PHYSICAL REVIEW MATERIALS **1**, 034604 (2017)

Electrical detection of spin accumulation and relaxation in *p*-type germanium

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(Received 17 April 2017; published 14 August 2017)

We report on electrical measurements of spin-dependent transport of holes in all-epitaxial CoFe/*p*-type germanium (*p*-Ge)/Fe_3Si spin valves, where the hole concentration (p_h) of the *p*-Ge layer is estimated to be $\sim 10^{18}$ cm^{-3}. Spin-accumulation output voltages can electrically be detected in the antiparallel magnetization state between CoFe and Fe_3Si ferromagnetic electrodes. The room-temperature spin lifetime of holes in the *p*-Ge layers can tentatively be discussed in terms of the theory by Fert and Jaffrès. We propose that the use of (111)-oriented *p*-Ge with a hole concentration of $\sim 10^{18}$ cm^{-3} enables the transport of spin-polarized holes in bulk Ge even at room temperature.

DOI: 10.1103/PhysRevMaterials.1.034604

I. INTRODUCTION

Transporting the spin angular momentum in a semiconductor (SC) is an important technology to achieve the integration of the nonvolatile memory effect with SC electronics devices [1,2]. Up to now, it has been proved that the electrically generated nonequilibrium spin accumulation enables diffusive transport of pure spin currents, the flows of the spin angular momentum, even in nonmagnetic SCs such as GaAs [3–5], Si [6–8], and Ge [9,10]. Also, there are lots of studies on the transport of spin-polarized electrons through SCs in ferromagnet (FM)-SC-FM two-terminal spin-valve device structures [11–20]. However, it is generally difficult to electrically detect the transport of spin-polarized holes even in SCs [12,13,16,21].

More than 10 years ago, Mattana *et al.* reported the electrical detection of the hole spin transport through a GaAs quantum well (QW) by injecting spin-polarized holes from the *p*-type ferromagnetic semiconductor (Ga,Mn)As via AlAs tunnel barriers [12]. They described that the spin splitting derived from the hole spin accumulation was created in the GaAs QW layer and the tunneling magnetoresistance (TMR) effect appeared via spin-dependent sequential tunneling. Since the spin relaxation of holes was intentionally suppressed by lifting the valence-band degeneracy at the Γ point in the QW layer, the spin relaxation phenomena of the injected spin-polarized holes could not be examined in this method [12]. Also, because the ferromagnetism of the (Ga,Mn)As electrodes was restricted in the low-temperature range, hole spin transport in the wide temperature range could not be observed [12,13,16]. Although other experimental techniques such as three-terminal Hanle measurements and the inverse spin Hall effect for detecting spin-polarized holes in SCs at room temperature have been reported [22–24], the spin accumulation and detection through these techniques still remain debatable [25] and there are large differences in the spin relaxation phenomena investigated by these methods and by optical ones [26–32] that have particularly been accessible for investigating spin dynamics in SCs.

In this article, we experimentally show electrical transport measurements of spin-polarized holes in all-epitaxial CoFe/*p*-type germanium (*p*-Ge)/Fe_3Si spin valves. Spin-accumulation output voltages can electrically be detected in the antiparallel magnetization state between CoFe and Fe_3Si ferromagnetic electrodes. Even at room temperature, we can see the spin-accumulation signals in the all-epitaxial CoFe/*p*-Ge/Fe_3Si spin valves. The room-temperature spin lifetime of holes in *p*-Ge layers can tentatively be discussed in terms of the theory by Fert and Jaffrès [33]. We propose that the use of (111)-oriented *p*-Ge with a hole concentration of $\sim 10^{18}$ cm^{-3} enables the transport of spin-polarized holes in bulk Ge even at room temperature.

