Spin Pumping and Inverse Spin Hall Effect in Germanium

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Abstract

We have measured the inverse spin Hall effect (ISHE) in $n$-Ge at room temperature. The spin current in germanium was generated by spin pumping from a CoFeB/MgO magnetic tunnel junction in order to prevent the impedance mismatch issue. A clear electromotive force was measured in Ge at the ferromagnetic resonance of CoFeB. The same study was then carried out on several test samples, in particular we have investigated the influence of the MgO tunnel barrier and sample annealing on the ISHE signal. First, the reference CoFeB/MgO bilayer grown on SiO$_2$ exhibits a clear electromotive force due to anisotropic magnetoresistance and anomalous Hall effect which is dominated by an asymmetric contribution with respect to the resonance field. We also found that the MgO tunnel barrier is essential to observe ISHE in Ge and that sample annealing systematically lead to an increase of the signal. We propose a theoretical model based on the presence of localized states at the interface between the MgO tunnel barrier and Ge to account for these observations. Finally, all of our results are fully consistent with the observation of ISHE in heavily doped $n$-Ge and we could estimate the spin Hall angle at room temperature to be $\approx 0.001$. 
I. INTRODUCTION

The first challenging requirement to develop semiconductor (SC) spintronics\textsuperscript{1,2} \textit{i.e.} using both carrier charge and spin in electronic devices consists in injecting spin polarized electrons in the conduction band of a SC at room temperature. SCs should be further compatible with silicon mainstream technology for implementation in microelectronics making silicon, germanium and their alloys among the best candidates.\textsuperscript{3} In Si, due to low spin-orbit coupling, very long spin diffusion lengths were predicted and measured experimentally.\textsuperscript{4–7} Germanium exhibits the same crystal inversion symmetry as Si, a low concentration of nuclear spins but higher carrier mobility and larger spin-orbit coupling which should allow in principle spin manipulation by electric fields such as the Rashba field.\textsuperscript{8–12} So far in order to perform spin injection from a ferromagnetic metal (FM) into Si or Ge, one needs to overcome at least three major obstacles: (i) the conductivity mismatch which requires the use of a highly-resistive spin-conserving interface between the FM and the SC,\textsuperscript{13} (ii) the Fermi level pinning at the SC surface due to the presence of a high density of interface states and the interface spin flips which are generally associated\textsuperscript{4,9,14} and finally (iii) the presence of random magnetic stray fields created by surface magnetic charges at rough interface\textsuperscript{9,15} around which the electrically injected spins are precessing and partly lost by decoherence. In this work, we have inserted a thin MgO tunnel barrier between Ge and the CoFeB ferromagnetic electrode in order to: (i) circumvent the conductivity mismatch and (ii) partly alleviate Fermi level pinning by strongly reducing the interface states density\textsuperscript{16–18} which leads to a modest Schottky barrier height at the MgO/$n$-Ge interface. We have then investigated the spin injection mechanisms using the so called three-terminal device.\textsuperscript{4} In this geometry, the same ferromagnetic electrode is used for spin injection and detection. This three-terminal device used in non-local geometry represents a simple and unique tool to probe spin accumulation both into interface states and in the SC channel.\textsuperscript{4,9,18} In particular, we could measure spin injection in the silicon and germanium conduction bands at room temperature.\textsuperscript{20,22} The spin Hall angle ($\theta_{\text{SHE}}$, ratio between the transverse spin current density and the longitudinal charge current density)\textsuperscript{23} is a key material parameter to develop new kinds of devices based on the spin Hall effect (SHE). The SHE is the conversion of a charge current into a spin current via the spin-orbit interaction. Conversely the inverse spin Hall effect (ISHE) is the conversion of a spin current into a charge current. Several methods have been developed to
determine quantitatively $\theta_{SHE}$: pure magnetotransport measurements on lateral spin valves (LSV)\textsuperscript{23–25} ferromagnetic resonance (FMR) along with spin pumping (SP-FMR)\textsuperscript{26–28} and spin torque FMR (ST-FMR)\textsuperscript{29} on ferromagnetic/non-magnetic bilayers (FM/N). Recently $\theta_{SHE}$ could be estimated in $n$ and $p$-GaAs\textsuperscript{40} as well as in $p$-Si\textsuperscript{30} by spin pumping and inverse spin Hall effect. The precession of the FM layer in direct contact with the SC has been excited by microwaves which pumps a spin current into the SC. The spin current was then detected by inverse spin Hall effect. In that case, the interface resistance to overcome the conductivity mismatch issue was given by the reminiscent Schottky barrier at the FM/SC interface. Here we have similarly used a combined SP-FMR method to study inverse spin Hall effect in germanium. In the first section, we describe the sample preparation and the experimental techniques. In the second section, the phenomenological models for the ferromagnetic resonance and electromotive force are presented. Finally the experimental results are shown and discussed in sections 3 and 4 respectively. In particular, we propose a microscopic model based on the presence of localized states at the MgO/Ge interface to explain the spin pumping mechanism in our system. We finally discuss about the influence of the MgO tunnel barrier and sample annealing on the ISHE signal.

II. SAMPLE PREPARATION

The multi-terminal device we initially used for electrical spin injection, detection and manipulation\textsuperscript{20} is made of a full stack Ta(5 nm)/CoFeB(5 nm)/MgO(3 nm) grown by sputtering on a 40 nm-thick germanium film on insulator (GOI).\textsuperscript{9} GOI wafers are made of a Si $p+$ degenerate substrate and a 100 nm-thick SiO$_2$ layer (BOX). They were fabricated using the Smart Cut\textsuperscript{TM} process and Ge epitaxial wafers.\textsuperscript{21} The transferred 40 nm-thick Ge film was $n$-type doped in two steps: a first step (phosphorus, $3 \times 10^{13}$ cm$^{-2}$, 40 keV, annealed for 1h at 550$^\circ$C) that provided uniform doping in the range of $10^{18}$ cm$^{-3}$, and a second step (phosphorus, $2 \times 10^{14}$ cm$^{-2}$, 3 keV, annealed for 10 s at 550$^\circ$C) that increased surface $n+$ doping to the vicinity of $10^{19}$ cm$^{-3}$. The thickness of the $n+$-doped layer is estimated to be 10 nm. The GOI surface was finally capped with amorphous SiO$_2$ to prevent Ge from surface oxidation. The tunnel barrier and ferromagnetic electrode were then fabricated from magnesium (Mg, 1.1nm) and cobalt-iron-boron (Co$_{60}$Fe$_{20}$B$_{20}$, 5nm) layers deposited by conventional DC magnetron sputtering onto germanium (Ge) after removing the SiO$_2$
capping layer using hydrofluoric acid and de-ionized water. The deposition rates were respectively of 0.02 nm.s$^{-1}$ and 0.03 nm.s$^{-1}$ at an argon pressure of $2 \times 10^{-3}$ mbar. The base pressure was $7 \times 10^{-9}$ mbar. All the depositions were performed at room temperature. The oxidation of the insulating barrier was performed by plasma oxidation, exposing the Mg metallic layer to a 30 seconds radio-frequency oxygen plasma at a pressure of $6 \times 10^{-3}$ mbar and a radio-frequency power of 100 W. Three successive Mg deposition plus oxidation steps were achieved ([Mg 1.1 / oxidation]$_3$) to grow a 3.3 nm thick MgO layer. The sample annealings were performed under $10^{-7}$ mbar at 300$^\circ$C for 90 minutes. The ferromagnetic layer is then capped with 5 nm of Ta to prevent oxidation. After depositing the spin injector, samples have been processed using standard optical lithography. In a first step, we define the ferromagnetic electrode (150 $\times$ 400 $\mu$m$^2$) and ohmic contacts (300 $\times$ 400 $\mu$m$^2$ on Ge and 100 $\times$ 100 $\mu$m$^2$ on top of the ferromagnetic electrode). In a second step, the germanium channel is etched down to the BOX to form a mesa of 1070 $\mu$m long and 420 $\mu$m large. Finally soft argon etching is used to remove the top 10 nm-thick $n$+-doped germanium layer. The whole device is shown in Fig. 1(a). In order to test the influence of the MgO tunnel barrier (resp. sample annealing), similar devices without MgO (resp. without annealing) were processed and studied. Ferromagnetic resonance and inverse spin Hall effect measurements were performed in a Bruker ESP300E X-band CW spectrometer with a cylindrical Bruker ER 4118X-MS5 cavity. The measurement geometry is depicted in Fig. 1(b). Complementary measurements of FMR lines at different frequencies between 2 and 24 GHz were performed in a stripe-line vector network analyzer system.

