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Critical effect of spin-dependent transport in a tunnel barrier on enhanced Hanle-type signals observed in three-terminal geometry

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The MgO thickness dependence of Hanle signals in $Co_{50}Fe_{50}$/MgO/n-Si tunnel junctions was investigated using a three-terminal geometry. The observed Hanle signal is several orders of magnitude stronger than the predicted value by conventional theory as reported in many literatures. Furthermore, the magnitude of the spin signal depends on the junction resistance rather than the channel resistance, implying that the largest part of the observed Hanle signal is not caused by spin accumulation in the semiconductor region. A possible origin of the observed strong Hanle signal is due to a modulation of the tunneling resistance by a magnetic field, which is induced by the spin precession in localized states formed in the vicinity of the $Co_{50}Fe_{50}$/MgO interface. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4754545]

The injection of spin-polarized electrons from a ferromagnet (F) into a semiconductor (SC) has attracted much interest for creating viable semiconductor-based spintronics. The Hanle effect measurement using a three-terminal geometry has been widely used to evaluate the spin accumulation in a semiconductor beneath the ferromagnetic electrode, and clear Hanle-type signals have been observed in several semiconductor materials, such as GaAs,[1] Si,[2–6] and Ge.[7–11] In most cases,[1–5,7–11] however, the magnitude of these Hanle-type signals was several orders of magnitude higher than those expected from theory.[12] This contrasted with the results for the four-terminal non-local geometry, in which the magnitude of the spin signal was modest.[13–16] Tran *et al.* proposed a sequential tunneling model to explain the enhanced Hanle-type signal observed in Co/AlO_x/GaAs junctions with the three-terminal geometry.[1] They assumed a sequential tunneling process via localized states (LS) formed in the vicinity of the semiconductor interface, and showed that the large resistance between the LS and the semiconductor channel, which was due to a depletion region existing in the interfacial region of SC facing the tunneling barrier, induced the enhancement of the Hanle signal. Dash *et al.*[2] and Iba *et al.*,[11] on the other hand, showed the Hanle signal could be enhanced even in the absence of a depletion region at the semiconductor interface. Thus, the details of the enhancement mechanism are not yet understood. Considering that the enhancement of the signals was observed in F/insulator (I)/SC junctions with relatively large junction resistance, systematic investigation of the junction-resistance dependence of the enhanced Hanle signals is necessary.

The purpose of this paper is to clarify the origin of the enhanced signal observed in the three-terminal Hanle-effect measurement through investigation of the MgO thickness (t_{MgO}) dependence of the signals in $Co_{50}Fe_{50}$(CoFe)/MgO/n-Si. As we explain in this paper, we experimentally found that the magnitude of the enhanced Hanle-type signal strongly depends on the junction resistance rather than on the channel resistance. This indicates that the largest part of the observed Hanle-type signal is not caused by spin accumulation in Si. Based on our experimental findings, we discuss the origin of the enhancement mechanism widely observed in F/I/SC junctions.

A heavily doped n-type silicon-on-insulator (SOI) substrate was used for the semiconductor channel. The resistivity (ρ) and the thickness of the SOI layer were 5 m$\Omega\cdot$cm at RT and 5 μm, respectively. The substrate was cleaned with HF solution, and heated at 550 °C for 1 h in an ultra-high vacuum chamber with a base pressure of $\sim 6\times10^{-8}$ Pa. A MgO wedge layer ranging from 1 to 3 nm was then deposited by electron beam evaporation at RT, and a 10-nm-thick CoFe layer was grown by magnetron sputtering at RT. Junctions with areas ranging from 50×50 to 250×250 μm^2 were fabricated by photolithography and Ar-ion milling techniques. The current (I)–voltage (V) characteristics and magnetoresistance (MR) of each junction were measured at 293 K using a three-terminal geometry (Fig. 1(a)). In the MR measurement, the voltage across the junction was measured