II. ALL-EPITAXIAL VERTICAL SPIN VALVES

The top illustration in Fig. 1(a) is a schematic of the vertically stacked spin valve consisting of a 20-nm-thick *p*-Ge layer sandwiched between CoFe (top) and Fe_3Si (bottom) electrodes. All-epitaxial (111)-oriented CoFe ($\sim$10 nm)/Ge ($\sim$20 nm)/Fe_3Si ($\sim$25 nm) trilayer films were grown on undoped Si(111) substrates ($\rho \sim 1000$ Ω cm) by low-temperature molecular beam epitaxy, where the detailed growth processes have been described in the literature [34,35]. Since free holes induced by the ionized acceptors due to the point defects [36] were detected by thermopower measurements of Ge/Fe_3Si bilayer structures on Si [21] we regarded the undoped Ge(111) epilayers used in this study as *p*-Ge. We subsequently processed the trilayer films into the vertical-type spin-valve structures shown in Fig. 1(a) by conventional electron-beam lithography and Ar ion milling techniques. To achieve the antiparallel magnetization state between CoFe and Fe_3Si for detecting spin accumulation in *p*-Ge, as shown in the bottom of Fig. 1(a), we fabricated the point-end-shaped CoFe layer and the wire-shaped Fe_3Si one with a junction size of $\sim$0.2 μm^2. An outward appearance is shown in Fig. 1(b). There are four Au bonding pads to measure electrical properties.

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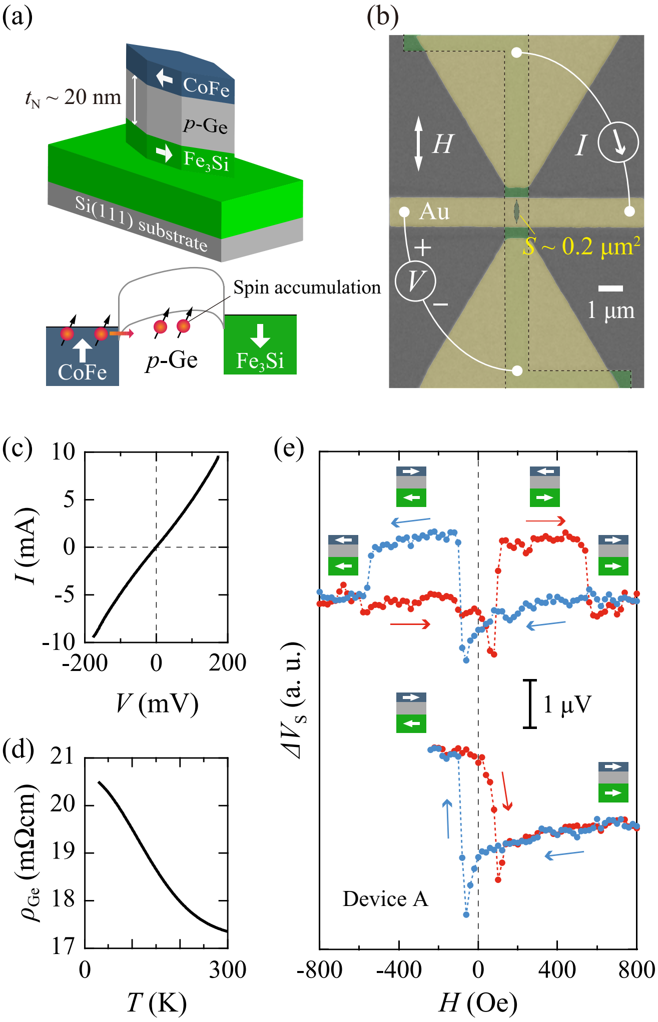

FIG. 1. (a) Schematic, (b) scanning electron micrograph, and (c) I-V and (d) ρ_{Ge}-T curves of CoFe/p-Ge/Fe_3Si spin valves. (e) Spin accumulation output voltage (ΔV_s) as a function of external magnetic field (H) for device A, measured at $T = 30$ K with $I_{dc} = 0.5$ mA. The scale bar shown is the magnitude of ΔV_s.