III. FERROMAGNETIC RESONANCE AND ELECTROMOTIVE FORCE

A. Ferromagnetic resonance (FMR)

Fig. 2(a) shows a schematic drawing of the reference sample and the definition of the magnetization and external magnetic field polar angles, $\theta_H$ and $\theta_M$, respectively. The reference sample is made of a single CoFeB layer grown on SiO$_2$ as follows: Ta(5nm)/CoFeB(5nm)/MgO(3.3nm)//SiO$_2$. We have inserted a thin MgO oxide layer between CoFeB and SiO$_2$ in order to make a comparison with the Ta(5nm)/CoFeB(5nm)/MgO(3.3nm)/Ge system studied in the next sections.
Figure 1: (Color online) (a) Schematic drawing of the multiterminal device used for SP-FMR measurements along with the definition of $\theta_H$ and $\theta_M$. $h_{rf}$ is the radiofrequency magnetic field. The thickness of each layer is given in nanometers between parenthesis. (b) Drawing of the device inserted into the cylindrical $X$-band electron paramagnetic resonance (EPR) cavity.

From FMR measurements (Fig. 2(b)), we can determine the peak-to-peak linewidth and the resonance field. By sweeping the external magnetic field $H$ under a microwave excitation of frequency $f$, the resonance condition is achieved in a ferromagnetic film when

$$
\left( \frac{\omega}{\gamma} \right)^2 = \frac{1}{M_s^2 \sin^2 \theta} \left[ \frac{\partial^2 F}{\partial \theta^2} \bigg|_{\theta_M, \phi_M} \frac{\partial^2 F}{\partial \phi^2} \bigg|_{\theta_M, \phi_M} - \left( \frac{\partial F}{\partial \theta \partial \phi} \bigg|_{\theta_M, \phi_M} \right)^2 \right] \tag{1}
$$

where $\omega = 2\pi f$ is the precession angular frequency, $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio with the Landé factor $g$, $\hbar$ is the reduced Planck constant, $\mu_B$ is the Bohr magnetron and $M_s$ is the saturation magnetization of the ferromagnetic film. The second order partial derivatives of the free energy density are evaluated at the equilibrium angles $\theta_M$ and $\phi_M$ of the magnetization $M$ for which: $\partial F/\partial \theta|_{\theta_M, \phi_M} = 0$ and $\partial F/\partial \phi|_{\theta_M, \phi_M} = 0$. The shape anisotropy in a ferromagnetic polycristalline film ($4\pi M_s$) is usually much larger than in-plane crystalline anisotropy. By recording FMR spectra with the DC magnetic field in the film plane at different azimuthal angles (not shown), we could demonstrate that in-plane anisotropy in the CoFeB electrodes used in this work is indeed negligible with respect to shape anisotropy. Thus we can consider that the free energy density is given by:
Figure 2: (Color online) (a) Schematic drawing of the reference sample Ta(5nm)/CoFeB(5nm)/MgO(3.3nm)//SiO$_2$. Electrical contacts are directly made on top of the metallic layer. The FMR spectrum (b) and the electromotive force (c) have been measured simultaneously by placing the sample in the EPR cavity. The red line in (c) is the fit according to Eq. (6). Symmetric and asymmetric contributions are also shown separately in blue dotted curves.

\[ F = -\mathbf{M} \cdot \mathbf{H} + 2\pi M_{\text{eff}}^2 \cos^2 \theta_M \]  

where the first term is the Zeeman energy and the second one accounts for shape anisotropy and any other perpendicular uniaxial anisotropy $H_{u\perp}$. The effective saturation magnetization $M_{\text{eff}}$ is thus defined as: $4\pi M_{\text{eff}} = 4\pi M_s + H_{u\perp}$. By minimizing numerically $F$ we can obtain the magnetization equilibrium angles: $\phi_{M0}$ and $\theta_{M0}$. The resonance field is then
Figure 3: Dispersion relationship in FMR experiments according to Eq. (1) and (2) by applying the external magnetic field at different polar angles $\theta_H$. Curves are shown every $2.5^\circ$ between $\mathbf{H}$ parallel ($\theta_H = 90^\circ$) and perpendicular ($\theta_H = 0$) to the film plane. The intercept of each curve with the dotted line ($f \approx 9.55$ GHz) yields the resonance field $H_{res}$.

given by combining Eq. (1) and (2), it can be plotted as a function of the external static field orientation ($\theta_H$) and the excitation frequency as shown in Fig. 3. $M_{eff}$ and $g$ are extracted from the out-of-plane (OOP) angular dependence of the resonance field using a least square fit (as shown for instance in Fig. 4(a,d)).

Analytical solutions of $H_{res}$ can be obtained for the parallel, $\theta_H = 90^\circ$, and perpendicular, $\theta_H = 0$, cases. In the parallel case, we find:

$$\left(\frac{\omega}{\gamma}\right)^2 = H_{res}(4\pi M_{eff} + H_{res})$$

(3)

The frequency dependence of the FMR linewidth also allows calculating the Gilbert damping constant $\alpha$ using the following expression:

$$\Delta H_{pp} = \Delta H_0 + \Delta H_G = \Delta H_0 + \frac{2}{\sqrt{3}} \frac{\omega}{\gamma}$$

(4)

where the peak-to-peak linewidth, $\Delta H_{pp}$, is measured when $\mathbf{H}$ is applied parallel to the film plane ($\theta_H = 90^\circ$). The $\Delta H_0$ term accounts for the frequency-independent contributions due to inhomogeneities in the ferromagnetic layer and $\Delta H_G$ is the FMR linewidth due to the Gilbert damping. As shown experimentally in section IV $\Delta H_0 << \Delta H_G$ at high frequency and we systematically neglect this contribution to the FMR linewidth. Moreover, the OOP angular dependence of the peak-to-peak linewidth $\Delta H_{pp}$ at a given frequency can be written:
\[ \Delta H_{pp} = \Delta H_G + \Delta H_\theta \]  

(5)

where the Gilbert contribution can be calculated from \( \Delta H_G = (2/\sqrt{3})\alpha (\omega/\gamma)/\cos(\theta_H - \theta_M) \), and \( \Delta H_\theta = |dH_{res}/d\theta_H|\Delta \theta \) is the angular dispersion of the perpendicular anisotropy and demagnetizing field \((4\pi M_{eff})\) due to inhomogeneities in the FM layer. We show in the following that \( \alpha \) and \( \Delta \theta \) can be extracted from the frequency and OOP angular dependences of \( \Delta H_{pp} \).

**B. Electromotive force measured on the reference sample**

The electromotive force generated in the ferromagnetic layer and shown in Fig. 2(c) is simultaneously recorded with the FMR spectrum. The origins of this electromotive force are the anisotropic magnetoresistance (AMR)\(^{26-28}\) and anomalous Hall effect which manifest at the resonance field. At the resonance field, the precessing magnetization induces a time varying resistivity of the ferromagnetic layer which combines with the radiofrequency induced currents (along \( x \) in Fig. 1(b)) to produce a DC voltage. The radiofrequency currents are likely produced within the metallic layer by the non-vanishing radiofrequency electric field in the cavity at the sample level. In our set-up geometry, the electromotive force is measured as a voltage along \( y \) (see Fig. 1(b)) and thus in the transverse Hall geometry. Therefore we measure the planar Hall effect (PHE) and the anomalous Hall effect (AHE) in CoFeB. It was proposed and shown by Azevedo et al.\(^{28}\) and Harder et al.\(^{31}\) that the resulting voltage is well described by both a symmetric and an asymmetric contributions. In the CoFeB reference film, the asymmetric component is dominant. The electromotive force can be written as (see Appendix A):

\[
V = V_{\text{offset}} + V_{\text{PHE}} + V_{\text{AHE}}  
= V_{\text{offset}} + V_{sAMR} \frac{\Delta H^2}{(H - H_{\text{res}})^2 + \Delta H^2} + V_{asAMR} \frac{-\Delta H(H - H_{\text{res}})}{(H - H_{\text{res}})^2 + \Delta H^2}  
\]

(6)

where \( V_{sAMR} \) (resp. \( V_{asAMR} \)) is the amplitude of the symmetric (resp. asymmetric) contribution to the electromotive force. We have taken into account a non-resonant offset voltage \( V_{\text{offset}} \), \( H_{\text{res}} \) is the resonance field and \( \Delta H = (\sqrt{3}/2)\Delta H_{pp} \) . \( V_{sAMR}/V_{asAMR} = -1/\tan \psi \) where \( \psi \) is the phase shift between the radiofrequency current and the magnetization.\(^{28,31}\)
The symmetric and asymmetric contributions as well as the offset voltage are proportional to the microwave power. It means that $V_{\text{offset}}$, $V_{\text{SAMR}}$, and $V_{\text{asAMR}}$ are proportional to $h_{rf}^2$, where $h_{rf}$ is the microwave magnetic field strength.

