

Recent Progress in Spin Transport Dynamics in Organic Semiconductors

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Abstract Organic semiconductors (OSCs) have emerged as a compelling materials platform for spintronic research due to their light-element composition generally gives rise to weak spin–orbit coupling and hyperfine interactions, thereby enabling long spin relaxation time and spin diffusion length even at room temperature. These features have stimulated extensive interest in spin transport dynamics in OSCs, and rapid progress in both experimental characterization and theoretical modeling has substantially deepened on the understanding of spin transport, spin relaxation, and spin-dependent interactions in OSCs. In this Review, we provide a comprehensive overview of recent advances in spin transport dynamics in OSCs, with emphasis on spin transport characteristics, spin relaxation strengths and mechanisms in organic films, and spin transport processes in representative device architectures. Finally, we outline the key challenges that remain and highlight emerging opportunities for advancing the fundamental understanding and practical applications of organic spintronics.

Keywords Organic spintronics; Organic semiconductor; Spin transport; Spin relaxation

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INTRODUCTION

Organic spintronics has emerged as an important research frontier in chemistry, physics, and materials science.^[1–4] By exploiting the spin degree of freedom of electrons in organic semiconductors (OSCs), this field aims to uncover the fundamental mechanisms governing spin injection, transport, relaxation, and manipulation while enabling novel device functionalities beyond conventional charge-based electronics.^[5,6] Compared with inorganic semiconductors, OSCs possess unique advantages for spintronic applications because their constituent elements are mainly light atoms, such as carbon (C), hydrogen (H), nitrogen (N), and oxygen (O). This light-element composition generally results in weak spin–orbit coupling (SOC) and relatively weak hyperfine interactions (HFI), which are conducive to maintaining spin polarization over long times and distances, even at room temperature.^[7–9] In particular, the predominance of the ¹²C isotope, which exhibits no nuclear spin, further suppresses HFI-induced spin decoherence. These intrinsic characteristics make OSCs a distinctive platform for fundamental spin studies and the development of next-generation spintronic materials

and devices.^[10]

Understanding the spin transport dynamics in OSCs is of central importance for establishing reliable structure–property relationships and for guiding the rational chemical structure design of high-performance organic spintronic materials and devices. Over the past decades, extensive experimental and theoretical studies have significantly advanced the understanding of spin transport dynamics in organic systems. Key spin transport parameters, such as spin diffusion length, spin relaxation time, and spin polarization, have been widely used to evaluate spin transport performance and probe the underlying spin dynamics.^[11–14] On this basis, increasing attention has been paid to the microscopic origin of spin relaxation in OSCs. Spin relaxation in OSCs is governed by multiple factors, including the molecular chemical structure, heavy-atom effects, HFI-active atom nuclei, intermolecular packing, film morphology, dynamic disorder, and interface-induced perturbations. For example, the incorporation of trace heavy atoms may significantly enhance SOC, whereas hydrogen- and nitrogen-containing groups can strengthen HFI and thus influence spin relaxation.^[15–17] These findings have substantially deepened our understanding of how the chemical structure and condensed phase together determine the spin transport dynamics in OSCs.

The investigation of spin transport in OSCs has also opened up new opportunities for integrating spin functionality with the intrinsic optoelectronic properties of OSCs. Unlike inor-

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ganic semiconductors, in which band transport is often dominant, charge and spin transport in OSCs commonly occurs through hopping between localized states, making spin transport strongly dependent on energetic disorder, molecular coupling, carrier localization, and interface effects. This distinctive transport scenario has stimulated the development of a variety of organic spintronic devices, including organic spin valves, spin photovoltaic devices, spin organic light-emitting diodes, and pure spin-current devices.^[18–20] Therefore, a systematic understanding of the spin transport dynamics in OSCs is essential not only for clarifying the fundamental physics of spin injection, transport, and relaxation, but also for promoting the development of multifunctional organic spintronic devices.

In this review, we summarize recent advances in the spin transport dynamics in OSCs, with an emphasis on the key spin transport parameters, spin relaxation strength and mechanisms, and spin transport processes in representative device architectures. First, we introduce the major experimental approaches applied to evaluate the spin diffusion length and spin relaxation time of OSCs. Next, we discuss the microscopic origins of spin relaxation in organic films, focusing on the respective roles of the SOC and HFI. Next, we review the dominant spin-transport mechanisms proposed for organic spintronic devices, including tunneling, hopping, and exchange-coupling-assisted transport. We further highlight the crucial roles of molecular structure, film morphology, intermolecular interactions, and interfaces in determining the spin transport dynamics. Finally, we outline the major challenges and future prospects for establishing robust structure-property relationships and advancing the rational design of high-performance organic spintronic materials and devices.

EXPERIMENTAL CHARACTERIZATION OF KEY SPIN TRANSPORT DYNAMICS PARAMETERS IN OSC FILMS

When spin-polarized carriers are injected into OSCs, spin polarization is progressively lost during transport owing to various spin relaxation factors, including SOC, HFI, defect-induced scattering, and other local environmental perturbations. Quantitative evaluation of the spin transport performance of different OSCs is essential for establishing reliable structure–property relationships.

In OSCs, the transport and relaxation of the injected spin-polarized carriers are commonly described by the spin diffusion length (λ_S), spin relaxation time (τ_S), and spin diffusion coefficient (D_S). To clarify the physical origin of these parameters, the spatiotemporal evolution of the spin magnetization vector (M) under an applied magnetic resonance field, spin relaxation, and spatial diffusion can be described using the Bloch diffusion equation:^[21,22]

$$\frac{dM}{dt} = \gamma M \times B - \frac{M_{xy}}{T_2} - \frac{M_z - M_0}{T_1} + D_S \nabla^2 M \quad (1)$$

where γ is the gyromagnetic ratio, B is the applied microwave radiation field, M_{xy} is the transverse component of the magnetization vector, T_2 is the spin-spin relaxation time and characterizes the spin relaxation along the transverse direction, M_z is the longitudinal component of the magnetization vector, T_1 is the

spin lattice relaxation time and characterizes the spin relaxation along the longitudinal direction, M_0 is the equilibrium magnetization, and D_S is the spin diffusion coefficient, which describes the spatial propagation capability of spin polarization in OSCs.^[23] Under a steady-state diffusive spin-transport framework and in the absence of spin precession, λ_S , τ_S , and D_S satisfy the relation $\lambda_S = \sqrt{D_S \tau_S}$.^[24]

This chapter focuses on these key parameters and reviews the principal experimental strategies used to characterize them, thereby laying the foundation for the subsequent discussion of spin relaxation mechanisms and spin transport mechanisms in OSCs.

Spin Diffusion Length of OSCs

As spin-polarized carriers propagate through OSCs, spin polarization progressively decays because the probability of spin relaxation increases with transport distance and film thickness. Accordingly, the determination of λ_S depends on quantitatively resolving the attenuation of spin-related signals as a function of the transport thickness. The thickness-dependent spin signals provide a direct basis for extracting λ_S and evaluating the spin-transport performance of OSCs. In this section, we summarize the representative methods used to extract λ_S for different device configurations.

Spin diffusion length extracted from vertical organic spin valve

Organic spin valves typically adopt a sandwich structure composed of two ferromagnetic (FM) electrodes and an intermediate organic layer (Fig. 1a). Owing to the different coercive fields of the two FM electrodes, their relative magnetization orientation can be reversibly switched between parallel and antiparallel states by sweeping the external magnetic field. In the antiparallel state, spin-polarized carriers undergo stronger spin-dependent scattering, leading to higher device resistance than that in the parallel state. Accordingly, MR is defined as

$$MR = \frac{R_{AP} - R_P}{R_P} \times 100\% \quad (2)$$

where R_P (R_{AP}) represents the device resistance in parallel (antiparallel) configuration (Fig. 1b).^[25] For sufficiently thin organic layers, carriers can tunnel directly between the two FM electrodes, giving rise to a TMR. According to Jullière's model, the TMR ratio can be written as

$$TMR = \frac{R_{AP} - R_P}{R_P} \times 100\% = \frac{2P_1 P_2}{1 + P_1 P_2} \quad (3)$$

where P_1 and P_2 are the spin polarizations of the two FM electrodes, respectively.^[26] In contrast to tunnel junctions, however, spin transport in organic spin valves is commonly mediated by the molecular energy levels of OSCs, and spin relaxation may occur during transport process. Meanwhile, owing to the impedance mismatch between the FM electrodes and the OSCs, as well as the formation of a Schottky interface induced by interfacial hybridization, the effective spin polarization of the interface injection and detection at the FM/OSC interfaces can deviate from the bulk spin polarization of the FM electrodes.^[27–29] Consequently, Jullière's formula should be modified when applied to OSC-based spin valves. Considering these interfacial effects, the spin polarization injected from the FM electrode into the OSC can be denoted as the effective interfacial spin polarization (P_1'). During spin transport through the organic layer, the

spin polarization decays exponentially with increasing transport thickness owing to the spin relaxation.^[17] Thus, after traversing the OSC layer with a thickness of d , the spin polarization decays to $P_1'P_2'e^{-(d/\lambda_s)}$. When spin-polarized carriers reach the detector electrode, the corresponding effective interfacial spin polarization is denoted as P_2' . By incorporating these factors into the Jullière formula, the thickness dependence of the MR in organic spin valves can be expressed as:

$$MR = \frac{2P_1'P_2'e^{-(d/\lambda_s)}}{1 - P_1'P_2'e^{-(d/\lambda_s)}} \quad (4)$$

It should be emphasized that Eq. (4) is an effective phenomenological model for analyzing thickness-dependent MR, rather than a complete microscopic description of spin injection across realistic FM/OSC interfaces. The exponential term $\exp(-d/\lambda_s)$ assumes a simplified independent decay of the spin polarization in the OSC layer. In real FM/OSC/FM junctions, the conductivity or impedance mismatch between the metallic FM electrodes and the OSCs layer can strongly affect spin injection, spin accumulation, and spin detection. The interfacial resistance, interfacial spin scattering, and metal/molecule hybridization can further modify the effective interfacial spin polarization. Therefore, $P_1'P_2'$ should be regarded as an effective interfacial spin polarization product that phenomenologically includes the combined effects of impedance mismatch, interface quality, and spinterface-induced hybridization.

Accordingly, fitting the experimental MR as a function of OSC thickness provides an effective way to estimate λ_s and $P_1'P_2'$ (Fig. 1c).^[30] Rather than merely being a fitting parameter, the extracted $P_1'P_2'$ has important physical significance for understanding the overall spin injection and detection efficiency, as well as the interfacial characteristics of the device. First, interfacial hybridization and the 'spinterface' effect can modulate P_1' and P_2' , which may enhance, suppress, or even reverse the effective interfacial spin polarization and thus change the MR response.^[1] Therefore, designing and tuning the interfacial hybridization is a feasible strategy for regulating the MR signals of organic spin valves.^[9,31] Second, for devices with identical material combinations and comparable architectures, comparison of the extracted $P_1'P_2'$ values can serve as an indicator of interfacial quality. In general, a flatter and well-defined FM/OSC interface can reduce the interfacial disorder and suppress unfavorable spin scattering, thereby yielding a larger effective $P_1'P_2'$ and more efficient spin injection and detection.^[25]

In addition to the MR signal, spin-polarized carriers induce a magnetic field signal proportional to spin polarization and decay with the increased thickness of OSCs, which can be detected by the low-energy muon spin rotation (LE-μSR) spectrum.^[32] The total detected magnetic field at distance z can be expressed as:

$$B(z) = B_{d1}\exp(-z/\lambda_{d1}) + B_{d2}\exp(-(z-d)/\lambda_{d2}) + B_s\exp(-z/\lambda_s) + B_a \quad (5)$$

where B_a is the applied magnetic field, B_{d1} and λ_{d1} (B_{d2} and λ_{d2}) are the magnitude and decay length of the average dipolar field at the top (bottom) interface, respectively, d is the thickness of the OSCs, and B_s is the magnitude of the magnetization induced by the injected spin-polarized carriers. The interface dipolar magnitude field and decay length can be obtained by

fitting the zero-current spectrum, and λ_s of the OSCs can be extracted by fitting the spectrum when the current is on (Fig. 1d).

Vertical organic spin valve devices provide a widely used platform for extracting λ_s from thickness-dependent spin-signal measurements. The main advantage of this configuration is its simple device geometry and the direct correspondence between the organic layer thickness and spin signal attenuation. However, this architecture is highly sensitive to the interfacial quality. During the deposition of the top electrode, metal atoms may penetrate the soft organic layer, leading to interfacial damage, the formation of a 'dead layer,' or even metallic filaments. These parasitic effects can reduce the effective transport thickness of the organic layer and may result in overestimated λ_s . Therefore, developing interface-damage-free fabrication methods is important for achieving robust and reproducible λ_s extraction from vertical organic spin-valve devices.^[33]

Spin diffusion length extracted from non-local spin valve

In a non-local spin-valve geometry, the injection of spin-polarized carriers from an FM electrode into a non-magnetic metal or semiconductor creates nonequilibrium spin polarization near the injection interface, resulting in spin accumulation in the nonmagnetic channel.^[34] The spin polarization gradient drives spins to diffuse away from the injector and induces a detectable spin-dependent voltage (V_S).^[35] The associated spin-dependent resistance is typically expressed as $R_S = V_S/I_S$, where I_S is the injected current (Fig. 1e).

To enhance the signal-to-noise ratio and accurately measure the spin-dependent voltage, a nonlocal spin-valve structure has been widely employed. In such a device, the change in the spin-dependent resistance between the parallel and antiparallel magnetization configurations of the FM electrodes can be expressed as

$$\Delta R = \frac{\alpha_F^2 \frac{\lambda_s}{\sigma_N A} e^{-(L/2\lambda_s)}}{(M+1)[M \sinh(L/2\lambda_s) + \cosh(L/2\lambda_s)]} \quad (6)$$

with

$$M = \frac{\lambda_s \sigma_F}{\lambda_F \sigma_N} (1 - \alpha_F^2), \quad \alpha_F = \frac{\sigma_\uparrow - \sigma_\downarrow}{\sigma_\uparrow + \sigma_\downarrow} \quad (7)$$

where α_F is the bulk spin polarization of the FM electrodes, L is the channel length, λ_s (λ_F) is the spin diffusion length of the non-magnetic (ferromagnetic) material, $\sigma_\uparrow$ ($\sigma_\downarrow$) is the majority (minority) spin conductivity in the ferromagnet, σ_N (σ_F) is the total conductivity of the nonmagnetic (ferromagnetic) material, and A is the cross-sectional area of the overlap.^[34] As shown in Fig. 1(f), λ_s can be obtained by fitting ΔR for different channel lengths. For the limit where L is much larger than λ_s , Eq. (6) can be simplified as an exponentially decaying function with respect to L .