Prior to the spin transport measurements, we evaluated current-voltage characteristics (I-V curves) in the temperature range from 10 to 300 K for various CoFe/p-Ge/Fe_3Si spin-valve devices. Symmetric I-V curves with a weak nonlinearity were obtained even at 10 K, as shown in Fig. 1(c), implying that the thermionic emission at both interfaces is negligibly small. This indicates that the tunneling conduction of holes through double Schottky tunnel barriers, CoFe/p-Ge and p-Ge/Fe_3Si, is achieved by reducing the strong Fermi level pinning (FLP) effect by forming high-quality interfaces [21,37], where the Schottky barrier height (ϕ_b) is expected to be ~0.1 eV at both CoFe/p-Ge and p-Ge/Fe_3Si interfaces. Figure 1(d) shows a temperature dependence of resistivity (ρ) measured by the terminal configuration depicted in Fig. 1(b). An evident semiconductor-like ρ-T nature can be seen, indicating that the measured resistivity (~20 mΩ cm) is governed by the intermediate Ge layer because those of CoFe (~40 $\mu\Omega$ cm) and Fe_3Si (~80 $\mu\Omega$ cm) are very small. Thus, we can roughly regard the measured data as the resistivity of the Ge layer (ρ_{Ge}) and can roughly estimate hole concentration (p_h) and hole mobility (μ_h) from the Irvin curve [38]. In this study, the epitaxial Ge layers have $p_h = 5.0 \times 10^{17} \sim 1.0 \times 10^{18}$ cm^{-3} and $\mu_h = 550 \sim 700$ cm^2/V s at room temperature, which are consistent with previous works [21,36].

Considering these electrical properties of the CoFe/p-Ge/Fe_3Si spin-valve devices, we can discuss the contribution of the field emission (FE) and/or thermionic field emission (TFE) to the tunneling of holes through the interfaces. If we compare the tunneling parameter ($E_{00} = qh/4\pi \times \sqrt{N_A/m^*\epsilon}$) with the thermal energy (k_BT/q) [39], we can find that FE is dominant at 10 K ($E_{00} \gg k_BT/q$) and TFE can contribute to the tunneling with increasing temperature ($E_{00} \sim k_BT/q$ at 300 K), where k_B is the Boltzmann constant, q is the electric charge, h is the Planck constant, N_A can be assumed to be p_h, m^* is the effective mass of holes in Ge, and ϵ is the permittivity in Ge. In this study, because the ϕ_b value is relatively low (~0.1 eV) and the total depletion layer width at zero bias is estimated to be less than ~20 nm, we can expect sufficiently narrow barrier thickness (~1 nm) for the tunneling at the low applied bias voltages used [19]. Thus, we can regard both barriers at CoFe/p-Ge and p-Ge/Fe_3Si interfaces as tunnel barriers to inject and detect spin-polarized holes by suppressing the spin resistance mismatch problem [33,40].

III. SPIN TRANSPORT THROUGH *p*-Ge

Using the all-epitaxial CoFe/p-Ge/Fe_3Si spin valves, we electrically measured transport properties as a function of in-plane external magnetic field (H) along the long axis of the wire-shaped electrodes. Here the output voltages (V) were measured by standard dc bias techniques (I_{dc}) at various temperatures. Figure 1(e) shows a representative output voltage change (ΔV_s) of device A at 30 K at $I_{dc} = 0.5$ mA (ΔV_s-H curve). As shown in the top figure, positive ΔV_s changes with clear hysteretic behavior are observed although small negative ΔV_s ones attributed to anisotropic magnetoresistance (AMR) effect in the bottom Fe_3Si electrode are simultaneously detected. Here we have checked that the fields showing the small negative signals correspond to the magnetization switching fields of the bottom Fe_3Si electrode from the AMR measurements. To understand the positive ΔV_s changes, we also conducted minor-loop measurements, as shown in the bottom figure in the same condition. Considering the AMR signals within ±100 Oe, we can expect that the magnetization of the bottom Fe_3Si layer is switched toward the opposite direction to that of the top CoFe layer. Thus, at $H \sim -250$ Oe, the antiparallel magnetization state between CoFe and Fe_3Si electrodes can be expected after H is decreased from +800 Oe to −250 Oe. Next, as H is increased toward positive values from $H \sim -250$ Oe, we can observe the remanent ΔV_s at $H = 0$ and steep reduction in ΔV_s at $H \sim +100$ Oe, at which the magnetic configuration between CoFe and Fe_3Si electrodes is also switched to be parallel. Thus, the hysteretic behavior in the bottom figure is an evident minor loop. This means that the positive ΔV_s feature in the top figure originates from the spin-dependent transport through the 20-nm-thick p-Ge layer detected by an all-electrical means. The above interpretation is consistent with our recent study that showed a spin diffusion length in the p-Ge