C. Spin pumping and inverse spin Hall effect in germanium

Here we consider the device shown in Fig. 1. Under radiofrequency excitation the magnetization precession of the ferromagnetic layer pumps spins to the non-magnetic germanium layer (N) and the corresponding spin current generates an electric field in Ge due to ISHE: $E_{\text{ISHE}} \propto J_S \times \sigma$. $J_S$ is the spin-current density along $z$ and $\sigma$ its spin polarization vector. This electric field $E_{\text{ISHE}}$ is converted into a voltage $V_{\text{ISHE}}$ between both ends of the Ge channel. In the case of germanium we overcome the conductivity mismatch issue by inserting a thin MgO tunnel barrier (I) between Ge and CoFeB. This additional interface resistance allows for spin accumulation in the germanium conduction band. As a consequence of spin pumping, the damping constant, $\alpha_{FM/I/N}$, is enhanced with respect to the one of the reference sample, $\alpha_{FM/I}$. The real part of the tunnel spin mixing conductance, $g_{t\uparrow\downarrow}$, is given by:

$$g_{t\uparrow\downarrow} = \frac{4\pi M_{\text{eff}} t_F}{g \mu_B} (\alpha_{FM/I/N} - \alpha_{FM/I})$$

(7)

where $t_F$ is the CoFeB thickness. When the static magnetic field is applied parallel to the interface ($\theta_H=90^\circ$), the spin-current density at the interface between CoFeB/MgO and Ge, $j_S^0$, is given by:

$$j_S^0 = \frac{g_{t\uparrow\downarrow}^4 \gamma^2 h h_{rf}^2}{8\pi \alpha^2} \left[ \frac{4\pi M_{\text{eff}} \gamma + \sqrt{(4\pi M_{\text{eff}} \gamma)^2 + 4\omega^2}}{(4\pi M_{\text{eff}} \gamma)^2 + 4\omega^2} \right]$$

(8)

where $h_{rf}$ is the strength of the microwave magnetic field into the resonance cavity. $h_{rf}$ is calculated by measuring the $Q$ factor of the resonance cavity $Q = f/\Delta f$, where $\Delta f$ is the width at half maximum of the frequency distribution when the sample is placed into the cavity. To measure $\Delta f$ we use a second frequencemeter in series with the first one. The voltage $V_{\text{ISHE}}$ due to the inverse spin Hall effect is always symmetric with respect to the resonance field and its amplitude is discussed in Refs.26–28. We then modify the equivalent circuit used in Ref.26 and refined the model used in Ref.20 to account for electron transport.
through the tunnel and Schottky barriers back to the FM (see Appendix B). Then the ISHE voltage in our system is given by:

\[ V_{ISHE} = \frac{w_F}{t\sigma + t_N\sigma_N} \left[ 1 + \frac{t\sigma}{t_N\sigma_N} \frac{2\lambda}{w_F} \tanh\left( \frac{w_F}{2\lambda} \right) \right] \theta_{SHE} l_{sf}^{cb} \tanh\left( \frac{t_N}{2l_{cb}^{ff}} \right) \frac{(2e)}{h} J_s^0 \]  

(9)

where \( w_F \) is the width of the ferromagnetic electrode (150 \( \mu \)m), \( t_N \) (resp. \( t \)) is the Ge (resp. Ta/CoFeB) thickness, \( \sigma_N \) (resp. \( \sigma \)) is the Ge (resp. Ta/CoFeB) conductivity. \( t\sigma = t_F\sigma_F + t_T a\sigma_T a \) where \( t_F \) and \( \sigma_F \) (resp. \( t_T a \) and \( \sigma_T a \)) are the thickness and conductivity of the CoFeB (resp. Ta) layer. \( \lambda \) depends on the resistance-area product \( RA \) of the interface between CoFeB/MgO and Ge as:

\[ \left( \frac{1}{\lambda} \right)^2 = \left( \frac{1}{t\sigma + t_N\sigma_N} \right) \frac{1}{RA} \]  

(10)

In order to estimate the \( V_{ISHE} \) magnitude, the electromotive force and the ferromagnetic spectrum are measured simultaneously. The measured voltage might have one symmetric, \( V_s \), one asymmetric, \( V_{asAMR} \), and one offset contributions. The raw data will be fitted with:

\[ V = V_{offset} + V_s \left( \frac{\Delta H^2}{(H - H_{res})^2 + \Delta H^2} \right) + V_{asAMR} \left( \frac{-\Delta H (H - H_{res})}{(H - H_{res})^2 + \Delta H^2} \right) \]  

(11)

Note that Eq. (11) is similar to Eq. (6) but in the presence of spin pumping the symmetric voltage is: \( V_s = V_{ISHE} + V_{asAMR} \).

IV. RESULTS

A. Reference sample

Fig. 4 shows the OOP dependence of the resonance field, peak-to-peak linewidth and electromotive force on the as-grown and annealed Ta(5nm)/CoFeB(5nm)/MgO(3.3nm)//SiO_2 reference samples. From the angular dependence of \( H_{res} \), we obtain the effective saturation magnetization \( (M_{eff}) \) and the \( g \) factor and from \( \Delta H_{pp} \) we obtain the damping constant \( (\alpha) \) and the angular dispersion \( (\Delta \theta) \). The angular dependence of the peak-to-peak linewidth can be calculated using the following method: after fitting numerically the OOP dependence of the resonance field (Fig. 4(a,d)) we use \( M_{eff} \) and \( g \) to calculate the theoretical dispersion relationship between \( f \) and the external magnetic field for different \( \theta_H \) angles. This is shown
in Fig. 3 where the dotted line corresponds to the frequency at which the measurements are performed. The intercept of each curve with the dotted line gives the value of the resonance field $H_{res}$ along with the equilibrium polar angle $\theta_{M0}$ of $M$ at different $\theta_H$ values. The OOP linewidth angular dependence is shown in Fig. 4(e) and fitted using Eq. (5). In addition, in Fig. 4(c), we have used the $V_{PHE}(\theta_H)$ formula of Appendix A to fit the OOP angular dependence of the symmetric voltage contribution to the electromotive force in the as-grown reference sample. The OOP angular dependence of the symmetric voltage in the Ge-based device ($V_{ISHE}$) clearly shows a different behavior (see Fig. 10(c) and 10(f)).

We have also recorded the power dependence of both the FMR signal and the electromotive force when $H$ is applied parallel to the film plane. The results are shown in Fig. 5. The electromotive force was fitted according to Eq. (6). $V_{offset}$ and $V_{asAMR}$ depend linearly on the applied power in the whole power range whereas $V_{sAMR}$ slightly deviates
from the linear behavior for high powers. Nevertheless we note that $V_{sAMR} \ll V_{asAMR}$ in both reference samples. The sample sizes are $\sim 2 \times 3.5 \text{ mm}^2$ for the as-grown sample and $\sim 2 \times 1.5 \text{ mm}^2$ for the annealed one. For the same RF power of 200 mW and the field applied parallel to the film plane ($\theta_H=90^\circ$), we found $V_{asAMR} \approx 159 \mu V$ for the as-grown sample and $V_{asAMR} \approx 64.2 \mu V$ for the annealed one. Since $V_{asAMR}$ depends linearly on the ferromagnetic electrode width, we can estimate the expected $V_{asAMR}$ value for the CoFeB bar of Fig. 1(a): $V_{asAMR} \approx 6.8 \mu V$ for the as-grown sample and $\approx 6.4 \mu V$ for the annealed one. The expected symmetric contribution to the electromotive force $V_{sAMR}$ will then be almost one order of magnitude less. Furthermore, in the device of Fig. 1 the electrical contacts are no more made on the metallic multilayer, as show in Fig. 2(a), but on Au/Ti ohmic contacts on top of Ge which would reduce the PHE and AHE contributions.

