx 10^{18}$ cm$^{-3}$) with a thickness of $\sim 50$ nm on undoped Ge(111) substrates ($\rho \approx 40 \Omega$cm) using an ion implantation technique and post-annealing at 700 °C. Then, an n$^+$.Ge(111) layer consisting of an Sb $\delta$-doped layer and a 5-nm-thick Ge epitaxial layer was grown by MBE at 400 °C,26) where the doping density of Sb was $2 \times 10^{14}$ cm$^{-2}$. After the fabrication of the n$^+$.Ge(111) layer, a 10-nm-thick Co$_2$FeSi epitaxial layer was grown on top of the n$^+$.Ge(111) layer by room-temperature MBE.27) Figure 1(b) shows a high-resolution cross-sectional transmission electron microscopy (TEM) image of the formed Co$_2$FeSi/n$^+$.Ge interface. The heterojunction is atomically flat, reducing the occurrence of interface states.28,29) We also observed (111) and (113) superlattice reflections in the nanobeam electron diffraction patterns of the Co$_2$FeSi layer [Fig. 1(c)], which result from the presence of L2$_1$-ordered structures. Thus, the Co$_2$FeSi Heusler compound electrodes were of high quality, as shown in our previous work.25) Figure 1(c) guarantees the quality of the n$^+$.Ge layer. To align the magnetic moments in the in-plane direction for the Hanle effect measurements, a polycrystalline Co layer with a thickness of $\sim 20$ nm was deposited on the Co$_2$FeSi layer by electron beam evaporation.

Conventional electron beam lithography, Ar$^+$ ion milling, and reactive ion etching processes were used to fabricate four-terminal LSVs. The size of each contact is presented in Fig. 1(a) (0.3 $\times$ 100 and 0.5 $\times$ 100 µm$^2$), and the center-to-center distance ($L$) between the Co$_2$FeSi/n$^+$.Ge contacts was 1.0 µm. The current–voltage ($I$–$V$) characteristics of the Co$_2$FeSi/n$^+$.Ge junctions showed almost no rectifying
behavior at room temperature, indicating tunneling conduction of electrons through the Co$_2$FeSi/n$^+$/Ge interface. Although we confirmed no change in the forward bias current with temperature variation, small decreases in the reverse bias current were observed with decreasing temperature. Very similar features were already seen in our previous work on the n$^+/Ge$ layer. To avoid changes in the interface resistance with changing temperature, we concentrate on measurements in the forward bias condition for the Co$_2$FeSi/n$^+$/Ge contacts. In this study, the resistance area product (RA) of the Co$_2$FeSi/n$^+$/Ge interface was nearly constant at $\sim 10^9 \Omega \mu m^2$. To precisely understand the spin-related phenomena, we also made a microfabricated Hall bar device with the same n-Ge channel and AuSb ohmic contacts. The electron carrier density ($n$) of the fabricated channel was experimentally estimated from electrical Hall effect measurements. The estimated values were $n \sim 2.5 \times 10^{18} \text{cm}^{-3}$ at 300 K and $n \sim 3.3 \times 10^{17} \text{cm}^{-3}$ at 150 K. As a result, the electron mobility ($\mu$) of this channel is $\sim 286 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ at 300 K and $\sim 665 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ at 150 K.

Figure 2(a) shows the nonlocal magnetoresistance ($\Delta R_{NL} = \Delta V_{NL}/I$) measured at $I = +1.0 \text{ mA}$ at 150 K, where the positive sign of $I$ ($I > 0$) means that the electrons are extracted from the Ge channel into Co$_2$FeSi (the spin extraction condition) through the Schottky tunnel barrier. By applying in-plane magnetic fields ($B_x$), a large spin-valve signal ($\sim 100 \text{ mO}$) can be seen even at 150 K. This spin-valve feature is attributed to the change in the magnetization direction of the two different Co$_2$FeSi contacts used here between nearly parallel and antiparallel configurations. Note that the magnitude of the nonlocal signal is almost compatible with that observed at 4 K for a device (Device B, reported by Zhou et al.$^7$) with Fe/MgO tunnel contacts and $L = 1.0 \mu m$. This feature implies that, even at a higher temperature, we demonstrated spin accumulation in the n-Ge channel equivalent to that obtained by Zhou et al.$^7$. Using this nonlocal four-terminal geometry, we also applied an out-of-plane magnetic field ($B_z$) under parallel and antiparallel magnetic configurations for the Co$_2$FeSi electrodes and recorded $\Delta R_{NL}$ as a function of $B_z$. As a result, clear Hanle-type spin precession curves, which are evidence of the generation, manipulation, and detection of pure spin currents in the n-Ge channel, can be seen in Fig. 2(b). At even higher temperatures, several devices exhibited such Hanle effect curves.

By fitting the Hanle effect curves with the one-dimensional spin drift diffusion model,$^5$ the approximate spin lifetime ($\tau_s$) in the channel region can be extracted. The model used for our device is

$$\Delta R_{NL}(B_z) \propto \pm \int_0^\infty \frac{1}{\sqrt{4\pi Dt}} \exp \left( -\frac{L^2}{4Dt} \right) \cos(\omega t) \exp \left( -\frac{t}{\tau_s} \right) dt,$$

where $\pm$ is the sign, which depends on the magnetization configuration (parallel or antiparallel); $D$ is the diffusion constant of the Ge channel; $\omega = \gamma_B B_z / h$ is the Larmor frequency; $g$ is the electron $g$-factor ($g = 1.56$);$^{31}$ and $\mu_B$ is the Bohr magneton. Representative fitting results are indicated by solid curves in Fig. 2(b). At 150 K, the estimated $\tau_s$
and $D$ values for the n-Ge channel used are $\sim$420 ps and $\sim$8.3 cm$^2$ s$^{-1}$, respectively. The obtained $D$ value is consistent with the experimentally estimated value of 8.6 cm$^2$ s$^{-1}$ at 150 K from the Einstein relation, $D = (k_B T/\mu_e)$, where $k_B$ is Boltzmann’s constant. We will comment on $\tau_s$ later.

