ORIGINAL ARTICLE

Room-temperature generation of giant pure spin currents using epitaxial Co_2FeSi spin injectors

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The generation, manipulation and detection of a pure spin current (i.e., the flow of spin angular momentum without a charge current) are prospective approaches for realizing next-generation spintronic devices with ultra-low electric power consumption. Conventional ferromagnetic electrodes such as Co and NiFe have been utilized as spin injectors to generate pure spin currents in nonmagnetic channels. However, the generation efficiency of pure spin currents is extremely low at room temperature, giving rise to a serious obstacle for device applications. Here we demonstrate the generation of giant pure spin currents at room temperature in lateral spin valve devices with a highly ordered Heusler-compound Co_2FeSi (CFS) spin injector. The generation efficiency of pure spin currents from the CFS spin injectors is 10 times greater than that of the NiFe injectors, indicating that Heusler compound spin injectors with high spin polarization enable us to materialize a high-performance lateral spin device. The present study is a technological jump in spintronics, and indicates the great potential of ferromagnetic Heusler compounds with half metallicity for generating pure spin currents.

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INTRODUCTION

Electrical spin injection from a ferromagnet (F) into a nonmagnet (N) can generate a spin current (i.e., the flow of spin angular momentum) even in a nonmagnetic channel.^{1–5} In general, the spin current is induced by diffusing non-equilibrium spin accumulations in the vicinity of the F/N interface under spin injection. However, because the difference in the density of states between the majority and minority spins (i.e., the spin polarization P) is not so large for a conventional F, such as Co or NiFe (Py), the induced spin current in the N mainly returns back to the F (Figure 1a). This phenomenon gives rise to an extremely low injection efficiency of the spin current in the N.^{6,7} If we utilize a highly spin-polarized F as a spin injector, the spin-polarized electrons can be efficiently injected into the N, and the backflow of the spin currents can be strongly suppressed, resulting in a dramatical improvement of the injection efficiency of the spin currents in the N (Figure 1b). A charge current can also be extracted with nonlocal electrical spin injection in a mesoscopic lateral geometry, and a spin current can be transferred without the charge current, that is, a pure spin current, in the nonmagnetic channel (Figures 1c and d). In this scheme,^{1,3,4,8–16} the use of highly spin-polarized F spin injectors is critical for generating a giant pure spin current in the N.

As materials with highly spin-polarized F, we focus on Co-based Heusler compounds in which the half-metallic properties are theore-

tically predicted.^{17,18} In fact, huge tunnel magnetoresistance and giant magnetoresistance effects have been reported in vertical stacking device structures.^{19–21} Therefore, the combination of the high-performance Co-based Heusler compounds with laterally configured device structures is a prospective challenge for highly efficient generation of pure spin currents. Recently, a nonlocal spin injection using polycrystalline CoFeAl was reported. However, the relatively large resistivity of the CoFeAl utilized in Bridoux *et al.*²² indicates that the crystal structure and the spin polarization are far from those of an ideal Heusler compound (see Supplementary information). In the present study, we show that an epitaxially grown Co-based Heusler compound, Co_2FeSi (CFS) with a highly ordered $L2_1$ structure, enables the highly efficient injection of pure spin currents.

MATERIALS AND METHODS

Our device structure is a lateral spin valve (LSV) consisting of a CFS spin injector and detector bridged by a Cu strip (Figure 2a), in which the CFS thin film with highly ordered $L2_1$ structures has been epitaxially grown on Si(111).²³ A conventional electron-beam lithography and Ar ion milling technique are employed to form the wire-shaped CFS spin injector and a detector with a width of 300 nm. Here, one CFS wire is connected to two square pads to facilitate domain wall nucleation, whereas the other has pointed-end edges. Using the two different wire shapes, we can control the magnetization

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Figure 1 Concept of efficient generation of a pure spin current. **(a, b)** Schematic diagrams of the electrical spin injection from a conventional ferromagnet (F) or a highly spin-polarized F (HSF) into a nonmagnet (N). For the conventional F, most of the original spin currents return back to the F (a backflow of the spin current), giving rise to a significant reduction in the injected spin current. For the HSF, the original spin current is effectively injected into the N by preventing the backflow. **(c)** Generation of a pure spin current with nonlocal spin injection. The electron charges are extracted toward left hand side while the spin currents diffuse into both sides symmetrically. **(d)** Spatial distributions of the spin-dependent electro-chemical potentials ($\mu_{\uparrow}$, $\mu_{\downarrow}$), in the N. Although the charge current $\propto \partial(\mu_{\uparrow} + \mu_{\downarrow})/\partial x$ is zero in the right hand side, a finite spin current $\propto \partial(\mu_{\uparrow} - \mu_{\downarrow})/\partial x$ is generated over the spin diffusion length. Thus, the pure spin current can be generated in the right hand side of the N.

