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Large impact of impurity concentration on spin transport in degenerate *n*-Ge

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We experimentally show that the spin relaxation in degenerate n-type germanium (n^+-Ge) depends strongly on the concentration of the donor impurity (N_d) at low temperatures. From measuring nonlocal spin signals for various lateral spin-valve devices at 8 and 77 K, the spin diffusion length (λ_{Ge}) of n^+-Ge can be estimated as a function of carrier concentration (n), i.e., $N_d \approx n$ ($\sim 10^{18}$ cm^{-3}). We clearly find a large change in λ_{Ge} from 1.43 to 0.56 μm within a relatively narrow range of n at 8 K. The experimental findings are interpreted quantitatively in terms of a recent theory based on donor-driven spin scattering in multivalley conduction bands in Ge.

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Spin injection, transport, and detection in semiconductors are important aspects for understanding spin-related physical phenomena in semiconductor spintronics applications [1–8]. Recently, there have been many reports on the spin injection into silicon (Si) and germanium (Ge) because these materials are compatible with the existing Si-based complementary metal-oxide-semiconductors [4–6,9–27]. For the case of Ge, optical methods for investigating the generation of spin polarization have particularly been utilized owing to the optically accessible multivalley nature of the conduction bands [28–30]. Also, studies of spin transport and its relaxation in Ge are fascinating because of its relatively high electron and hole mobility compared to Si [13,17,20,26]. For intrinsic Ge, Li *et al.* theoretically reported the detailed contribution of phonon-induced intervalley and intravalley spin scattering to the temperature-dependent spin lifetime [31]. In addition, they experimentally showed anisotropic spin relaxation due to g-factor anisotropy and intervalley scattering at low temperatures by combining a ballistic hot electron spin injection-detection technique with changing in-plane applied magnetic field directions [17]. For heavily doped (degenerate) Ge (n^+-Ge), however, there are few reports on spin relaxation through spin transport in n^+-Ge [26].

So far, spin relaxation in degenerate Si (n^+-Si) and n^+-Ge has been investigated by electron spin resonance (ESR) experiments [32–35]. The spin lifetime obtained by ESR was generally considered by the Elliott picture, indicating that the spin lifetime depends on the host material [36,37]. However, the reported spin lifetime also depended strongly on the dopant species [6,35], which cannot be quantitatively explained by the Elliott picture. With respect to this open question, Pifer proposed that spin-orbit coupling to the impurities is a controlling factor in the spin relaxation [35]. Recently, Song *et al.* theoretically interpreted the dominant spin relaxation in n^+-Si and n^+-Ge as an intervalley spin-flip scattering induced by the central-cell potential of the impurities [38]. This mechanism, called donor-driven spin relaxation [38], is short-range spin scattering due to the spin-orbit coupling of impurities rather than the spin mixing of states from spin-orbit coupling in the host materials. In short, a new mechanism differing from the Elliott process was recently proposed for n^+-Si and n^+-Ge [38]. They described that the spin scattering rate ($\frac{1}{\tau}$) depends on the concentration of the donor impurity (N_d) in degenerate conditions as the Fermi energy (ϵ_F) is larger than the thermal energy ($k_B T$) at low temperatures [38], where k_B is the Boltzmann's constant. The modified equation for degenerate Ge (n^+-Ge) can be roughly expressed as

$$\frac{1}{\tau} \approx \frac{4\pi n m_e a_B^6}{27\hbar^3}(3\pi^2 n)^{\frac{1}{3}}\Delta_{so}^2, \tag{1}$$

where a_B is the Bohr radius in Ge, m_e is the electron effective mass in Ge, and Δ_{so} is the spin-orbit coupling induced splitting of the triply degenerated 1s (T_2) donor state in Ge. Here, we assigned $\epsilon_F \approx \frac{\hbar^2}{2m_e}(3\pi^2 n)^{2/3}$ to the conduction electron energy ($\epsilon_{\mathbf{k}}$) from Eq. (4) in Ref. [38] and we regarded the value of the donor impurity concentration N_d as the carrier concentration n. Recently, we have verified temperature-independent spin scattering in n^+-Ge ($n \sim 8.2 \times 10^{18}$ cm^{-3}) at low temperatures [26], derived from the large influence of the donor-driven spin relaxation mechanism.

