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Temperature-independent spin relaxation in heavily doped *n*-type germanium

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We experimentally study the spin relaxation mechanism in heavily doped *n*-type germanium (Ge) layers by electrically detecting pure spin current transport. The spin diffusion length (λ_{Ge}) in heavily doped *n*-type Ge layers at 125 K is less than 0.7 μm, much shorter than that expected in the recent study by Dushenko *et al.* We find that the spin relaxation time τ_{s} is independent of temperature in the range of 8 to 125 K, which can be interpreted by the recent theory by Song *et al.* This study clarifies that the spin-relaxation mechanism at low temperatures in degenerate Ge is dominated by extrinsic scattering with impurities.

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Spin relaxation in semiconductors has been investigated by using electron spin resonance [1–3], optical spin generation and detection [4], and electrical spin-transport [5] measurements. For spintronics [6], the mechanism of spin relaxation is also important to understand the physical origin of the spin-related phenomena in device structures. Thanks to the development of the electrical spin transport measurements with hot-electron spin injection [7], even spin relaxation in group-IV semiconductors such as undoped silicon (Si) [8–10] and germanium (Ge) [11–13] was intensively explored and excellent agreement between experiment and theory was reported. In addition, for the case of Ge, the optical techniques enabled researchers to systematically examine the spin-relaxation times (τ_{s}) of both electrons and holes in various conditions [14–18].

Because there are few reports on the pure spin current transport in doped Ge [19–22], the spin-relaxation mechanism of diffusive spin currents in Ge indicates a puzzling situation. By electrically measuring nonlocal (NL) magnetoresistance in lateral spin valves (LSVs) [19–21], pure spin current transport was observed only at low temperatures. For *n*-type Ge (*n*-Ge) layers with an electron concentration *n* of $\sim 10^{16}$ cm^{-3} [19,20], weak temperature dependence of τ_{s} ($\sim$1 ns) was seen in the range of 4 to 10 K and phonon-induced spin relaxation was observed at higher temperatures. For *n*-Ge layers with $n \sim 10^{18}$ cm^{-3} [21], nearly constant τ_{s} values ($\sim$0.4 ns) were obtained in the range of 150 to 225 K. Recently, Dushenko *et al.* reported pure spin current transport detected through the inverse spin Hall effect in the lateral Py/*n*-Ge/Pd devices with an *n* of $\sim 10^{19}$ cm^{-3} [22]. They claimed that room-temperature spin diffusion length (λ_{Ge}) is $\sim$0.66 μm and λ_{Ge} is enhanced up to $\sim$1.3 μm at 130 K in heavily doped *n*-Ge (n^+-Ge) layers, meaning the presence of a temperature dependent spin-relaxation mechanism. They also showed that τ_{s} in the range of 130 to 297 K showed weak temperature dependence with the $1/\sqrt{T}$ behavior, explained by the theory of the donor-induced spin-relaxation mechanism in multivalley semiconductors, reported by Song *et al.* [23]. However, the theory in Ref. [23] claims that τ_{s} depends only on the donor concentration in such degenerate ($n \sim 10^{19}$ cm^{-3}) conditions because the Fermi energy ϵ_{F} in n^+-Ge is larger than the conduction-electron energy. In this context, the relaxation mechanism with respect to the diffusive spin currents in heavily doped Ge is still an open question.

In this article, using pure spin current transport measurements in LSVs, we study the spin-relaxation mechanism in n^+-Ge layers. We find that the estimated λ_{Ge} at 125 K is less than 0.7 μm. Unlike the previous aspects in Ref. [22], the τ_{s} values are independent of temperature in the range of 8 to 125 K. We discuss the spin-relaxation mechanism in n^+-Ge layers in detail.

