Spin mixing conductance in Cu–Ir dilute alloys

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We have investigated the spin mixing conductance at the interface of Py and Cu–Ir dilute alloys by means of spin pumping and inverse spin Hall effect (ISHE) measurements for Cu–Ir. From systematic studies of the effective spin mixing conductance as a function of Ir concentration, we found that the interfacial spin mixing conductance $g^{11}$ was proportional to the Ir concentration. This result is consistent with a scenario based on the impurity-density dependent density of states at the Fermi energy. From the ISHE measurements as a function of Ir concentration, we show that the mechanism of the spin Hall effect is skew scattering and that the spin Hall angle is comparable to the previously determined value by using the lateral spin valve structures. © 2016 The Japan Society of Applied Physics

![Diagram](https://example.com/diagram.png)

**Fig. 1.** Schematic illustrations of the device structures used for the FMR and spin-pumping experiments: (a) detection scheme of ISHE and (b) conversion process from spin current generated by spin-pumping to charge current.

Electric spin injection1) and spin pumping2) are representative spin-injection methods employed for determining the spin Hall angle and the spin diffusion length, both of which are important characteristic parameters for spin transport. In particular, the latter spin pumping technique has been widely accepted, since its first demonstration, as a method for spin injection because its simple device structure requires no microfabrication. In addition, spin pumping is a versatile method applicable to a variety of materials, including metals,3) semiconductors,2) and even organic conductors.4) In this paper, we focus on spin mixing conductance, which determines the amount of injected spin current. The spin current originates from the magnetization precession in a ferromagnetic (F) layer exchange coupled with a nonmagnetic metal (NM) layer; thus, the spin mixing conductance at the interface should correlate with the density of states (DOS) of the NM. In order to verify this assumption, we chose Cu–Ir dilute alloys in which Ir impurities promote 5d–4s hybridization, increasing their DOS systematically as a function of the Ir concentration. In Cu-5d transition metal binary alloys, resonant scattering causes the spin Hall effect (SHE).5) Cu–Ir alloys are expected to induce the largest spin Hall angle dominantly caused by skew scattering, which is also confirmed with the non-local spin valve method.6)

In general, the total amount of spin current crossing the F/NM interface is determined from the effective spin mixing conductance $g^{11}_{\text{eff}}$, given by the difference in the linewidth of ferromagnetic resonance (FMR) spectra for the F layer with and without the NM. This spin current includes the backflow driven by the spin accumulation built up in the NM layer. In contrast, the spin mixing conductance is the interfacial parameter $g^{11}$, which is determined by the transmission and reflection coefficients of conduction channels at the interface.7) We can therefore deduce $g^{11}$ from $g^{11}_{\text{eff}}$ by taking into account the backflow.8)

This work reports our investigation of the development of $g^{11}$ by systematically varying the Ir concentration to clarify how Ir impurities affect the amount of injected spin current at the F/Cu–Ir interface.

Figures 1(a) and 1(b) show schematic illustrations of our measurement configuration for inverse SHE (ISHE) using a strip-shaped bilayer structure consisting of a 5-nm-thick permalloy (Py: Ni81Fe19) and a 20-nm-thick Cu–Ir layer. The sample has a rectangular shape with dimensions of 40 $\times$ (900–1100) $\mu$m$^2$. The Py layer was grown on a thermally oxidized silicon substrate by electron-beam evaporation with a rate of about 0.6 Å/s, while the Cu and Cu–Ir layers with different Ir concentrations (1, 3, 6, and 9%) were deposited by magnetron sputtering. All deposition processes were performed without air exposure. The bilayer stripe was then wired using silver pastes and inserted into the center of a TE102 rectangular cavity. The operation frequency was approximately 9.45 GHz. At the FMR resonance field, pure spin current was injected by means of interfacial exchange interaction, that is, spin pumping, and was then converted to the transverse electrical current by means of ISHE in Cu–Ir alloy, as shown in Fig. 1(b). In Fig. 2, we show typical FMR spectra of Py for Py/MgO and Py/Cu–Ir 1% bilayers. The FMR linewidth of Py/Cu–Ir 1% becomes broader than the linewidth of the Py/MgO bilayers. The observed difference indicates that additional damping is caused by the pure spin current into Cu–Ir, while no spin current is injected to the insulating MgO layer.