Figure 5: (Color online) Power dependence of the symmetric ($V_{sAMR}$) and asymmetric ($V_{asAMR}$) contributions to the electromotive force according to eq. (6). Samples are multilayers of Ta(5nm)/CoFeB(5nm)/MgO(1.1nm)/SiO2 as grown (squares) and annealed (circles). Solid lines are guides for the eyes and dashed lines in (a) show the non linear behavior of $V_{sAMR}$ in all the experimental frequency range. The insets show the electromotive force measured under different power excitations.

Fig. 6 shows the frequency dependence of the resonance field (a) and peak-to-peak linewidth (b) of the as-grown and annealed reference samples. In both figures, we have used
the $g$ factors deduced from the OOP angular dependence of $H_{res}$ (see Fig. [4](a) and [4](d)) and adjusted the $M_{eff}$ and $\alpha$ values according to eq. (3) and (4) respectively to fit the curves. The frequency independent part of the peak-to-peak linewidth $\Delta H_0$ which is due to inhomogeneities in the magnetic layer is very weak in both samples. We find 1.1 Oe for the as-grown sample and 2.3 Oe for the annealed one which confirms that $\Delta H_0 << \Delta H_G$ at a frequency close to 9 GHz. Moreover the effective perpendicular anisotropy $(4\pi M_{eff})$ increases from 1.175 T up to 1.545 T upon annealing as recently reported. Both samples exhibit very low damping constants comparable to the ones found in Ref.37. Interestingly the peak-to-peak linewidth and damping constants decrease upon annealing in contrast with other results. It means that we have effectively reduced the intrinsic inhomogeneities of the CoFeB electrode by annealing. In particular, the annealing process did not promote chemical inter-diffusion at the interfaces with CoFeB as found in thinner CoFeB films in magnetic tunnel junctions.39

Figure 6: (Color online) FMR dispersion relationship (a) and frequency dependence of the peak-to-peak linewidth (b) for the parallel case. Black dots are for the annealed sample and squares for the as-grown one. Solid red lines are fits according to Eq. (3) in (a), and Eq. (4) in (b).
B. Spin pumping at the ferromagnet/Germanium interface

1. CoFeB/Ge Interface

In this section, we consider the device shown in Fig. 1(a) where the CoFeB electrode has been directly grown on the Ge film without tunnel barrier. The FMR line and the corresponding electromotive force are shown in Fig. 7(a). A clear absorption is observed in the FMR spectrum whereas the electromotive force at the resonance field is negligible. Hence, in the measuring geometry of Fig. 1(a) where the voltage is directly probed on the germanium layer, we do not detect the PHE and the AHE in the CoFeB ferromagnetic layer at the resonance. The angular dependence of the resonance field and peak-to-peak linewidth are displayed in Fig. 7(b). The frequency dependence of $\Delta H_{pp}$ and $H_{res}$ are shown in Fig. 8. First, the effective CoFeB saturation magnetization $M_{eff}$ is lower than in the reference sample: this is probably due to the intermixing between CoFeB and Ge at the interface. In the same way, the larger damping constant $\alpha$ may be due to interface inhomogeneities as a consequence of intermixing and not to spin pumping since no electromotive force is observed. We have then performed the same measurements on the annealed sample. In that case, both the ferromagnetic resonance signal and the electromotive force vanish and the CoFeB film has completely diffused into the Ge layer. These results show that the MgO tunnel barrier is not only necessary to overcome the conductivity mismatch issue but also to prevent the intermixing between CoFeB and Ge at the interface.

2. CoFeB/MgO/Ge Interface

We now consider the same device as in the previous section but with a thin MgO tunnel barrier inserted between CoFeB and Ge as shown in Fig. 1(a). The FMR spectrum and the corresponding electromotive force are shown in Fig. 9(a) for the as-grown sample and Fig. 9(b) for the annealed one. Here a clear electromotive force is detected at the resonance field in both cases. The red line is the fit according to Eq. (11) considering a single symmetric contribution. Moreover by annealing we observe an enhancement of the electromotive force signal. In Fig. 10, the OOP angular dependence of the resonance field, peak-to-peak linewidth and the amplitude of the electromotive force of both samples are displayed. Like in the previous section, the complete analysis of these
Figure 7: (Color online) Results on the device without MgO tunnel barrier and without annealing: (a) FMR line along with the voltage measured in the parallel case. (b) OOP angular dependence of the resonance field and peak-to-peak linewidth with their numerical fits. The inset shows the equilibrium angle of the magnetization as a function of $\theta_H$. Similar devices without MgO barrier and after annealing process do not exhibit ferromagnetic resonance.

The data yields $M_{eff}$, $g$, $\alpha$, and $\Delta H_0$. The solid lines in Fig. 10(c) and Fig. 10(f) are fits according to the formula:

$$V_{ISHE}(\theta_H) \propto \sin(\theta_{M_0}) \left\{ \left[ \frac{(H_{res}/(4\pi M_{eff})) \cos(\theta_{M_0} - \theta_H) - \cos(2\theta_{M_0})}{(2H_{res}/(4\pi M_{eff})) \cos(\theta_{M_0} - \theta_H) - \cos(2\theta_{M_0}) - \cos^2(\theta_{M_0})} \right] \right\}.$$

We also measured the FMR spectrum in the parallel case at different frequencies on both devices (not shown). The frequency dependence of $\Delta H_{pp}$ always shows a linear behavior with a very low $\Delta H_0$ value showing that the Gilbert-type effect is the dominating contribution to the damping in all the samples studied.

In Table I, we can clearly see that the annealing process increases the perpendicular magnetic anisotropy of the system (enhancement of $M_{eff}$) and reduces the intrinsic damping constant. Spin pumping in Ge leads to an increase of the damping constant ($\alpha_{CoFeB/MgO/Ge}$) with respect to that of the reference system ($\alpha_{CoFeB/MgO}$).

The power dependence of the $V_{ISHE}$ amplitude when the external DC magnetic field is applied parallel to the FM layer is shown in Fig. 11 where the solid line is a linear fit. Such
Figure 8: (Color online) FMR dispersion relationship (a) and frequency dependence of the peak-to-peak linewidth (b) in the parallel case. The sample is a Ta(5nm)/CoFeB(5nm)/Ge device without MgO tunnel barrier. The red solid lines are fits according to Eq. (3) in (a), and Eq. (4) in (b).

|              | $\alpha \, (10^{-3})$ | $M_{\text{eff}} \, (\text{emu/cm}^3)$ |
|--------------|------------------------|--------------------------------------|
| CoFeB/MgO ref. n-Ge device | CoFeB/MgO ref. n-Ge device |
| as-grown     | $8.1 \pm 0.08$         | $8.3 \pm 0.06$                       |
| annealed     | $7.2 \pm 0.14$         | $7.5 \pm 0.27$                       |
|              | $935 \pm 20$           | $940 \pm 15$                        |
|              | $1230 \pm 12$          | $1040 \pm 20$                       |

Table I: Damping constant and effective saturation magnetization of the CoFeB/MgO/Ge system and CoFeB/MgO reference sample.

linear behavior accounts well for the $h_{rf}^2$ dependence of the $V_{\text{ISHE}}$ since the microwave power is proportional to the square of the rf magnetic field ($P \propto h_{rf}^2$).

All these results support the fact that the measured electromotive force is due to spin pumping from the CoFeB electrode and inverse spin Hall effect in germanium.
Figure 9: (Color online) FMR spectrum and electromotive force measured simultaneously on the as-grown CoFeB/MgO/Ge sample (a) and the annealed one (b). The magnetic field is applied parallel to the CoFeB bar. There is clearly a voltage peak at the resonance condition and an enhancement of that peak by annealing.