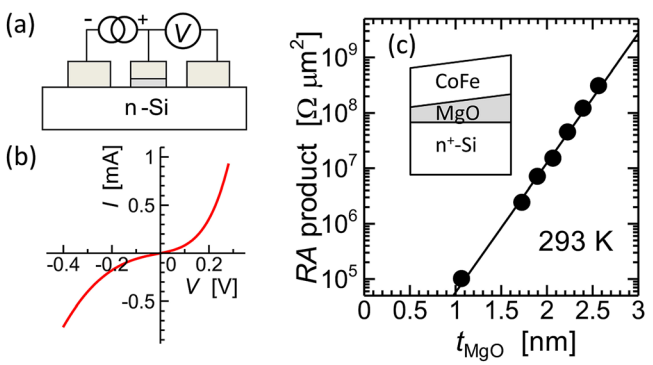


FIG. 1. (a) Schematic diagram for three-terminal geometry. The central electrode is made of CoFe/MgO and the outer electrodes are made of Au. (b) I–V characteristics at 293 K for a CoFe/MgO/n-Si junction with a MgO thickness (t_{MgO}) of 2.2 nm. (b) Resistance-area products of CoFe/MgO/n-Si single junctions as a function of t_{MgO}.

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under a constant bias current (I_{bias}) as functions of both in-plane and out-of-plane magnetic field. The bias polarity was defined with respect to the Si channel.

Figure 1(b) shows *I–V* characteristics for a CoFe/MgO/n-Si junction with t_{MgO} of 2.2 nm. The *I–V* curve exhibits nonlinear characteristics and is almost symmetric against the bias polarity, indicating that the tunneling conduction is dominant. A slightly larger current in the positive bias region is most probably due to a thermionic emission current through a Schottky barrier at the MgO/Si interface. Figure 1(c) shows the resistance-area products ($R \cdot A$) of CoFe/MgO/n-Si single junctions as a function of t_{MgO}, where R is the resistance which was evaluated from the slope of the *I–V* curve at $V = 0$ V, and A is the junction area. The $R \cdot A$ increases exponentially with increasing t_{MgO}. The value of $m^*\phi$ estimated from the slope of $\ln(R \cdot A)$ versus t_{MgO} according to the Wenzel-Kramer-Brillouin (WKB) approximation is 0.28 eV, where m^* is the effective mass of a tunneling electron normalized by the bare electron mass and ϕ is the potential barrier height. This value is comparable to the 0.32 eV obtained in $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/Co_{50}Fe_{50}$ magnetic tunnel junctions with relatively high tunnel magnetoresistance ratios of 90% at RT and 240% at 4.2 K.[17] These results indicate that the MgO layer grown on Si works properly as a tunnel barrier.

Figure 2(a) shows the change in the junction voltage at $I_{\text{bias}} = -20\ \mu$A for a CoFe/MgO/n-Si junction with t_{MgO} of 2.2 nm as functions of both in-plane and out-of-plane magnetic fields. Under this bias condition, electrons tunnel from CoFe to Si (spin injection). The junction resistance decreases as the out-of-plane magnetic field increases, while it increases as the in-plane magnetic field increases. Similar out-of-plane MR characteristics[1–11,18] and in-plane MR characteristics[5,7–9,11,18] have been reported in many literatures. The out-of-plane MR was explained by the Hanle effect in the semiconductor channel. The in-plane MR, on the other hand, was explained by the inverted Hanle effect, in which the in-plane magnetic field suppresses the spin precession induced by the stray field in junctions with a finite interface roughness, resulting in increased junction resistance with an in-plane magnetic field.[18] If we assume that the observed MR characteristics come from the Hanle effect and the inverted Hanle effect for the injected spins into Si, the spin lifetime (τ_{sf}) and the spin diffusion length (λ_{sf}) are estimated to be approximately 150 ps and 200 nm at 293 K. These values are comparable to those obtained in similar structures.[2,5] The magnitude of the spin signal (ΔV) defined by the sum of the voltage change in the Hanle curve and the inverted Hanle curve is 0.31 mV, and the spin resistance-area product ($\Delta R_S \cdot A$), where ΔR_S is defined by $\Delta V/I_{\text{bias}}$, is 350 kΩ · μm^2. In the case of $R \cdot A \gg \rho\lambda_{sf}$, a condition satisfied in our devices, $\Delta R_S \cdot A$ predicted by the conventional theory of transport across a single F/I/SC junction is[12]