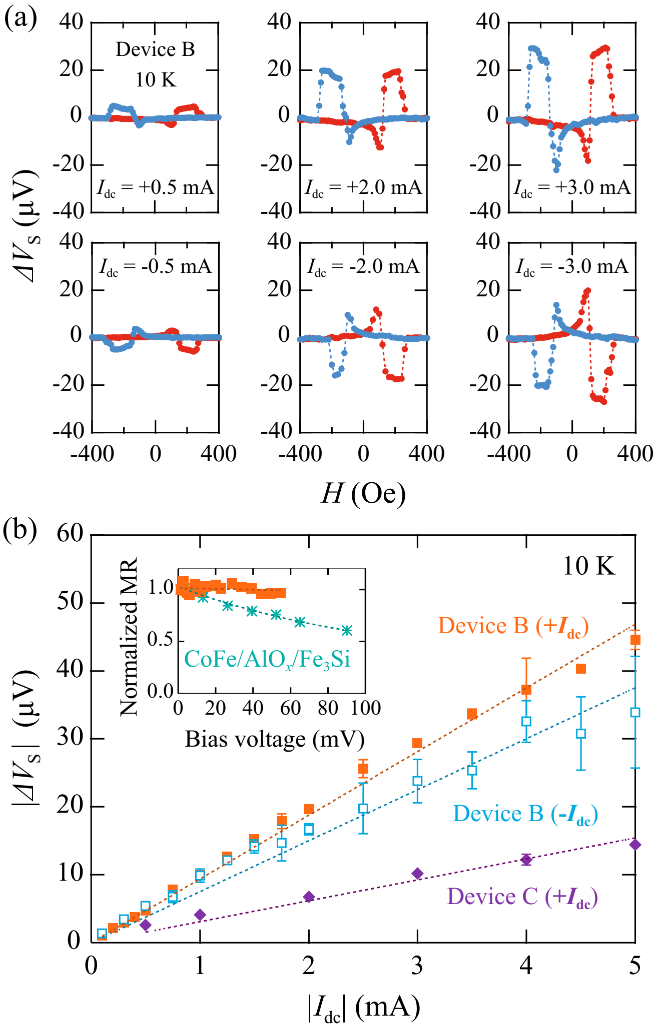

FIG. 2. (a) ΔV_s as a function of H measured at $T = 10$ K with $I_{dc} = \pm 0.5$, ± 2.0, and ± 3.0 mA for device B. (b) Bias current dependence of $|\Delta V_s|$ for device B and device C measured at $T = 10$ K. The inset shows the bias voltage dependence of normalized magnetoresistance for device B, together with that for a CoFe/AlO$_x$($\sim$2 nm)/Fe$_3$Si MTJ.