3. Estimation of the spin Hall angle in n-Ge at room temperature

In order to estimate the spin Hall angle $\theta_{\text{SHE}}$ in n-Ge, we have calculated the tunnel spin mixing conductance according to Eq. (7). We found: $g_{\uparrow\downarrow} = 6.1 \times 10^{17} \, m^{-2}$ for the device with the as-grown CoFeB layer and $1.2 \times 10^{18} \, m^{-2}$ for the device with the annealed CoFeB layer. The spin-current density at the interface $j_S^0$, when $\theta_H=90^\circ$, is calculated using Eq. (8) where the CoFeB effective saturation magnetization $M_{\text{eff}}$, the gyromagnetic ratio $\gamma$, and the damping factor $\alpha$ were deduced from FMR measurements. The results are reported in Table II. For a power of 200 mW, the microwave magnetic field $(h_{rf})$ was measured with the sample inside the resonator cavity. The $V_{\text{SHE}}$ amplitude is calculated according to Eq. (9) with the width of the ferromagnetic electrode $w_F=150 \, \mu m$, the FM thickness $t_F = 5 \, nm$, the Ge channel thickness $t_{Ge} = 40 \, nm$ and the conductivities and the resistance-area product of the interface $RA_{CoFeB/MgO/Ge}$ given in Table II. The spin diffusion length in the semiconductor channel is $l_{sf}^{Ge} \approx 1.3 \, \mu m$ (Ref. 20). The conductivities (including the interface
Figure 10: (Color online) Angular out-of-plane dependence of the resonance field (a,d), peak-to-peak linewidth (b,e), and ISHE voltage (c,f) on the as-grown CoFeB/MgO/Ge sample (a-c) and the annealed one (d-f). The numerical fits are in solid lines. The insets in (b,e) show the equilibrium angle of the magnetization as a function of $\theta_H$.

Figure 11: (Color online) Power dependence of the ISHE voltage measured on the Ta/CoFeB/MgO/Ge device with and without annealing. The voltage depends linearly on the excitation power.

$RA$ value) and the spin diffusion length were measured independently on the same device (Ref.20).

We then estimate the spin Hall angle in $n$-Ge from the annealed sample at room temperature: $\theta_{SHE} \approx 0.0011$, which is of the same order of magnitude as in $n$-GaAs (0.007 in Ref.40) and one order of magnitude larger than in $p$-Si (0.0001 in Ref.41). In a similar way
Table II: Measured and calculated parameters on the CoFeB/MgO/Ge sample from spin pumping and inverse spin Hall effect measurements in order to estimate $\theta_{SHE}$ of $n$-Ge according to Eq. (9). The conductivities and $RA$ products were measured separately.

we could estimate the spin Hall angle in $n$-Ge using the data from the as-grown sample and found: $\theta_{SHE} \approx 0.00044$. Such a difference might come either from the error bars and/or from the phenomenological model we have used here. We have measured several annealed devices from the same batch and found $\theta_{SHE}$ between 0.0010 and 0.0012 which gives an estimation of the error bar. We thus conclude that the phenomenological model we use to estimate $\theta_{SHE}$ is not adapted to our system. In particular, this model does not account for the presence of interface states between MgO and Ge. We have shown in a previous work that interface states play a crucial role in the spin injection mechanism. Electrical spin injection into Ge proceeds by two-step tunneling: the electrons tunnel from the FM to the localized interface states (IS) through the MgO barrier and from the IS to the Ge conduction band through the Schottky barrier. Because spin flips occur into interface states, the spin accumulation (hence the spin current) is drastically reduced in the Ge conduction band. By annealing, the density of interface states is reduced and direct spin injection into the Ge conduction band is favored. As a consequence, the spin current in the as-grown sample $j_s^0$ is reduced as compared to the spin current in the annealed sample which leads to the underestimation of $\theta_{SHE}$ as found experimentally. We thus give in the next section a microscopic model accounting for the presence of the tunnel barrier and interface states to accurately describe spin pumping and ISHE in germanium.
V. DISCUSSION

In the as-grown and annealed CoFeB/MgO/Ge samples, we could clearly measure an electromotive force due to ISHE at the ferromagnetic resonance of CoFeB. This photovoltage has a symmetric Lorentzian shape. Furthermore we have shown that all our findings are in good agreement with the observation of ISHE: symmetrical behaviour of $V_{ISHE}$ around the resonance field $H_{res}$, $V_{ISHE}=0$ when the external magnetic field is applied perpendicular to the film ($\theta_H=0$), $V_{ISHE}$ changes its sign when crossing $\theta_H=0$ (Fig. 10), and finally the linear dependence of its amplitude with the microwave power excitation (Fig. 11). This result clearly demonstrates the presence of both spin accumulation and related spin current in the Ge conduction band at room temperature. It was also supported by temperature dependent measurements in a previous work.\textsuperscript{20} In order to confirm that the photovoltage we measure is really due to ISHE and rule out any spurious effects, we carried out complementary measurements. First we studied the photovoltage in millimeter-sized reference samples (both as-grown and annealed) made of CoFeB/MgO/SiO$_2$ with the voltage probes directly connected to the CoFeB film. In that case, we found a dominant asymmetric voltage contribution with respect to the resonance field. It corresponds to the planar Hall effect in the ferromagnet as a combination of anisotropic magnetoresistance and the rf current induced in the ferromagnet by the non-vanishing electric field from the cavity (see Appendix A). This asymmetric voltage contribution due to PHE could not be detected on the device of Fig. 1 with Ge. Moreover the out-of-plane angular dependence of this weak symmetric voltage on the reference sample is different from that of the symmetric voltage we detected in the device of Fig. 1 with Ge. To summarize this study on the reference sample, we can claim that the symmetric photovoltage observed in CoFeB/MgO/Ge samples is due to ISHE and not to PHE in the ferromagnet. We also carried out the same measurements on CoFeB/Ge samples to study the effect of the MgO tunnel barrier. Without MgO tunnel barrier, we never detected a photovoltage in Ge. It first proves that the photovoltage due to the PHE in CoFeB is undetectable in Ge. It also shows that the MgO tunnel barrier is necessary to perform spin injection in Ge by spin pumping. Furthermore, as shown in Fig. 12, we have recorded several voltages on the same CoFeB/MgO/Ge device at the ferromagnetic resonance in order to estimate the tunneling spin Seebeck effect.\textsuperscript{43,44} Indeed at the ferromagnetic resonance, the CoFeB electrode absorbs part of the incident microwave power which
increases its temperature. As a consequence, a vertical temperature difference may appear between the CoFeB electrode and the Ge layer. This temperature gradient may create a tunneling Seebeck voltage and a tunneling spin Seebeck voltage which is only a few percents of the Seebeck voltage. The resulting spin current gives rise to ISHE in germanium just like spin pumping. In order to discriminate between spin pumping and the tunneling spin Seebeck effect, we measured the following voltages at the FMR: $V_{12}$, $V_{1F}$ and $V_{F2}$ shown in Fig. 12. The sum of the tunneling Seebeck and tunneling spin Seebeck voltages ($V_{Sb}$) is given by: $V_{Sb} = V_{F2} - \frac{1}{2}V_{12}$ or $V_{Sb} = V_{1F} - \frac{1}{2}V_{12}$. As shown in Fig. 12, $V_{Sb}$ is negligible (below the noise level) which rules out the presence of tunneling spin Seebeck at the ferromagnetic resonance in our system. Therefore spin injection in Ge proceeds by spin pumping and not by tunneling spin Seebeck effect.

Figure 12: (Color online) FMR espectrum along with the transverse voltage measured to study ISHE ($V_{12}$) and the voltage between the FM layer and one of the ohmic contacts ($V_{F2}$). On the left are shown the contacts geometries. Finally, the expected tunneling spin Seebeck voltage is shown at the bottom right of the figure.