$$\Delta R_S \cdot A = \gamma^2 \rho \lambda_{sf}, \tag{1}$$

where γ is the spin polarization for spin-dependent tunneling. However, the observed value of $\Delta R_S \cdot A$ (350 kΩ · μm^2) for $t_{\text{MgO}} = 2.2$ nm is more than four orders of magnitude larger than the maximum theoretical value of $\rho\lambda_{sf}$ (approximately 10 Ω · μm^2), indicating the enhancement effect. Thus, our CoFe/MgO/n-Si junctions also showed the enhanced Hanle signals measured with the three-terminal geometry as often reported in the literature.[1–5,7–11] Figure 2(b) shows ΔV along with the junction voltage V as a function of I_{bias} for the same junction as in Fig. 2(a). The magnitude of V is scaled by a factor of 0.008. In the negative bias region, where the tunneling conduction is dominant, the ΔV–I_{bias} curve matches well with the scaled V–I_{bias} curve, indicating that ΔV is proportional to V in the relation of $\Delta V/V = 0.8\%$. In the positive bias region, on the other hand, both curves are slightly deviated most probably due to a thermionic emission current along with the tunneling current, suggesting that the ΔV is originating from tunnel conduction.

Figure 3 shows $\Delta R_S \cdot A$ at $I_{bias} = -20\ \mu$A as a function of t_{MgO}. The value of $R \cdot A$ is also plotted for comparison. Most importantly, the observed $\Delta R_S \cdot A$ values show exponential dependence on t_{MgO}, and ΔR_S is proportional to R over a relatively wide range of $R \cdot A$ from 3×10^6 to 5×10^8 Ω · μm^2. Note that if Eq. (1) of the model of Fert and Jaffrès[12] is applicable, the maximum theoretical value for $\Delta R_S \cdot A$ is given by $\rho\lambda_{\text{sf}}$, and $\Delta R_S \cdot A$ should be constant against the junction resistance. Because of the exponential dependence of $\Delta R_S \cdot A$ on t_{MgO}, a value of about 10^4 for the ratio of $\Delta R_S \cdot A$ to the theoretical value of $\rho\lambda_{\text{sf}}$ for the junction with t_{MgO} of 2.2 nm mentioned above is a typical value for the given t_{MgO} value. We also observed exponential dependence of $\Delta R_S \cdot A$ on t_{MgO} for CoFe/MgO/n-Ge junctions,[24] indicating that our results are valid for a wide range of F/I/SC junctions with relatively high junction resistance.

Now, we will discuss the possible origin of the enhancement mechanism. We first discuss if the sequential tunneling model proposed by Tran *et al.*[1] can explain our results.

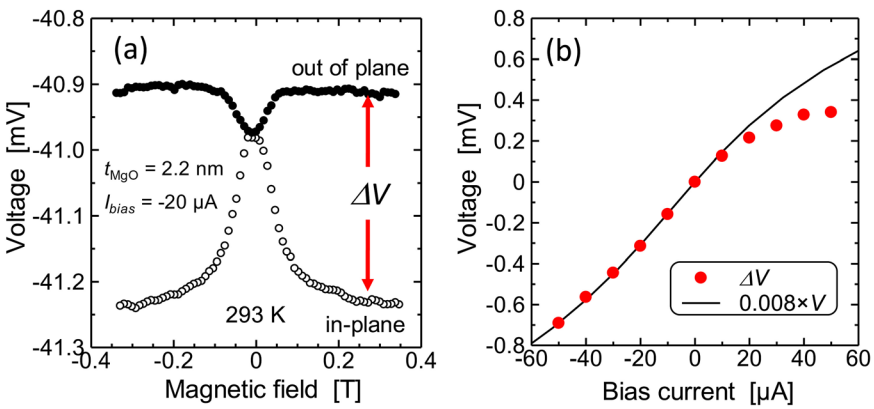


FIG. 2. (a) Junction voltage (V) at $I_{\text{bias}} = -20\ \mu$A as functions of both in-plane and out-of-plane magnetic fields, and (b) I_{bias} dependence of ΔV for a CoFe/MgO/n-Si junction with t_{MgO} of 2.2 nm. ΔV is defined by the sum of the voltage change in the Hanle curve and the inverted Hanle curve. In (b), the magnitude of V is scaled by a factor of 0.008 and also plotted as a function of I_{bias}.