Notably, the application of nonlocal spin valves to OSCs critically depends on efficient spin injection at the FM/OSC interface, which requires impedance matching between the FM electrodes and OSCs.^[36] However, the FM/OSC interface generally exhibits low spin injection efficiency owing to impedance mismatch caused by the much lower electrical conductivity of OSCs than FM electrodes, which severely restricts the realization of non-local spin accumulation measurements in organic systems.^[37,38] Moreover, because the spin diffusion length of OSCs is typically below 100 nm, the

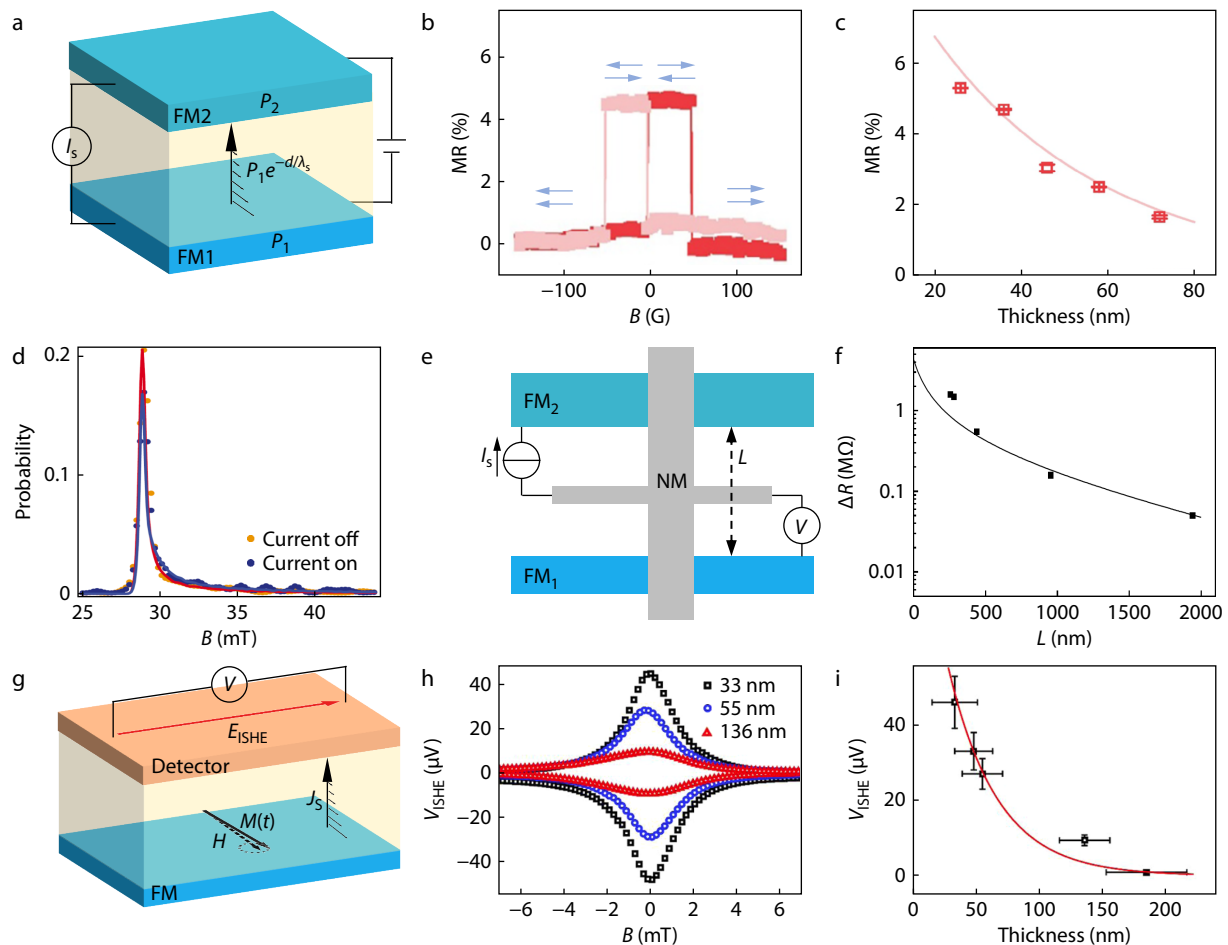


Fig. 1 Representative methods for extracting the spin diffusion length (λ_s) in organic semiconductors (OSCs): (a) schematic diagram of a spin valve. The orange curve represents the decay of spin polarization with increasing OSC thickness. (b) Magnetoresistance (MR) curve of Co/AIO_x/reg-poly(2-ethynyl-1H-indene)/NiFe spin valve and (c) corresponding thickness dependence for extracting $\lambda_s = (40.5 \pm 1.6)$ nm by fitting the thickness dependence of MR according to Eq. (4). The blue arrows in (b) indicate the relative magnetization direction of FM electrodes (Reproduced with permission from Ref. [11]; Copyright (2025), Wiley-VCH GmbH). (d) Magnetic-field distribution in CoFe/TPD/8-hydroxy-quinoline aluminium (Alq₃)/NiFe spin valve detected by LE-μSR with current on (current density 3 mA·cm⁻²) and off. Solid lines show the result of model calculations for a dipolar field distribution due to rough interfaces (red) and spin-polarized current (blue) (Reproduced with permission from Ref. [32]; Copyright (2009), Nature Publishing Group). (e) Schematic diagram of a non-local spin valve. (f) Dependence of ΔR on channel length for a NiFe/Cu/NiFe spin valve measured at 4.2 K, from which $\lambda_s = (1000 \pm 200)$ nm is extracted by fitting according to Eq. (6) (Reproduced with permission from Ref. [34]; Copyright (2001), Nature Publishing Group). (g) Schematic diagram of the generation, transport and detection of pumped pure spin current. (h) Typical p-ISHE(B) response in the NiFe/Super yellow poly-phenylenevinylene (SY-PPV)/Pt polymer device with various d and (i) the corresponding thickness dependence. $\lambda_s = (39 \pm 6)$ nm is extracted by fitting the thickness dependence according to exponential decay (Reproduced with permission from Ref. [20]; Copyright (2020), American Physical Society).

fabrication of lateral devices with sufficiently short transport channels remains highly challenging. Therefore, addressing these issues is essential for the future development of non-local organic spin-valve devices.

Spin diffusion length extracted from pure spin current devices

Pure spin current devices provide an ideal platform for investigating spin transport in OSCs owing to almost 100% spin polarization. Typically, pure spin currents are generated by magnetization precession in the FM layer under ferromagnetic resonance (FMR) conditions, and then, a spin current diffuses into adjacent nonmagnetic layers through dynamic exchange coupling, which is called spin pumping.^[39,40] Because there is no charge current injected across the FM/OSC interface, it circum-

vents the impedance mismatch of electric-field-driven spin injection and exhibits high spin injection efficiency.^[41,42] According to the inverse spin Hall effect (ISHE), the SOC field generates an inverse spin Hall current (I_{ISHE}) and voltage (V_{ISHE}) at the detector, $I_{\text{ISHE}} \propto V_{\text{ISHE}} \propto J_s \times \sigma$, where σ is the spin polarization direction, and J_s is the spin current propagation direction (Fig. 1g).^[43,44]

Under the FMR condition, the magnetization dynamics of the FM electrode under a magnetic field H can be described by the Landau-Lifshitz-Gilbert equation (LLG) Gilbert equation:

$$\frac{dM(t)}{dt} = -\gamma M(t) \times H + \frac{\alpha}{M_s} M(t) \times \frac{dM(t)}{dt} \quad (8)$$

where γ is the gyromagnetic ratio, α is the Gilbert damping con-

stant, and M_s is the saturation magnetization. α is directly proportional to the linewidth ΔH_{FM} of the microwave absorption at a fixed microwave frequency. When the spin is pumped into the non-magnetic material (NM) layer, owing to the FM/NM interface coupling, the spin angular momentum will attenuate and the effective damping will increase, which is manifested as an increase in linewidth $\Delta H_{\text{FM/NM}}$, the additional damping $\Delta\alpha$ can be expressed as $\Delta\alpha = (\gamma/\omega) (\Delta H_{\text{FM/NM}} - \Delta H_{\text{FM}})$.^[45] The additional damping $\Delta\alpha$ derived from the LLG equation can also be expressed as

$$\Delta\alpha = B \left(1 + \frac{C}{\tanh(d_N/\lambda_S)} \right) \quad (9)$$

where B and C are material-dependent constants.^[46] Therefore, λ_S of OSCs can be obtained by fitting the thickness dependence of the FM/NM microwave absorption spectrum linewidth ΔH .

Another way to obtain λ_S is to fit the thickness dependence of V_{ISHE} , which V_{ISHE} can be expressed as

$$V_{\text{ISHE}} = \frac{w\theta_{\text{SH}}\lambda_S \tanh\left(\frac{d_N}{2\lambda_S}\right)}{(d_N\sigma_N + d_F\sigma_F)} \left(\frac{2e}{h}\right) J_S \quad (10)$$

where d_N (d_F) and σ_N (σ_F) are the thickness and conductivity of NM (FM) layer, w is the FM width parallel to the interface, $\hbar$ is reduced Planck constant, θ_{SH} is the spin Hall angle of NM layer, J_S is the spin current density injected into the non-magnetic layer, which can be calculated from the FMR(H) response.^[47] It is also worth noting that fitting the thickness dependence of the V_{ISHE} yields λ_S and θ_{SH} simultaneously.

It should be noted that Eq. (10) is derived under simplified assumptions, including efficient absorption of the injected spin current at the NM or OSC interface and a single-exponential spin relaxation profile inside the transport layer. When the film thickness d_N is significantly smaller than λ_S , the hyperbolic tangent term in Eq. (10) approximates a linear function: Under this condition, the spin backflow effect that reduces the net spin current absorbed by the adjacent NM layer should be considered, especially for OSCs with a relatively long λ_S .^[48] In addition, when the pulsed ferromagnetic resonance is used, the corresponding correction to Eq. (10) is necessary.^[49]

In addition to the above methods, λ_S can also be extracted by exponentially fitting the thickness dependence of I_{ISHE} or V_{ISHE} , $I_{\text{ISHE}} \propto V_{\text{ISHE}} \propto e^{-(d/\lambda_S)}$, as shown in Figs. 1(h) and 1(i).^[20,50,51]

It should be noted that V_{ISHE} is highly sensitive to the spin-to-charge conversion coefficient of the detector material, which is governed by SOC strength. Owing to the intrinsically weak SOC of OSCs, the spin-to-charge conversion coefficient is generally small, making the detection of ISHE signals more difficult. In addition, FMR absorption is often accompanied by considerable heat generation; therefore, the thermoelectric voltage arising from the Seebeck effect may interfere with, or even overwhelm, the genuine ISHE response.^[52–54] Furthermore, reliable signal analysis requires careful exclusion of spurious contributions, including the anomalous Hall effect (AHE), spin backflow, and anisotropic magnetoresistance (AMR) etc.^[55–58]

Section summary

The thickness dependence of the vertical spin valves, non-local

spin valves, and spin pumping devices provides a reliable means for characterizing the λ_S of OSCs. However, certain differences exist in the applicable ranges and the underlying physical mechanisms.

First, in terms of applicability, although non-local spin valve devices offer higher signal-to-noise ratios and avoid overestimation of spin relaxation lengths owing to interfacial metal penetration, the lateral channel distances are typically several hundred nanometers, making them more suitable for materials with long λ_S . In contrast, the organic layer thicknesses in vertical spin valves and spin pumping devices can be easily tuned to tens of nanometers, enabling the characterization of OSCs with shorter λ_S , even below 10 nm. Additionally, the inverse spin Hall effect relies on a strong SOC, which contradicts the generally weak SOC strength of OSCs, making them better suited for studying materials with strong SOC and short λ_S .

Second, in terms of the physical mechanism, the MR signal is affected by factors such as the interface spin polarization, contact resistance, impedance mismatch, interface hybridization, interface spin scattering, and interface quality. Therefore, λ_S extracted through MR-thickness-dependent measurements can represent the effective spin transport performance of OSCs in a real electric injection device. In contrast, the nonlocal spin valve measurement extracts the λ_S of the bulk OSCs under the diffusion mechanism because the nonlocal device configuration spatially separates the charge current and spin current, thereby reducing the parasitic MR effect and the influence of the interface pseudo-signal. In contrast, spin pumping devices provide relaxation evolution of the pure spin current injected under ferromagnetic resonance conditions, and the characterization results reflect the dynamic spin relaxation process of OSCs.

Spin Relaxation Time of OSCs

τ_S is one of the fundamental parameters for characterizing the spin relaxation in OSCs. While λ_S describes the spatial attenuation of spin polarization, τ_S characterizes the timescale of spin relaxation and, therefore, directly reflects the persistence of the nonequilibrium spin polarization of OSCs. As such, τ_S is widely used to assess the spin relaxation strength of OSCs and to analyze the influence of the molecular composition, chemical structure, intermolecular interactions, and film microstructure on spin relaxation.^[59] Reliable determination of τ_S is essential for understanding spin evolution in OSCs. In this section, we review the principal methods used to determine τ_S in OSC systems.