From the $|\Delta R_{NL}|$ shown in Fig. 2(b), we also estimate the spin generation efficiency of this device with the Co$_2$FeSi/n$^+$-Ge contacts. The $|\Delta R_{NL}|$ detected by nonlocal four-terminal geometry can generally be expressed as:

$$|\Delta R_{NL}| = \frac{P_{gen} P_{det} \rho_N \Delta N}{S} \exp\left(-\frac{L}{\lambda_N}\right),$$  

(2)

where $P_{gen}$ and $P_{det}$ are the generated and detected spin polarizations, respectively, at the ferromagnetic electrodes. Thus, $(P_{gen} \times P_{det})^{1/2}$ can be regarded as the approximate spin generation efficiency. Further, $\rho_N$ and $\lambda_N$ are the resistivity and spin diffusion length of the nonmagnetic channel. In this study, $\rho_N$ and $\lambda_N$ at 150 K are 28.2 m$\Omega$ cm and 0.59 $\mu$m, respectively, where $\lambda_N = \sqrt{D/\mu_e}$. $S$ is the cross section of the Ge channel ($\sim$5.0 $\mu$m$^2$), and we used $L = 1.0$ $\mu$m. Using these parameters, we can obtain $(P_{gen} \times P_{det})^{1/2} \approx 0.12$ ($\sim$12%), which is about two orders of magnitude larger than that for an Fe/MgO/GaAs device (Device B) at 4 K in Ref. 7. Although $\rho_N$ is about $\sim$100 $\Omega$ cm in Ref. 7 because of the carrier freeze-out at 4 K, we infer that $\rho_N$ at 150 K in Ref. 7 can become several tens of m$\Omega$ cm, which is the same order of magnitude. Because they have not observed Hanle effect curves above 100 K, we can judge that the spin generation efficiency in this study is sufficiently higher than that in Ref. 7. Note that the order of the spin generation efficiency in this study is consistent with that for a GaAs-based device with L$_21$-ordered Co$_2$FeSi electrodes ($\sim$0.16). Even for Ge, the L$_21$-ordered Co$_2$FeSi electrodes can open a possible path to highly efficient spin generation even in semiconductor channels.

Next, we discuss the spin signals of this device with temperature evolution. Because the temperature-dependent $RA$ can affect the spin signals, as shown in our previous work, we simply focus on the data that can be measured under the terminal geometry can generally be expressed as:

$$\tau_s = \frac{D}{\pi D/\mu_e},$$

(3)

We should also recognize that spin-flip scattering due to the heavily doped Sb near the Co$_2$FeSi/n$^+$-Ge interface strongly influences the spin generation and detection, giving rise to much shorter $\tau_s$ values. Taking this special situation for our devices into account, we infer that the $\rho_N$ value with temperature evolution is limited by extrinsic factors rather than the conventional spin relaxation mechanism.

According to Eq. (2), the magnitude of $\Delta R_{NL}$ depends not only on $\lambda_N (\sqrt{D/\mu_e})$ but also on $\rho_N$. Thus, we should reconsider the change in $\rho_N$ with temperature evolution. The inset of Fig. 3(c) shows the temperature dependence of $\rho_N$ for the n-Ge channel used in this study. Because the electron density of the Ge channel used here decreases with decreasing temperature, $\rho_N$ decreases monotonically. As expected, this feature is close to the temperature-dependent $\Delta R_{NL}$ in the main panel of Fig. 3(c). From these considerations, we can judge that the contribution of $\rho_N$ to $\Delta R_{NL}$ is greater than that of the spin relaxation in the Ge channel in this study.

We finally comment on the short $\lambda_N$ of $\sim 0.6$ $\mu$m at low temperatures. As described above, our Ge channels were fabricated by a conventional ion implantation technique,
and we then used etching processes to fabricate devices for transport measurements. Thus, the lack of optimization of these processes yielded damaged channels with relatively poor mobility (≈300 cm² V⁻¹ s⁻¹ at 300 K) compared to bulk wafers (more than ≈1000 cm² V⁻¹ s⁻¹). As a result, the D values of ≈10 cm² s⁻¹ are relatively low at low temperatures, leading to short \( \Delta G \). Even though we detected the lateral transport of pure spin currents in n-Ge at higher temperatures by using Heusler compound Co₂FeSi electrodes because of the large enhancement in the spin generation efficiency, a marked improvement of \( \Delta G \) might also be required for achieving Ge spintronics. The fabrication processes for lateral structures are currently being optimized to obtain a long \( \Delta G \) in Ge channels for actual device applications.

In summary, we presented a greatly enhanced spin generation efficiency in n-Ge by using L₂₁−ordered Co₂FeSi Heusler compound electrodes. Thanks to this technological development, we demonstrated the lateral transport of pure spin currents in n-Ge up to 225 K. For actual Ge spintronic applications, improvements in the device fabrication processes are also required.

Acknowledgment
This work was supported in part by the Industrial Technology Research Grant Program from NEDO and a Grant-in-Aid for Scientific Research (A) (No. 25246020) from the Japan Society for the Promotion of Science (JSPS). S.Y. acknowledges a JSPS Research Fellowship for Young Scientists.

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