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ABSTRACT

Spin relaxation properties of π -conjugated organic semiconductors are key indicators of the performance of organic spintronic devices. However, reliable determination of spin relaxation parameters in organic materials is hindered by complex interfacial phenomena at organic/ferromagnetic metal interfaces that couple spin injection with charge injection. Here, we study the spin pumping induced pure spin transport in Permalloy/rubrene/Pt trilayers and determine the spin diffusion length $\lambda_s = 132 \pm 9$ nm and the spin relaxation time $\tau_s = 3.8 \pm 0.5$ ms in rubrene films at room temperature by using the inverse spin Hall effect. The determined spin diffusion length λ_s is found to be almost two times larger than that of ~ 46.3 nm at 100 K extracted from rubrene spin valve devices in which charge carrier injection/detection occurs at organic/ferromagnetic metal interfaces. Our results demonstrate experimentally that the efficiency and the rate of spin polarized charge transport through the organic/ferromagnetic metal interface play a dominant role in determining the spin relaxation process of spin valve devices in which charge and spin currents are coupled.

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Organic semiconductors (OSCs), including π -conjugated polymers and small molecules, are characterized by extremely weak spin-orbit coupling (SOC) and hyperfine interaction (HFI),¹ which have made them ideal materials for various spintronic applications, including organic spin valves (OSVs),² spin-polarized organic light-emitting diodes (spin-OLEDs),^{3,4} and spin transistors.⁵ In spintronic devices that essentially exploit the spin degree of freedom of electrons for information processing and storage,⁶ the suppression of spin relaxation is essential for enhancing the device performance. For instance, long spin relaxation times, in particular at room temperature, are a prerequisite for effective spin manipulation. As a consequence, quantifying the spin relaxation length and time (λ_s and τ_s) in OSCs not only provides fundamental

insights into the spin transport mechanism in OSCs, but also serves as an important gauge for the rational design of spintronic devices.

Several techniques have been implemented for determining the spin relaxation (or spin diffusion) length λ_s in OSCs, including spin valve technique,⁷ low-energy muon spin rotation technique,⁸ two-photon photoemission technique,⁹ and spin pumping induced inverse spin Hall effect (ISHE).^{10–13} Among them, organic spin valves (OSVs) with a sandwiched structure consisting of ferromagnetic metal (FM)/organic spacer/FM are widely employed for the determination of λ_s by measuring the thickness-dependent magnetoresistance (MR) and fitting it with Jullière's formula. For instance, λ_s in the tris-(8-hydroxyquinolinolato)aluminum (Alq₃) film and amorphous rubrene has been

determined to be ~ 45 nm at 11 K (Ref. 7) and ~ 46.3 nm at 100 K,¹⁴ respectively. However, OSV devices require efficient electrical injection of spin polarized charge carriers from FM to organic spacers, which is inhibited by the well-known conductance mismatch hurdle at organic/FM interfaces that results in the loss of injected spin polarization.¹⁵ In addition, the so-called spinterface effect resulting from strong hybridization of electronic states between organic molecules and FM could alter the spin injection efficiency by providing additional spin polarized electronic states for charge injection or reversing the spin polarization.^{16,17} For example, spin polarized interface states at the Alq_3/Co interface result in a spin-dependent trapping of electrons,^{18,19} which in turn makes this spinterface a spin filter leading to enhanced MR even at room temperature. The inherently coupled charge and spin in OSVs give rise to the question: How significant is the influence of charge/spin relaxation at the interface on the measured spin relaxation in organic films? This question is intriguing because practical spintronic devices are driven by an electric current and organic/electrode interfaces are ubiquitous in those devices.