Derivation of Spin Relaxation Time from Spin Diffusion Length

In diffusion spin transport, λ_S and τ_S are linked by $\lambda_S = \sqrt{D_S\tau_S}$, where D_S is the spin diffusion coefficient.^[24] This relation provides an indirect route for estimating τ_S from λ_S , or *vice versa*, provided that D_S can be reasonably determined.^[60–62] In OSCs, where spin transport is strongly coupled to charge transport, D_S is often approximated by the charge diffusion coefficient D , which, under (quasi-) quasi-equilibrium conditions, may be described by the Einstein relationship $D_S = D = k_B T \mu / q$, where T is the temperature, q is elementary charge, and μ is the carrier mobility.^[63–67] In this way, τ_S can be extracted from the experimentally obtained λ_S .

However, $\lambda_s = \sqrt{D_s \tau_s}$ should be used cautiously. First, the Einstein relation describes the charge-diffusion coefficient rather than the spin-diffusion coefficient. Therefore, the approximation $D_s \approx D$ is only reasonable when spin transport is coupled with charge transport, as in hopping-dominated spin transport.^[68] In contrast, when spin and charge transport are partially decoupled, for example, in exchange-coupling-mediated spin transport, D_s may be much larger than D , and an independent description of spin diffusion is required.^[69] Indeed, recent studies on disordered polymer films have revealed that D_s may be much larger than D , which has been attributed to exchange-coupling-assisted spin transport.^[20] Under such circumstances, extracting τ_s from λ_s and the Einstein relation may lead to significant uncertainties. Second, the Einstein relation in disordered OSCs is generally valid under (quasi-) quasi-equilibrium states. Under strong electric fields or far-from-equilibrium transport conditions, significant deviations may occur.^[70] Accordingly, if μ is used to estimate D , then the mobility should be determined in equilibrium states.^[71] Overall, although the relationship between λ_s and τ_s provides a useful route for parameter estimation, its validity depends critically on the underlying spin transport mechanism and transport conditions of OSCs.

Determination of spin relaxation time based on Hanle effect

The Hanle effect is one of the most important experimental signatures for evaluating spin relaxation in spin-transport systems and provides a direct route for determining τ_s . When a magnetic field is applied perpendicular to the spin polarization direction, the injected spins undergo Larmor precession during transport, which obeys the Lorentz expression $1/(1+(\omega_L \tau_s)^2)$, where $\omega = g\mu_B B/\hbar$ is spin precession frequency, g is electron g -factor, μ_B is the Bohr magneton, and B is the applied transverse magnetic field.^[72,73] τ_s can be obtained by fitting the decay of the spin polarization with respect to the transverse magnetic field.

Based on the coupling between charge and spin, Johnson and Silsbee proposed an electrical detection scheme for nonequilibrium magnetization at FM/paramagnetic interfaces.^[74,75] In this description, spin transport is governed by a one-dimensional Bloch drift-diffusion equation containing diffusion, spin relaxation, and Larmor precession terms, with an additional drift contribution included when the charge current flows through the device.^[76] In a non-local transverse spin geometry, the solution for steady-state spin polarization is given by

$$S_x(x_1, x_2, B) = \int_0^\infty \frac{S_0}{\sqrt{4\pi D_s t}} e^{-(x_2 - x_1 + v_d t)^2 / 4D_s t - t/\tau_s} \times \cos(g\mu_B B t / \hbar) dt \quad (11)$$

where S_x is the x component of the steady-state spin polarization, x_1 is the position of the spin injection electrode, x_2 is the position of the spin detection electrode, D_s is the spin diffusion coefficient, t is time, and v_d is the drift velocity, which is non-zero in the crossover configuration with electron transport.^[77,78] The value of τ_s can then be extracted by fitting the perpendicular magnetic-field dependence of the nonlocal resistance or voltage.^[79,80] In the presence of electrodes for both spin injection and detection, if the current density and drift velocity in the semiconductor are both small (the drift term can be ignored) and the contact is much larger than λ_s , the fit of Eq. (11) can be simplified to a Lorentzian line fit.^[81,82]

In inorganic semiconductors, the Hanle effect is often hindered by parasitic effects such as spin filtering, Hall voltage, and the magnetoresistance effect caused by body scattering in FM films. Moreover, the impedance mismatch between FM metals and semiconductors frequently suppresses electrical spin injection efficiency, making Hanle detection more challenging. To avoid interference from undesirable effects, Appelbaum employed hot electron injection to realize a high spin-polarized injection and detected the Hanle effect in undoped single-crystal float-zone (FZ)-Si by detecting the secondary collector current.^[83,84] The spin precession frequency can be calculated from the magnetic field when the secondary collector current reaches the first extreme value, and the average precession angle can be approximated as π rad, from which the spin transit time (τ_{transit}) of float-zone (FZ)-Si can be derived. τ_{transit} is controlled by the voltage V_{C1} . Assuming an exponential decay of spin polarization, τ_s can be calculated using the following formula:

$$\frac{\Delta I_{C2}(V_{C1})}{I_{C1}(V_{C1})} \propto \exp\left[-\frac{\tau_{\text{transit}}(V_{C1})}{\tau_s}\right] \quad (12)$$

where I_{C2} and ΔI_{C2} are the normalized magnetic current and its variation, respectively, I_{C1} is the current of the first collector.^[85]

In the early stages of organic spintronics, the lack of an unambiguous Hanle signal in OSC systems triggered persistent debate over whether spin-polarized carriers could be efficiently injected into and transported through OSCs.^[86] However, the absence of the Hanle effect should not simply be interpreted as direct evidence of spin injection or spin transport.

In general, the failure to observe a resolvable Hanle response in organic spintronic devices is commonly associated with low spin injection efficiency caused by impedance mismatch at the FM/OSC interface, as well as rapid spin relaxation or dephasing during transport.^[87] These factors can suppress spin accumulation and obscure precession-induced variation. In addition to these conventional factors, the underlying spin-transport mechanism may also affect the Hanle response. For OSCs in which exchange-coupling-assisted spin transport is involved, the spin transit time may become sufficiently short, such that the accumulated precession angle remains too small to generate a clearly resolvable Hanle response within the experimentally accessible magnetic-field window.^[88,89] Therefore, a reduced precession angle may be an additional factor hindering the detection of the Hanle effect in organic systems.

Recently, Yang *et al.* reported a high MR ratio of up to 440% in a $(\text{La}_{2/3}\text{Pr}_{1/3})_{5/8}\text{Ca}_{3/8}\text{MnO}_3$ (LPCMO)/8-hydroxy-quinoline aluminum (Alq_3)/Co spin valve with high spin injection efficiency. By monitoring the resistance change (ΔR) under a perpendicular magnetic field, they extracted a hole spin relaxation time of $\tau_s \approx 45$ ps (Figs. 2a and 2b).^[14] This value is much shorter than that reported for bulk Alq_3 , suggesting that exchange-coupling-assisted transport may play an important role in Alq_3 films. Nevertheless, this interpretation remains under active discussion and more systematic experimental and theoretical studies are required to verify its general applicability.

Extraction of spin relaxation time of OSCs by electron spin resonance (ESR)

ESR provides a powerful tool for simultaneously extracting T_1

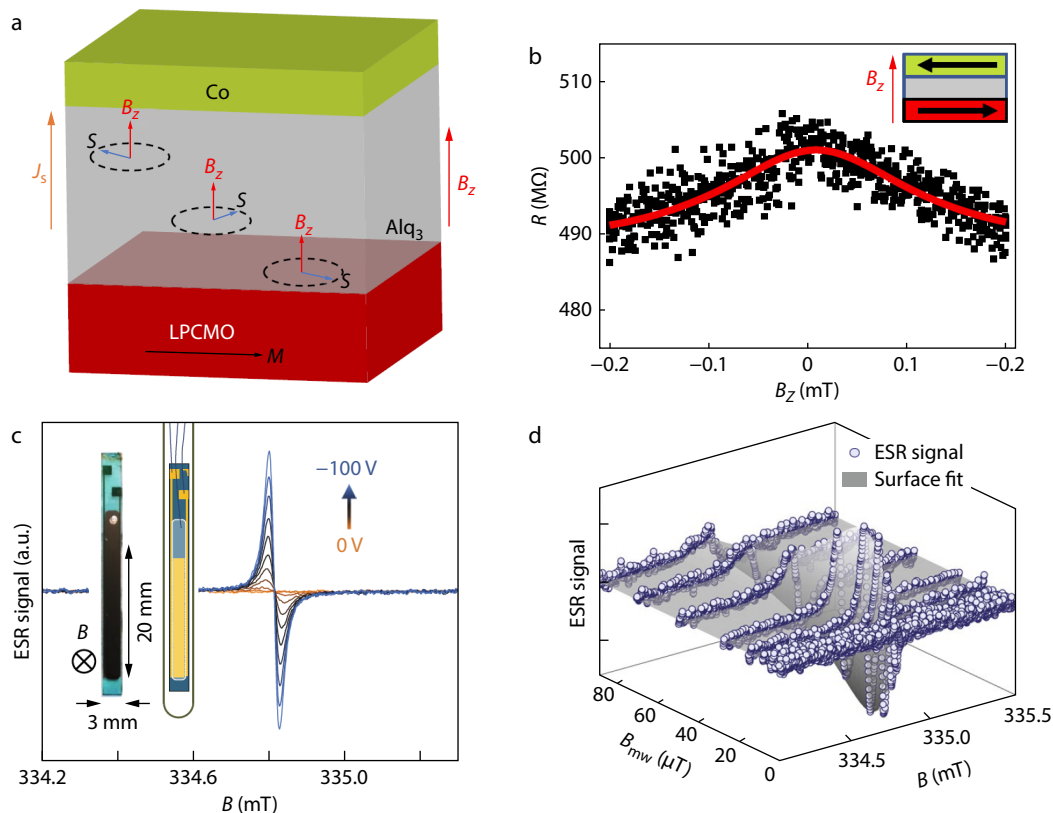


Fig. 2 Different methods for extracting spin relaxation time. (a) Schematic of Hanle effect detected in $(\text{La}_{2/3}\text{Pr}_{1/3})_{5/8}\text{Ca}_{3/8}\text{MnO}_3$ (LPCMO)/Alq₃/Co spin valve and (b) corresponding Hanle effect detected under antiparallel magnetic configuration (Reproduced with permission from Ref. [14]; Copyright (2019), Nature Publishing Group). (c) Field induced ESR spectrum of amorphous indacenodithiophene-co-benzothiadiazole copolymer (IDTBT) and a photograph and schematic diagram of the device (Reproduced with permission from Ref. [63]; Copyright (2019), Nature Publishing Group). (d) ESR spectrum of IDTBT with increasing microwave power at $V_G = -70$ V, 140 K, and 2D fit to determine spin relaxation time (Reproduced with permission from Ref. [63]; Copyright (2019), Nature Publishing Group).

and T_2 from OSCs or single-ion radicals.^[90] Under an external static magnetic field B_0 , the Zeeman splitting of the electronic energy levels in the OSCs creates an energy level space $\Delta E = g\mu_B B_0$. When a microwave radiation field with a frequency of ν perpendicular to B_0 satisfies $h\nu = g\mu_B B_0$, the unpaired electrons of the low-energy level undergo resonance absorption and transition to a high-energy level. Owing to spin-spin relaxation and hyperfine interactions, non-uniform broadening or splitting of the absorption lines occurred (Fig. 2c). T_2 can be obtained by fitting the absorption spectrum with the Lorentz function, and then T_1 can be calculated based on T_2 and the saturated absorption spectrum.^[91,92] By two-dimensional fitting of the ESR spectrum with different microwave powers, the error can be minimized, particularly for T_1 (Fig. 2d).^[63]

Notably, ESR measurements rely on unpaired electrons in the molecular orbitals, which can be obtained by field-induced or dilute solution doping to form free radicals.^[93–95] Meanwhile, field-induced ESR measurements require a sufficiently high spin density in the sample, which requires the integration of multiple field-effect transistor devices in the resonant cavity or optimized device design, posing challenges to device fabrication and measurement.^[62,63] While inducing organic free radicals by doping in dilute solutions, it is necessary to pay attention to maintain a low solution concentration to prevent interactions between spins on different

molecules, and to monitor the changes in free radical concentration during measurement to ensure the accuracy of the results.^[95]

Section summary

This section introduces three common methods for extracting the spin relaxation time of OSCs, providing a reference for analyzing spin relaxation strength and mechanisms. However, the application scenarios and characterization of the physical mechanisms of the three methods vary.

First, $\lambda_S = \sqrt{D_S \tau_S}$ characterizes the spin relaxation process of the equilibrium state of bulk OSCs. It can be used for a simple correlation analysis of specific relaxation factors and spin relaxation strength, such as comparing the spin relaxation strength of annealed and as-cast OSC films. However, owing to certain errors in the characterization of λ_S and D_S , the derivation process amplifies the measurement error, resulting in low accuracy and making it difficult to achieve quantitative analysis.

The Hanle effect can accurately characterize the relaxation process of spin transport in bulk OSCs in real devices. However, the Hanle effect is susceptible to impedance mismatch, low spin-injection efficiency, and interference from rapid spin relaxation, making its detection difficult.

Unlike the two aforementioned methods for characterizing the spin-relaxation time of bulk OSCs, ESR characterizes the

microscopic dynamic spin-relaxation process of spin carriers induced at the FM/OSC interface by field or in dilute solutions by doping. This can be used to study the influence of the molecular structure and composition on the spin relaxation strength, but deviates from the spin transport process of the solid-state bulk.

Therefore, it is inappropriate to evaluate the spin-relaxation strength of OSCs by directly comparing the results of the three methods. This should be noted when using the spin relaxation time to analyze the causes of relaxation in OSCs.

SPIN RELAXATION STRENGTH AND MICROSCOPIC MECHANISMS IN OSC FILMS

Spin relaxation is a central process in spin transport dynamics because it determines the loss of non-equilibrium spin polarization during transport in OSCs. The microscopic origin of spin relaxation is essential for establishing reliable structure–property relationships between molecular characteristics and spin transport performance. In OSCs, spin relaxation is strongly influenced by multiple structural factors, including the molecular composition, configuration, molecular conformation, intermolecular packing, and film microstructure. Revealing how these factors govern spin relaxation is therefore of great importance for the molecular design of high-performance OSCs for spin-transport applications.