We fabricated LSVs with ferromagnet (FM)/n^+-Ge Schottky-tunnel contacts, as illustrated in Fig. 1(a). First, we formed an undoped Ge(111) layer ($\sim$28 nm) grown at 350 °C (LT-Ge) on the commercial undoped Si(111) substrate ($\rho \sim$ 1000 Ω cm), followed by an undoped Ge(111) layer ($\sim$70 nm) grown at 700 °C (HT-Ge), where we utilized the two-step growth technique by molecular beam epitaxy (MBE) [24,25]. These layers on Si(111) have *p*-type conduction and relatively high resistivity compared with the following n^+-Ge channel layer. Then, as a channel layer, a 70-nm-thick phosphorous (P)-doped n^+-Ge(111) layer (doping concentration $\sim 10^{19}$ cm^{-3}) was grown by MBE at 350 °C on top of it. Since there exists a *p*- *n* junction with a depletion layer, we can ignore the parallel conduction and spin diffusion from the channel layer into the HT-Ge layer at low temperatures. To promote the tunneling conduction at the FM/n^+-Ge interface, a P δ-doped Ge layer with an ultrathin Si insertion layer was grown on top of the n^+-Ge layer [26]. As a spin injector and detector, we grew a Co$_2$FeSi$_{0.5}$Al$_{0.5}$(9 nm)/CoFe(1 nm) bilayer on top of it by well-established low-temperature MBE techniques [27,28]. There was almost no reaction layer between FM and Ge layers, as shown in our previous work [21]. Conventional processes were used to fabricate LSVs, as also mentioned in Ref. [21]. The sizes of the FM/n^+-Ge contacts are 0.4×5.0 μm^2 and

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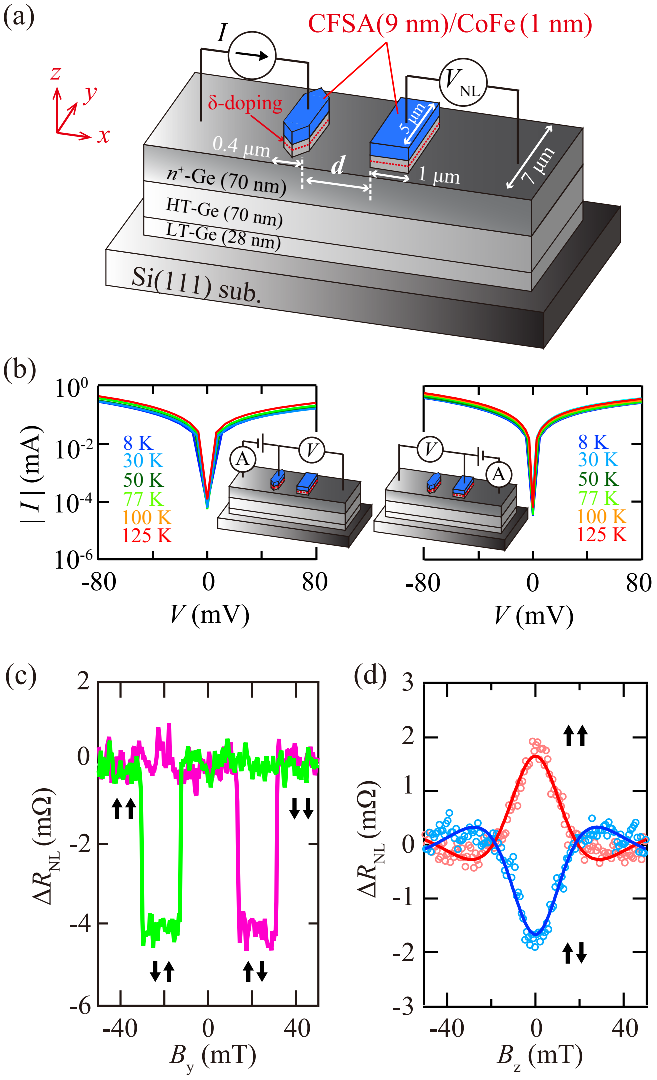

FIG. 1. (a) Schematic illustration of the fabricated n^+-Ge based LSV. (b) I-$V_{\rm int}$ characteristics at various temperatures for spin injector and detector. (c) Nonlocal magnetoresistance curve and (d) Hanle-effect curves for the parallel and antiparallel magnetization configurations at 8 K at $I = -1.0$ mA.