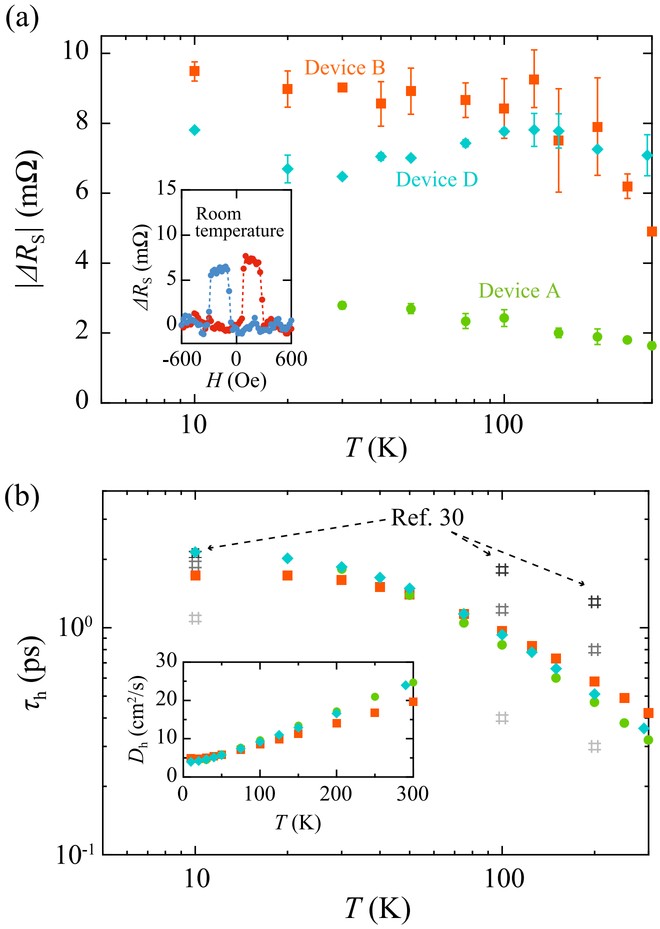

FIG. 3. (a) $|\Delta R_s|$ as a function of temperature for device A, device B, and device D. The inset shows a representative room-temperature spin signal for device D. (b) τ_h as a function of temperature for device A, device B, and device D, together with the results in Ref. [30] with photon fluences of 4.4×10^{14} (black), 1.0×10^{15} (dark gray), and 9.0×10^{15} cm^{-2} (light gray). The inset shows the calculated D_h value as a function of temperature.

layers (λ_{Ge}) of ~40 nm at low temperatures [21]. Because the ΔV_s values still fluctuated due to the slight difference of ρ_{Ge}, we would like to call the next four devices A ($\rho_{Ge} \sim 17$ mΩ cm at 300 K), B ($\rho_{Ge} \sim 9.4$ mΩ cm at 300 K), C ($\rho_{Ge} \sim 24$ mΩ cm at 10 K), and D ($\rho_{Ge} \sim 17$ mΩ cm at 290 K).

If the spin-dependent transport of holes seen in Fig. 1(e) arises from the spin-accumulation conservation in the p-Ge layer including CoFe/p-Ge and p-Ge/Fe$_3$Si interfaces, further injecting spin-polarized currents can create more ΔV_s [33]. Also, the polarity of ΔV_s can be controlled by that of the injecting spin-polarized currents [33]. Thus, we measured ΔV_s-H curves for various values of I_{dc} or switching the polarity of I_{dc}. Figure 2(a) shows the data for device B, measured at I_{dc} of ± 0.5, ± 2.0, and ± 3.0 mA at 10 K. For both I_{dc} polarities, the amplitude of ΔV_s in the antiparallel magnetization state, $|\Delta V_s|$, is enhanced with increasing $|I_{dc}|$ although the AMR signals of the bottom Fe$_3$Si electrode are still superimposed on the spin signals. Figure 2(b) summarizes $|\Delta V_s|$ as a function of $|I_{dc}|$ for device B and device C. Irrespective of the polarity of I_{dc}, $|\Delta V_s|$ increases linearly with increasing $|I_{dc}|$. In short, this behavior can be interpreted as a consequence of the spin-accumulation conservation in the p-Ge layer [33]. It should be noted that the magnetoresistance (MR) amplitude ($\Delta V_s / I_{dc}$) is nearly constant with respect to bias voltage (see inset). This tendency is largely different from that observed in (Ga,Mn)As/AlAs/GaAs-QW/AlAs/(Ga,Mn)As structures, indicating spin-dependent sequential tunneling of holes [12]. We also plotted the bias-voltage dependence of TMR in a CoFe/AlO$_x$/Fe$_3$Si magnetic tunnel junction (MTJ) in the same inset [41]. Comparing a CoFe/p-Ge/Fe$_3$Si spin-valve with a CoFe/AlO$_x$/Fe$_3$Si MTJ, we can clearly recognize the difference in the mechanism of the spin-dependent transport. From these considerations, the observed bias dependencies in Figs. 2(a) and 2(b) are not TMR but are caused by the creation and detection of the spin accumulation in the p-Ge layer including CoFe/p-Ge and p-Ge/Fe$_3$Si interfaces.