For OSCs, spin relaxation is primarily governed by the SOC and HFI.^[96,97] As OSCs are typically composed of light elements, their intrinsic SOC is generally much weaker than that of metals or inorganic semiconductors. However, the SOC strength can be modulated by heavy-atom substitution, molecular orbital overlap, and intermolecular interactions, which enhance spin relaxation.^[98] Meanwhile, the HFI arising from the interaction between electron spins and nuclear spins provides another important spin relaxation channel in OSCs.^[99] In this chapter, we review the main approaches used to evaluate the spin relaxation strength in OSC films and discuss the microscopic mechanisms underlying spin relaxation in these systems.

SOC Strength of OSCs Films

In terms of molecular composition, heavy metal atoms are one of the major contributors to the SOC strength of OSCs.^[100–102] In contrast to single atoms, bond formation and orbital hybridization of OSCs, which are related to molecular configuration and conformation, respectively, also significantly affect the SOC strength. This section summarizes the quantitative and semi-quantitative parameters that characterize the SOC strength of OSCs, such as θ_{SH} , V_{ISHE} , phosphorescence (Ph) fluorescence (FL) ratio (Ph/FL) of the electroluminescence spectrum, g -factor shift (Δg), and spin admixture parameters (γ^2).

θ_{SH} is a quantitative parameter for evaluating the SOC strength of OSCs, which can be experimentally extracted by fitting the thickness dependence of the V_{ISHE} or I_{ISHE} signals of spin-pump devices (Fig. 3a).^[49,103,104] As derived from Eq. (10), V_{ISHE} is proportional to θ_{SH} , unlike θ_{SH} , V_{ISHE} also depends on the device structure. Hence, under the same device structure, V_{ISHE} can serve as a semi-quantitative parameter of SOC strength in OSCs. Sun *et al.* extracted the θ_{SH} of several Pt-containing polymers with tunable SOC strengths.^[49] Based on

the extracted θ_{SH} , they found that as the Pt content decreased, the θ_{SH} of Pt-containing polymers decreased from about 10^{-3} for Pt-1 and Pt-3 to about 10^{-4} for Pt-Q, but was still much larger than that of metal-free OSCs.^[64] This indicates that trace heavy metal atoms significantly enhance the SOC strength of OSCs, and θ_{SH} and V_{ISHE} can serve as reliable SOC strength indicators.^[105,106]

Δg is another parameter for characterizing the SOC strength of OSCs and can be expressed as

$$\Delta g = \Delta g^{\text{RMC}} + \Delta g^{\text{GC}} + \Delta g^{\text{OZ/SOC}} \quad (13)$$

where Δg^{RMC} is a relativistic mass correction term, Δg^{GC} is a diamagnetic correction, and $\Delta g^{\text{OZ/SOC}}$ is a cross-term between the orbital Zeeman and SOC operators, and is related to the mixed derivative of the molecular total energy with respect to the electron magnetic moment and external magnetic field.^[107] Because the values of Δg^{RMC} and Δg^{GC} are negligibly small and have opposite signs that nearly cancel each other, $\Delta g^{\text{OZ/SOC}}$ dominates the contribution Δg in almost all cases, which depends on the SOC.^[108] Therefore, Δg can reflect the SOC strength of OSCs, which can be measured using ESR or DFT calculations. Based on the extracted Δg values of different molecules, Schott *et al.* established the effect of molecular composition and configuration on SOC strength.^[95] They verify that the introduction of heavy atoms significantly enhanced the SOC strength of the OSCs. For example, by replacing the S atoms in [1]benzothieno[3,2-*b*][1]benzothiophene (BTBT) ($\Delta g = 2.0 \times 10^{-3}$) molecules with Se atoms in [1]benzoseleno[3,2-*b*][1]benzoselenophene (BSBS) ($\Delta g = 1.4 \times 10^{-2}$), Δg increased nearly sevenfold. They also found that the introduction of alkyl chains at heavy atomic sites may cause spin density depletion in heavy atoms (Fig. 3b), thereby reducing SOC strength. For instance, 2,7-diethyl[1]benzoseleno[3,2-*b*][1]benzoselenophene (C8-BSBS) ($\Delta g = 6.8 \times 10^{-3}$), where the introduced alkyl chain located at the Se atom of BSBS, has a much smaller Δg than BSBS ($\Delta g = 1.4 \times 10^{-2}$), while the Δg of 3,8-diethyl[1]benzoseleno[3,2-*b*][1]benzoselenophene (C8s-BSBS) with an introduced alkyl chain far away from the Se atom ($\Delta g = 1.7 \times 10^{-2}$) was enhanced.

However, Δg solely accounts for intra-orbital spin mixing and neglects inter-orbital spin mixing, which underestimates the SOC strength of the OSCs.^[109] To this end, Yu proposed γ^2 to quantitatively evaluate the SOC strength of the OSCs.^[110] Because interorbital mixing is closely related to the molecular configuration, γ^2 varies with the molecular torsion angle. Taking biphenyl as an example, γ^2 increases exponentially with increasing molecular torsion angle and reaches a maximum value near $\pi/2$ (Fig. 3c), which means that molecules with orthogonal configurations may have stronger SOC strength. This explains why Alq₃, which has a lower atomic number but orthogonal three ligands, exhibits a higher spin relaxation strength and a lower λ_s than sexithiophene (T₆).^[32,111] This theory also explains the curvature-enhanced SOC strength due to intermolecular orbital mixing in C₆₀ and C₇₀ molecules.^[49,112,113] Those results imply that molecular configuration fluctuations may significantly affect the spin relaxation strength, which is common in disordered polymer films. Therefore, regulating the molecular packing structure may be an effective means to further enhance the spin transport performance of OSCs.

In addition, the strong SOC strength facilitates intersystem

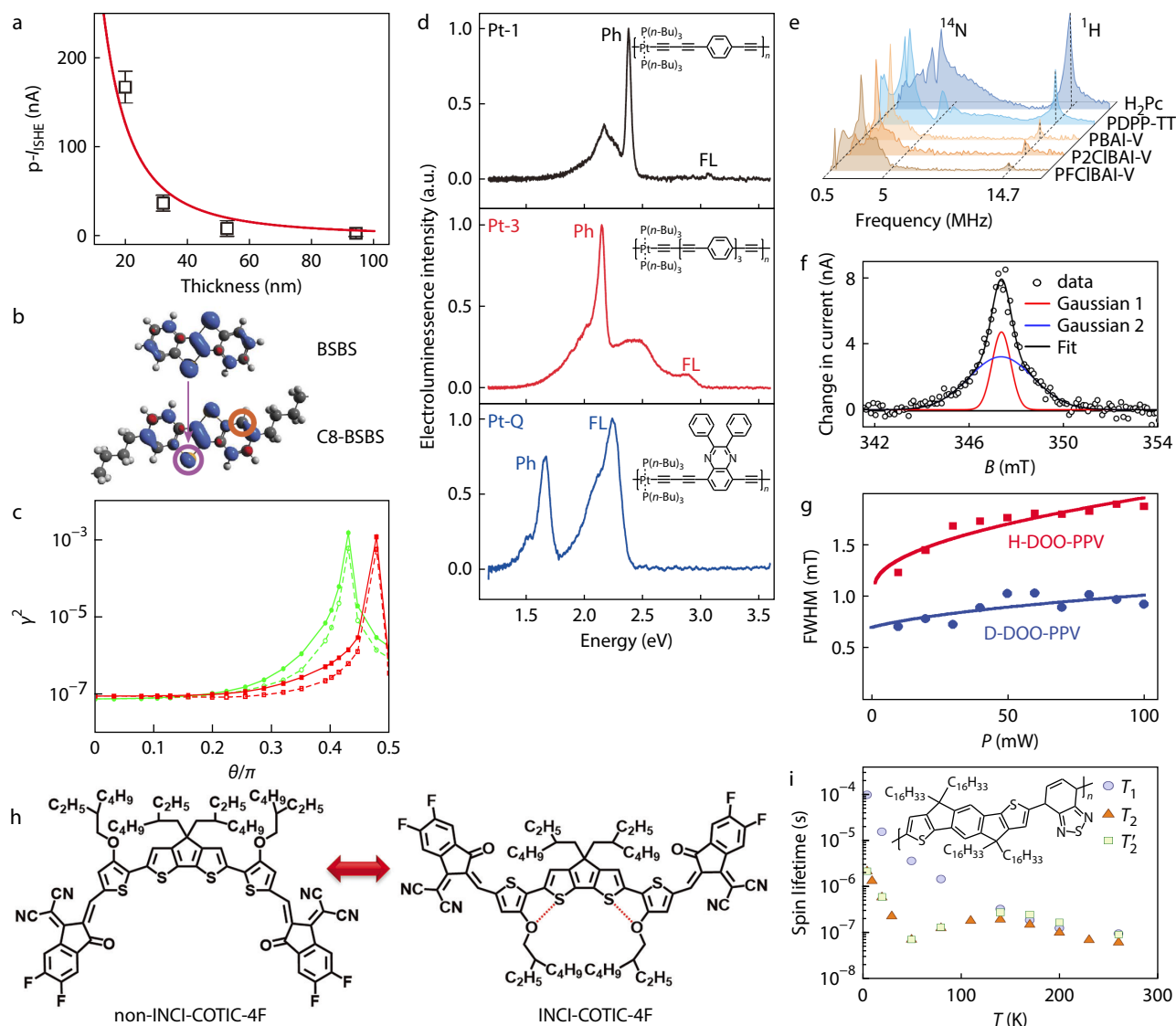


Fig. 3 The characterization of spin relaxation strength in OSCs. (a) Thickness dependence of the I_{ISHE} responses in NiFe/Pt-1/Cu spin pumping device. The extracted $\lambda_5 = (1.5 \pm 0.5)$ nm, spin Hall angle $\theta_{\text{SH}} = (-2.7 \pm 1.0) \times 10^{-3}$ (Reproduced with permission from Ref. [49]; Copyright (2016), Nature Publishing Group). (b) Spin isodensity contours of BSBS and C8-BSBS. Spin maximum and minimum are shown in blue and red, respectively. The shifted bonding site at the phenyl rings has been labelled in yellow, and the observed spin depletion at heavy atoms highlighted in purple (Reproduced with permission from Ref. [95]; Copyright (2017), Nature Publishing Group). (c) Spin admixture parameters γ^2 as a function of torsion angle θ in biphenyl. Green (light gray) and red (dark gray) lines are for the electron and hole polarons, respectively (Reproduced with permission from Ref. [110]; Copyright (2011), American Physical Society). (d) Normalized electroluminescence spectrum for Pt-1 (black), Pt-3 (red) and Pt-Q (blue), respectively. Inserts show the corresponding molecular structure. The Ph/FL ratio can be used to qualitatively estimate SOC strength (Reproduced with permission from Ref. [49]; Copyright (2016), Nature Publishing Group). (e) Frequency domain ESEEM spectrum of echo signal after Fourier transform (Reproduced with permission from Ref. [108]; Copyright (2024), Nature Publishing Group). (f) Two Gaussian lines fitted EDMR spectrum of MEH-PPV OLED devices (Reproduced with permission from Ref. [120]; Copyright (2010), American Physical Society). (g) The microwave power dependence of the ODMR spectrum FWHM for D-DOO-PPV (blue) and H-DOO-PPV (red) (Reproduced with permission from Ref. [119]; Copyright (2010), Nature Publishing Group). (h) Molecular structures and conformation transformation of non-INCI-COTIC-4F (left) and INCI-COTIC-4F (right) (Reproduced with permission from Ref. [92]; Copyright (2024), Wiley-VCH GmbH). (i) The temperature-dependent of T_1 for hole polarons in IDTBT measured by ESR at $V_G = -70$ V (Reproduced with permission from Ref. [63]; Copyright (2019), Nature Publishing Group).

crossing from singlet to triplet manifolds, thus promoting Ph emission.^[114] The Ph/FL ratio of the electroluminescence spectrum can serve as a semi-quantitative parameter for characterizing the SOC strength of OSCs.^[115] As shown in Fig. 3(d), with the increase of Pt content in the Pt-containing polymer, Ph/FL increases from 0.75 for Pt-Q molecules to 12 for Pt-3, and finally up to 27 for Pt-1 molecules.^[49] This also indicates

that an increase in Pt content significantly enhances the SOC intensity of the OSCs.

It is worth noting that although Δg may underestimate the SOC strength of OSCs, it is proportional to γ^2 and can serve as a semiquantitative parameter to evaluate the SOC strength. Meanwhile, γ^2 depends on theoretical calculations, and the analysis combined with the experimentally extracted θ_{SH} ,

Ph/FL, Δg can more comprehensively characterize the SOC strength of OSCs.

HFI Strength of OSCs Films

The HFI constitutes another major microscopic mechanism governing spin relaxation in OSCs, originating from the coupling between electron spins and nuclear spins. Because the most abundant carbon isotope of ^{12}C indicates no nuclear spin, the HFI in OSCs is often dominated by ^1H nuclei, and in some molecules may also involve contributions from other spin-active atoms, such as ^{14}N .^[116] In this section reviews the principal experimental approaches used to evaluate HFI strength in OSCs, including electron spin-echo envelope modulation (ESEEM), optically detected magnetic resonance (ODMR), electrically detected magnetic resonance (EDMR), and magnetoconductivity (MC). We also discuss the influence of molecular vibrations and conformational changes on the dynamic HFI strength. In addition, we summarize representative strategies for weakening the HFI, such as promoting 'motional narrowing,' enhancing charge delocalization by annealing, and using isotope substitution.