We now address the important issue of the microscopic origin of spin pumping effects in Ge through a MgO tunnel barrier from a theoretical point of view. As demonstrated below, the origin of spin pumping into SCs through a tunnel barrier lies in the evanescent but however non-zero exchange coupling between a band of localized states (LS) and the
ferromagnet through the tunnel barrier, nonetheless sufficiently transparent. Indeed, spin-pumping\textsuperscript{32,46} in metallic tunnel junctions is expected to fall-off in the absence of any exchange field experienced from the ferromagnet (FM) by the delocalized carriers injected in the non-magnetic metal (N). On the other hand, spin injection into a SC by electrical means, as well as by spin-pumping, requires a tunnel barrier at the interface between both types of materials\textsuperscript{13,47} in order to overcome the impedance mismatch issue\textsuperscript{48} describing a total diffusive spin current backflow towards the FM. As shown in our experiments, spin-pumping in a semiconductor with a tunnel barrier can be recovered with some conditions. First, the carriers injected by tunneling from a FM contact have to remain localized at the interface between the tunnel barrier and the semiconductor in the timescale of a single magnetization precession. Second, the effective \textit{tunnel exchange} field experienced by the carriers, that we call hereafter $\tilde{J}$, has to be large enough for the spin to rotate in a timescale of a magnetization precession. These two necessary conditions may be fulfilled within a two-step tunneling picture of spin injection into evanescent (or localized states)\textsuperscript{14} and in the limit of an effective exchange field larger than a certain lower bound. This will be demonstrated below. The third condition to observe spin-pumping in FM/tunnel barrier/SC systems is a minimum value for the conductance of the Schottky barrier delimiting the two regions \textit{i.e} the evanescent states and the SC channel. A thermal activation may be needed to fulfill this third condition. We thus give an analytical expression for the source term taking into account a two-step tunneling process.

Let us consider the standard theory of spin-pumping at the FM/N interface. The source term is known to be equal to\textsuperscript{32,46}

$$I_s^p = \frac{\hbar}{4\pi} \left( \Re g^{\uparrow\downarrow} \mathbf{m} \times \frac{d\mathbf{m}}{dt} + \Im g^{\uparrow\downarrow} \frac{d\mathbf{m}}{dt} \right)$$  \hspace{1cm} (12)$$

in the case of a FM/N ohmic contact where $\mathbf{m}$ is the unit magnetization vector and $g^{\uparrow\downarrow}$ the complex spin mixing conductance. The spins pumped into N then create a \textit{diffusive} spin current backflow to the FM according to the three-dimensional spin-dependent transmission matching at the FM/N interface\textsuperscript{49}

$$I_s^b = \frac{g}{8\pi} \left[ 2p(\mu_0^F - \mu_0^N) + \mu_s^F - \mu_s^N \right] \mathbf{m}$$

$$- \frac{\Re g^{\uparrow\downarrow}}{4\pi} \mathbf{m} \times (\mu_s^N \times \mathbf{m}) + \frac{\Im g^{\uparrow\downarrow}}{4\pi} (\mathbf{m} \times \mu_s^N)$$  \hspace{1cm} (13)$$
where $\mu_0^N, \mu_0^s$ in N and $\mu_0^F, \mu_s^F m$ in FM are respectively the charge and spin accumulations at the interface. $g$ is the sum of spin-up and spin-down conductances and $p$ is the interfacial spin asymmetry coefficient. This backflow of spin current results in a down-renormalization of the spin current pumped in the non magnetic material as shown in a recent couple of papers. The exact form of the corresponding *down renormalization* has to be considered case by case.

The new source term describing spin-pumping in a broad band of evanescent states at the tunnel barrier/SC interface has to involve a small but however non-zero exchange interaction $\tilde{J}_{exc}$ between localized states and the magnetization $M$ (of unit vector $m$) of the FM through the tunnel barrier; this exchange interaction couples evanescent wavefunctions inside the barrier. In the following, we will define $\tilde{J}_{exc}$ in the form: $\tilde{J}_{exc} = J_0 \exp -2\kappa d \approx J_0 T$ where $J_0$ is the bare *on-site* exchange interaction of the order of the exchange interaction in FM or even larger (about 1 eV), $\kappa$ is the imaginary electronic wavevector in the barrier and $d$ the barrier thickness. $T$ is the tunnel transmission coefficient. We note $\Gamma = \hbar/\tau_n$ the mean energy broadening of the localized states due to the finite carrier lifetime ($\tau_n$) through their escape towards the ferromagnetic reservoir FM. This energy broadening can be expressed vs. the localization energy $\epsilon_n$ within the centers and $T$ according to $\Gamma \approx \epsilon_n T$ (Ref.$^{52}$). Note that the escape towards the semiconductor channel, moderately doped, is generally prohibited in an energy band located downward the Fermi energy.

If one defines three different components of the carrier spin vector $s$ injected in the evanescent states by $s_z$, $s_+ = s_x m_x + s_y m_y$ and $s_- = s_y m_x - s_x m_y$, the equation of motion for the injected spin, along the $x$ direction at time $t = 0$, in a localized state in the exchange field of the magnetization $m$ rotating in the $(x,y)$ plane follows the Heinsenberg evolution for the spin-operator $(i\hbar)ds/dt = [s, \tilde{J}_{exc} s]_-$; thus giving in fine:

\[
\begin{align*}
    s_+ & = s_x m_x + s_y m_y = \frac{\omega_{exc}^2}{\omega_{eff}^2} + \frac{\omega_{rf}^2}{\omega_{eff}^2} \cos(\omega_{eff} t) \\
    s_- & = s_y m_x - s_x m_y = \frac{\omega_{rf}}{\omega_{eff}} \sin(\omega_{eff} t) \\
    s_z & = \frac{\omega_{exc} \omega_{rf}}{\omega_{eff}^2} [1 - \cos(\omega_{eff} t)]
\end{align*}
\]

with $\omega_{rf}$ the RF pulsation frequency, $\omega_{exc} = \frac{J_{exc}}{\hbar}$ the *exchange* pulsation and $\omega_{eff} = \sqrt{\omega_{rf}^2 + \omega_{exc}^2}$ the effective pulsation of the spin during its rotation.
In an homogeneous FM layer, the precession frequency due to the exchange interaction \( \omega_{\text{exc}} \approx 10^{15} \text{ rad.s}^{-1} \) is very large compared to the RF frequency. This results in a small average component of the spin vector pumped along \( z \): \( s_z = \frac{\omega_{\text{rf}}}{\omega_{\text{exc}}} \), of the order of \( 10^{-5} \). However, this small spin rotation is counterbalanced by a large number of uncompensated spins due to the strong exchange and whose number equals \( N_{\text{DOS}}J_{\text{exc}} \) (\( N_{\text{DOS}} \) is the density of states). The total spin along the \( z \) direction then writes \( S_z \approx N_{\text{DOS}}J_{\text{exc}}s_z = N_{\text{DOS}}\hbar \omega_{\text{rf}} \).

One recovers the standard formula for the spin-current pumped at the ohmic FM/N interface if the interfacial spin-mixing conductance \( g_{\uparrow\downarrow} \) is introduced hereafter. In the case of spin-pumping into evanescent states, the exchange pulsation \( \omega_{\text{exc}} \) can be of the order of magnitude of the RF pulsation or even smaller. To derive the average \( s_z \) component pumped in a localized center, one has to perform a time average of \( s_z \) on the carrier lifetime \( \tau_n \) to give:

\[
s_z = \frac{\omega_{\text{exc}}\omega_{\text{rf}}}{\omega_{\text{exc}}^2 + \omega_{\text{rf}}^2} \left[ 1 - \frac{\sin(\omega_{\text{eff}}\tau_n)}{\omega_{\text{eff}}\tau_n} \right]
\]

(17)

By analogy with the previous calculations relative to the bulk FM layer, and taking into account that the total number of uncompensated spins introduced by the tunneling exchange interactions, \( N_{\text{DOS}}\tilde{J}_{\text{exc}} \), one can generalize the total spin accumulation \( (\Delta \mu_z) \) pumped along the \( z \) direction as:

\[
\Delta \mu_z = \frac{(J_0T)^2}{(J_0T)^2 + (\hbar \omega_{\text{rf}})^2} \left[ 1 - \text{sinc}(\sqrt{\frac{(\hbar \omega_0)^2 + (J_0T)^2}{\epsilon_nT}}) \right] \times \left[ \hbar \mathbf{m} \times \frac{d\mathbf{m}}{dt} \right]
\]

(18)

for any rotation \( d\mathbf{m}/dt \) vector. It comes two important conditions on the effective exchange \( \tilde{J}_{\text{exc}} \) to generate significant spin-pumping at the FM/tunnel barrier/SC interface:

1) \( \tilde{J} \) must be larger than the intrinsic energy broadening \( \Gamma \) (or equivalently \( J_0 > \epsilon_n \)) corresponding to a time of interaction larger than the time of the spin precession.

2) \( \tilde{J} \) must be larger than the RF frequency energy \( \hbar \omega_{\text{rf}} \) of the order of \( \approx 40 \mu\text{eV} \) in the present case. This condition corresponds to a characteristic spin precession time due to exchange, and necessary for any spin rotation, smaller than the magnetization \( \mathbf{m} \) precession time itself.