We focus on the influence of external temperatures on the spin transport of holes in p-Ge. Figure 3(a) shows the temperature dependence of the spin-signal amplitude, $|\Delta R_s|$ ($|\Delta V_s/I|$), for devices A, B, and D. Unfortunately, device C was broken at 10 K under the bias-current dependence measurements. Despite elevating external temperatures, we cannot see a drastic change in $|\Delta R_s|$ values. These features are different from the strong reduction of the spin signals in the lateral spin valves with the 43-nm-thick *p*-Ge layer in Ref. [21]. Very interestingly, we can observe $|\Delta R_s|$ even at room temperature, as shown in the inset, for all the devices with 20-nm-thick *p*-Ge layer. This fact indicates that spin-polarized holes can be transported in SC even at room temperature.

IV. SPIN RELAXATION IN *p*-Ge

To discuss the spin relaxation of holes in *p*-Ge, we tentatively utilize the theory by Fert and Jaffrès [33]. As discussed in Sec. II, the formed barriers at CoFe/*p*-Ge and *p*-Ge/Fe$_3$Si interfaces at the low bias voltage range can be regarded as ultrathin tunnel barriers the same as the insulating barriers in the theory by Fert and Jaffrès [33]. Thus, we can roughly express the spin accumulation voltage obtained here, ΔV_s, as follows:

$$\Delta V_s = \frac{2I(\beta r_F + \gamma r_b^*)^2}{(r_b^* + r_F)\cosh\left(\frac{t_N}{\lambda_N}\right) + \left(\frac{r_N}{2}\right)\left\{1 + \left(\frac{r_b^*}{r_N}\right)^2\right\}\sinh\left(\frac{t_N}{\lambda_N}\right)}, \tag{1}$$

where β and γ are spin-asymmetry coefficients of the FM bulk and FM/SC interface, respectively; r_b^*, r_F, and r_N are the spin resistances of the FM/SC interface, FM bulk, and SC bulk, respectively; t_N and λ_N are thickness and spin-diffusion length of the SC layer, respectively. Here since the room-temperature β of CoFe ($\beta \sim 0.2$) and Fe$_3$Si ($\beta \sim 0.2$) have been obtained [42], we can estimate r_F and r_b^* to be $\sim 3.1 \times 10^{-15}\ \Omega\ \text{m}^2$ and $1.9 \sim 3.5 \times 10^{-12}\ \Omega\ \text{m}^2$, respectively. From $R_{\text{parallel}} = r_b^*(1 - \gamma^2)$ [33], γ is tentatively determined to be ~ 0.02. Thus, when we use Eq. (1) with $r_N = 2.7 \sim 5.1 \times 10^{-12}\ \Omega\ \text{m}^2$ and $t_N = 20$ nm, we can roughly obtain the spin diffusion length λ_N of *p*-Ge (λ_{Ge}) to be ~ 30 nm at room temperature, consistent with the the framework of the model by Fert and Jaffrès ($t_N \leqslant \lambda_N$) [33]. In the previous work on the spin transport in *p*-Ge [21], the thickness of the *p*-Ge layer was larger (~ 43 nm) than the room-temperature λ_{Ge}, giving rise to the strong temperature dependence of the spin signals. In this work, the small thickness of the *p*-Ge layer in the all-epitaxial spin valves is relatively advantageous to observe the room-temperature spin transport compared to the previous work in Ref. [21].

Using the room-temperature μ_h value ($550 \sim 700\ \text{cm}^2/\text{V s}$) described in Fig. 1, we can calculate the diffusion constant, D_h, from Eq. (4) in Ref. [43]. As a result, room-temperature D_h values of the *p*-Ge layers are estimated to be $20 \sim 25\ \text{cm}^2/\text{s}$. To extract the spin lifetime of holes (τ_h), we used the following relation, $\lambda_N = \sqrt{D_h \tau_h}$. From these considerations, the room-temperature τ_h value of the *p*-Ge layers is determined to be $0.3 \sim 0.4$ ps, which is as long as those in optically probed hole spin dynamics in Ge-QW structures with comparable photoinduced carrier concentration [30].