The pure spin injection and detection can avoid the aforementioned interfacial conductance mismatch problem and the spinterface effect, and they provide alternative means to investigate spin transport.²⁰ Recently, a method combining spin pumping and ISHE has been demonstrated to generate and detect the pure spin current. This method has been applied to determine λ_s in OSCs at room temperature in trilayer architecture of FM/organic interlayer/non-FM with a large SOC (cf. Fig. 1). The spin relaxation time τ_s is subsequently computed using the relationship $\tau_s = \lambda_s^2/D_s$, in which D_s denotes the spin diffusion coefficient. For a detailed mechanism of spin pumping technique, readers are referred to the excellent review by Tserkovnyak *et al.*,²¹ and research reports by Ando *et al.*²² Using this method, λ_s and τ_s in pentacene films are estimated to be 42 ± 10 nm and 0.15 ± 0.12 μs , respectively, at room temperature.^{12,13} $\lambda_s = 50$ nm has been obtained in Alq_3 films at room temperature.²³ For the poly(2,5-bis(3-alkylthiophen-2-yl)thieno[3,2-b]thiophene) (PBTtT) polymer, exceptionally large $\lambda_s = 200 \pm 30$ nm and $\tau_s = 20 \pm 13$ ms are obtained.¹¹ Although SOC is expected to be weak in these molecules containing no heavy elements, the discrepancy among these reported values indicates that other factors, such as molecular geometry/structure, film morphology, and packing structure, are also responsible for the strength of SOC and HFI, which in turn affects the spin transport behaviors in OSCs.^{2,24–26}

Rubrene with its molecular structure shown in Fig. 1(a) is being extensively employed in organic electronic devices, in particular, in (all-)organic spin valves,^{27–32} due to its high charge mobility, long exciton lifetime, and exceptional chemical stability.³³ λ_s in amorphous films was theoretically predicted to be ~ 1 μm ,²⁵ making rubrene a highly attractive material for spintronic applications. Recently, amorphous rubrene based spin valves have been demonstrated to exhibit a large room temperature MR ($\sim 6\%$).²⁸ However, the maximum λ_s of ~ 46.3 nm at 100 K extracted from Jullière's formula is much smaller from the theoretical prediction.³⁴ In this work, spin pumping induced ISHE and current density-voltage (J - V) characteristics are used to examine the pure spin injection and transport properties in rubrene films. $\lambda_s = 132 \pm 9$ nm and $\tau_s = 3.8 \pm 0.5$ ms are extracted from thickness dependent ISHE current. By comparing the λ_s value obtained in the present study with those yielded from MR effects in OSVs, it is found that spin relaxation at the interface dominates over spin relaxation in rubrene-based OSVs.

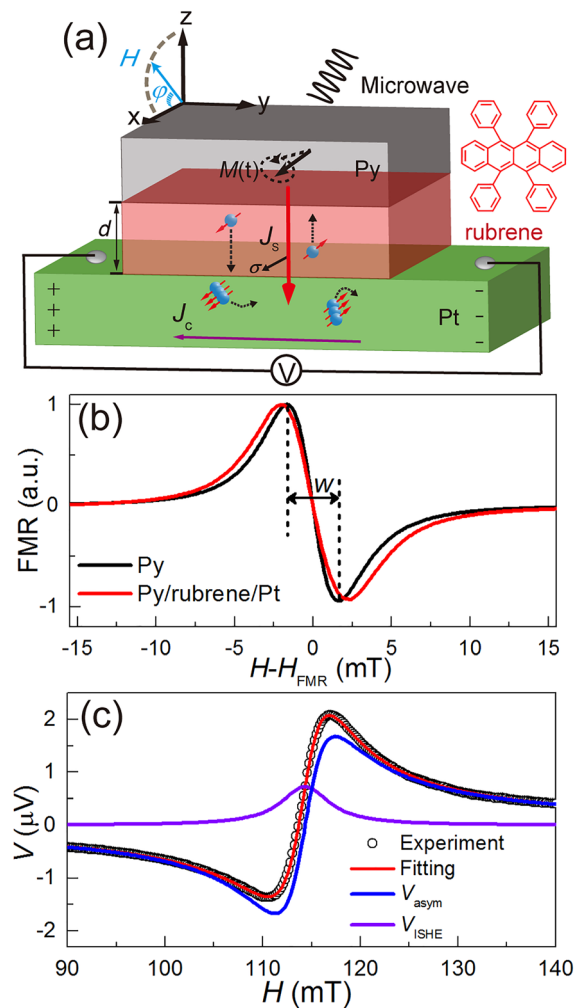


FIG. 1. (a) A schematic illustration of spin pumping and ISHE in a trilayer. ϕ is the angle between the external magnetic field H and the x -axis. (b) FMR derivative of the absorption spectra of a bare Py film and a Py/rubrene ($d = 138$ nm)/Pt device for $\phi = 0^\circ$. (c) The detected DC voltage V vs H spectrum and its decomposition for the same trilayer device with a positive magnetic field.