Figure 2 Enhanced nonlocal spin valve effect. **(a)** A scanning electron microscope image of the fabricated $\text{Co}_2\text{FeSi(CFS)/Cu}$ lateral spin valve (LSV). **(b)** Schematic of a nonlocal spin valve measurement. Spin-polarized electrons are injected from contact 2, and electron charges are extracted from contact 1. A nonlocal voltage is measured between contact 3 and contact 4. **(c)** A room-temperature nonlocal spin-valve signal for the CFS/Cu LSV, together with that for the Py/Cu LSV. The signal varies according to the relative magnetization orientation of two wire-shaped CFS electrodes, as shown in the inset illustrations. **(d)** A room-temperature local spin-valve signals for the Py/Cu and CFS/Cu LSVs. The inset shows the current–voltage probe configuration (i.e., the current is injected from contact 2 and extracted from contact 3), and the voltage is measured between contact 5 and contact 6. The low and high resistance states correspond to the parallel and anti-parallel magnetization alignments, respectively. The expected magnetization configurations agree with those observed in the nonlocal spin valve signal. **(e)** Nonlocal spin signal ΔR_S as a function of J_{inj} at the injecting junction, normalized by ΔR_S at a small bias current density of $J_0 \sim 10^9 \text{ A m}^{-2}$, for the CFS/Cu LSV (red solid squares) and the Py/Cu LSV (blue open circles). **(f)** Temperature dependence of ΔR_S for the CFS/Cu LSV (red solid squares) and the Py/Cu LSV (blue open circles), normalized by ΔR_S at 20 K. The inset shows a nonlocal spin-valve effect of the CFS/Cu LSV at $T = 70 \text{ K}$. The scale bars in **(c)**, **(d)**, and the inset of **(f)** are 1, 2 and 5 mΩ, respectively.

configuration by adjusting the external magnetic fields (H), where H is applied along the CFS wires. Finally, the top Cu strips (200 nm wide and 100 nm thick) bridging the CFS wires and bonding pads were patterned by a conventional lift-off technique. Before the Cu deposition, the surfaces of the CFS wires are well cleaned by Ar ion milling with a low accelerating voltage, producing in low resistive ohmic interfaces with $R_{\text{CFS/Cu}} \approx 1 \text{ f}\Omega\text{m}^2$. Here, the optimization for the milling conditions of the CFS surface is performed by changing the milling time and acceleration voltage. The transport measurements are carried out by a conventional current-bias lock-in technique ($\sim 200 \text{ Hz}$). The resistivities for the prepared CFS and Cu wires are 90.5 and $2.5 \mu\Omega\text{cm}$, respectively, at room temperature (RT), and 54.6 and $1.2 \mu\Omega\text{cm}$, respectively, at 80 K. As shown in Figure 2b, a pure spin current generated by the nonlocal spin injection from CFS1 can be detected by CFS2 after propagation through the 600-nm distance in the Cu strip.

RESULTS AND DISCUSSION

Figure 2c shows the nonlocal magnetoresistance of the CFS/Cu LSV measured at RT, together with that of a Py/Cu LSV. Here, the size of the CFS/Cu junction is three times as large as that of the Py/Cu junction. A giant spin signal (ΔR_S) of $2.3 \text{ m}\Omega$ is seen for the CFS/Cu LSV (Figure 2c), which is approximately 10 times as large as that of the Py/Cu LSV. As the spin injection efficiency is inversely proportional to the size of the F/N junctions,⁶ the large ΔR_S demonstrated in the CFS/Cu LSV with larger junctions implies a significant possibility for the present CFS/Cu LSV.

