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Photon energy dependence of photo-induced inverse spin-Hall effect in Pt/GaAs and Pt/Ge

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We report the photon energy dependence of photo-induced inverse spin Hall effect (ISHE) in Pt/GaAs and Pt/Ge Schottky junctions. The experimental results are compared with a spin drift-diffusion model, which highlights the role played by the different spin lifetime in the two semiconductors, in determining the energy dependence of the ISHE signal detected in the Pt layer. The good qualitative agreement between experiments and modelling indicates that photo-induced ISHE can be used as a tool to characterize spin lifetime in semiconductors. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4922290]

The capability of generating and detecting pure spin currents is essential in spintronics.[1] In semiconductors, it is possible to exploit *optical spin orientation*[2] to generate a spin-unbalanced population of electrons in the conduction band, with a net spin polarization defined as $\mathbf{P} = (n_\uparrow - n_\downarrow)/(n_\uparrow + n_\downarrow)\mathbf{u_k}$, being $n_\uparrow$ ($n_\downarrow$) the up- (down-) spin densities with respect to the quantization axis, given by the direction of the incident light $\mathbf{u_k}$. In bulk GaAs and Ge, P reaches the maximum value of 0.50 for a photo-excitation energy $h\nu$ resonant with the direct bandgap E_d.[3] For $h\nu > E_d + \Delta_0$, being Δ_0 the spin-orbit splitting, P sharply decreases in both semiconductors.[4,5]

Optically oriented spins can be detected using spin-resolved photoemission,[3] which requires the establishment of a *negative electron affinity* condition at the semiconductor surface, in order to allow the photoemission of electrons excited for $h\nu \approx E_d$. While negative electron affinity at the Γ point is attainable in GaAs,[3] this is not the case for Ge and Ge-based heterostructures.[6,7] Photoluminescence (PL) can also be used to detect spin orientation and relaxation in semiconductors.[8] Yet, in continuous wave-PL, spin lifetimes shorter than the carrier lifetimes cannot be measured, an issue that can hinder application of PL at room temperature in Ge.[9]

An alternative route for the detection of optically-oriented spins relies on the inverse spin Hall effect[10] (ISHE). By means of the ISHE, a spin current J_s is converted into a charge current J_c with an efficiency given by the spin-Hall angle $\gamma = J_c/J_s$, thus allowing for the (indirect) electrical detection of pure spin currents. Spin-to-charge current conversion can take place within the semiconductor[11] or inside a high atomic-number metal layer, such as Pt, deposited on the semiconductor surface.[12] Since γ in Pt is much higher than in doped GaAs[13] and Ge, Pt can be used as a non-magnetic electrode, sensitive to pure spin-currents. Moreover, the optical-injection/electrical-detection scheme relying on the ISHE favours the design of devices merging spintronics and photonics as recently demonstrated by the fabrication of a spin photo-voltage generator.[14]

Photo-induced ISHE has been first observed by Ando *et al.*[12,15] in Pt/GaAs for an excitation energy of $h\nu = 1.85$ eV and by Bottegoni *et al.* in Pt/Ge in the 1–1.8 eV energy range.[16] The temperature dependence of the ISHE signal has also been investigated in the direct gap system Au/InP in Ref. 17 for $h\nu = 2.33$ eV. The photon-energy dependence of the ISHE signal in Pt/GaAs has been modelled by Khamari *et al.* in Ref. 18, but no experimental investigation has been carried out so far.

In this paper, we report the photon-energy dependence of the ISHE signal measured at room temperature, for the prototypical cases of a direct (GaAs) and an indirect (Ge) gap semiconductors. Spin detection by the ISHE in both semiconductors has been interpreted by means of a spin drift-diffusion model which qualitatively reproduces the experimental data.

The two analyzed samples are a Pt/GaAs and a Pt/Ge junction. The substrates consist, respectively, of a 350 μm-thick Si-doped GaAs (donor concentration $N_d \approx 2 \times 10^{18}$ cm^{-3}) and 450 μm-thick As-doped Ge ($N_d \approx 1.7 \times 10^{16}$ cm^{-3}). Schottky junctions were formed by depositing 4 nm-thick, 5×5 mm^2 wide Pt layers using e-beam evaporation.