The Permalloy (Py, $\text{Ni}_{80}\text{Fe}_{20}$)/rubrene/Pt trilayers with geometry illustrated in Fig. 1(a) were fabricated under ultra-high-vacuum conditions. The cross-section area is 0.6×0.6 mm^2 . Spin pumping experiments were performed by using an X-band Bruker EMX-10/12 plus electron spin resonance (ESR) spectrometer. The electrical transport measurements on the same trilayers were carried out in an Ar protected glovebox. The detailed procedures on fabrication and measurements are described in the [supplementary material](#).

To evidence the spin injection and detection, the ferromagnetic resonance (FMR) spectra and the ISHE voltage of the trilayer device of Py/rubrene/Pt [Fig. 1(a)] were measured at a microwave frequency of $f = 9.4$ GHz with a power of 100 mW under both negative and positive fields, respectively. Figure 1(b) shows the field dependence of FMR spectra for a bare Py film and the trilayer of Py/rubrene/Pt with 138 nm of rubrene collected with a positive field, that is, H applied

along the x -axis with $\varphi = 0^\circ$ [cf. Fig. 1(a)]. For clarity, the applied H is rescaled with respect to the resonance field (H_{FMR} , 106 mT for the Py film and 114 mT for the trilayer), and the absorption intensity is normalized to the maximum value. The peak-to-peak linewidth, defined as w in Fig. 1(b), of 4.3 mT for the trilayer is slightly larger than that of 3.2 mT for the bare Py film, suggesting that a pure spin current (J_s) generated by the precession of Py magnetization (M), or the spin pumping process, is pumped into the adjacent rubrene layer due to the strong spin-exchange coupling at the organic/FM interface, and transmitted through an organic layer to the Pt layer.²² The SOC in the Pt layer leads to the conversion of the pure spin current J_s into a charge current (J_c). An electromotive force along the y -axis (E) is thus generated by ISHE following the relation $E = \rho J_c = \theta_{\text{sh}} J_s \times \sigma$, where ρ and θ_{sh} denote the electric resistivity and the spin Hall angle of Pt, and J_s and σ represent the spin current along the z -axis and the spin-polarization direction, respectively.

The voltages $V(H)$ in the Pt layer were detected along the y -axis during FMR measurements, which are shown in Fig. 1(c) as a function of positive sweeping field. In order to extract the electromotive force referred to as the V_{ISHE} signal, the voltage spectrum is fitted by a combination of a symmetric Lorentzian voltage (V_{ISHE} , purple line) profile arising from ISHE and an asymmetric Lorentzian profile (V_{asym} , blue line) due to anisotropic magnetoresistance (AMR) and the anomalous Hall effect (AHE), which is solely observed in the bare Py film³⁵ [cf. Fig. S1(a) in the [supplementary material](#)], according to the following equation:

$$V(H) = V_{\text{ISHE}} \frac{\Gamma^2}{(H - H_{\text{RES}})^2 + \Gamma^2} - V_{\text{asym}} \frac{2\Gamma(H - H_{\text{RES}})}{(H - H_{\text{RES}})^2 + \Gamma^2}, \quad (1)$$

where Γ is the damping constant and H_{RES} is the resonance field. A resonant peak at $H_{\text{RES}} = 114$ mT with a maximum V_{ISHE} of 0.72 μV , as derived from the best fitting shown in Fig. 1(c) (red line), is in excellent agreement with $H_{\text{FMR}} = 114$ mT from the FMR measurements, confirming that the detected V_{ISHE} signal is driven exclusively by FMR-induced spin pumping. In addition, by using the Kittel formula $\omega/\gamma = [H_{\text{FMR}}(H_{\text{FMR}} + 4\pi M_s)]^{1/2}$, in which ω represents the angular frequency $2\pi f$ ($f = 9.4$ GHz) and γ is the gyromagnetic constant which is reported to be $1.86 \times 10^{11}/\text{T}$ for the Py thin film,^{12,13} the saturation magnetization M_s of the Py film is estimated to be 63 mT, which is comparable to the reported value of 59 mT.²²