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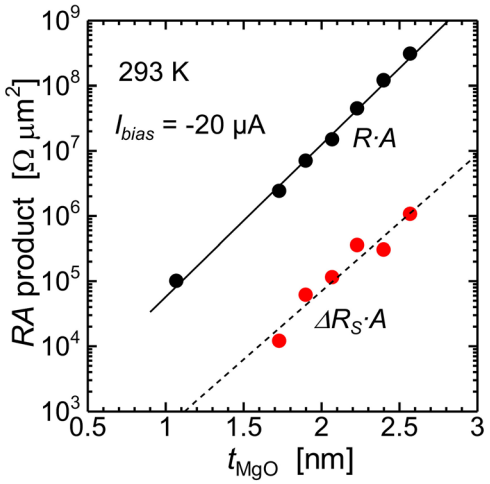


FIG. 3. t_{MgO} dependence of $\Delta R_S \cdot A$ at $I_{bias} = -20\,\mu$A for CoFe/MgO/n-Si junctions. The value of $R \cdot A$ was also plotted for comparison.

Based on the Based on the sequential tunneling model proposed by proposed by Tran *et al.*,[1] $\Delta R_S \cdot A$ is given by

$$\Delta R_S \cdot A = \gamma^2 \frac{r_{LS}(r_{ch} + r_b)}{r_{LS} + r_{ch} + r_b}, \tag{2}$$

where r_b is leakage resistance between the LS and the semiconductor channel, r_{LS} is spin-flip resistance associated with the LS, and r_{ch} is semiconductor resistance defined by $\rho\lambda_{sf}$. In the case of $r_{LS} \gg r_b$, r_{ch}, $\Delta R_S \cdot A$ is enhanced by a factor of $1 + r_b/r_{ch}$, compared with the case without LS. If r_b is much larger than r_{ch}, one can explain the enhancement effect of the Hanle signals. Tran *et al.*[1] assumed that the LS is formed in the vicinity of the semiconductor interface to produce a large r_b due to depletion of the semiconductor surface. If the LS exist at the MgO/Si interface, however, r_b is independent of t_{MgO} because r_b is the resistance between the LS and Si channel; from Eq. (2), this means that $\Delta R_S \cdot A$ should be independent of t_{MgO}. This assumption cannot explain our experimental results that $\Delta R_S \cdot A$ does depend on t_{MgO}. Thus, the sequential tunneling model cannot explain our results consistently.

We now propose a model of the modulation of spin-dependent tunneling resistance by a magnetic field. Based on the conventional theory,[12,19] ΔV should be equal to $\gamma\Delta\mu/2$, where $\Delta\mu$ is a split of the chemical potentials in the SC. In this formula, however, the effect of modulation of tunneling resistance by a magnetic field is not taken into account. Here, we extend this formula under the assumption that the tunneling probability of electrons depends on the magnetic field. Figures 4(a) and 4(b) show a schematic band diagram

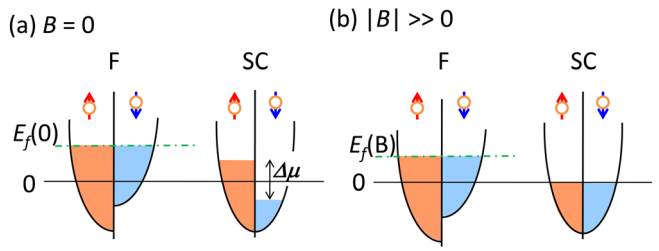


FIG. 4. Schematic band diagram of F/I/SC junctions under an out-of-plane magnetic field (B) of (a) $B = 0$ and (b) $|B| \gg 0$.