The experimental geometry is shown in Fig. 1(a). Spin polarized electrons are excited by using a collimated monochromatic beam from a Ti:sapphire tunable laser, which provides photons in the spectral range from 1.2 to 1.8 eV. Continuous-wave fiber pigtailed Si and InAs-quantum dot lasers were used for measurements at 0.8 and 1 eV, respectively. The laser spot size was large enough to completely illuminate the Pt pad. A multiaxial stage allows the rotation around the polar angle θ and the azimuthal angle φ defined in Fig. 1(a).

Under excitation with circularly polarized light, spin-oriented carriers are generated around the Γ point of the Brillouin zone and subsequently diffuse into the Pt layer where the conversion of spin current into charge current generates an electromotive field $\mathbf{E}_{ISHE} = \frac{\gamma}{\sigma_c}\mathbf{J}_s \times \mathbf{u_k}$, being σ_c the electrical conductivity of the Pt layer. Under open circuit conditions, a voltage $\Delta V_{ISHE} = E_{ISHE} d$ appears between two 50 nm-thick Au electrodes evaporated at the edges of the Pt pad and separated by a distance $d \approx 4$ mm. Light helicity is modulated by means of a photo-elastic modulator and

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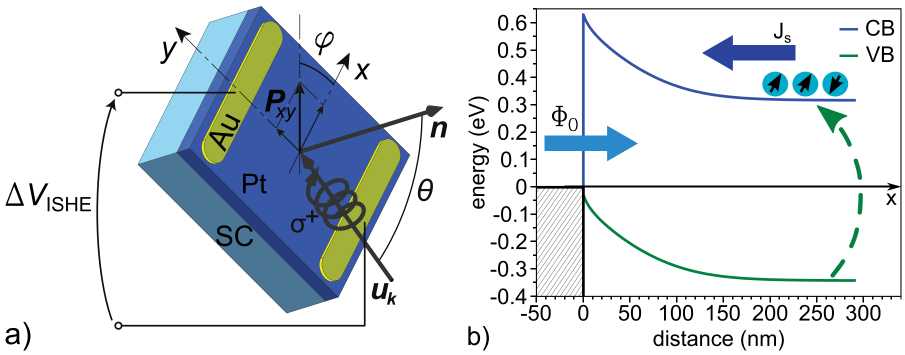

FIG. 1. (a) Sketch of the experimental configuration. θ is the angle between the direction of the incident photons $\mathbf{u_k}$ and the normal to the sample surface, while φ is the angle between the projection of $\mathbf{u_k}$ in the xy-plane and the x-axis. (b) Schematic representation of the spin current J_s induced by a photon flux Φ_0 at the Pt/Ge Schottky junction.

ΔV_{ISHE} is acquired through a lock-in amplifier. Under open circuit conditions, an equal number of electrons and holes must be injected in the Pt film, however, since the electron spin lifetime in both bulk GaAs[19] and Ge[20] exceeds by orders of magnitude the hole spin lifetime,[19,21] we assume the ISHE signal to be associated solely to electron transport. All the measurements have been performed at room temperature. The vectorial product in the expression of E_{ISHE} implies that the ISHE signal is maximized for $\varphi = 0$; however, the expected $\cos(\varphi)$ dependence of ΔV_{ISHE} was routinely checked to confirm the ISHE origin of the measured photovoltage, while $\theta = 65°$ was used to maximize[15] the in-plane component of $\mathbf{P}$.