A series of trilayer devices with varying rubrene thickness (d) ranging from 41 nm to 200 nm were fabricated to quantify the spin relaxation length λ_s in the rubrene film by examining the thickness dependence of V_{ISHE} at room temperature. The thickness d of rubrene films was given by a quartz crystal microbalance during deposition and calibrated independently by scanning electron microscopy (SEM, cf. Fig. S2 in the [supplementary material](#)). As seen from Fig. 2(a), the resonance field H_{FMR} obtained from FMR spectra and saturation magnetization M_s calculated by the Kittel formula remain almost constant with increasing thickness d , evidencing the reproducibility of the Py films in different devices. In addition, the sign of V_{ISHE} changes upon the reversal of the magnetic field from positive ($\varphi = 0^\circ$) to negative ($\varphi = 180^\circ$) [cf. Figs. S1(b) and S1(c) in the [supplementary material](#)] due to the reversed direction of injected spins σ , consistent with ISHE theory. The average current value of $I_{\text{ave}} = [V_{\text{ISHE}}(\varphi = 0^\circ) - V_{\text{ISHE}}(\varphi = 180^\circ)]/2/R$ is thus obtained and plotted in Fig. 2(b) as a function of d , where R represents the resistance of the Pt layer. I_{ave}

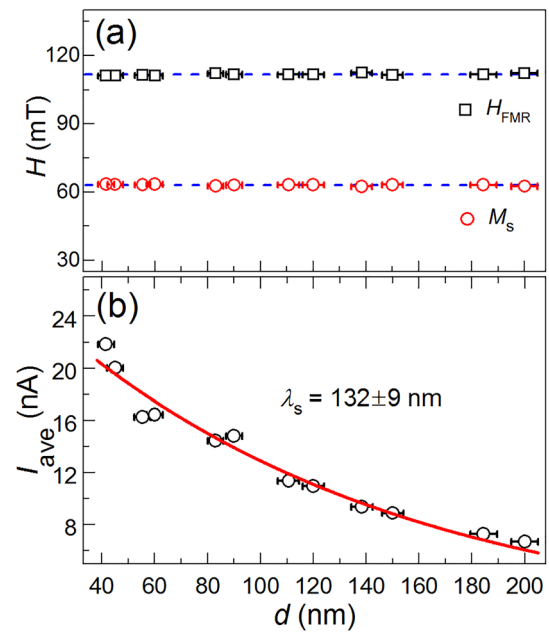


FIG. 2. (a) H_{FMR} and M_s for a series of trilayers with various d . The horizontal dashed-lines are guides for the eye. (b) The evolution of I_{ave} as a function of d . The solid line represents the fitting curve.

decreases exponentially with increasing rubrene thickness, which corresponds well with exponential decay of the spin current with distance by diffusion mode.¹¹ Considering a constant injected spin current $[J_s(0)]$ at Py/rubrene interfaces,^{10–12} an exponential fit of $I_{\text{ave}}(d) \propto J_s(0)e^{-d/\lambda_s}$ thus yields the spin relaxation length $\lambda_s = 132 \pm 9$ nm in the rubrene films. This experimental λ_s is one magnitude less than the theoretically predicted value of $\sim 1 \mu\text{m}$, most likely due to other factors that influence electron spins and their transport, such as HFI and structural disorder in organic films, being out of consideration in the theoretical calculation process.²⁵ This value is nearly two times larger than the previously reported value of 46.3 nm at 100 K by the spin valve method,¹⁴ and almost one order of magnitude higher than that of 13.3 nm at 0.45 K for amorphous rubrene films estimated using the spin polarized tunneling effect.³⁶