By detecting the spin echo with an increasing pulse time interval, the ESEEM spectrum is used to characterize the HFI strength of the nuclear spin of specific atoms.^[15] This is based on the principle that if the atoms exhibit no nuclear spin, the electron spin echo decays exponentially with the pulse time interval, whereas the presence of magnetic nuclei induces characteristic oscillations in the echo intensity. These oscillations occur at characteristic frequencies corresponding to nuclear spin transitions, allowing us to distinguish the contribution of a specific nuclear spin to the HFI strength. Yang *et al.* investigate the HFI-induced spin relaxation in bay-annulated indigo (BAI)-based polymers using ESEEM, and the spin echo shows two distinct characteristic frequencies 2.5 MHz and 14.7 MHz, respectively (Fig. 3e), indicating that the spin-polarized carriers undergo two types of HFI modulation field of nuclear spins in the thin film.^[108] By comparing the characteristic Larmor frequency of atoms, the echo signal of 14.7 MHz is attributed to ^1H atoms located at the side of BAI unit, while the echo signal of 2.5 MHz is attributed to ^{14}N atom located at the conjugate center of BAI unit. Moreover, the modulation intensity of ^{14}N atom is much more robust than that of ^1H atom, indicating that the atoms within the conjugated unit contribute more to the HFI strength of the polymer than the edge atoms.

The EDMR, ODMR, and MC spectra can also be used to characterize the comprehensive HFI strength of the OSCs.^[16,117] Since the spin mixing caused by the local HFI field, the magnetic resonance spectrum and magnetic response broadened inhomogeneously.^[118,119] The HFI field strength can be semi-quantitatively characterized by Gaussian fitting of the half-width at half-maximum (HWHM) of the magnetic resonance spectrum. By fitting the EDMR spectrum of polymer MEH-PPV OLED devices with two Gaussian lines, McCamey *et al.* extract the HFI fields of two polaron types as 2.7 and 0.8 mT, respectively (Fig. 3f), indicating that the HFI field strength is related to the polaron type.^[120] This result is similar to the results of the MC HMHW of a poly(3-hexylthiophene) (P3HT) device discovered by Geng *et al.*^[30] The HMHW of electron devices is approximately three times wider than that of hole devices, indicating that the electrons experience

3 times a localized HFI field that is three times stronger than that of holes, which is thought to be related to the stronger charge localization of electrons in disordered polymer films. Through annealing, the HMHW of the MC of the P3HT film gradually decreases from 10.6 G to 7.7 G as the annealing temperature increased to 160 °C. This indicates that the disordered molecular packing may be the culprit of charge localization, and increasing the degree of molecular packing order can reduce the strength of the local HFI field.

In addition to static nuclear spin distributions, dynamic HFI should also be considered in OSCs. Techniques such as ESEEM and EDMR mainly provide information on static HFI components, whereas molecular vibrations, conformational fluctuations, and rotational or segmental motions can cause fluctuations in the local HFI field experienced by the spin carriers. This dynamic HFI is particularly relevant in polymeric semiconductors, where torsional motion and segmental relaxation can continuously modulate the molecular conformation, orbital overlap, spin density distribution, and local nuclear spin environment.

For polymer molecules, conformational fluctuations can alter the overlap between the molecular orbitals, thereby changing the local HFI strength. Yu's theoretical study showed that when the torsion angle of a biphenyl molecule increased from 0° to 90°, the HFI strength increased by approximately 20%, indicating that molecular conformational changes can significantly influence the dynamic HFI.^[116] Moreover, the dynamic disorder induced by thermal vibrations or phonons can lead to continuous torsional and vibrational fluctuations, generating randomly fluctuating local HFI fields and enhancing phonon-assisted spin scattering.^[121]

It should also be noted that a fast carrier motion may reduce the effective HFI strength through the motional narrowing effect.^[122] When charge carriers move sufficiently fast through multiple local hyperfine fields with different orientations, these local fields can be averaged into a weaker effective hyperfine field. This motional averaging can suppress hyperfine-induced spin dephasing and may prolong the apparent spin relaxation time. Recently, Carey *et al.* showed that a high carrier mobility in organic single-crystal systems can promote motional narrowing and weaken HFI-related spin relaxation.^[123] In rubrene single crystals, where the structural disorder is largely reduced, a microsecond-scale spin relaxation time was observed even at room temperature.

Because the HFI of OSCs is mainly contributed by hydrogen nuclear spin, Nguyen *et al.* prepared D-DOO-PPV by replacing hydrogen atoms ^1H of H-DOO-PPV with deuterium atoms ^2D to reduce the HFI strength.^[119] D-DOO-PPV shows a narrower full-width at half maximum (FWHM) of the ODMR spectrum than H-DOO-PPV and yields the HFI constant $a(D) = a(H)/6.5$ (Fig. 3g), validating the effectiveness of isotope substitution in reducing HFI strength.

Overall Spin Relaxation Strength and Identification of Dominant Spin Relaxation Mechanisms

In OSC films, spin relaxation is generally governed by the joint action of the SOC and HFI. As these two mechanisms have distinct microscopic origins and cannot be quantified on the same basis, their relative importance cannot be assessed by direct comparison. Therefore, it is necessary to employ parameters

that reflect the overall spin relaxation strength and, in combination with complementary analyses, to identify the dominant spin relaxation mechanism in a specific OSC system.

Since spin relaxation arises from the coupling of the spin to its surrounding environment, fluctuations in the local magnetic fields can induce transitions between Zeeman levels, leading to energy dissipation and spin depolarization. Within this framework, T_1 is a key parameter for evaluating the overall spin-relaxation strength of OSCs. Based on T_1 values extracted from ESR measurements, Yang *et al.* investigated the influence of intramolecular noncovalent interactions (INCI) on the spin relaxation strength.^[12] As shown in Fig. 3(h), INCI can regulate the molecular conformation, backbone planarity, and electronic delocalization, thereby redistributing the spin density along the conjugated framework. In conformationally locked molecules, heteroatoms such as S and O are directly involved in the spin transport route. As S and O atoms possess stronger SOC than C and H atoms, their enhanced participation can promote SOC-mediated spin scattering and increase the probability of spin-flip processes. Consequently, films with INCI exhibit a shorter T_1 and reduced spin diffusion length compared to control films without such interactions.

The dominant spin relaxation mechanism in OSCs can be distinguished by temperature-dependent analysis of the spin relaxation time.^[62] It is worth noting that although T_1 can quantitatively compare the spin relaxation strength of OSCs, T_1 usually exhibits an SOC-mediated Elliot-Yafet relaxation mechanism that monotonically decreases with increasing temperature,^[124,125] leading to the details of spin relaxation being masked.^[90] Therefore, the microscopic details of the spin-relaxation mechanism cannot be resolved from T_1 . Therefore, the temperature dependence of T_2 was adopted for the analysis of the dominant spin-relaxation mechanism in OSCs.

Based on T_2 of the high-mobility polymer semiconductor amorphous indacenodithiophene-co-benzothiadiazole copolymer (IDTBT), Schott *et al.* identified three characteristic temperature intervals of the spin relaxation mechanism, as shown in Fig. 3(i).^[63] First, below 50 K, T_2 decreases with an increase in temperature, accompanied by a distinctly inhomogeneous broadening of the ESR spectrum, indicating that inhomogeneous static local fields, such as HFI fields, act as dominant fluctuating fields causing spin relaxation. Second, in the range of 50–140 K, T_2 increases with temperature, and the resonance spectrum exhibits a Lorentz line shape. This phenomenon is attributed to motional narrowing, in which faster carrier motion averages out randomly distributed local HFI fields, thereby suppressing spin dephasing. Third, above 150 K, T_2 is approximately equal to T_1 and decreases with increasing temperature, consistent with the SOC-mediated Elliot-Yafet relaxation mechanism. In this high-temperature range, incoherent hopping transport becomes dominant, and the SOC-induced mixing of spin states during hopping enhances the spin-flip process, which dominates the spin relaxation.

In some scenarios, λ_S can be used as a semiquantitative parameter to characterize the spin relaxation strength of OSCs according to $\lambda_S = \sqrt{D_S T_S}$. However, λ_S is not determined solely by spin relaxation because it also depends on the spin diffusion coefficient D_S , which is closely related to carrier mobility,

molecular packing structure, and film microstructure.^[126] Therefore, λ_S is more suitable for semi-quantitative comparisons among systems with similar transport pathways and comparable structural characteristics, such as isotopically substituted molecules (e.g., H-DOO-PPV and D-DOO-PPV),^[119] chemically related fullerene systems (e.g., C_{60} and C_{70}),^[112] or the same material in different packing structures, such as as-cast and annealed P3HT,^[30] single crystals, and polycrystals of TIPS-pentacene.^[127]

In addition, the MR of spin valves can be used to evaluate the spin relaxation strength of OSCs. However, according to Eq. (4), the MR depends not only on the spin relaxation in OSCs, but also on the spin polarization of the FM electrodes, OSC thickness, interfacial properties, and device fabrication conditions. Therefore, MR should only be used with caution for semi-quantitative comparisons and is most meaningful when comparing OSCs measured in the same device architecture and under closely matched experimental conditions.^[128,129]

It is still challenging to directly compare the relative strengths of SOC and HFI in OSC molecules. The relationship between the molecular structure and spin relaxation strength as well as the spin transport performance still needs to be established. Fortunately, through a summary of studies on specific relaxation factors, researchers have identified the main sources of the SOC and HFI. Generally, SOC contributes significantly to the spin relaxation strength of molecules containing metal atoms, (such as Pt-containing polymers), with S, O, N, and other heteroatoms (such as INCI), conjugated groups in an orthogonal configuration (such as Alq_3), and large molecular curvature (such as C_{60}). HFI has a greater contribution to molecules with N atoms as heteroatoms at the conjugated center or H atoms contained in the carrier delocalization range. The design strategies, including light element composition, deuterium substitution, planar delocalization of conjugated structures, and avoiding the introduction of heteroatoms, point to efficient spin-transport materials.

SPIN TRANSPORT MECHANISMS OF OSC FILMS

The spin-transport mechanism is a key issue in organic spintronics because it determines both the physical nature of spin propagation in OSCs and the extent to which spin transport can be integrated with the intrinsic electronic and optoelectronic functionalities of OSCs. In contrast to conventional inorganic systems, spin transport in OSCs has multiple pathways depending on the molecular electronic structure, degree of carrier localization, intermolecular coupling, and device architecture. Therefore, identifying the dominant transport mechanism is essential for understanding the spin transport dynamics and rationally designing functional organic spintronic devices.

Different transport mechanisms imply fundamentally different spin manipulation routes. In hopping-mediated transport, spin-polarized carriers propagate through molecular energy levels and remain closely coupled to charge transport, making spin transport strongly dependent on the carrier motion and external stimulation. This feature is particularly important for the realization of multifunctional spintronic devices that combine charge, spin, and photoresponses. In contrast, in tunneling-dominated devices, spins traverse the or-

ganic spacer through direct tunneling, which bypasses the photoelectric functionalities of the organic layer. Exchange-coupling-assisted transport has also emerged as an important concept for explaining spin propagation in OSCs, particularly in systems with high carrier concentrations. In this chapter, we review the main spin-transport mechanisms proposed for OSC-based spintronic devices, including tunneling, hopping-mediated transport, and exchange-coupling-assisted transport.

Tunneling Spin Transport

Tunneling spin transport is based on the quantum tunneling of spin-polarized carriers across an ultrathin spacer layer. In organic tunnel junctions, when the OSC layer is sufficiently thin (typically below approximately 5 nm), it can function as a tunneling barrier that permits direct spin-polarized tunneling between the two FM electrodes.^[130] In this case, the device resistance changes sharply when the relative magnetization alignment of the FM electrodes switches between parallel and antiparallel states, leading to TMR. As expressed in Eq. (3), TMR is primarily governed by the spin polarization of the two FM electrodes.

A key feature of tunneling-dominated transport is weak thermal activation. Accordingly, the current-voltage characteristics and resistance of such junctions typically exhibit only a weak dependence on temperature (Fig. 4a).^[131,132] Other commonly signatures of tunneling transport include parabolic differential conductance and an exponential increase in resistance with increasing spacer thickness.^[133,134]

Under the tunneling mechanism, spin transport predominantly occurs through direct quantum tunneling between the

two electrodes, which can avoid spin relaxation within the OSC bulk and is favorable for obtaining TMR signals with a relatively high signal-to-noise ratio. Therefore, tunneling transport is particularly suitable for ultrathin organic layers and junction-type devices, where a clear MR response is the primary objective, which can be activated by selecting a thin spin-transport layer. However, because spin carriers do not substantially propagate through the molecular energy levels of OSCs, their ability to regulate spin signals through OSC-specific optical, electrical, or excitonic responses is limited. This restricts the integration of intrinsic optoelectronic functionalities and spin transport in tunnel-dominated organic spintronic devices.

Hopping Spin Transport

In hopping spin transport, spin-polarized carriers are injected into OSCs under an applied electric field and propagate through molecular energy levels *via* site-to-site hopping.^[135] This is the dominant transport regime in disordered polymer films when the thickness of the organic spacer exceeds the tunneling limitation.^[136] Unlike inorganic semiconductors, where carriers typically move through delocalized band states, OSCs often exhibit energetic and structural disorder and charge carriers are localized.^[137] Therefore, carrier transport in OSCs proceeds through thermally assisted hopping between localized states, which exhibits pronounced temperature dependence in device resistance and *I-V* characteristics. In some cases, the *I-V* curve exhibits an onset voltage, below which the current is very small, and above which the current increases rapidly, and will increase as the temperature decreases.^[138]

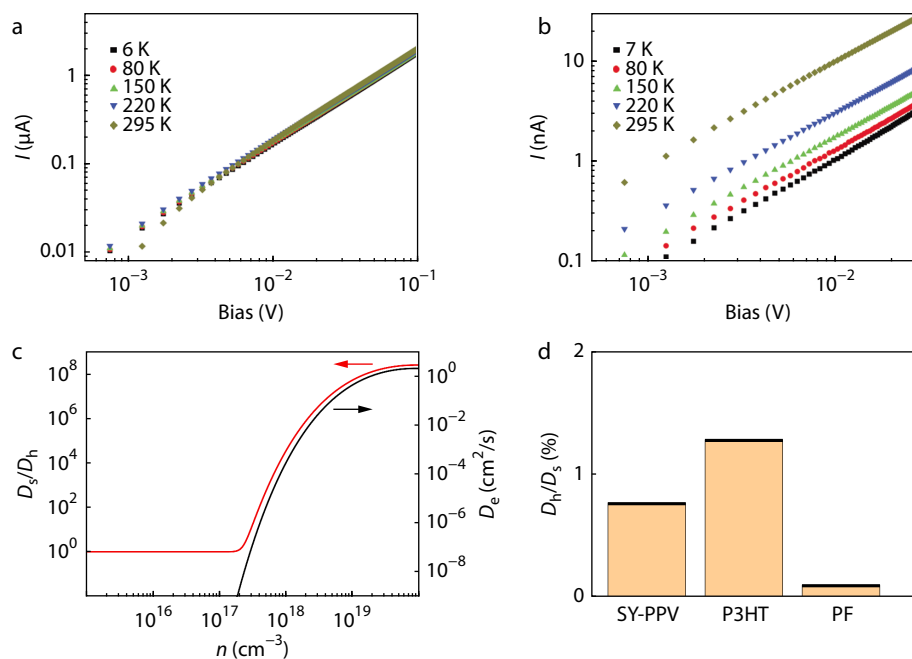


Fig. 4 Different spin transport mechanisms in OSCs. (a) The *I-V* curves of a Co/Al₂O₃/F₁₆CuPc/NiFe spin valve with weak temperature dependence, which exhibits tunneling mechanism (Reproduced with permission from Ref. [139]; Copyright (2016), Wiley-VCH GmbH). (b) The *I-V* curves of a Co/AlO_x/F₁₆CuPc/NiFe spin valve with strong temperature dependence, which exhibits hopping transport mechanism (Reproduced with permission from Ref. [139]; Copyright (2016), Wiley-VCH GmbH). (c) The ratio of the spin diffusion coefficient to the hopping diffusion coefficient (D_s/D_h) and the exchange-coupling diffusion coefficient (D_e) as a function of carrier concentration (Reproduced with permission from Ref. [89]; Copyright (2013), American Physical Society). (d) The ratio of the hopping diffusion coefficient to the exchange-coupling diffusion coefficient (D_h/D_s) of SY-PPV, P3HT and polyfluorene (PF).