Considering Eq. (1), we can guess that the impact of the donor impurity concentration ($n \approx N_d$) on the spin scattering rate in Ge is relatively large compared to that in Si because a_B (=6.45 nm [39]) and Δ_{so} ($\sim$0.11 meV [26]) in Ge are larger than those in Si ($a_B = 2$ nm and $\Delta_{so} \sim 0.03$ meV [40]). More than 40 years ago, although an ESR experiment for the spin lifetime in n^+-Ge was reported [34], the broad linewidth of the ESR signals was analyzed. Considering the recent substantial progress of spin transport measurements in n^+-Ge [26], we should utilize the electrical measurements to explore the influence of N_d on the spin relaxation in n^+-Ge. In this Rapid Communication, we experimentally find a large change in the spin diffusion length (λ_{Ge}) of n^+-Ge at low temperatures with increasing n. The experimental findings are interpreted quantitatively in terms of a recent theory based on donor-driven spin scattering in multivalley semiconductors, predicted by Song *et al.* [38]. This study experimentally

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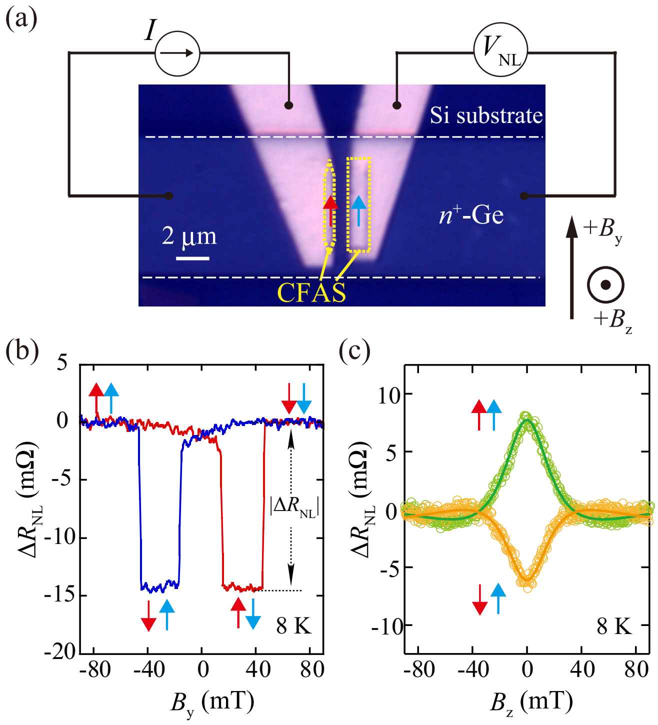

FIG. 1. (a) Optical micrograph of a fabricated CFAS/n^+-Ge based LSV. (b) Nonlocal magnetoresistance curve and (c) nonlocal Hanle effect curves at 8 K for an LSV with $d = 0.5\ \mu$m.

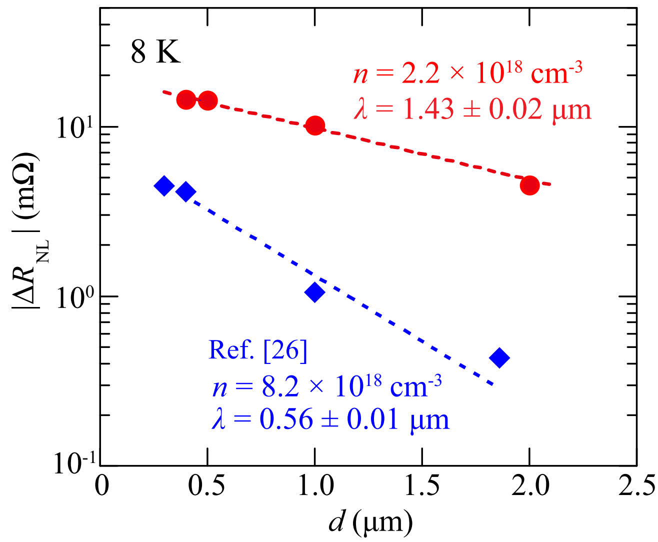

FIG. 2. d dependences of $|\Delta R_{NL}|$ for the LSV with $n = 2.2 \times 10^{18}$ cm^{-3} (red), together with that for the LSV with $n = 8.2 \times 10^{18}$ cm^{-3} (blue) in Ref. [26]. The dashed lines indicate the results of fitting to Eq. (2).

presents information on spin relaxation in heavily doped multivalley semiconductors.