Once these two conditions are satisfied, the spin-pumping effect at the FM/tunnel barrier/SC interface becomes efficient, a large rotation angle of the spin \( s_z = \frac{\omega_{\text{exc}}\omega_{\text{rf}}}{\omega_{\text{exc}}^2 + \omega_{\text{rf}}^2} \) compensating the small number of uncompensated spins \( N_{\text{DOS}}\tilde{J}_{\text{exc}} \).
The total spin-current pumped \((I^p_s)\) at the LS/SC channel interface, that is the source term, equals \(I^p_s = G_{sh}\Delta \mu_z\) where \(G_{sh}\) is the Schottky conductance playing the role of the mixing conductance \(g^{\uparrow \downarrow}\) for FM/N interfaces. We now proceed to the down-renormalization of the spin-current pumped in the SC as described previously. In the light of the recent published works\(^{50,51}\) this total spin-current has then to be decomposed into the real spin-current injected in the Ge channel added to a backflow of spin-current relaxing either into the localized states or into the FM reservoir by back-absorption. We have:

\[
P_s^p = G_{sh}\Delta \mu_z = G_{Ge}\Delta \mu_{Ge} + \frac{\Delta \mu_{LS}}{R_{LS}}
\]

\[
G_{sh}(\Delta \mu_{Ge} - \Delta \mu_{LS}) = \frac{\Delta \mu_{LS}}{R_{LS}}
\]

where \(G_{Ge} = \tanh(t_N/l_{sf}^{Ge})/R_{s}^{Ge}\) is the spin conductance of the Ge layer of thickness \(t_N\) and spin diffusion length \(l_{sf}^{Ge}\) (Ref. \(^{51}\)) and where \(R_{s}^{Ge} = \rho_{Ge} \times l_{sf}^{Ge}\) is the corresponding bulk spin resistance. \(\Delta \mu_{Ge}\) (resp. \(\Delta \mu_{LS}\)) is the spin accumulation generated in the Ge layer (resp. in the LS) and \(R_{LS} = \tau_{sF}^{LS}/(e^2N^{2D}DOS)\) is the spin resistance of the LS (\(\tau_{sF}^{LS}\) is the corresponding spin lifetime). Eq. (20) describes the continuity of the spin-current backflow through the Schottky barrier. It results from these calculations that the effective spin-current \(I^{Ge}_s\) injected in the Ge channel writes:

\[
I^{Ge}_s = \frac{1 + R_{LS}G_{sh}}{1 + G_{Ge}^{-1} + R_{LS}^{-1}} \Delta \mu_z
\]

with \(\Delta \mu_z\) the spin accumulation generated in the localized states by spin-pumping like calculated previously. A zero Schottky conductance leads to zero spin-current. On the opposite case of a large Schottky conductance e. g. on increasing the temperature, the spin current pumped in the Ge channel writes \(\frac{R_{LS}G_{sh}}{1 + R_{LS}G_{Ge}} \times G_{sh}\Delta \mu_z\) i.e. it corresponds to the maximum spin-current pumped weighted by the ratio of spin-flips occurring in the channel itself over the total number of spin-flips also possible in the band of LS and parameterized by \(1/R_{LS}\). Consequently, the real spin current pumped into the Ge layer depends on the Schottky conductance and on the different spin-resistances involved in the spin-relaxation process. The main question that has to be addressed in the future is the fraction of the spin-current pumped and relaxing in the LS by spin-flip. Indeed, this part of the spin-current would contribute to the broadening of the FMR spectra but not to the ISHE voltage. Finally
these parameters have to be determined experimentally in order to relate this microscopic model to our data.

VI. CONCLUSION

We have demonstrated that at the FM/I/N interface where N is a non-magnetic semiconductor channel we could inject a spin current by spin pumping from the FM layer into the N channel at the ferromagnetic resonance. We have also shown that the MgO tunnel barrier is useful not only to overcome the conductivity mismatch between CoFeB and Ge but also to keep the magnetic properties of the FM after annealing the samples. There is no spin pumping nor inverse spin Hall effect voltage signal on devices without barrier while it clearly appears on the devices with the MgO barrier. Moreover there is an enhancement of the ISHE signal and consequently of the spin Hall angle when the device is annealed. A microscopic model involving interface states and evanescent tunnel exchange coupling has been developed in order to explain spin pumping into Ge from a FM electrode through a tunnel barrier. We could finally find and discuss the spin Hall angle in $n$-Ge: $\sim 0.0011$ from the annealed sample and $\sim 0.00044$ from the as-grown one.

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Appendix A: Angular dependence of the planar Hall effect and anomalous Hall effect

The sample is rotated in the electromagnet with the static magnetic field $\vec{H}_{DC}$ applied perpendicular ($\theta_H=0^\circ$) to parallel to the film plane ($\theta_H=90^\circ$). We define the sample magnetization: $\vec{M} = \vec{M}_s + \vec{m}$ and the current in the ferromagnetic electrode: $\vec{J} = \vec{J}_0 + \vec{j}$ where $\vec{M}_s$ is the equilibrium magnetization in static conditions. It makes an angle $\theta_M$ with the normal to the sample $\hat{z}$. $\vec{m}(t)$ is the time-varying part of the magnetization. We do not apply any bias current to the system: $\vec{J}_0 = 0$ and consider the RF current created in the ferromagnetic...
layer by the non-vanishing RF electric field: $\vec{j} = j_0 \cos(\omega t + \psi) \hat{x}'$ where $\psi$ is the constant phase difference between the current and the magnetization precession at resonance. The generalized Ohm's law then writes:

$$\vec{E} = \rho \vec{j} + \frac{\Delta \rho}{M^2} (\vec{j} \cdot \vec{M}) \vec{M} - R_H \vec{j} \times \vec{M}$$

(A1)

$\rho$, $\Delta \rho$ and $R_H$ are the resistivity, the anisotropic magnetoresistance and the anomalous Hall constant of the ferromagnetic electrode. Since we experimentally measure a DC voltage, we calculate the time average of $\vec{E}$:

$$\langle \vec{E} \rangle = \frac{\Delta \rho}{M_s^2} \left[ \left\langle (\vec{j} \cdot \vec{m}) \vec{M}_s \right\rangle + \left\langle (\vec{j} \cdot \vec{M}_s) \vec{m} \right\rangle \right] - R_H \left\langle \vec{j} \times \vec{m} \right\rangle$$

(A2)

where: $\vec{M}_s = M_s \sin(\theta_M - \theta_H) \hat{x} + M_s \cos(\theta_M - \theta_H) \hat{z}$, $\vec{m} = m_x \cos(\theta_M - \theta_H) \hat{x} + m_y \hat{y} - m \sin(\theta_M - \theta_H) \hat{z}$ and $\vec{j} = j \cos \theta_H \hat{x} + j \sin \theta_H \hat{z}$. We then obtain the voltage:
\[ V = \int_{-w_F/2}^{w_F/2} \langle E_y \rangle \, dy = \frac{w_F \Delta \rho}{M_s} \langle jm_y \rangle \sin \theta_M - \frac{w_F r_H}{M_s} \langle jm_\theta \rangle \sin \theta_M \] 

(A3)

where \( r_H = M_s R_H \). The first term corresponds to the planar Hall effect and the second one to the anomalous Hall effect. \( m_\theta \) and \( m_y \) are then determined by solving the Landau-Lifschitz-Gilbert (LLG) equation:

\[ \frac{d\vec{M}}{dt} = -\gamma (\vec{M} \times \vec{H}_{eff}) + \frac{\alpha}{M_s} \vec{M} \times \frac{d\vec{M}}{dt} \] 

(A4)

where \( \gamma \) is the gyromagnetic ratio, \( \alpha \) the damping factor and \( \vec{H}_{eff} \) the effective magnetic field given by:

\[ \vec{H}_{eff} = H_{DC} \hat{z} + h_{rf} \hat{y} - 4\pi (\vec{M} \cdot \hat{z}') \hat{z}' \] 

(A5)