of F/I/SC junctions under an out-of-plane magnetic field (B) of (a) $B = 0$ and (b) $|B| \gg 0$. In Fig. 4(a), the chemical potentials are split by $\Delta\mu$ in the SC due to the spin injection, and in Fig. 4(b) the splitting is reduced by B due to the Hanle effect. The experiments are carried out with a constant current supply. This condition requires that the tunneling current density J at $B = 0$ and at $|B| \gg 0$ be equal. From this requirement, we get

$$\begin{aligned}&(E_f(0) - \Delta\mu/2)D_\uparrow T_\uparrow(0) + (E_f(0) + \Delta\mu/2)D_\downarrow T_\downarrow(0)\\ &\quad = E_f(B)D_\uparrow T_\uparrow(B) + E_f(B)D_\downarrow T_\downarrow(B),\end{aligned} \tag{3}$$

where $\Delta\mu = 2e\gamma\rho\lambda_{sf}J$, e (< 0) is an electron charge, $E_f(B)$ is the electrical chemical potential of the ferromagnetic electrode as a function of B, $D_{\uparrow(\downarrow)}$ is the effective density of states for majority (minority) spins at the Fermi level, and $T_{\uparrow(\downarrow)}(B)$ is the tunneling probability of majority (minority) spins, which is assumed to depend on B. The left-hand side and the right-hand side of Eq. (3) represent the tunneling current densities at $B = 0$ and at $|B| \gg 0$, respectively. In the three-terminal geometry, $E_f(B)$ is measured with respect to the electrochemical potential of the reference electrode. From Eq. (3), $\Delta R_S \cdot A$ defined by $[E_f(0) - E_f(B)]/eJ$ is given by

$$\begin{aligned}\Delta R_S \cdot A &= \frac{\Delta\mu}{2eJ}\frac{D_\uparrow T_\uparrow(0) - D_\downarrow T_\downarrow(0)}{D_\uparrow T_\uparrow(0) + D_\downarrow T_\downarrow(0)}\\ &+ \frac{E_f(B)}{eJ}\frac{D_\uparrow[T_\uparrow(B) - T_\uparrow(0)] + D_\downarrow[T_\downarrow(B) - T_\downarrow(0)]}{D_\uparrow T_\uparrow(0) + D_\downarrow T_\downarrow(0)}\\ &= \gamma^2\rho\lambda_{sf} + R \cdot A\frac{D_\uparrow[T_\uparrow(B) - T_\uparrow(0)] + D_\downarrow[T_\downarrow(B) - T_\downarrow(0)]}{D_\uparrow T_\uparrow(0) + D_\downarrow T_\downarrow(0)}.\end{aligned} \tag{4}$$

The first term of the right-hand side of Eq. (4) gives the spin accumulation signal, as predicted in the conventional theory,[12,19] while the second term gives the signal caused by the B-dependent tunneling probability. In the conventional theory, the second term becomes zero, because the B-dependent tunneling probability is not taken into account, resulting in $\Delta R_S \cdot A = \gamma^2\rho\lambda_{sf}$. However, if $T_{\uparrow(\downarrow)}$ depends on B and $R \cdot A$ is much larger than $\rho\lambda_{sf}$, the second term becomes dominant, resulting in $\Delta R_S \cdot A$ being proportional to $R \cdot A$. This is exactly what we observed, as shown in Figs. 2(b) and 3. Thus, we conclude that the main origin of the enhanced Hanle-type signals widely observed in F/I/SC junctions, including our samples, is a modulation of the tunneling resistance by a magnetic field. The fact that the enhancement of the Hanle signals was observed in the junctions with relatively large resistance also supports this picture. Thus, the Hanle signal measurement using three-terminal geometry is not suitable for evaluating the exact value of $\Delta\mu$ in the channel, in particular, for junctions with a large $R \cdot A$ value. The four-terminal non-local geometry, on the other hand, has no such problem, because the detector contact is unbiased, resulting in that the second term of Eq. (4) is negligibly small. Therefore, $\Delta\mu$ in the semiconductor channel can be properly measured through the non-local voltage. The in-plane magnetic field dependence of the junction resistance (inverted Hanle curve) for the three-terminal geometry also can be explained consistently

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with this model, because the in-plane magnetic field reduces the out-of-plane component of the stray field arising from the interface roughness.