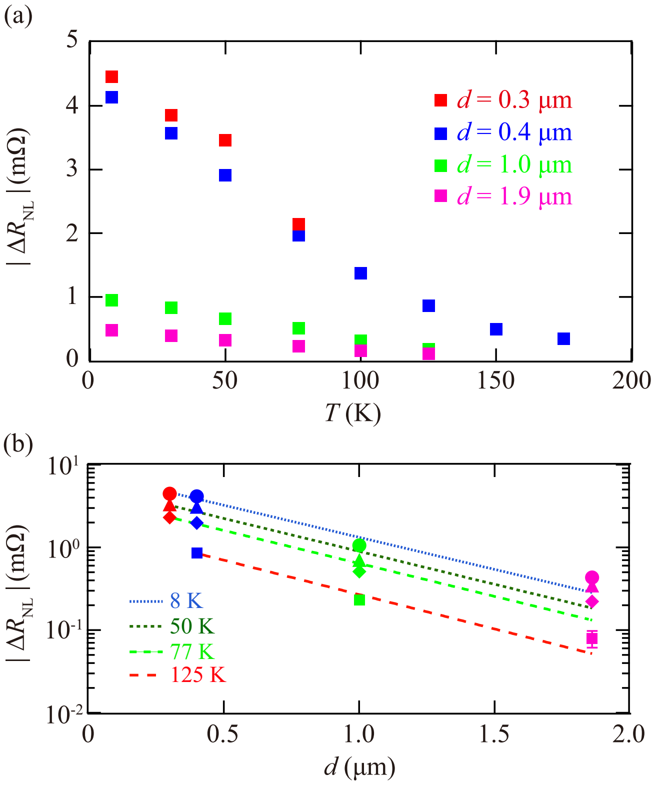

FIG. 2. (a) Temperature-dependent $|\Delta R_{\rm NL}|$ for the LSVs with $d = 0.3$, 0.4, 1.0, and 1.9 μm. (b) d dependencies of $|\Delta R_{\rm NL}|$ at various temperatures.

1.0×5.0 μm^2, and the edge-to-edge distances d between the FM/n^+-Ge contacts are designed to be 0.3, 0.4, 1.0, and 1.9 μm.

We confirmed that the current-voltage (I-V) characteristics of the FM/n^+-Ge junctions, as presented in Fig. 1(b), show almost no rectifying behavior at various temperatures, indicating the demonstration of the tunneling conduction of electrons through the FM/n^+-Ge interfaces. Thus, we can conduct NL magnetoresistance measurements [29–31] even at low temperatures irrespective of the influence of the strong Fermi-level pinning at FM/n-Ge [32]. Figure 1(c) shows a NL magnetoresistance ($\Delta R_{\rm NL} = \Delta V_{\rm NL}/I$) for the LSV with $d = 0.4$ μm, measured at $I = -1.0$ mA at 8 K, where the negative sign of I ($I < 0$) means that the spin-polarized electrons are injected from FM into n^+-Ge. A clear spin-valve signal with a magnitude ($|\Delta R_{\rm NL}|$) of 4.13 mΩ can be seen by applying an in-plane magnetic field B_y. We also applied an out-of-plane magnetic field B_z under parallel and antiparallel magnetization configurations between the FM electrodes to record $\Delta R_{\rm NL}$ as a function of B_z. In Fig. 1(d) we see evident Hanle-type spin precession curves, indicating the generation, manipulation, and detection of pure spin currents in the n^+-Ge layer by all-electrical means [5]. This means that the data shown in the following are reliable to discuss the spin-relaxation phenomena in n^+-Ge layers.

The temperature dependence of the NL spin signals is explored for various LSVs in Fig. 2(a). The $|\Delta R_{\rm NL}|$ values are gradually decreased with raising temperature in all the LSVs with various d. Although the spin-injection contact of the LSV with $d = 0.3$ μm was broken at 100 K and we could not measure the NL spin signals at higher temperatures, the spin transport could not be seen at room temperature. For the LSVs with $d = 1.0$ μm and $d = 1.9$ μm, since we observed both NL spin signals and Hanle curves up to 125 K, we can guarantee the reliable spin transport from 8 to 125 K to deduce the spin-diffusion length $\lambda_{\rm Ge}$. In Fig. 2(b) we plot $|\Delta R_{\rm NL}|$ versus d at various temperatures. In this temperature regime, since exponential reductions in the $|\Delta R_{\rm NL}|$ values are observed, we can estimate $\lambda_{\rm Ge}$ by using the following equation [29,30]:

$$|\Delta R_{\rm NL}| = \frac{|P_{\rm inj}||P_{\rm det}|\rho_{\rm Ge}\lambda_{\rm Ge}}{S}\exp\left(-\frac{d}{\lambda_{\rm Ge}}\right), \qquad (1)$$

where $P_{\rm inj}$ and $P_{\rm det}$ are spin polarizations of the electrons in Ge created by the spin injector and detector, respectively, $\rho_{\rm Ge}$ is the resistivity (1.74 mΩ cm $\leqslant \rho_{\rm Ge} \leqslant$ 1.83 mΩ cm), and S is the cross section (∼0.49 μm^2) of the n^+-Ge layer. So far,

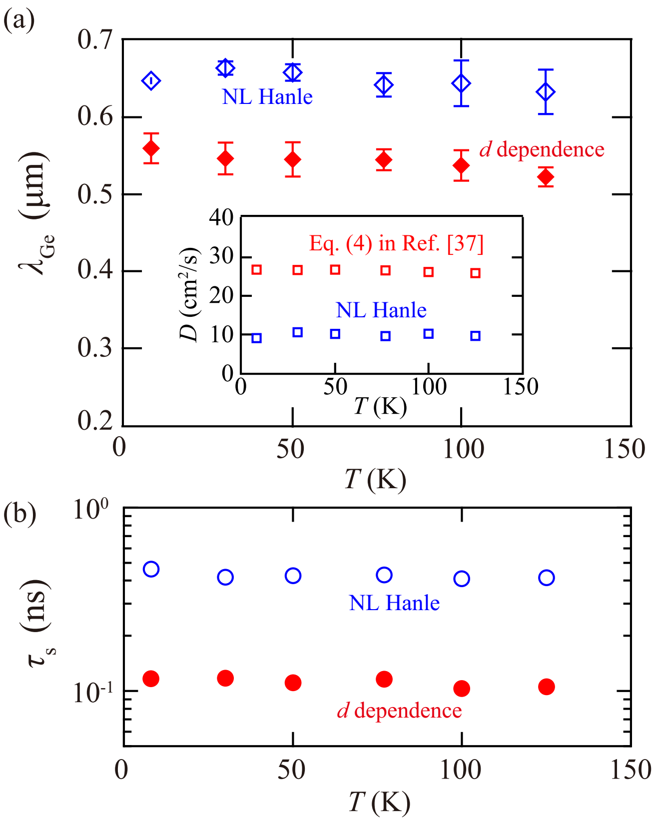

FIG. 3. Temperature dependencies of (a) $\lambda_{\rm Ge}$ and (b) τ_s, estimated by d dependencies (red) and Hanle-effect curves (blue) of NL spin signals. The inset of Fig. 3(a) shows the temperature dependence of D, estimated from Eq. (4) in Ref. [37] (red squares) and nonlocal Hanle-effect curves (blue squares).

while the estimations of $\lambda_{\rm GaAs}$ or $\lambda_{\rm Si}$ from the contact-distance dependence of NL spin signals have already been reported in GaAs- or Si- based LSVs [33–36], it has never been reported in Ge-based LSVs because of experimental difficulties. As a consequence, reliable $\lambda_{\rm Ge}$ values were given by all-electrical means and we clarified $\lambda_{\rm Ge}$ at various temperatures as follows:

The red symbols in Fig. 3(a) are $\lambda_{\rm Ge}$ estimated from the d dependence of NL spin signals. The $\lambda_{\rm Ge}$ value slightly decreases with rising temperature from 8 to 125 K and $\lambda_{\rm Ge}$ at 125 K is $0.52 \pm\ 0.01$ μm. The observed weak temperature dependence and $\lambda_{\rm Ge} \sim\ 0.52$ μm at 125 K are inconsistent with the temperature dependence and $\lambda_{\rm Ge} \sim 1.3$ μm at 130 K in Ref. [22]. Taking into account the weak temperature dependence of $\lambda_{\rm Ge}$, we can recognize the decay of the spin signals in Fig. 2(a) as a consequence of the reduction in $P_{\rm inj}$ and $P_{\rm det}$ with increasing temperature. Surely, the value of $|P_{\rm inj}| \times |P_{\rm det}|\ = \sim 0.0004$ at 8 K was down to $\sim$0.0001 at 125 K. From the $\lambda_{\rm Ge}$ value at each temperature, we can also deduce τ_s by using the relation, $\lambda_{\rm Ge}\ =\ \sqrt{D\tau_s}$, where D is the diffusion constant. Because degenerate semiconductors were used in this work, we should estimate D values from Eq. (4) in Ref. [37]. The estimated D as a function of temperature is shown in the inset of Fig. 3(a), where the electron mobility in the n^+-Ge layer was measured at each temperature. Using $\lambda_{\rm Ge}$ and D in Fig. 3(a), we can calculate τ_s at each temperature, as displayed in the red symbols in Fig. 3(b). Note that we see the temperature-independent τ_s and the obtained τ_s values ($\sim$0.1 ns) are slightly smaller than those in previous works [19–22].

We can also extract the τ_s and D values by fitting the Hanle-effect curves with the one-dimensional spin drift diffusion model [30], which is expressed as follows:

$$\Delta R_{\rm NL}(B_z) = \pm A \int_0^\infty \phi(t)\cos(\omega_L t)\exp\left(-\frac{t}{\tau_s}\right)dt, \tag{2}$$

where $A = (P_{\rm inj}P_{\rm det}\rho_{\rm Ge}\lambda_{\rm Ge})/S$, $\phi(t) = \frac{1}{\sqrt{4\pi Dt}}\exp(-\frac{d^2}{4Dt})$, ω_L $(=g\mu_{\rm B}B_z/\hbar)$ is the Larmor frequency, g is the electron g factor ($g = 1.56$) in Ge [38], and $\mu_{\rm B}$ is the Bohr magneton. The solid curves in Fig. 1(d) show representative results of the fitting with Eq. (2). The τ_s and D values at 8 K are estimated to be 0.46 ± 0.04 ns and 9.0 ± 0.8 cm^2/s, respectively, for the n^+-Ge layer. We also plot $\lambda_{\rm Ge}$ and τ_s values estimated from the Hanle-effect curves as blue symbols in Figs. 3(a) and 3(b), respectively, at each temperature. Although there are slight differences between the two estimation methods, we can judge that the τ_s values are independent of temperature in the range of 8 to 125 K. From these analyses, the temperature dependence of τ_s measured by electrical spin injection and detection is indeed inconsistent with Ref. [22].

In regards to this study, we discuss the spin-relaxation mechanism in detail. The recent theory by Song *et al.* [23] suggests that, in addition to the Elliott–Yafet-type phonon-induced spin relaxation [11], the central-cell potential of impurities can cause the short-range scattering of conduction-electron spins in multivalley semiconductors. Considering these two contributions of electron spin relaxation in doped Ge, we can roughly assume the following expression associated with scattering rate $\frac{1}{\tau_s}$:

$$\frac{1}{\tau_s} = \frac{1}{\tau_{\rm imp}} + \frac{1}{\tau_{\rm phon}}, \tag{3}$$