Here we consider the observation of room-temperature spin transport of holes in all-epitaxial CoFe/*p*-Ge/Fe$_3$Si spin valves. First, in previous works on optical pump-probe methods, Lange *et al.* [30] and Pezzoli *et al.* [31] reported relatively long τ_h ($0.3 \sim 2.1$ ps) by lifting the heavy hole (HH)–light hole (LH) degeneracy at the Γ point in Ge-QW structures, where the splitting energy between HH and LH bands is ~ 100 meV. To generate hole spins at the Γ point, (100)-oriented QW structures have so far been utilized [30,31]. On the other hand, the estimated τ_h ($0.3 \sim 0.4$ ps) in this study was the same order of the time scale at room temperature without using the QW structure and strain effects. We now infer that (111)-oriented epitaxial structures are useful to electrically observe the spin transport of holes. Since we have epitaxially grown the CoFe/*p*-Ge/Fe$_3$Si trilayer along the [111] direction, the spin-polarized holes can be injected toward the L point in the valence bands of Ge. At the L point, there is a large energy splitting between HH and LH bands (~ 200 meV) even for bulk [44]. This energy splitting is much larger than $k_B T/q \sim 26$ meV at room temperature (~ 300 K). In short, the phonon-induced spin relaxation by HH intraband and/or HH-LH interband scatterings, as discussed in Refs. [30,31], should be reduced at the L point even at room temperature. For the creation of the spin accumulation in *p*-Ge, the (111)-oriented epitaxial spin-valve structures are effective to demonstrate the transport of spin-polarized holes.

To further take into account the weak temperature dependence of spin signals, we finally comment on τ_h as a function of temperature in the *p*-Ge layers. As the external temperature was lowered to 10 K, the β values of CoFe and Fe$_3$Si were increased to ~ 0.5 [45,46]. Accordingly, we can roughly estimate λ_{Ge} at 10 K to be ~ 30 nm by using the observed ΔV_s value at 10 K. Using the calculated D_h values shown in the inset of Fig. 3(b), we can plot τ_h versus temperature in Fig. 3(b), together with those in Ge-QW, measured by an optical method [30]. The temperature dependence of τ_h in this study is likely to be similar to that in Ge-QW [30]. In general, if the Elliott-Yafet spin relaxation mechanism is dominant in the Ge valence band [2,47], τ_h can be proportional to the momentum relaxation time of holes, depending on the hole mobility μ_h. According to Golikova *et al.* [48] the μ_h value for *p*-Ge with $p_h \sim 1.0 \times 10^{18}\ \text{cm}^{-3}$ showed a weak temperature dependence. We infer that the weak temperature-dependent μ_h results in the weak phonon-induced spin relaxation in *p*-Ge with $p_h \sim 1.0 \times 10^{18}\ \text{cm}^{-3}$. When we utilize the (111)-oriented epitaxial *p*-Ge layers with $p_h = 5.0 \times 10^{17} \sim 1.0 \times 10^{18}\ \text{cm}^{-3}$, both relatively long τ_h ($0.3 \sim 0.4$ ps) and weak temperature dependence can be expected. As there is excellent agreement between experimental and theoretical studies of the electron spin transport in Ge [10,49–51], further studies of the influence of the valence band structures on the spin relaxation in Ge are desirable.

V. CONCLUSION

We presented electrical transport measurements of spin-polarized holes in all-epitaxial CoFe/*p*-Ge/Fe$_3$Si spin valves. Spin accumulation output voltages in *p*-Ge were electrically detected in the antiparallel magnetization state between CoFe and Fe$_3$Si ferromagnetic electrodes up to 300 K. The room-temperature hole spin lifetime in *p*-Ge layers can tentatively be discussed in terms of the theory by Fert and Jaffrès [33]. We propose that the use of (111)-oriented *p*-Ge with

$p_{\mathrm{h}} \sim 1.0 \times 10^{18}$ cm^{-3} enables the transport of spin-polarized holes in bulk Ge even at room temperature.

ACKNOWLEDGMENTS

This work was partly supported by Grants-in-Aid for Scientific Research (A) (No. 25246020 and No. 16H02333) from the Japan Society for the Promotion of Science (JSPS) and a Grant-in-Aid for Scientific Research on Innovative Areas "Nano Spin Conversion Science" (No. 26103003) from the Ministry of Education, Culture, Sports, Science, and Technology (MEXT). M.K. acknowledges support from JSPS Research Fellowships for Young Scientists.

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