Figure 2(a) shows the photon energy dependence of the Pt/GaAs ISHE in the 1.5–1.9 eV energy range. The excitation power I_{inc} varied from 20 to 8 mW for the investigated energy range. Therefore, the data-points of Fig. 2(a) have all been rescaled to an incident photon rate $\Phi_0 = 5\times10^{17}\,\mathrm{s^{-1}\,cm^{-2}}$. The linear dependence of the ΔV_{ISHE} on I_{inc} is confirmed by experimental data not presented here. The spin current density J_s injected from the semiconductor into the Pt layer can be estimated from ΔV_{ISHE} by means of the expression

$$J_s = \frac{t}{d}\frac{\sigma_c}{\gamma}\left(\int_0^t \frac{\sinh\left[(t-z)/\lambda\right]}{\sinh(t/\lambda)}\,dz\right)^{-1}\Delta V_{\mathrm{ISHE}}, \qquad (1)$$

where t and λ are the Pt layer thickness and the spin diffusion length in Pt, respectively. Equation (1) has been first proposed by Ando *et al.* in Ref. 12 and relies on a diffusion model to treat the spin relaxation in Pt. The same approach has also been used in our previous work on Pt/Ge[16] and by Khamari *et al.* in Ref. 17 with the modifications required by the fact that the ISHE current and not the ISHE voltage was measured in this case. Yet, in all cases, Eq. (1) overestimates J_s. Indeed, if we define the spin quantum efficiency as $SQE = J_s/q\Phi_0$ being q the elementary charge, a SQE exceeding one by more than one order of magnitude is obtained in Refs. 12 and 17. The same issue arises when Eq. (1) is used to extract J_s from the data reported in Fig. 2. Such an inconsistency might be due to several reasons: the application of diffusion equations to model spin relaxation in layers thinner than the diffusion length, the large scattering of values reported for γ in the literature, or the use of the bulk value of σ_c to treat extremely thin films. All these are still open issues and require further investigation in order to attain a quantitative relationship between J_s and ΔV_{ISHE}. It is worth noticing that a similar inconsistency has been recently reported also for the ISHE obtained by spin-pumping in Pt[22] and for spin injection through magnetic semiconductors tunnel contacts.[23] Having clarified this point, we can still make a relative comparison between the J_s value of $1.2\times10^4\,\mathrm{A/m^2}$ we have obtained from Eq. (1) for $h\nu = 1.82\,\mathrm{eV}$, $I_{\mathrm{inc}} = 10$ mW, and $\theta = 65°$ by using the same values of σ_c, γ, and λ employed in Ref. 12, where $J_s = 2.1\times10^4\,\mathrm{A/m^2}$ is estimated for $h\nu = 1.85\,\mathrm{eV}$ and identical values of θ and I_{inc}.[24]

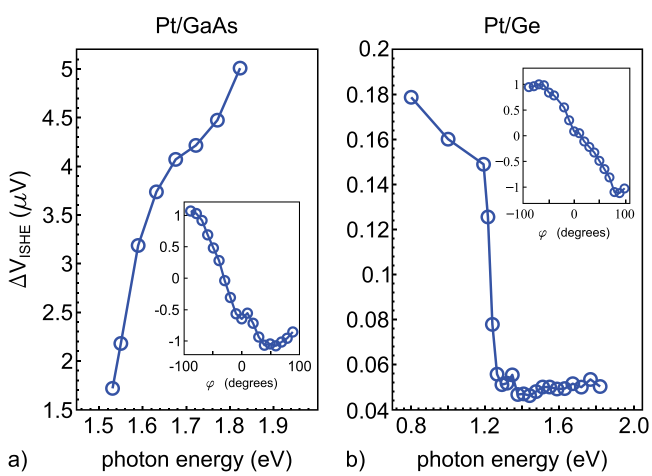

FIG. 2. (a) Photon energy dependence of ΔV_{ISHE}, rescaled to an incident photon flux $5\times10^{17}\,\mathrm{s^{-1}\,cm^{-2}}$ for the Pt/GaAs junction. The inset shows the relative variation of ΔV_{ISHE} as a function of φ for at $\theta = 65°$ and $h\nu = 1.77\,\mathrm{eV}$. (b) Energy spectrum of ΔV_{ISHE} obtained under similar experimental condition as for (a) in the case of Pt/Ge.