As described above, our experimental result that $\Delta R_S \cdot A$ is proportional to $R \cdot A$ can be explained if the tunnel resistance is modulated by a magnetic field. However, the microscopic physical model for this is not clarified at present. We provide here one possible physical model. Considering that ΔV vs. B curve can be fitted well by Lorentzian function, we assume that the tunneling electrons are depolarized to some extent due to a precession, and this induces the modulation of tunneling probability. However, it is impossible that the tunneling electrons precess in the MgO barrier since the tunneling time, which is estimated from the tunneling barrier thickness and the Fermi velocity of electrons, is too short. Therefore, we introduced LS at CoFe/MgO interface so that electrons are trapped and depolarized in the LS. We have calculated the tunnel current for a CoFe/MgO/Si junction using Slonczewski's model,[20–22] in which the tunnel probabilities for both majority and minority electrons have been calculated using an exact wave function within the free electron model. The applied magnetic field was treated as a precession field which induces a rotation of spins trapped in LS at CoFe/MgO interface and depolarizes the spin states. The steady-state spin polarization at LS under continuous injections is proportional to the time integral $\int_0^\infty \gamma \cos \omega_L t \exp(-t/\tau_{sf})\mathrm{d}t$, where ω_L is the Larmor frequency. The spin polarization is incorporated into the Fermi wave numbers for both majority and minority electrons in Slonczewski's model. Thus, we have also taken into consideration the spin depolarization by changing the Fermi wave numbers injected at CoFe/MgO interface. Figure 5(a) shows calculated tunnel resistance normalized by the value at $B = 0$ as a function of B, and Figure 5(b) shows $\Delta R_S \cdot A$ and $R \cdot A$ as a function of t_{MgO}, respectively. The typical parameters used in the calculation are as follows: $m^* \phi = 0.28\,\mathrm{eV}$, $\tau_{sf} = 150$ ps, $k_{\mathrm{F}\uparrow} = 1.05 \times 10^{10}\,\mathrm{m}^{-1}$, and $k_{\mathrm{F}\downarrow} = 0.44 \times 10^{10}\,\mathrm{m}^{-1}$,[23] which correspond to $\gamma = 41\%$, where $k_{\mathrm{F}\uparrow(\downarrow)}$ is Fermi wave number of majority(minority) spin in the ferromagnetic Fe electrode. Here, we have assumed that the polarization and wave numbers for CoFe are almost equal to those for Fe. Although the calculated value for $\Delta R_S/R$ of approximately 8.5% is almost ten-times larger than the experimental value, the proposed model can reproduce our experimental results qualitatively.

In conclusion, we have experimentally found that $\Delta R_S \cdot A$ is almost proportional to $R \cdot A$, suggesting that the largest part of the observed Hanle-type signal using a three-terminal geometry is not caused by spin accumulation in the semiconductor. We have discussed the origin of the Hanle-type signal, and indicated that the spin-dependent transport properties in the tunnel barrier play a crucial role for the enhanced value of the three-terminal Hanle-type signal. Thus, the three-terminal measurement is not suitable to detect exactly spin accumulation in the semiconductor for junctions with a large junction resistance, and the four-terminal non-local measurement is indispensable.

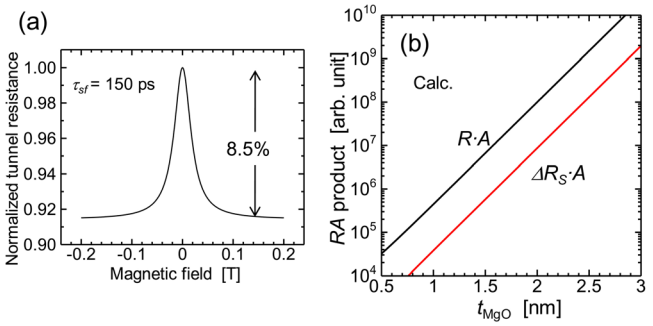


FIG. 5. (a) Calculated tunnel resistance as a function of out-of-plane magnetic field (B). The resistance was normalized by the value at $B = 0$. (b) Calculated $\Delta R_S \cdot A$ and $R \cdot A$ values as a function of t_{MgO}.

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