where $\tau_{\rm imp}$ and $\tau_{\rm phon}$ are donor-induced and phonon-induced spin-relaxation times. Here the $\tau_{\rm imp}$ and $\tau_{\rm phon}$ values include material and physical properties of Ge [11,23]. $\frac{1}{\tau_{\rm phon}}$ is directly proportional to $\sum \frac{1}{\sqrt{b}} \frac{a\{(k_{\rm B}T/b)+1\}}{\exp(b/k_{\rm B}T)-1}$ [11], here a and b are the material- and crystal-related spin-flip scattering constant and phonon energy, respectively, and $k_{\rm B}$ is Boltzmann's constant. Also, $\frac{1}{\tau_{\rm imp}}$ is temperature independent ($\epsilon_{\rm F} \gtrsim k_{\rm B}T$) or is directly proportional to $\sqrt{T}$ ($\epsilon_{\rm k} \approx\ k_{\rm B}T$), as predicted in Ref. [23]. For n^+-Ge ($n \sim\ 10^{19}$ cm^{-3}), $\epsilon_{\rm F}$ is evidently larger than $k_{\rm B}T$ in the range of 8 to 125 K, so that we should not consider the $\sqrt{T}$ behavior [23]. Because this study clearly indicates no contribution of $\tau_{\rm phon}$ to τ_s in Fig. 3(b), we can understand that τ_s within a range of 8 K $\leqslant T \leqslant$ 125 K is only attributed to the donor-induced spin-relaxation mechanism in Ref. [23]. The experimental data in Fig. 3(b) are approximately consistent with the theoretically calculated $\tau_{\rm imp}$ value of $\sim$0.12 ns from Eq. (4) in Ref. [23] when we assume $a_{\rm B} = 6.45$ nm [2], $n = 8.2 \times 10^{18}$ cm^{-3}, obtained by Hall-effect measurements [39], $m_e\ =\ 0.16m_0$ [40], and $\Delta_{\rm so} = 0.11$ meV, where $a_{\rm B}$, m_e, and $\Delta_{\rm so}$ are the electron Bohr radius, the electron effective mass in heavily doped Ge, and the spin-orbit-coupling–induced splitting of the triply degenerate $1s(T_2)$ donor state in Ge, respectively. When we assume $\Delta_{so} = 0.062$ meV, we can also obtain $\tau_{imp} \sim 0.47$ ns. Here, the assumed Δ_{so} values are much smaller than the valley-orbit–induced singlet-triplet splitting in P-doped Ge of $\sim$2.82 meV [41]. From these considerations, this study clarifies that the temperature-independent τ_s is dominated by the short-range spin scattering due to the central-cell potential of impurities in Ge [23]. Experimentally, the temperature-independent D, which is strongly related to the channel mobility, results in the temperature-independent τ_s. To achieve longer λ_{Ge} values, one should use high-mobility Ge layers as a spin-transport channel.

Finally, we comment on the experimental difficulties of the investigation of the n dependence of τ_s and λ_{Ge} by all-electrical means. When we used moderately doped Ge layers ($n \leqslant 10^{18}$ cm^{-3}), the device resistance including the designed contact resistance became two or three orders of magnitude larger than that with an n of 10^{19} cm^{-3}. This situation is compelled to the formation of the large-sized contacts to reduce the electrical noises in the measurements. Unfortunately, this limitation causes the poor control of the magnetization configuration between spin injector and detector [21]. Also, such large devices could not guarantee the validity of the d dependence of spin signals based on the one-dimensional spin diffusion model. Therefore, for n-Ge ($n \leqslant 10^{18}$ cm^{-3}), the reliable report on the electrical measurements is likely to be an only method for the hot-electron techniques [12]. In Ref. [21], we already reported $\lambda_{Ge} \sim 0.59$ μm and $\tau_s \sim 0.42$ ns at 150 K, estimated from the Hanle-curve measurements, for n-Ge with $n \leqslant 10^{18}$ cm^{-3}. By a comparison of these values and Fig. 3(b), we can explain that the data in Ref. [21] are influenced by the interfacial heavily doped layer with an n of 10^{19} cm^{-3} between the ferromagnetic material and the n-Ge layer. In the optical experiment for n-Ge with an $n \sim 10^{17}$ cm^{-3} [17], both the donor-induced and phonon-induced spin relaxations were considered and τ_s was $\sim$5 ns [17] at around 100 K. We can tentatively compare the data with those in this work from the perspective of the donor concentration dependence of τ_s. As a result, τ_s for $\sim 10^{17}$ cm^{-3} is $\sim$5 ns [17] and τ_s for $\sim 10^{19}$ cm^{-3} is $\leqslant$0.5 ns, respectively, at around 100 K. This relationship is likely to be consistent with the presence of the impurity induced spin-relaxation mechanism in n-Ge [23].

In conclusion, we experimentally studied the spin-relaxation mechanism in n^+-Ge layers at low temperatures. We found that, unlike the previous report in Ref. [22], the temperature-independent τ_s from 8 to 125 K was obtained, consistent with the recent theory in Ref. [23]. In a future work, the difference in λ_{Ge} between this work and Ref. [22] should be discussed.

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