In organic spintronic devices, the interfacial injection barrier can strongly influence whether the spin transport is dominated by direct tunneling or hopping. In general, a relatively high injection barrier tends to favor tunnel-dominated transport, whereas a lower barrier is more conducive to hopping-mediated injection and transport. Sun *et al.* found that the interfacial barrier height can be tuned by adjusting the oxidation time of the AlO_x interfacial layer, thereby enabling a transition between spin transport regimes.^[19,139] Longer oxidation times tend to produce a higher injection barrier, leading to tunneling transport with a weak temperature dependence of the I - V curves. As the oxidation time decreased, the injection barrier was gradually reduced and finally governed by the energy level gap between the molecular transport levels and the Fermi level. Under these conditions, spin transport was dominated by hopping, as reflected by the much stronger temperature dependence of the I - V characteristics (Fig. 4b).

Recent studies have revealed that hopping transport in OSCs may not be limited to nearest-neighbor processes. For example, long-range hopping can be promoted by super-exchange contributions to the transfer integral, which may enhance the electric-field dependence of the carrier mobility.^[140] In addition, variable-range hopping has been proposed as an important transport mode in disordered OSC systems and may become dominant at room temperature.^[141]

Unlike tunneling transport, hopping spin transport involves carrier propagation through the molecular energy levels of OSCs, thereby providing a route for coupling spin transport with the electronic and optoelectronic functionalities of OSCs. In this regime, the motion of injected spin-polarized carriers is accompanied by charge transport, exciton-related processes, and photoelectric conversion.^[142] Therefore, hopping-mediated spin transport is particularly suitable for the development of multifunctional organic spintronic devices by selecting OSCs with optical, electrical, or excitonic responses as the spin transport layer, such as spin organic light-emitting diodes (spin-OLEDs),^[143,144] and spin photovoltaic devices.^[18,145] In these systems, spin-dependent functionalities can be realized through the interplay between spin transport and charge-related processes. Nevertheless, hopping transport is also more susceptible to energetic disorder, trap states, hyperfine interactions, and thermally activated spin relaxation, which may limit the spin diffusion length and reduce the spin signal.

Exchange-coupling-assisted Spin Transport

Exchange-coupling-assisted spin transport is an important regime for spin propagation in OSCs, especially in pure-spin-current devices and systems with abundant impurity states or high carrier concentrations. When neighboring occupied states are sufficiently close, the overlap of their electronic wave functions enables exchange between localized spins, allowing spin propagation through OSC films without requiring charge-carrier transport.

In Yu's theory, the spin diffusion coefficient in OSCs contains both the hopping contribution D_h and the exchange-coupling contribution D_e ,^[89] that is, $D_s = D_h + D_e$. $D_h = k_B T \mu / q$ and D_e can be expressed as

$$D_e = \frac{1.6J(R)R^2}{\hbar} \quad (14)$$

$$J(R) = \frac{0.821R^{5/2}e^{2-2R/\xi}}{4\pi\epsilon_0\epsilon_r\xi^{7/2}} \quad (15)$$

where $R = n^{-1/3}$ is the radius of spin, $n = \sigma/q\mu$; ϵ_0 is the vacuum dielectric constant, ϵ_r is the relative dielectric constant, σ is the conductivity of OSCs; ξ is polaron localization length and $\hbar$ is reduced Planck constant. Here, D_h is linear with the carrier mobility, whereas D_e increases exponentially with increasing carrier concentration and polaron localization length. As a result, when the carrier concentration reaches the range of 10^{17} – 10^{18} cm^{-3} or the polaron localization length reaches approximately 1 nm, exchange-coupling transport may become the dominant spin transport mechanism (Fig. 4c).^[89]

Exchange-coupling-assisted spin transport mainly occurs in disordered OSC films. Owing to the large number of defects and localized states introduced by the disordered molecular arrangement, an impurity band forms and exchange-mediated spin transport is activated.^[146,147] Consistent with this picture, Groesbeck *et al.* recently observed spin-charge-separated transport in several OSC systems, including super yellow poly-phenylenevinylene (SY-PPV), P3HT, and polyfluorene (PF), and found that the extracted D_s significantly exceeded D_h by several orders of magnitude (Fig. 4d),^[20] indicating that exchange coupling can dominate spin transport in disordered organic films.

According to Eqs. (14) and (15), the carrier concentration is an important tuning parameter for the exchange coupling mechanism. By doping poly(2,5-bis(3alkylthiophen-2-yl)thieno [3,2-*b*]thiophene) (PBTtT) films with F4-TCNQ to increase the carrier concentration, Wang *et al.* obtained a λ_s up to $(1.2 \pm 0.1) \mu\text{m}$, and concluded that exchange-coupling-assisted transport was the dominant spin transport pathway in this system.^[51] This finding indicates that carrier concentration engineering is a promising route for enhancing spin diffusion in OSCs.

Exchange-coupling-assisted transport provides a potential pathway for achieving long-range spin transport by enhancing the electronic coupling and increasing the polaron localization length through molecular packing regulation. In principle, this mechanism may be activated in doped OSCs, organic single crystals, covalent organic frameworks (COFs), or other highly ordered materials with extended and rigid π -conjugated planes. Compared to hopping transport, exchange-coupling-assisted transport may reduce the influence of site-to-site hopping disorder and enable more efficient spin propagation. However, its experimental realization remains challenging because it requires precise control of molecular stacking, high-quality spin device fabrication, mechanically robust molecular crystals, and reliable characterization methods for the polaron localization length. Therefore, although regulating molecular stacking to increase the polaron localization length and induce exchange-coupling-assisted spin transport represents a promising strategy for improving spin transport performance in OSCs, further experimental verification and theoretical modeling are still needed.

CHALLENGES AND OUTLOOK

Significant progress has been made in understanding the spin

transport dynamics in OSCs, from quantitative evaluation of the spin diffusion length and spin relaxation time to identification of microscopic spin relaxation mechanisms and transport pathways. In particular, deeper insights have been gained into the roles of SOC, HFI, disorder, molecular packing, and motional narrowing as well as into the distinct contributions of tunneling, hopping-mediated transport, and exchange-coupling-dominated transport in different OSC systems.

However, several key challenges remain. Direct and unambiguous verification of spin injection and spin transport in OSCs is difficult in many cases, and the interpretation of Hanle effect signals remains under investigation.^[116] Meanwhile, a general structure-property relationship linking the molecular structure, solid-state molecular arrangement, and spin transport dynamics has yet to be established. Addressing these issues will require a closer integration of multimodal characterization, interface engineering, mechanistic modeling, and material design^[148–150].

In conclusion, high-mobility single-crystal OSCs with weak HFI and SOC, optimized interfaces, and large polaron localization length to activate exchange-coupling-dominated transport represent promising directions for the development of efficient spin-transport OSCs. The features of organic single crystals, such as long-range order, low defect states, and trap density, provide an ideal material platform for exploring the intrinsic spin transport dynamics mechanisms of organic molecules, as well as for verifying and improving spin transport and relaxation theoretical models. Meanwhile, its millisecond-scale spin relaxation time and the expected micrometer-scale spin diffusion length provide a physical basis for the development of nonvolatile, ultralow-power spin logic and storage devices, which will drive organic spintronics for practical device applications. However, owing to the bottleneck of metal atom penetration during the preparation of organic single-crystal spin devices, research on the spin transport dynamics of organic single crystals based on these devices is still in its early stages. In the future, the application of damage-free device fabrication techniques may be an important pillar for advancing the research on the spin dynamics of organic single crystals.

More broadly, future efforts should move beyond simply extending the spin diffusion length or spin relaxation time and instead focus on actively regulating the dominant spin transport mechanism and coupling spin transport with the intrinsic optoelectronic functionalities of OSCs. Such advances are essential for deepening the fundamental understanding of spin transport in organic materials and enabling next-generation multifunctional organic spintronic devices.

BIOGRAPHIES

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Conflict of Interests

The authors declare no interest conflict.

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REFERENCES

- 1 Xiong, Z. H.; Wu, D.; Vardeny, Z. V.; Shi, J. Giant magnetoresistance in organic spin-valves. *Nature* **2004**, *427*, 821–824.
- 2 Xie, Z.; Liu, D.; Gao, C.; Zhang, X.; Dong, H.; Hu, W. High mobility emissive organic semiconductors for optoelectronic devices. *J. Am. Chem. Soc.* **2025**, *147*, 2239–2256.
- 3 Meng, K.; Li, M.; Guo, L.; Zhang, R.; Guo, A.; Liu, M.; Gu, X.; Qin, Y.; Yang, T.; Yang, X.; Hu, S.; Zhang, C.; Zheng, R.; Wu, M.; Sun, X. Room-temperature organic spintronic devices with wide range magnetocurrent tuning and multifunctionality via electro-optical compensation strategy. *Adv. Mater.* **2025**, *37*, 2417995.
- 4 Fu, D.; Liu, J.; Hou, F.; Chang, X.; Qu, T.; Félisaz, J.; Krishnaswamy, G. K.; Grebenchuk, S.; Jie, Y.; Watanabe, K.; Taniguchi, T.; Pereira, V. M.; Novoselov, K. S.; Koperski, M.; Yakovlev, N. L.; Soumyanarayanan, A.; Avsar, A.; Yazyev, O. V.; Lin, J.; Özyilmaz, B. Electric field-tunable ferromagnetism in a van der Waals semiconductor up to room temperature. *Nat. Commun.* **2025**, *16*, 10197.
- 5 Cornia, A.; Seneor, P. Spintronics: the molecular way. *Nat. Mater.* **2017**, *16*, 505–506.
- 6 Vardeny, V. Organics strike back. *Nat. Mater.* **2009**, *8*, 91–93.
- 7 Dediu, V. A.; Hueso, L. E.; Bergenti, I.; Taliani, C. Spin routes in organic semiconductors. *Nat. Mater.* **2009**, *8*, 707–716.

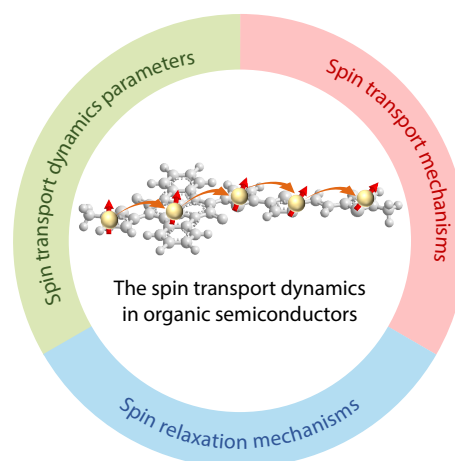
Graphical Abstract

Recent Progress in Spin Transport Dynamics in Organic Semiconductors

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This review elucidates spin transport dynamics in organic semiconductors by integrating key parameters, relaxation, and transport mechanisms to guide the molecular design of high-performance spintronic materials.