Fig. 2(b) shows ΔV_{ISHE}, normalized to $5\times10^{17}\,\mathrm{s^{-1}\,cm^{-2}}$, measured in Pt/Ge using the same procedure described for the case of Pt/GaAs. We notice that the Pt/GaAs ISHE signal is 10 to 50 times higher than the Pt/Ge signal. Moreover, the ΔV_{ISHE} dependence on $h\nu$ is quite different for the two semiconductors. For the energy range accessible with our experimental set up, which covers the spectral region from the bandgap ($E_{\mathrm{GaAs}} = 1.43\,\mathrm{eV}$) to energies slightly above the threshold for split-off state excitation ($E_{\mathrm{GaAs}} + \Delta_{\mathrm{GaAs}} = 1.77\,\mathrm{eV}$), ΔV_{ISHE} monotonically increases in the case of Pt/GaAs, in qualitative agreement with the modelling of Ref. 18. An opposite trend is observed between $E_{\mathrm{Ge}} = 0.8\,\mathrm{eV}$ and $E_{\mathrm{Ge}} + \Delta_{\mathrm{Ge}} = 1.1\,\mathrm{eV}$ for Pt/Ge. In this case, excitation energies well above the split-off threshold could also be achieved and a sharp decrease of ΔV_{ISHE} is observed for $h\nu > E_{\mathrm{Ge}} + \Delta_{\mathrm{Ge}}$. In the attempt to explain the different behaviours of the two semiconductors, we have numerically solved the spin drift-diffusion equation at a metal/semiconductor (M/S) Schottky junction.

The drift-diffusion of optically-oriented spins in a $p-n$ junction has been treated in Ref. 25: here we have applied a similar approach to the Schottky barrier case. Spin (and charge) drift-diffusion equations can be solved analytically only in a few limited cases,[26] therefore we relied on the Nextnano software[27] to numerically solve the coupled drift-diffusion-Poisson equations for charge current. In the case of Ge, recombination-generation (R-G) has been modelled by using the Shockley-Read-Hall theory, while a bimolecular R-G term has been employed for GaAs. Electron-hole pair photo-generation was described by the Lambert-Beer term $G = \Phi_0 \alpha \exp(-\alpha x)$, where α is the absorption coefficient and x is the coordinate axis used in our 1D simulation as shown in Fig. 1(b). The integration domain was 40 μm long and bounded by a Schottky contact at $x = 0$ and by an Ohmic contact at $x = 40\,\mu$m. For a given value of $h\nu$, the electric field $E(x)$ and the R-G rate have been calculated by assuming open-circuit operation, thus matching the zero-current condition of the ISHE measurements. In the hypothesis that only electrons are polarized, spin transport can be described by two sets of equations[25]

$$J_{\uparrow(\downarrow)} = -D_{\rm n}\frac{\partial n_{\uparrow(\downarrow)}}{\partial x} - \mu_{\rm n} n_{\uparrow(\downarrow)} E, \tag{2a}$$

$$\frac{\partial J_{\uparrow(\downarrow)}}{\partial x} = \frac{n_{\uparrow(\downarrow)}}{2\tau_{\rm s}} - w(x)\left(n_{\uparrow(\downarrow)}p - n_0 p_0/2\right) + G_{\uparrow(\downarrow)}, \tag{2b}$$

where $n_{\uparrow(\downarrow)}$ is the up(down) spin density, $J_{\uparrow(\downarrow)}$ is the spin *particle* current (which flows in opposite directions to those of *charge* currents), $\tau_{\rm s}$ is the spin-lifetime, and $G_{\uparrow(\downarrow)}$ is the optical generation rate of spin-polarized electrons. The R-G rate $w(x)$ and $E(x)$ have been obtained by solving Eqs. (2). As a consequence, the electrostatic effects due to Schottky barrier formation, the photovoltaic effect and the internal field (Dember field), associated with the ambipolar diffusion of electrons and holes, are all properly taken into account. From Eqs. (2), two coupled differential equations for the spin density $s = n_\uparrow - n_\downarrow$ and spin current density $J_{\rm s} = q(J_\uparrow - J_\downarrow)$ can be obtained