For diffusive spin transport, spin relaxation time τ_s and λ_s can be related to the spin diffusion coefficient D_s by $\tau_s = \lambda_s^2/D_s$. Assuming that D_s is the same as the charge diffusion coefficient D_c in particular, when the population of spin-up and spin-down carriers are equal for organic molecules,³⁷ τ_s can be roughly determined by using $\tau_s \approx \lambda_s^2/D_c$. D_c is related to the mobility μ by the Einstein relationship $D_c = k_B T/\mu q$ for nondegenerate semiconductors,³⁸ where k_B , T , μ , and q represent the Boltzmann constant, temperature, carrier mobility, and elementary charge, respectively. To examine the carrier mobility μ , so that the spin relaxation time τ_s can be obtained, the current density-voltage (J - V) characteristics of a trilayer Py/rubrene (138 nm)/Pt were measured at room temperature, and are presented in Fig. 3(a). The J - V curve initially exhibits a linear behavior resulting from the Ohmic diffusion current at low bias (< 0.5 V),³⁹ and a transition into a quadratic voltage dependence region with $\log(J)/V$ vs $\log V$ having a slope of ~ 2 corresponding to the space-charge-limited-current (SCLC) region, in

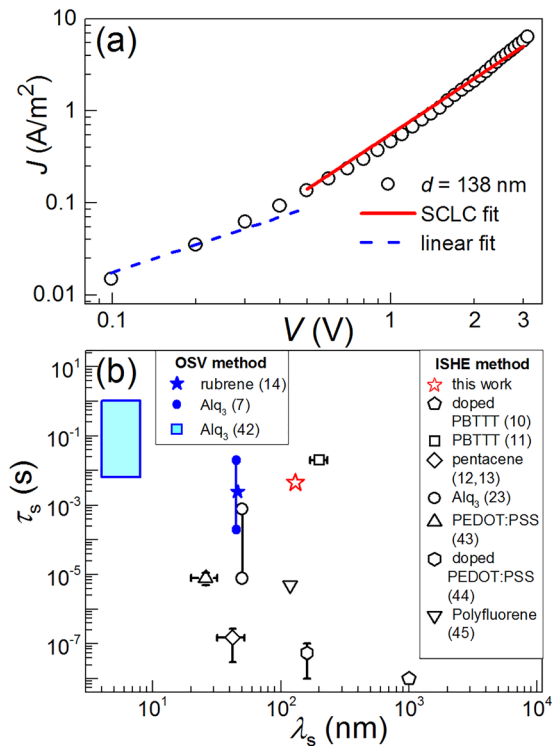


FIG. 3. (a) J - V curve (logarithmic scale) of a trilayer with a rubrene thickness of 138 nm. The red solid line and the blue dashed line represent the best fitting results by the Mott-Gurney law and Ohm's law, respectively. (b) Calculated λ_s vs τ_s for several organic materials. The numbers in brackets refer to the reference values. The blue rectangular region represents the possible values of Alq₃ described in the previous report.⁴²

which the mobility is almost constant. Noticeable deviations from the quadratic voltage dependence occur at relatively high voltages (>3 V, cf. Fig. S3 in the [supplementary material](#)) because the dependence of the mobility on the electric field and the carrier-density has to be taken into account. In the SCLC region (>0.5 V), the carrier mobility can be determined by the Mott-Gurney law⁴⁰

$$J = \frac{9}{8} \mu \epsilon_r \epsilon_0 \frac{V^2}{d^3}, \quad (2)$$

where ϵ_r is the relative permittivity which is ~ 3.5 for amorphous rubrene films⁴¹ and ϵ_0 is vacuum permittivity. The fitting result [cf. Fig. 3(a)] yields a space charge limited carrier mobility of 1.76×10^{-6} cm²/V s at room temperature, consistent with previously reported values of 2.2×10^{-6} cm²/V s for rubrene thin films.³³ D_c is thus calculated to be 4.6×10^{-8} cm²/s. Taking $\lambda_s = 132 \pm 9$ nm from ISHE measurements, the spin relaxation time τ_s in rubrene films is estimated to be 3.8 ± 0.5 ms at room temperature.