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- 8 Galbiati, M.; Barraud, C.; Tatay, S.; Bouzehouane, K.; Deranlot, C.; Jacquet, E.; Fert, A.; Seneor, P.; Mattana, R.; Petroff, F. Unveiling self-assembled monolayers' potential for molecular spintronics: spin transport at high voltage. *Adv. Mater.* **2012**, *24*, 6429–6432.
- 9 Hu, S.; Liu, W.; Guo, L.; Zhang, R.; Gu, X.; Meng, K.; Qin, Y.; Guo, A.; Yang, T.; Zhang, C.; Yang, X.; Lu, S.; Wu, M.; Lu, K.; Tan, T.; Zhou, E.; Wei, Z.; Sun, X. Continuous room-temperature spin-injection modulation achieved by spin-filtering competition in molecular spin valves. *Adv. Mater.* **2023**, *35*, 2300055.
- 10 Rybicki, J.; Lin, R.; Wang, F.; Wohlgenannt, M.; He, C.; Sanders, T.; Suzuki, Y. Tuning the performance of organic spintronic devices using X-ray generated traps. *Phys. Rev. Lett.* **2012**, *109*, 076603.
- 11 Qin, Y.; Wu, M.; Xiang, J.; Yang, T.; Guo, L.; Gu, X.; Zhang, R.; Zhang, X.; Meng, K.; Hu, S.; Zheng, R.; Li, M.; Wang, Y.; Zou, Y.; Zhang, J.; Gao, X.; Sun, X. Enhancing room-temperature spin lifetimes in molecular semiconductors by designing intramolecular dipole orientations. *Adv. Mater.* **2025**, *37*, 2500521.
- 12 Yang, T.; Qin, Y.; Wu, M.; Guo, L.; Gu, X.; Meng, K.; Hu, S.; Zhang, C.; Zheng, R.; Zhang, R.; Sun, X. Structural isomeric effect on spin transport in molecular semiconductors. *Adv. Mater.* **2024**, *36*, 2402001.
- 13 Hu, S.; Qin, Y.; Lu, S.; Guo, L.; Gu, X.; Yang, T.; Zhang, R.; Meng, K.; Zhang, C.; Wu, M.; Sun, X. Interface engineering for enhancing air-stable spin-charge interaction in molecular spin-photovoltaic devices. *Adv. Funct. Mater.* **2024**, *34*, 2315239.
- 14 Yang, W.; Shi, Q.; Miao, T.; Li, Q.; Cai, P.; Liu, H.; Lin, H.; Bai, Y.; Zhu, Y.; Yu, Y.; Deng, L.; Wang, W.; Yin, L.; Sun, D.; Zhang, X.-G.; Shen, J. Achieving large and nonvolatile tunable magnetoresistance in organic spin valves using electronic phase separated manganites. *Nat. Commun.* **2019**, *10*, 3877.
- 15 Xu, J.; Jarocha, L. E.; Zollitsch, T.; Konowalczyk, M.; Henbest, K. B.; Richert, S.; Golesworthy, M. J.; Schmidt, J.; Déjean, V.; Sowood, D. J. C.; Bassetto, M.; Luo, J.; Walton, J. R.; Fleming, J.; Wei, Y.; Pitcher, T. L.; Moise, G.; Herrmann, M.; Yin, H.; Wu, H.; Bartölke, R.; Käsehausen, S. J.; Horst, S.; Dautaj, G.; Murton, P. D. F.; Gehrckens, A. S.; Chelliah, Y.; Takahashi, J. S.; Koch, K.-W.; Weber, S.; Solov'yov, I. A.; Xie, C.; MacKenzie, S. R.; Timmel, C. R.; Mouritsen, H.; Hore, P. J. Magnetic sensitivity of cryptochrome 4 from a migratory songbird. *Nature* **2021**, *594*, 535–540.
- 16 Geng, R.; Mena, A.; Pappas, W. J.; McCamey, D. R. Sub-micron spin-based magnetic field imaging with an organic light emitting diode. *Nat. Commun.* **2023**, *14*, 1441.
- 17 Devkota, J.; Geng, R.; Subedi, R. C.; Nguyen, T. D. Organic spin valves: a review. *Adv. Funct. Mater.* **2016**, *26*, 3881–3898.
- 18 Sun, X.; Vélez, S.; Atxabal, A.; Bedoya-Pinto, A.; Parui, S.; Zhu, X.; Llopis, R.; Casanova, F.; Hueso, L. E. A molecular spin-photovoltaic device. *Science* **2017**, *357*, 677–680.
- 19 Sun, X.; Gobbi, M.; Bedoya-Pinto, A.; Txoperena, O.; Golmar, F.; Llopis, R.; Chuvilin, A.; Casanova, F.; Hueso, L. E. Room-temperature air-stable spin transport in bathocuproine-based spin valves. *Nat. Commun.* **2013**, *4*, 2794.
- 20 Groesbeck, M.; Liu, H.; Kavand, M.; Lafalce, E.; Wang, J.; Pan, X.; Tannahwa, T. H.; Popli, H.; Malissa, H.; Boehme, C.; Vardeny, Z. V. Separation of spin and charge transport in pristine π -conjugated polymers. *Phys. Rev. Lett.* **2020**, *124*, 067702.
- 21 Shah, B.; Anderson, S. W.; Scalera, J.; Jara, H.; Soto, J. A. Quantitative MR imaging: physical principles and sequence design in abdominal imaging. *Radiographics* **2011**, *31*, 867–880.
- 22 Awojoyogbe, O. B.; Dada, O. M.; Faromika, O. P.; Dada, O. E. Mathematical concept of the Bloch flow equations for general magnetic resonance imaging: a review. *Concepts Magn. Reson. Part A* **2011**, *38A*, 85–101.
- 23 Bhalekar, S.; Daftardar-Gejji, V.; Baleanu, D.; Magin, R. Fractional Bloch equation with delay. *Comput. Math. Appl.* **2011**, *61*, 1355–1365.
- 24 Jiang, S. W.; Wang, P.; Chen, B. B.; Zhou, Y.; Ding, H. F.; Wu, D. Tuning carrier mobility without spin transport degrading in copper-phthalocyanine. *Appl. Phys. Lett.* **2015**, *107*, 042407.
- 25 Guo, L.; Gu, X.; Hu, S.; Sun, W.; Zhang, R.; Qin, Y.; Meng, K.; Lu, X.; Liu, Y.; Wang, J.; Ma, P.; Zhang, C.; Guo, A.; Yang, T.; Yang, X.; Wang, G.; Liu, Y.; Wang, K.; Mi, W.; Zhang, C.; Jiang, L.; Liu, L.; Zheng, K.; Qin, W.; Yan, W.; Sun, X. Strain-restricted transfer of

- ferromagnetic electrodes for constructing reproducibly superior-quality spintronic devices. *Nat. Commun.* **2024**, *15*, 865.
- 26 Julliere, M. Tunneling between ferromagnetic films. *Phys. Lett. A* **1975**, *54*, 225–226.
 - 27 Sarkar, S.; Oh, S.; Newton, P. J.; Li, Y.; Zhu, Y.; Ghani, M. A.; Yan, H.; Jeong, H. Y.; Wang, Y.; Chhowalla, M. Spin injection in graphene using ferromagnetic van der Waals contacts of indium and cobalt. *Nat. Electron.* **2025**, *8*, 215–221.
 - 28 Liu, H.; Zhang, C.; Malissa, H.; Groesbeck, M.; Kavand, M.; McLaughlin, R.; Jamali, S.; Hao, J.; Sun, D.; Davidson, R. A.; Wojcik, L.; Miller, J. S.; Boehme, C.; Vardeny, Z. V. Organic-based magnon spintronics. *Nat. Mater.* **2018**, *17*, 308–312.
 - 29 Atodiresi, N.; Brede, J.; Lazić, P.; Caciuc, V.; Hoffmann, G.; Wiesendanger, R.; Blügel, S. Design of the local spin polarization at the organic-ferromagnetic interface. *Phys. Rev. Lett.* **2010**, *105*, 066601.
 - 30 Geng, R.; Subedi, R. C.; Luong, H. M.; Pham, M. T.; Huang, W.; Li, X.; Hong, K.; Shao, M.; Xiao, K.; Hornak, L. A.; Nguyen, T. D. Effect of charge localization on the effective hyperfine interaction in organic semiconducting polymers. *Phys. Rev. Lett.* **2018**, *120*, 086602.
 - 31 Sun, D.; Fang, M.; Xu, X.; Jiang, L.; Guo, H.; Wang, Y.; Yang, W.; Yin, L.; Snijders, P. C.; Ward, T. Z.; Gai, Z.; Zhang, X.-G.; Lee, H. N.; Shen, J. Active control of magnetoresistance of organic spin valves using ferroelectricity. *Nat. Commun.* **2014**, *5*, 4396.
 - 32 Drew, A. J.; Hoppler, J.; Schulz, L.; Pratt, F. L.; Desai, P.; Shaky, P.; Kreouzis, T.; Gillin, W. P.; Suter, A.; Morley, N. A.; Malik, V. K.; Dubroka, A.; Kim, K. W.; Bouyanfif, H.; Bourqui, F.; Bernhard, C.; Scheuermann, R.; Nieuwenhuys, G. J.; Prokscha, T.; Morenzoni, E. Direct measurement of the electronic spin diffusion length in a fully functional organic spin valve by low-energy muon spin rotation. *Nat. Mater.* **2009**, *8*, 109–114.
 - 33 Ding, S.; Tian, Y.; Wang, H.; Zhou, Z.; Mi, W.; Ni, Z.; Zou, Y.; Dong, H.; Gao, H.; Zhu, D.; Hu, W. Reliable spin valves of conjugated polymer based on mechanically transferrable top electrodes. *ACS Nano* **2018**, *12*, 12657–12664.
 - 34 Jedema, F. J.; Filip, A. T.; van Wees, B. J. Electrical spin injection and accumulation at room temperature in an all-metal mesoscopic spin valve. *Nature* **2001**, *410*, 345–348.
 - 35 Yu, Z. G.; Baker, J.; Krishnamurthy, S. Transfer lengths and spin injection from a three-dimensional ferromagnet into graphene. *Phys. Rev. B* **2010**, *82*, 035425.
 - 36 Yang, T.; Kimura, T.; Otani, Y. Giant spin-accumulation signal and pure spin-current-induced reversible magnetization switching. *Nat. Phys.* **2008**, *4*, 851–854.
 - 37 Rashba, E. I. Theory of electrical spin injection: tunnel contacts as a solution of the conductivity mismatch problem. *Phys. Rev. B* **2000**, *62*, R16267–R16270.
 - 38 Schmidt, G.; Ferrand, D.; Molenkamp, L.; Filip, A.; van Wees, B. Fundamental obstacle for electrical spin injection from a ferromagnetic metal into a diffusive semiconductor. *Phys. Rev. B Condens. Matter Mater. Phys.* **2000**, *62*, R4790–R4793.
 - 39 Ando, K.; Watanabe, S.; Mooser, S.; Saitoh, E.; Sirringhaus, H. Solution-processed organic spin-charge converter. *Nat. Mater.* **2013**, *12*, 622–627.
 - 40 Kajiwara, Y.; Harii, K.; Takahashi, S.; Ohe, J.; Uchida, K.; Mizuguchi, M.; Umezawa, H.; Kawai, H.; Ando, K.; Takanashi, K.; Maekawa, S.; Saitoh, E. Transmission of electrical signals by spin-wave interconversion in a magnetic insulator. *Nature* **2010**, *464*, 262–266.
 - 41 Ando, K.; Takahashi, S.; Ieda, J.; Kurebayashi, H.; Trypiniotis, T.; Barnes, C. H. W.; Maekawa, S.; Saitoh, E. Electrically tunable spin injector free from the impedance mismatch problem. *Nat. Mater.* **2011**, *10*, 655–659.
 - 42 Tran, T. L. A.; Le, T. Q.; Sanderink, J. G. M.; van der Wiel, W. G.; de Jong, M. P. The multistep tunneling analogue of conductivity mismatch in organic spin valves. *Adv. Funct. Mater.* **2012**, *22*, 1180–1189.
 - 43 Kimura, T.; Otani, Y.; Sato, T.; Takahashi, S.; Maekawa, S. Room-temperature reversible spin Hall effect. *Phys. Rev. Lett.* **2007**, *98*, 156601.
 - 44 Saitoh, E.; Ueda, M.; Miyajima, H.; Tataru, G. Conversion of spin current into charge current at room temperature: inverse spin-Hall effect. *Appl. Phys. Lett.* **2006**, *88*, 182509.
 - 45 Wittmann, A.; Schweicher, G.; Broch, K.; Novak, J.; Lami, V.; Cornil, D.; McNellis, E. R.; Zadvorna, O.; Venkateshvaran, D.; Takimiya, K.; Geerts, Y. H.; Cornil, J.; Vaynzof, Y.; Sinova, J.; Watanabe, S.; Sirringhaus, H. Tuning spin current injection at ferromagnet-nonmagnet interfaces by molecular design. *Phys. Rev. Lett.* **2020**, *124*, 027204.
 - 46 Tserkovnyak, Y.; Brataas, A.; Bauer, G. E. W. Spin pumping and magnetization dynamics in metallic multilayers. *Phys. Rev. B* **2002**, *66*, 224403.
 - 47 Ando, K.; Takahashi, S.; Ieda, J.; Kajiwara, Y.; Nakayama, H.; Yoshino, T.; Harii, K.; Fujikawa, Y.; Matsuo, M.; Maekawa, S.; Saitoh, E. Inverse spin-Hall effect induced by spin pumping in metallic system. *J. Appl. Phys.* **2011**, *109*, 103913.
 - 48 Jiao, H.; Bauer, G. E. W. Spin backflow and AC voltage generation by spin pumping and the inverse spin Hall effect. *Phys. Rev. Lett.* **2013**, *110*, 217602.
 - 49 Sun, D.; van Schooten, K. J.; Kavand, M.; Malissa, H.; Zhang, C.; Groesbeck, M.; Boehme, C.; Vardeny, Z. V. Inverse spin Hall effect from pulsed spin current in organic semiconductors with tunable spin-orbit coupling. *Nat. Mater.* **2016**, *15*, 863–869.
 - 50 Li, T.; Xu, L.; Xiao, X.; Chen, F.; Cao, L.; Wu, W.; Tong, W.; Zhang, F. Enhanced spin transport of conjugated polymer in the semiconductor/insulating polymer blend. *ACS Appl. Mater. Interfaces* **2020**, *12*, 2708–2716.
 - 51 Wang, S.-J.; Venkateshvaran, D.; Mahani, M. R.; Chopra, U.; McNellis, E. R.; Di Pietro, R.; Schott, S.; Wittmann, A.; Schweicher, G.; Cubukcu, M.; Kang, K.; Carey, R.; Wagner, T. J.; Siebrecht, J. N. M.; Wong, D. P. G. H.; Jacobs, I. E.; Aboljadayel, R. O.; Ionescu, A.; Egorov, S. A.; Mueller, S.; Zadvorna, O.; Skalski, P.; Jellett, C.; Little, M.; Marks, A.; McCulloch, I.; Wunderlich, J.; Sinova, J.; Sirringhaus, H. Long spin diffusion lengths in doped conjugated polymers due to enhanced exchange coupling. *Nat. Electron.* **2019**, *2*, 98–107.
 - 52 Uchida, K. I.; Adachi, H.; Kikkawa, T.; Kirihaara, A.; Ishida, M.; Yoroza, S.; Maekawa, S.; Saitoh, E. Thermoelectric generation based on spin seebeck effects. *Proc. IEEE* **2016**, *104*, 1946–1973.
 - 53 Slachter, A.; Bakker, F. L.; Adam, J. P.; van Wees, B. J. Thermally driven spin injection from a ferromagnet into a non-magnetic metal. *Nat. Phys.* **2010**, *6*, 879–882.
 - 54 Wang, P.; Zhou, L. F.; Jiang, S. W.; Luan, Z. Z.; Shu, D. J.; Ding, H. F.; Wu, D. Unidirectional spin-wave-propagation-induced seebeck voltage in a PEDOT:PSS/YIG bilayer. *Phys. Rev. Lett.* **2018**, *120*, 047201.
 - 55 Mosendz, O.; Pearson, J. E.; Fradin, F. Y.; Bauer, G. E. W.; Bader, S. D.; Hoffmann, A. Quantifying spin hall angles from spin pumping: experiments and theory. *Phys. Rev. Lett.* **2010**, *104*, 046601.
 - 56 Wang, P.; Jiang, S. W.; Luan, Z. Z.; Zhou, L. F.; Ding, H. F.; Zhou, Y.; Tao, X. D.; Wu, D. Spin rectification induced by spin Hall magnetoresistance at room temperature. *Appl. Phys. Lett.* **2016**, *109*, 112406.
 - 57 Miao, B. F.; Huang, S. Y.; Qu, D.; Chien, C. L. Inverse spin Hall effect in a ferromagnetic metal. *Phys. Rev. Lett.* **2013**, *111*, 066602.
 - 58 Jiang, S. W.; Liu, S.; Wang, P.; Luan, Z. Z.; Tao, X. D.; Ding, H. F.;