This additional damping was interpreted as the dissipation of spin current. Therefore, we estimated the amount of spin current from this additional damping parameter. The amount of injected spin current at the interface is expressed5) as

$$j_s^0 = \frac{h}{4\pi} g^{11}_{\text{eff}} P_{\text{cont}} \sin^2 \Theta. \quad (1)$$

Here, $g^{11}_{\text{eff}}$ is the effective spin mixing conductance, and $\Theta$ is the precession cone angle, which was calculated from the rf magnetic field ($H_{rf}$) determined from the $Q$-factor of the
$P_e$ is the correction factor originating from the elliptical precession of ferromagnetic magnetization, $\delta_{\text{eff}}$ is the driving frequency, and $\hbar$ is the reduced Planck constant. The effective spin mixing conductance is obtained from the difference of the FMR linewidths,

$$s_{\text{eff}}^{11} = \frac{4\pi g M_{\text{Py}}}{g g_{\text{eff}}} (\Delta H_{\text{Py/Cu-Ir}} - \Delta H_{\text{Py/MgO}})$$  \tag{2}

using the linewidths of FMR spectra for the Py/Cu–Ir ($\Delta H_{\text{Py/Cu-Ir}}$) and Py/MgO ($\Delta H_{\text{Py/MgO}}$) bilayers. In this expression, $g$, $\gamma$, $M_s$, and $\mu_B$ are the $g$-factor, gyromagnetic ratio, saturation magnetization, and Bohr magneton, respectively. As expressed by Eq. (1), the effective spin mixing conductance determines the net injected spin current, which includes the backflow into the ferromagnet and interfacial loss of the spin current, that is, the spin memory loss (SML). As discussed in Ref. 8, the effective spin mixing conductance can be expressed as

$$s_{\text{eff}}^{11} = \left[ \frac{1}{g^2} + \frac{e^2}{h \rho \lambda_{\text{sd}} \delta \sinh \delta + R^* \cosh \delta \tanh (\delta/\lambda_{\text{sd}})} \right]^{-1}$$  \tag{3}

where $g^{11}$, $\rho$, $\lambda_{\text{sd}}$, $R^*$, and $\delta$ are the interfacial spin mixing conductance, electrical resistivity, spin diffusion length of Cu–Ir, interfacial specific resistance, and SML parameter, respectively. The SML is understood by presuming a fictitious interfacial layer in the NM layer, which is attached to the F layer. Assuming that this fictitious layer has a different spin-diffusion length $\lambda$ and thickness $t$, $\delta$ is defined as $\delta \equiv \lambda / \lambda_{\text{sd}}$. We then obtain the spin memory-loss parameter $\delta = 0.4$ from the relation $P = 1 - \exp(-\delta)$ with the interfacial spin-flip probability $P = 0.33$ determined by Manchon et al.\textsuperscript{12} Note that the parameter $P$ in our discussion corresponds to $\delta$ in Ref. 12. $\lambda_{\text{sd}}$ of Cu at room temperature (120 nm) was calculated by using $\lambda_{\text{sd}}$ at 4 K (300 nm) obtained from weak antilocalization measurements of Cu nanowire\textsuperscript{13} and by assuming the same phonon contribution to $\lambda_{\text{sd}}$ as in Ref. 14. $R^*$ used in this calculation is a reported value from the giant magnetoresistance (GMR) experiment.\textsuperscript{10} Substitution of all the parameters into Eq. (3) yields $g^{11}$ for each Ir concentration.

In Fig. 3, both the effective spin mixing conductance ($s_{\text{eff}}^{11}$) and the spin mixing conductance ($g^{11}$) are plotted as functions of the Ir impurity concentration in Cu. In this figure, $s_{\text{eff}}^{11}$ shows a tendency to saturate in the high Ir concentration, while $g^{11}$ lineally increases with the Ir concentration. In the high Ir concentration region, most of the spin current flows back into the ferromagnetic layer; thus, $g^{11}$ largely deviates from $g_{\text{eff}}^{11}$. The linear increase of $g^{11}$ with the Ir concentration can be interpreted as the increase of DOS by Ir doping at the Fermi level. The 5d orbital of Ir atoms forms a resonance level near the Fermi level; thus, the DOS at the Fermi level is expected to increase proportionally with the Ir concentration. According to theoretical predictions,\textsuperscript{16,17} $g^{11}$ is proportional either to the number of conduction channels at the interface\textsuperscript{16} or to the Pauli paramagnetic susceptibility\textsuperscript{17} of the NM. Both discussions agree with our conclusion, where $g^{11}$ is proportional to the DOS of the NM.