$$\frac{1}{q}J_{\rm s} = -D_{\rm n}\frac{\partial s}{\partial x} - \mu_{\rm n} s{\rm E}(x), \tag{3a}$$

$$\frac{1}{q}\frac{\partial J_{\rm s}}{\partial x} = \frac{s}{\tau_{\rm s}} - w(x)sp + P\Phi_0\alpha e^{-\alpha x}, \tag{3b}$$

where P was taken from Ref. 4 for GaAs and Ref. 5 for Ge. Equations (3) have then been numerically integrated using a boundary value problem solver implemented in Matlab by imposing $s = 0$ at $x = 0$ and $J_{\rm s} = 0$ at $x = 40\,\mu$m.

The model describes the spatial distribution of spins residing at the Γ valley for GaAs and at the L valleys for Ge under the approximation that all the electrons are thermalized. This also implies that $\tau_{\rm s}$ does not depend on photon energy. Moreover, in the case of Ge, the initial electron polarization P at the L valleys has been taken equal to the one at Γ, thus assuming that spin is preserved during the fast Γ to L scattering, an hypothesis supported by photoluminescence studies in bulk Ge.[9,28]

The inset of Fig. 3 shows an example of $J_{\rm s}$ and s as obtained from Eqs. (3) in the case of Ge for $\tau_{\rm s} = 10^{-9}$ s, the

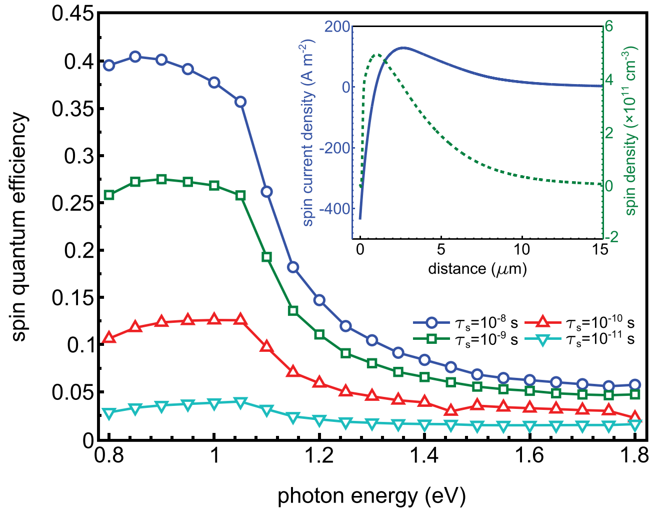

FIG. 3. Photon energy dependence of the *SQE* calculated from Eqs. (3) for a Pt/Ge Schottky barrier. The inset shows the spatial dependence of $J_{\rm s}$ and s for $h\nu = 0.85$ eV and $\tau_{\rm s} = 10^{-9}$ s.

value expected at room temperature.[29,30] $J_{\rm s}$ becomes negative approaching the M/S interface, indicating that indeed spins are injected in Pt. The absolute value of $J_{\rm s}$ at $x = 0$ is therefore the spin current density collected in the Pt layer and can be considered proportional to $\Delta V_{\rm ISHE}$.

Figure 3 shows the calculated *SQE* obtained at different photon energies for $\tau_{\rm s}$ varying from 10^{-8} to 10^{-11} s and can therefore be compared with the experimental data reported in Fig. 2. Since the physical modelling of spin dynamic within a thin Pt layer is far from being adequate in all the different experimental set-ups (i.e., spins injected into Pt from a semiconductor by optical orientation,[12,15–17] spin pumping[22] or magnetic tunnel junctions[23]), our data cannot be used to extract an experimental value of γ in Pt.