We fabricated lateral spin valves (LSVs) to observe spin transport in n^+-Ge as follows. First, we grew an undoped Ge(111) layer (∼28 nm) at 350 °C [low-temperature Ge (LT-Ge)] on commercial undoped Si(111) substrates ($\rho \sim 1000\ \Omega$ cm), followed by an undoped Ge(111) layer (∼70 nm) grown at 700 °C [high-temperature Ge (HT-Ge)], where we utilized a two-step growth technique by molecular beam epitaxy (MBE) [41,42]. Then, 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 as spin transport layers. By slightly changing the impurity doping concentration, N_d can be tuned, leading to spin transport layers with various n ranging from 2.2 to 8.2 × 10^{18} cm^{-3}, which were estimated from Hall effect measurements. The obtained n values are reliable to consider spin relaxation in n^+-Ge [43–45]. To promote the tunneling conduction for spin injection and detection, a P δ-doped Ge layer with an ultrathin Si insertion layer was grown on top of the n^+-Ge layer [46]. As spin injectors and detectors, we grew a $Co_2FeAl_{0.5}Si_{0.5}$ (CFAS) layer on top of it by a nonstoichiometric growth technique with Knudsen cells by MBE at room temperature [47–50]. High quality heterointerfaces between CFAS and Ge were guaranteed elsewhere [51]. Finally, the CFAS/n^+-Ge layers were patterned into the contacts with $0.4 \times 5.0\ \mu$m^2 and $1.0 \times 5.0\ \mu$m^2, where the edge-to-edge distances (d) between the CFAS/n^+-Ge contacts are designed to be 0.4–2.0 μm. Schottky-tunnel conduction through the CFAS/n^+-Ge interfaces has been reproducibly confirmed [15,16,20,26,50].

An optical micrograph of a fabricated LSV is shown in Fig. 1(a). Nonlocal (NL) voltage measurements were performed by the terminal configuration shown in this figure [52–54]. Figure 1(b) shows a representative NL magnetoresistance ($\Delta R_{NL} = \Delta V_{NL}/I$) measured at $I = -1$ mA at 8 K, where the negative sign of I means the spin injection condition, i.e., electrons are injected from CFAS to the conduction band of n^+-Ge. The LSV measured here has an n^+-Ge layer with $n = 2.2 \times 10^{18}$ cm^{-3}, where the resistivity of the Ge layers (ρ_{Ge}) and n were determined by Hall effect measurements. When in-plane magnetic fields (B_y) are swept along the longitudinal axis of the CFAS contacts, a clear hysteretic behavior of ΔR_{NL} can be observed, depending on the magnetization configuration between the two CFAS contacts (see the arrows). Owing to the high quality CFAS/Ge heterointerface [50,51], relatively large spin signals (∼15 mΩ) were seen.

To check for reliable spin transport in n^+-Ge, we have also applied out-of-plane magnetic fields (B_z) under parallel and antiparallel magnetic configurations of the two CFAS contacts and recorded ΔR_{NL} as a function of B_z. As shown in Fig. 1(c), distinct NL Hanle effect curves owing to the spin precession in n^+-Ge are observed for both magnetic configurations, indicating the generation, manipulation, and detection of pure spin current through the n^+-Ge layer by all electrical means [3]. Accordingly, these reliable spin transports in n^+-Ge enable us to examine the spin relaxation mechanism in n^+-Ge layers with various n.

To examine λ_{Ge} of the used n^+-Ge layers, we measured ΔR_{NL} for the LSVs with various d at 8 K. Figure 2 shows the d dependence of $|\Delta R_{NL}|$ for the LSVs with $n = 2.2 \times 10^{18}$ cm^{-3} at 8 K, together with our previous result for the LSVs with $n = 8.2 \times 10^{18}$ cm^{-3} in Ref. [26]. Although there is a large difference in $|\Delta R_{NL}|$ between this study and Ref. [26], the cause of this difference is due to the difference in the spin injector and detector used. The growth temperature of the spin injector/detector and the fabrication processes of the LSVs in this study are identical to those in Ref. [26]. A detailed discussion about the spin injector and detector is given elsewhere [50]. In Fig. 2, $|\Delta R_{\rm NL}|$ values decay exponentially with increasing d for both LSVs.