We also show the local spin valve signals at RT for the same CFS/Cu and Py/Cu LSVs (Figure 2d). The values are almost twice those of the non-local ΔR_S , which are in reasonable agreement with the values of the previous reports.^{2,6} This result means that the one-dimensional spin diffusion model provides a good description of the spin transport in the present CFS/Cu LSV. Figure 2e shows the dependence of ΔR_S on the bias current density (J_{inj}) at the injecting junction for the CFS/Cu LSV, which is almost the same as that of the Py/Cu LSV. The reduction of ΔR_S is less than 20% even under a high-bias current density ($\sim 10^{11} \text{ A m}^{-2}$), indicating much superior properties to those of the LSV consisting of the high-resistive tunnel junctions, in which ΔR_S drastically decreases even at low-bias current density ($\sim 10^8 \text{ A m}^{-2}$).¹¹ The temperature dependence of ΔR_S for the CFS/Cu LSV is also almost the same as that of the Py/Cu LSV, in which ΔR_S takes its maximum value around 20 K, below which the ΔR_S decreases with decreasing temperature (Figure 2f). This behavior can be explained by an enhancement of the spin-flip scattering at the Cu surface for the CFS/Cu LSV below 20 K, as discussed in Kimura *et al.*¹⁰ Surprisingly, the ΔR_S for the CFS/Cu exceeds $10 \text{ m}\Omega$ below 70 K (inset of Figure 2f). From these results, we recognize that the present CFS/Cu LSV can be treated as a conventional ohmic LSV, and it can generate a giant pure spin current with a much less electric power than the previously reported LSVs.^{1,2,6–14}

To quantitatively evaluate the device performance of the present LSVs from their nonlocal spin signals, we measure ΔR_S of the CFS/Cu LSV devices with various distances (d ; where d is the center–center distance between spin injector and detector) and Py/Cu LSV devices as references. Here we introduce a characteristic value in the LSV devices by extending the resistance change area product, which is commonly utilized to characterize the device performance of the vertical spin devices.^{24,25} The resistance change area product for the nonlocal spin signal ($\Delta R_S A$) is defined as $\Delta R_S (S_{\text{inj}} S_{\text{det}} / S_N)$, where S_{inj} , S_{det} and S_N are the junction sizes in the spin injector and detector, and the cross section of the nonmagnetic strip. This $\Delta R_S A$ allows us to equivalently compare the device performances between our CFS/Cu and the Py/Cu LSV devices. The plot of $\Delta R_S A$ versus d at RT for the CFS/Cu LSVs and Py/Cu LSVs is shown in Figure 3, and the plot at 80 K is shown in

Figure 3 Scaling plot for lateral spin valve (LSV) devices with metallic junctions. The resistance change area product for the nonlocal spin signal ($\Delta R_S A$) as a function of d for CFS/Cu LSVs (filled squares), together with that for Py/Cu LSVs (open circles). The main panel and inset show the data at RT and 80 K, respectively. The solid curves are fitting results with $\lambda_{\text{Cu}} = 500 \text{ nm}$ at RT (red line) and $\lambda_{\text{Cu}} = 1300 \text{ nm}$ at 80 K (blue line), and the dashed curves are theoretical upper limits using $P = 1$, calculated from the equation, $\Delta R_S A = P^2 (S_{\text{inj}} S_{\text{det}} / S_N) \rho_{\text{Cu}} \lambda_{\text{Cu}} e^{-d/\lambda_{\text{Cu}}}$.

the inset. The value of $\Delta R_S A$ increases with decreasing d for both the CFS/Cu and Py/Cu LSV devices. By solving the one-dimensional spin diffusion equation^{6,26} (see Supplementary information), $\Delta R_S A$ can be expressed as follows:

$$\Delta R_S A \approx \frac{\left(\frac{P_F}{(1-P_F^2)} \rho_F \lambda_F + \frac{P_I}{(1-P_I^2)} R A_{\text{F/N}} \right)^2}{\rho_N \lambda_N \sinh(d/\lambda_N)}, \quad (1)$$