We notice that despite the several simplifications already mentioned, Eqs. (3) capture the main features of the ISHE energy spectrum. A closer comparison between the calculated $J_{\rm s}$ and $\Delta V_{\rm ISHE}$ is reported in Fig. 4. For the

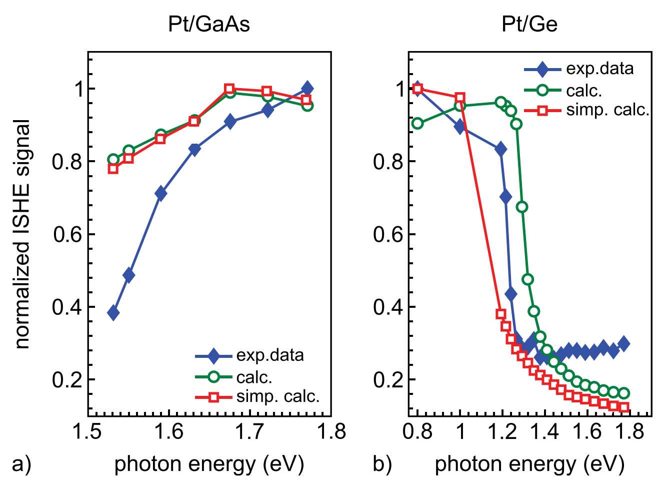

FIG. 4. (a) Comparison between the ISHE signal measured for Pt/GaAs and the calculated spin current for $\tau_{\rm s} = 10^{-11}$ s. (b) Same as (a) but for Pt/Ge and $\tau_{\rm s} = 10^{-9}$ s. Calc. and simp. calc. refer to the solution of Eqs. (3) and (4), respectively.

reasons already discussed, it is not possible to make any quantitative analysis, yet a qualitative comparison between experiments and modelling is still possible. The calculated ISHE signal displayed in Fig. 4(a) has been obtained by solving Eqs. (3) assuming $\tau_s = 10^{-11}$ s, which corresponds with the order of magnitude expected in n-type doped GaAs.[13] A similar comparison is made for Ge in Fig. 4(b) and $\tau_s = 10^{-9}$ s.

It is interesting to underline the role played by τ_s and, as a consequence, by the spin diffusion length $L_s = \sqrt{D_n \tau_s}$, in determining the ISHE energy spectrum. As seen in Fig. 3, in the region comprised between E_d and $E_d + \Delta_0$, the *SQE* monotonically increases as $h\nu$ increases for τ_s comprised between 10^{-10} and 10^{-11} s, as observed in Pt/GaAs. Instead, a monotonic decrease is observed (with the exception of $h\nu \approx E_d$) for τ_s in the 10^{-8}–10^{-9} s range as in the case of Pt/Ge. This can be better understood in terms of a simple model which assumes that all spins generated within a distance L_s from the M/S interface are injected in the Pt contact. J_s can then be written as

$$J_s = q\Phi_0 P[1 - \exp(-\alpha L_s)]. \tag{4}$$

For $h\nu \approx E_d$, highly polarized spins ($P \approx 0.5$) cannot be collected if $\alpha L_s \ll 1$ and a higher value of J_s is obtained at higher photon energy where, despite the lower value of P, α increases resulting in a more efficient spin collection ($\alpha L_s \gg 1$). This is essentially what we have observed in Pt/GaAs. When $\alpha L_s \gg 1$ already at $h\nu \approx E_d$, J_s reproduces the energy dependence of P as seen in Pt/Ge. The comparison between the ISHE signal obtained by using such simplified model and the experimental data is also shown in Fig. 4. It is interesting to note that in the Pt/Ge case, the simplified model better reproduces the experimental data for $h\nu \approx E_d$, where a larger fraction of the photoexcited spins are generated deeper in the semiconductor, i.e., outside the depletion region. Since in Eq. (4) all electric effects are completely neglected, our findings might indicate that the effects of the electric field within the depletion region are overestimated in Eqs. (2) and a more refined analysis including image force lowering of the barrier and the presence of surface states should be considered.

In summary, we have performed photon-energy dependent ISHE measurements on bulk Pt/GaAs and Pt/Ge at room temperature. The two semiconductors clearly show distinct spectral behaviours, which can be qualitatively understood by means of a drift-diffusion model, highlighting the potentialities of photo-induced ISHE as a tool to probe spin physics in semiconductors.

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