In general, the reduction of $|\Delta R_{\rm NL}|$ with increasing d can be represented by [52,53]

$$|\Delta R_{\rm NL}| = \frac{|P_{\rm inj}||P_{\rm det}|\rho_{\rm Ge}\lambda_{\rm Ge}}{S}\exp\left(-\frac{d}{\lambda_{\rm Ge}}\right), \quad (2)$$

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 and S is the cross section of the n^+-Ge layer. Here, we used $\rho_{\rm Ge}$ values of 1.74 and 4.32 mΩ cm for $n = 8.2$ and 2.2×10^{18} cm^{-3}, respectively, and an S value of $\sim$0.49 μm^2. By fitting the decay of $\Delta R_{\rm NL}$ with Eq. (2), $\lambda_{\rm Ge}$ can be estimated, as shown in Fig. 2 (see the dashed line). We note that a $\lambda_{\rm Ge}$ of 1.43 μm, nearly three times as long as that of Ref. [26], is obtained for the LSV with $n = 2.2 \times 10^{18}$ cm^{-3}. It is quite interesting to experimentally observe such a large change in $\lambda_{\rm Ge}$ just by changing n from 8.2×10^{18} to 2.2×10^{18} cm^{-3} in the n^+-Ge layer. This strongly implies that there is a large impact of $N_{\rm d}$ on spin relaxation in n^+-Ge.

Figure 3(a) displays the plots of the estimated $\lambda_{\rm Ge}$ vs n ($\approx N_{\rm d}$) at 8 and 77 K. As shown in Fig. 2, the data shown here are estimated from the d dependence of $|\Delta R_{\rm NL}|$ for various LSVs. Interestingly, the $\lambda_{\rm Ge}$ value is systematically decreased from 1.43 to 0.56 μm with increasing n within the range measured here. To take the reduction in $\lambda_{\rm Ge}$ into account, we examine the spin lifetime ($\tau_{\rm Ge}$) as a function of n. Here, the $\tau_{\rm Ge}$ values are able to be calculated from the next relation, $\lambda_{\rm Ge} = \sqrt{D\tau}$, where D is the diffusion constant. In this study, we used the D values estimated from Eq. (4) in Ref. [55]. Here, the carrier mobility (425 cm^2/V s $\leqslant \mu \leqslant$ 430 cm^2/V s) obtained by Hall effect measurements was in good agreement with the literature [43–45]. The estimated D values as a function of n are shown in the inset of Fig. 3(a). From these data, we can obtain $\tau_{\rm Ge}$ for various n, as shown in Fig. 3(b). When n is reduced from 8.2×10^{18} to 2.2×10^{18} cm^{-3}, the $\tau_{\rm Ge}$ value is drastically enhanced from $\sim$0.1 to 2.0 ns. In this context, we find a large impact of the donor impurity concentration on spin relaxation in n^+-Ge even within a relatively narrow range of n (2.2×10^{18} cm$^{-3} \leqslant n \leqslant 8.2 \times 10^{18}$cm^{-3}).