Figure 3(b) summarizes the reported values of λ_s and τ_s for several organic molecules determined similarly by a combination of spin pumping and ISHE at room temperature (black open symbols) including the current work. They generally lie in the range of 10–1000 nm for the spin diffusion length and 10^{-6} – 10^{-2} s for the spin relaxation time, which are in line with theoretical calculations.³⁴ Rubrene exhibits

significantly larger λ_s and τ_s than other small OSC molecules such as Alq₃²³ and pentacene,^{12,13} as well as polymers such as poly(3,4-ethylenedioxythiophene):poly(4-styrenesulphonate) (PEDOT:PSS)^{43,44} and polyfluorene;⁴⁵ this is most likely due to its weak SOC, HFI, and moderate carrier mobility. Theoretical calculations demonstrate that the SOC strength in rubrene (3.25×10^{-4}) is one order of magnitude lower than that in Alq₃ (6.91×10^{-3}) due to the absence of metal atoms.³⁴ Although the SOC strength in rubrene is comparable to that in pentacene, HFI strength in rubrene (0.39 mT) is smaller than that in pentacene (1.00 mT) resulting from the more delocalized nature of the electronic wave function of rubrene molecules.²⁶ This exceptional suppression of spin relaxation in rubrene may explain its widespread application in OSV devices^{27–32} and the record-high MR of $\sim 6\%$ at room temperature.²⁸ Only the PBTTT and the doped one have their reported spin transport characteristics superior to those of rubrene because of their enhanced spin exchange coupling resulting from high carrier concentration.^{10,11} However, there are no reports on its spin valve effect.

To understand the influence of organic/FM interfaces on the spin transport in OSV devices, Fig. 3(b) also presents the spin relaxation lengths/times (solid symbols) of a widely used organic spacer of rubrene and Alq₃ molecules for spin valve devices extracted from Jullière's formula. Although the spin relaxation time (>2.5 ms) in rubrene estimated by the spin valve effect is comparable to our results herein, the spin diffusion length is two third less, implying that Jullière's formula may not be suitable to model the FM/rubrene interfaces. A similar underestimation in the spin diffusion length is observed for Alq₃ as well.^{23,42} When the charge and spin currents are coupled, the spin relaxation (or spin memory loss) at organic/metal interfaces is predominant due to the conductance mismatch hurdle,² which suppresses the polarized spin injection/detection efficiency. On the other hand, enhanced spin polarization at the interfaces due to hybridization of electronic states between the metal substrates and organic molecules change the effective spin polarization of FM electrodes, which in turn may increase the spin injection rate.¹⁶ Consequently, tuning both the interfacial energetics by inserting a tunneling barrier and/or reducing a charge injection barrier via interfacial engineering² and interfacial spin polarized electronic states by controlling the spin interface effect are valuable strategies to enhance the spin valve effect.⁴⁶ It is worth noting that the interfacial electronic states' hybridization and energy level alignment, which does not always follow the vacuum level alignment (Schottky-Mott limit) due to the presence of interfacial dipoles, are not independent of each other,^{47,48} and therefore can be optimized simultaneously. On the other hand, developing spintronic devices by manipulating pure spin current rather than spin polarized charge current as in conventional OSVs will likely lead to a paradigm shift in the application of organic molecules in spintronics.

In summary, herein, the spin transport properties of rubrene thin films at room temperature are studied by spin pump induced ISHE, and the room temperature spin diffusion length λ_s has been quantified to be 132 ± 9 nm. In conjunction with the carrier mobility μ of 1.76×10^{-6} cm²/V s in rubrene as extracted from J - V characteristics, the spin relaxation time τ_s is estimated to be 3.8 ± 0.5 ms. The large λ_s and τ_s in rubrene determined in this study help to explain the excellent performance of rubrene-based spin valve devices as reported earlier. By achieving pure spin transport measurements and removing any

interface effects, we have obtained λ_s and τ_s values that are significantly higher than those obtained from conventional spin valve devices. Our study suggests that organic/FM metal interfaces play a dominant role in determining the spin injection/collection. Tuning the charge injection barrier and the spinterface effect are therefore highly sought after to enhance spin injection at the organic/metal interface for high performance organic spin valves. Measuring the intrinsic spin relaxation properties of OSCs has important implications for further optimization of organic spintronic devices by providing an exclusive gauge for the rational design and engineering of organic layers.

See the [supplementary material](#) for experimental details, DC voltage of a bare Py film, DC voltage and extracted V_{ISHE} of a trilayer device with $d = 138$ nm, and SEM cross section images of a device with $d \sim 80$ nm.

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