Here we only consider the shape anisotropy field. The radiofrequency magnetic field can be written as: \( h_{rf} = h_{rf0} \cos(\omega t) \hat{y} \), \( \omega = 2\pi f \) where \( f=9.4 \text{ GHz} \) is the X-band cavity frequency. In static conditions, the magnetization equilibrium angle \( \theta_M \) is found by solving:

\[ \vec{M} \times \vec{H}_{eff} = \vec{0} \] i.e. \( 4\pi M_s \sin(2\theta_M) - 2H_{DC} \sin(\theta_M - \theta_H) = 0 \). The resolution of the LLG equation yields:

\[ m_\theta \cos(\theta_M - \theta_H) = -\gamma A m_y + \gamma M_s h_{rf} \cos(\theta_M - \theta_H) - \alpha m_y \cos(\theta_M - \theta_H) \] 

(A6)

\[ m_y = -\gamma B m_\theta + \alpha m_\theta \] 

(A7)

\[ -\dot{m}_\theta \sin(\theta_M - \theta_H) = -\gamma M_s h_{rf} \sin(\theta_M - \theta_H) + \gamma C m_y + \alpha m_y \sin(\theta_M - \theta_H) \] 

(A8)

where: \( A = H_{DC} - 4\pi M_s \cos \theta_H \cos \theta_M \), \( B = 4\pi M_s \cos(2\theta_M) - H_{DC} \cos(\theta_M - \theta_H) \) and \( C = 4\pi M_s \sin \theta_H \cos \theta_M \). By using: \( m_\theta = \text{Re}(m_\theta e^{i\omega t}) \), \( m_y = \text{Re}(m_y e^{i\omega t}) \) and \( h_{rf} = \text{Re}(h_{rf0} e^{i\omega t}) \), we finally find:

\[ m_\theta = \frac{M_s h_{rf} \cos(\theta_M - \theta_H)}{a^2 + b^2} [a \cos(\omega t) + b \sin(\omega t)] \] 

(A9)

\[ m_y = \frac{M_s h_{rf} \cos(\theta_M - \theta_H)}{a^2 + b^2} [(\alpha a + \frac{\gamma}{\omega} B b) \cos(\omega t) - (\frac{\gamma}{\omega} B a - \alpha b) \sin(\omega t)] \] 

(A10)

where: \( a = \alpha [A - B \cos(\theta_M - \theta_H)] \) and \( b = (\gamma/\omega) [AB + (\omega/\gamma)^2 (1 + \alpha^2) \cos(\theta_M - \theta_H)] \). The FMR spectrum is defined by \( m_y \). Then, after time averaging, we obtain:
Here we point out that the rf electric field $e_{\text{rf}}$ induces an additional angular dependence because $j_0$ change with the DC magnetic field angle as $\sin(\theta_H + \theta_E)$ where $\theta_E$ is the direction of the rf electric field in the cavity (see Fig. 13). Note that $j_0$ is proportional to the strength of the rf electric field, i.e. to the strength of the rf magnetic field. As a consequence the magnitude of the electromotive forces $V_{\text{PHE}}$ and $V_{\text{AHE}}$ are proportional to $h_{\text{rf}}^2$. Hence they exhibit a linear dependence with the microwave power as shown in the main text. Such linear dependence might allow to deduce either the ratio $\Delta \rho/r_H$ or the phase shift $\psi$. Since the Hall coefficient is of the order of $10^{-12} \ \Omega \text{cm}/G$ and $M_s$ is of the order of $10^3 \ \text{G}$, then $r_H = M_s R_H$ is much smaller than the anisotropic magnetoresistance, $\Delta \rho$.

The OOP angular dependence of the symmetric component of either $V_{\text{PHE}}$ or $V_{\text{AHE}}$ clearly shows a behavior different from that of the ISHE out-of-plane angular dependence ($V_{\text{ISHE}} \propto \sin \theta_M$) as shown in Fig. 4(c) where we considered $\theta_E = -30^\circ$ and $\psi = 15^\circ$.

Appendix B: Charge backflow into the ferromagnet by the ISHE in Ge

\begin{align*}
V_{\text{PHE}} &= \frac{w_F \Delta \rho j_0 h_{\text{rf}} \cos(\theta_M - \theta_H) \sin \theta_M}{2(a^2 + b^2)} \\
&\times \left[ (aa + \frac{\gamma}{\omega} Bb) \cos \psi + (\frac{\gamma}{\omega} Ba - ab) \sin \psi \right] \quad (A11) \\
V_{\text{AHE}} &= \frac{w_F r_H j_0 h_{\text{rf}} \cos(\theta_M - \theta_H) \sin \theta_M}{2(a^2 + b^2)} \times [b \sin \psi - a \cos \psi] \quad (A12)
\end{align*}
At the ferromagnetic resonance of the CoFeB electrode, the combination of spin pumping and ISHE creates a charge current (in A/m) $I_{ISHE}$ in the Ge layer. Part of this charge current flows back to the ferromagnetic and tantalum capping layers which affects the estimation of $V_{ISHE}$ and $\theta_{SHE}$ in germanium. In the following, we make an estimation of this backflow current. In Fig. 14, the current density crossing the interface at $x$ corresponds to the variation of the current in the layers:

$$
t\delta j(x) = \frac{V_N - V}{RA} \delta x \quad (B1)
$$

$$
t_N \delta j_N(x) = \frac{V - V_N}{RA} \delta x \quad (B2)
$$

where $t$, $j$ and $V$ are the thickness, current density and potential in the Ta/CoFeB bilayer; $RA$ is the resistance-area product of the interface between CoFeB/MgO and Ge. The current densities in each layer with conductivities $\sigma$ and $\sigma_N$ can be written:

$$
\begin{align*}
  j(x) &= -\sigma \partial_x V(x) \quad (B3) \\
  j_N(x) &= -\sigma_N \partial_x V_N(x) \quad (B4)
\end{align*}
$$

the current conservation involving the current source due to spin pumping and ISHE gives:

$$
tj(x) + t_Nj_N(x) = -I_{ISHE} \quad (B5)
$$

which can also be written:

$$
t\sigma \partial_x V(x) + t_N \sigma_N \partial_x V_N(x) = I_{ISHE} \quad (B6)
$$

by using the symmetry of the system, we set the origin of $x$ in the middle of the trilayer and find:

$$
t\sigma V(x) + t_N \sigma_N V_N(x) = I_{ISHE}x \quad (B7)
$$

Using Eq. $B1$ and $B3$ we can write:

$$
\partial_x^2 (V(x) - V_N(x)) = \left(\frac{1}{t\sigma} + \frac{1}{t_N\sigma_N}\right) \frac{V(x) - V_N(x)}{RA} \quad (B8)
$$
which gives the following solution:

\[ V(x) - V_N(x) = asinh\left(\frac{x}{\lambda}\right) \]  

(B9)

with:

\[ \left(\frac{1}{\lambda}\right)^2 = \left(\frac{1}{t\sigma} + \frac{1}{t_N\sigma_N}\right) \frac{1}{RA} \]  

(B10)

combining Eq. (B7) and (B9) yields the potentials:

\[ V(x) = \frac{I_{ISHE}}{t\sigma + t_N\sigma_N} x - asinh\left(\frac{x}{\lambda}\right) \frac{t_N\sigma_N}{t\sigma + t_N\sigma_N} \]  

(B11)

\[ V_N(x) = \frac{I_{ISHE}}{t\sigma + t_N\sigma_N} x + asinh\left(\frac{x}{\lambda}\right) \frac{t\sigma}{t\sigma + t_N\sigma_N} \]  

(B12)

The current in the Ta/CoFeB bilayer (proportional to the derivative of \( V \)) vanishes at the edges \((x = \pm w_F/2)\) which gives access to the constant \( a \):

\[ a = \frac{\lambda I_{ISHE}}{t_N\sigma_N\cosh((w_F/2)/\lambda)} \]  

(B13)

Then the ratio between the induced voltage \( U \) in Ge and the current \( I_{ISHE} \) is given by:

\[ \frac{U}{I_{ISHE}} = \frac{2V_N(w_F/2)}{I_{ISHE}} = \frac{w_F}{t\sigma + t_N\sigma_N} \left[ 1 + \frac{t\sigma}{t_N\sigma_N} \frac{2\lambda}{w_F} \tanh\left(\frac{w_F}{2\lambda}\right) \right] \]  

(B14)

where: \( t\sigma = t_F\sigma_F + t_{Ta}\sigma_{Ta} \). \( t_{Ta} \) (resp. \( \sigma_{Ta} \)) is the thickness (resp. conductivity) of the tantalum capping layer.

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