where P_F and P_I are the bulk and interface spin polarizations for F, respectively; λ_F and λ_N are the spin diffusion lengths for F and N, respectively; and ρ_F and ρ_N are the resistivities for F and N, respectively. As shown in Figure 3, the plots of $\Delta R_S A$ versus d for both the CFS/Cu and Py/Cu LSV devices are well reproduced by the fitting curves with $\lambda_{\text{Cu}} = 500 \text{ nm}$ and $\lambda_{\text{Cu}} = 1300 \text{ nm}$ at RT and 80 K,^{1,2,6,7} respectively. For the Py/Cu LSVs, assuming that $\lambda_{\text{Py,RT}} = 3 \text{ nm}$ and $\lambda_{\text{Py,80K}} = 5 \text{ nm}$, we obtain a reasonable P_{Py} values of 0.3 and 0.35 at RT and 80 K, respectively.^{1,2,6} Thus, the above equation reliably expresses the generation efficiency of the pure spin current among various LSVs. We then estimate the spin polarization for CFS (P_{CFS}). As it is impossible to determine the spin polarization and the spin diffusion length independently from the present results, we assume that λ_{CFS} is of the same order as that of $\text{Co}_2\text{FeSi}_{0.5}\text{Al}_{0.5}$ (see Supplementary information). Using the experimentally obtained values of ρ_{CFS} , ρ_{Cu} and λ_{Cu} and assuming that $\lambda_{\text{CFS,RT}} = 3 \text{ nm}$ and $\lambda_{\text{CFS,80K}} = 4 \text{ nm}$,^{24,25} P_{CFS} can be estimated to be 0.55 at RT and 0.73 at 80 K. These values for CFS are larger than those for Py, and they are consistent with the relatively large value of 0.59 obtained in one of our CFS films with a point-contact Andreev reflection measurement.²⁵ Although the present CFS epitaxial layers have highly ordered structures with a high magnetic moment above $5 \mu_B/\text{f.u.}$,²⁴ the value is still smaller than $6 \mu_B/\text{f.u.}$ in the perfectly ordered CFS.^{20,21} As further

enhancement of the P_{CFS} will be achieved by improving the crystal growth technique, a scaling characteristic with $P=1$ will ultimately be obtainable (see dashed line).

In the present CFS/Cu LSV, the non-negligible backflow of the spin current remains. However, the results described above indicate that the device performance of the LSV is strongly improved with the CFS spin injector instead of the Py injector. To evaluate the device performance of our developed CFS/Cu LSV quantitatively, we compare the generation efficiency of the pure spin current in the CFS/Cu LSV to that in the Py/Cu LSV. Using Eq. (1), the injection efficiency of the pure spin current η_{Is} , which is defined by the ratio of the spin current (I_{S}) injected into the ferromagnetic detector to the excited charge current (I_{C}), can be calculated as follows:

$$\eta_{\text{Is}} \equiv \frac{I_{\text{S}}}{I_{\text{C}}} \approx \frac{1}{2} \frac{S_{\text{N}}}{S_{\text{inj}}} \frac{\left(\frac{P_{\text{F}}}{(1-P_{\text{F}}^2)} \rho_{\text{F}} \lambda_{\text{F}} + \frac{P_{\text{I}}}{(1-P_{\text{I}}^2)} R_{\text{AF/N}} \right)}{\rho_{\text{N}} \lambda_{\text{N}} \sinh(d/\lambda_{\text{N}})}. \quad (2)$$

The injection efficiency of the present CFS/Cu LSV with $d=200$ nm is estimated to be 0.27, which is much larger than the value of 0.022 for the Py/Cu LSV. Thus, the injection efficiency is strongly improved with the CFS spin injector, although the non-negligible backflow of the spin current remains. We then evaluate the power consumption for switching the magnetization by injecting the pure spin current with CFS and Py spin injectors. The injection power for switching can be estimated as $(I_{\text{SCR}}/\eta_{\text{Is}})^2 R$, where I_{SCR} is the critical spin current for switching and R is the device resistance, which is mainly dominated by the resistance of the ferromagnetic injector. Assuming that I_{SCR} is the same in devices with either spin injector (see Supplementary information), we find that the CFS injector substantially reduces the power consumption for switching to 3% of that required for the Py injector. The present result represents a significant technological advance in spintronics using pure spin currents, which are generated by Heusler-compound spin injectors with high spin polarization.

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ERRATUM

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