Finally, we discuss spin relaxation in n^+-Ge within 10^{18} cm$^{-3} \leqslant n \leqslant 10^{19}$ cm^{-3} at low temperatures. As we consider n^+-Ge with $n \sim 10^{18}$ cm^{-3} (degenerate condition), $\epsilon_{\rm F}$ is still much larger than the energy $k_{\rm B}T$ because the Fermi level is located inside the multivalley conduction band. In our previous study [26], the temperature-independent spin relaxation in n^+-Ge with $n = 8.2 \times 10^{18}$ cm^{-3} below 125 K can be explained by the donor-driven spin scattering suggested by Song *et al.* [38]. According to Eq. (1), the donor-driven spin relaxation mechanism enables us to meet the relation of $\tau \propto n^{-\frac{4}{3}}$. In Fig. 3(b), we also plot a theoretical curve described in Eq. (1) as a red dashed one, in which $\Delta_{\rm so} = 0.11$ meV [38]. Note that the experimental data can be quantitatively explained by Eq. (1), indicating that the donor-driven spin relaxation mechanism is dominant for the n^+-Ge layer within a range of 10^{18}–10^{19} cm^{-3} [38]. This means that not only the theory but also an experiment clarify the presence of donor-driven spin relaxation at low temperatures in n^+-Ge. If we take the Elliott process for degenerate semiconductors ($N_{\rm d} \approx n$) into account, the relation of $\tau \propto n^{-\frac{4}{3}}$ can also be obtained theoretically, as described in Eq. (19) of Ref. [37]. However, the theoretical curve on the basis of the Elliott process largely deviates from the experimental data, as denoted by the blue dotted curve in Fig. 3(b). That is, the Elliott process cannot quantitatively explain the experimental data obtained by electrical spin transport measurements. In addition, as plotted in Fig. 3(b), the n dependence of τ estimated from ESR measurements [34] largely deviates from our data in this study and the donor-driven spin relaxation theory [38]. From these considerations, spin relaxation in n^+-Ge should be reexamined in detail by measuring electrical spin transport [26,50,56] and considering the donor-driven spin relaxation mechanism [38].

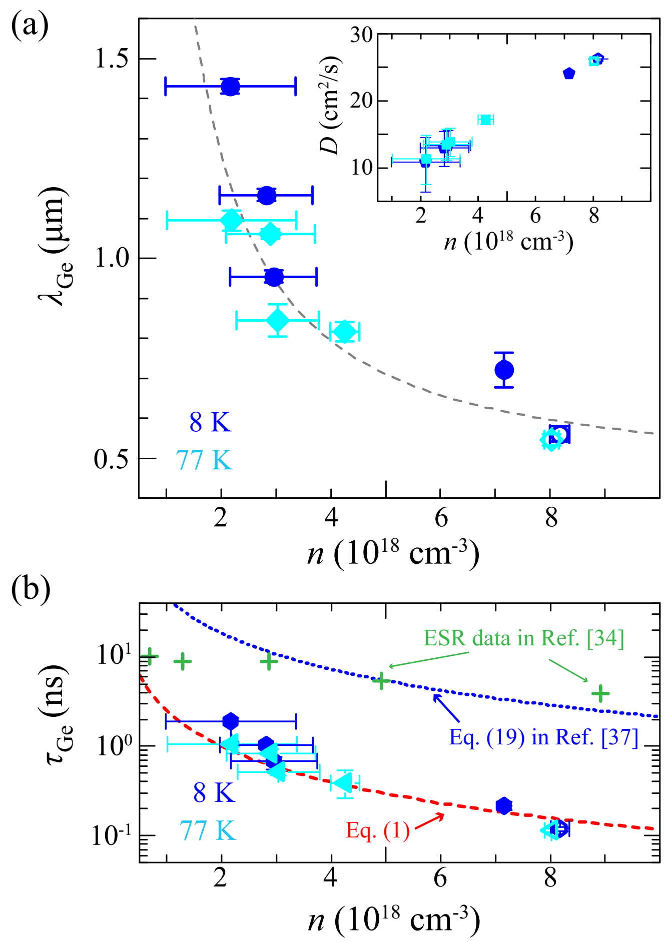

FIG. 3. (a) $\lambda_{\rm Ge}$ and (b) $\tau_{\rm Ge}$ vs n at 8 (blue) and 77 (light blue) K. The inset in (a) shows n dependence of D calculated from Eq. (4) in Ref. [55]. The red and blue dashed curves in (b) show theoretical curves from Eq. (1) and the Elliott process in Ref. [37], respectively, and the cross symbols in (b) are $\tau_{\rm Ge}$ measured by ESR in Ref. [34].

In conclusion, we experimentally found a large change in $\lambda_{\rm Ge}$ from 1.43 to 0.56 μm within a relatively narrow range of n (2.2×10^{18} cm$^{-3} \leqslant n \leqslant 8.2 \times 10^{18}$ cm^{-3}) at 8 K. The experimental findings can be interpreted quantitatively in terms of a recent theory based on donor-driven spin scattering in multivalley conduction bands in Ge. The content of this study means that donor-driven spin relaxation in doped multivalley semiconductors should be studied in more detail by electrically measuring the spin transport for spintronics devices.

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