Next, we will discuss the SHE in Cu–Ir alloys. The inset of Fig. 4 shows the raw voltage signal observed in the Py/Cu–Ir 9% bilayer. As illustrated in Fig. 1(b), the injected spin current is converted to the charge current by means of the ISHE in Cu–Ir; thus, we can observe the voltage signal in an open-circuit configuration. Since the voltage signal due to the ISHE has a symmetric Lorentzian function with the FMR field, we extracted the symmetric voltage contribution from the raw voltage signal using a combination of symmetric and asymmetric Lorentzian functions expressed as

$$V(H) = \left[ \frac{V_S (\Delta H / 2)^2}{(H - H_0)^2 + (\Delta H / 2)^2} - \frac{V_{AS} \Delta H (H - H_0)}{(H - H_0)^2 + (\Delta H / 2)^2} \right] \left( \frac{1}{L} \right)$$  \tag{4}

where $\Delta H$ is the spectral full width at half maximum and $H_0$.
The main panel of Fig. 4 shows the extracted distance between two voltage electrodes for the normalized impurities. The dotted line is a linear fit to the data. This is consistent with the expected behavior of ISH voltage induced by spin pumping. In this study, we also considered a DC-voltage contribution coming from the Py layer during the FMR excitation. The mechanisms for this voltage, as discussed in several sources in the literature, are not fully understood. Actually, we observed the typical value of the normalized voltage of −0.001 V/m in the measurement of a Py single layer ($V^P_{S}$). However, for the Py/Cu–Ir bilayer, this voltage signal is significantly reduced by the shunting effect of the Cu–Ir layer. Thus, the observed $V_S$ for the Py/Cu–Ir bilayer sample was dominated by the ISHE of Cu–Ir. To obtain the precise value of $V^ISH_{S}$, we subtracted the contribution of $V^P_{S}$ from the observed $V_S$ based on the model, where the resistances of the Py and Cu–Ir layers were connected in parallel. The resulting voltage signal ($V^ISH_{S}$) was used to calculate the spin Hall resistivity. The spin Hall resistivity ($\rho^ISH$) is given by $V^ISH_{S}$ and $j_S$, and thus, we can calculate $\rho^ISH$ using

$$\rho^ISH = \frac{2eV^ISH_{S}/L}{\hbar j_S}. \quad (5)$$

Here, $j_S^eff$ is the total amount of spin current contributing to the ISHE. In this study, the analysis of spin mixing conductance suggests the presence of non-negligible SML effects that affect the spin-current contribution to the ISHE. Thus, the spin accumulation diminishes as the spin-flip probability increases with $1 - e^{-\delta}$. Because the spin current is proportional to the gradient of spin accumulation, the spin current at the interface ($z = 0$) is given by $j^eff_{S}(z = 0) = e^{-\delta} j^0_{S}$. Here we assume that the thickness of the fictitious layer is much smaller than that of the NM layer, that is, $t_l \ll t_N$. $j^eff_{S}(z = 0)$ relaxes in the Cu–Ir layer as

$$j^eff_{S}(z) = j^0_{S}(z = 0) \frac{\sinh[(t_N - z)/\lambda_{sd}]}{\sinh(t_N/\lambda_{sd})},$$

with the boundary condition of $j^eff_{S}(z = 0) = 0$ at $z = t_N$. Therefore, $j^0_{S}$ is calculated as

$$\int_0^{t_N} j^eff_{S}(z)dz = j^eff_{S}(z = 0) = \lambda_{sd} \cdot \tanh\left(\frac{t_N}{2\lambda_{sd}}\right).$$

Figure 5 shows the spin Hall resistivity as a function of the impurity contribution of the resistivity $\rho^{Cu-Ir}/\rho^{Cu}$, indicating clearly the linear dependence. This linear dependence suggests that the responsible mechanism of SHE is the skew scattering. The spin Hall angle estimated from the slope is $1.4 \pm 0.3\%$. This is comparable to the previously reported value obtained by performing non-local spin valve measurements.

In summary, we have performed spin-pumping experiments for Cu–Ir dilute alloys. Our systematic study of the dependence of the effective spin mixing conductance on Ir doping revealed that the interfacial spin mixing conductance $g^{11}$ varies proportionally with the Ir concentration, reflecting the Ir concentration-dependent DOS at the Fermi energy. In addition, the ISHE measurements clearly showed that skew scattering is the mechanism of the spin Hall effect. The spin Hall angle obtained from the ISHE measurements is comparable to the previously reported value estimated by means of the spin-absorption method using lateral spin valve structures.

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