- Wu, D. Exchange-dominated pure spin current transport in Alq₃ molecules. *Phys. Rev. Lett.* **2015**, *115*, 086601.
- 59 Zhang, C.; Guo, L.; Cong, C.; Zhou, Q.; Sun, X. Organic spintronic materials: recent progress and emerging multifunctionalities. *CCS Chem.* **2026**, *8*, 716–735.
 - 60 Shikoh, E.; Ando, K.; Kubo, K.; Saitoh, E.; Shinjo, T.; Shiraishi, M. Spin-pump-induced spin transport in p-type Si at room temperature. *Phys. Rev. Lett.* **2013**, *110*, 127201.
 - 61 Pramanik, S.; Stefanita, C.-G.; Patibandla, S.; Bandyopadhyay, S.; Garre, K.; Harth, N.; Cahay, M. Observation of extremely long spin relaxation times in an organic nanowire spin valve. *Nat. Nanotechnol.* **2007**, *2*, 216–219.
 - 62 Tsurumi, J.; Matsui, H.; Kubo, T.; Häusermann, R.; Mitsui, C.; Okamoto, T.; Watanabe, S.; Takeya, J. Coexistence of ultra-long spin relaxation time and coherent charge transport in organic single-crystal semiconductors. *Nat. Phys.* **2017**, *13*, 994–998.
 - 63 Schott, S.; Chopra, U.; Lemaury, V.; Melnyk, A.; Olivier, Y.; Di Pietro, R.; Romanov, I.; Carey, R. L.; Jiao, X.; Jellett, C.; Little, M.; Marks, A.; McNeill, C. R.; McCulloch, I.; McNellis, E. R.; Andrienko, D.; Beljonne, D.; Sinova, J.; Sirringhaus, H. Polaron spin dynamics in high-mobility polymeric semiconductors. *Nat. Phys.* **2019**, *15*, 814–822.
 - 64 Watanabe, S.; Ando, K.; Kang, K.; Mooser, S.; Vaynzof, Y.; Kurebayashi, H.; Saitoh, E.; Sirringhaus, H. Polaron spin current transport in organic semiconductors. *Nat. Phys.* **2014**, *10*, 308–313.
 - 65 Mooser, S.; Cooper, J. F. K.; Banger, K. K.; Wunderlich, J.; Sirringhaus, H. Spin injection and transport in a solution-processed organic semiconductor at room temperature. *Phys. Rev. B* **2012**, *85*, 235202.
 - 66 Giannini, S.; Carof, A.; Blumberger, J. Crossover from hopping to band-like charge transport in an organic semiconductor model: atomistic nonadiabatic molecular dynamics simulation. *J. Phys. Chem. Lett.* **2018**, *9*, 3116–3123.
 - 67 Nguyen, T. D.; Basel, T. P.; Pu, Y. J.; Li, X. G.; Ehrenfreund, E.; Vardeny, Z. V. Isotope effect in the spin response of aluminum tris(8-hydroxyquinoline) based devices. *Phys. Rev. B* **2012**, *85*, 245437.
 - 68 Wang, G.; Liu, B. L.; Balocchi, A.; Renucci, P.; Zhu, C. R.; Amand, T.; Fontaine, C.; Marie, X. Gate control of the electron spin-diffusion length in semiconductor quantum wells. *Nat. Commun.* **2013**, *4*, 2372.
 - 69 Meng, K.; Guo, L.; Sun, X. Strategies and applications of generating spin polarization in organic semiconductors. *Nanoscale Horiz.* **2023**, *8*, 1132–1154.
 - 70 Li, L.; Lu, N.; Liu, M.; Bässler, H. General Einstein relation model in disordered organic semiconductors under quasiequilibrium. *Phys. Rev. B* **2014**, *90*, 214107.
 - 71 Wetzelaer, G. A. H.; Koster, L. J. A.; Blom, P. W. M. Validity of the Einstein relation in disordered organic semiconductors. *Phys. Rev. Lett.* **2011**, *107*, 066605.
 - 72 Hanle, W. Über magnetische beeinflussung der polarisation der resonanzfluoreszenz. *Z. Für Phys.* **1924**, *30*, 93–105.
 - 73 Parsons, R. R. Band-to-band optical pumping in solids and polarized photoluminescence. *Phys. Rev. Lett.* **1969**, *23*, 1152–1154.
 - 74 Johnson, M.; Silsbee, R. Interfacial charge-spin coupling: injection and detection of spin magnetization in metals. *Phys. Rev. Lett.* **1985**, *55*, 1790–1793.
 - 75 Johnson, M.; Silsbee, R. H. Spin-injection experiment. *Phys. Rev. B* **1988**, *37*, 5326–5335.
 - 76 Johnson, M.; Silsbee, R. Coupling of electronic charge and spin at a ferromagnetic-paramagnetic metal interface. *Phys. Rev. B* **1988**, *37*, 5312–5325.
 - 77 Lou, X.; Adelmann, C.; Furis, M.; Crooker, S. A.; Palmström, C. J.; Crowell, P. A. Electrical detection of spin accumulation at a ferromagnet-semiconductor interface. *Phys. Rev. Lett.* **2006**, *96*, 176603.
 - 78 Lou, X.; Adelmann, C.; Crooker, S. A.; Garlid, E. S.; Zhang, J.; Madhukar Reddy, K. S.; Flexner, S. D.; Palmström, C. J.; Crowell, P. A. Electrical detection of spin transport in lateral ferromagnet–semiconductor devices. *Nat. Phys.* **2007**, *3*, 197–202.
 - 79 Jedema, F. J.; Heersche, H. B.; Filip, A. T.; Baselmans, J. J. A.; van Wees, B. J. Electrical detection of spin precession in a metallic mesoscopic spin valve. *Nature* **2002**, *416*, 713–716.
 - 80 Tombros, N.; Jozsa, C.; Popinciuc, M.; Jonkman, H. T.; van Wees, B. J. Electronic spin transport and spin precession in single graphene layers at room temperature. *Nature* **2007**, *448*, 571–574.
 - 81 Dash, S. P.; Sharma, S.; Patel, R. S.; de Jong, M. P.; Jansen, R. Electrical creation of spin polarization in silicon at room temperature. *Nature* **2009**, *462*, 491–494.
 - 82 Han, W.; Jiang, X.; Kajdos, A.; Yang, S.-H.; Stemmer, S.; Parkin, S. P. Spin injection and detection in lanthanum- and niobium-doped SrTiO₃ using the Hanle technique. *Nat. Commun.* **2013**, *4*, 2134.
 - 83 Appelbaum, I. Introduction to spin-polarized ballistic hot electron injection and detection in silicon. *Phil. Trans. R. Soc. A.* **2011**, *369*, 3554–3574.
 - 84 Huang, B.; Monsma, D. J.; Appelbaum, I. Coherent spin transport through a 350 micron thick silicon wafer. *Phys. Rev. Lett.* **2007**, *99*, 177209.
 - 85 Appelbaum, I.; Huang, B.; Monsma, D. J. Electronic measurement and control of spin transport in silicon. *Nature* **2007**, *447*, 295–298.
 - 86 Grünewald, M.; Göckeritz, R.; Homonnay, N.; Würthner, F.; Molenkamp, L. W.; Schmidt, G. Vertical organic spin valves in perpendicular magnetic fields. *Phys. Rev. B* **2013**, *88*, 085319.
 - 87 Huang, B.; Jang, H. J.; Appelbaum, I. Geometric dephasing-limited Hanle effect in long-distance lateral silicon spin transport devices. *Appl. Phys. Lett.* **2008**, *93*, 162508.
 - 88 Riminucci, A.; Yu, Z. G.; Prezioso, M.; Cecchini, R.; Bergenti, I.; Graziosi, P.; Dediu, V. A. Controlling magnetoresistance by oxygen impurities in Mq₃-based molecular spin valves. *ACS Appl. Mater. Interfaces* **2019**, *11*, 8319–8326.
 - 89 Yu, Z. G. Suppression of the Hanle effect in organic spintronic devices. *Phys. Rev. Lett.* **2013**, *111*, 016601.
 - 90 Carey, R. L.; Giannini, S.; Schott, S.; Lemaury, V.; Xiao, M.; Prodhon, S.; Wang, L.; Bovoloni, M.; Quarti, C.; Beljonne, D.; Sirringhaus, H. Spin relaxation of electron and hole polarons in ambipolar conjugated polymers. *Nat. Commun.* **2024**, *15*, 288.
 - 91 Li, D.; Yu, G. Innovation of materials, devices, and functionalized interfaces in organic spintronics. *Adv. Funct. Mater.* **2021**, *31*, 2100550.
 - 92 Yang, T.; Qin, Y.; Wu, M.; Gu, X.; Meng, K.; Hu, S.; Zhang, C.; Guo, A.; Zheng, R.; Zhang, R.; Guo, L.; Sun, X. Spin-lifetime probe for detecting intramolecular noncovalent interaction in organic semiconductors. *Adv. Mater.* **2024**, *36*, 2410695.
 - 93 Marumoto, K.; Kuroda, S.-I.; Takenobu, T.; Iwasa, Y. Spatial extent of wave functions of gate-induced hole carriers in pentacene field-effect devices as investigated by electron spin resonance. *Phys. Rev. Lett.* **2006**, *97*, 256603.
 - 94 Matsui, H.; Hasegawa, T.; Tokura, Y.; Hiraoka, M.; Yamada, T. Polaron motional narrowing of electron spin resonance in organic field-effect transistors. *Phys. Rev. Lett.* **2008**, *100*, 126601.
 - 95 Schott, S.; McNellis, E. R.; Nielsen, C. B.; Chen, H.-Y.; Watanabe, S.; Tanaka, H.; McCulloch, I.; Takimiya, K.; Sinova, J.; Sirringhaus, H. Tuning the effective spin-orbit coupling in molecular

- semiconductors. *Nat. Commun.* **2017**, *8*, 15200.
- 96 Harmon, N. J.; Flatté, M. E. Distinguishing spin relaxation mechanisms in organic semiconductors. *Phys. Rev. Lett.* **2013**, *110*, 176602.
 - 97 Bandyopadhyay, S. Dominant spin relaxation mechanism in compound organic semiconductors. *Phys. Rev. B* **2010**, *81*, 153202.
 - 98 Harmon, N. J.; Flatté, M. E. Spin relaxation in materials lacking coherent charge transport. *Phys. Rev. B* **2014**, *90*, 115203.
 - 99 Sheng, Y.; Nguyen, T. D.; Veeraraghavan, G.; Mermer, Wohlgemant, M.; Qiu, S.; Scherf, U. Hyperfine interaction and magnetoresistance in organic semiconductors. *Phys. Rev. B* **2006**, *74*, 045213.
 - 100 Nuccio, L.; Willis, M.; Schulz, L.; Fratini, S.; Messina, F.; D'Amico, M.; Pratt, F. L.; Lord, J. S.; McKenzie, I.; Loth, M.; Purushothaman, B.; Anthony, J.; Heeney, M.; Wilson, R. M.; Hernández, I.; Cannas, M.; Sedlak, K.; Kreouzis, T.; Gillin, W. P.; Bernhard, C.; Drew, A. J. Importance of spin-orbit interaction for the electron spin relaxation in organic semiconductors. *Phys. Rev. Lett.* **2013**, *110*, 216602.
 - 101 Chen, B. B.; Wang, S.; Jiang, S. W.; Yu, Z. G.; Wan, X. G.; Ding, H. F.; Wu, D. The role of heavy metal ions on spin transport in organic semiconductors. *New J. Phys.* **2015**, *17*, 013004.
 - 102 Zhang, Y.; Bai, H.; Han, L.; Chen, C.; Zhou, Y.; Back, C. H.; Pan, F.; Wang, Y.; Song, C. Simultaneous high charge-spin conversion efficiency and large spin diffusion length in altermagnetic RuO₂. *Adv. Funct. Mater.* **2024**, *34*, 2313332.
 - 103 Wang, H. L.; Du, C. H.; Pu, Y.; Adur, R.; Hammel, P. C.; Yang, F. Y. Scaling of spin hall angle in 3d, 4d, and 5d metals from Y₃Fe₅O₁₂/metal spin pumping. *Phys. Rev. Lett.* **2014**, *112*, 197201.
 - 104 Sun, R.; Park, K. S.; Comstock, A. H.; McConnell, A.; Chen, Y.-C.; Zhang, P.; Beratan, D.; You, W.; Hoffmann, A.; Yu, Z.-G.; Diao, Y.; Sun, D. Inverse chirality-induced spin selectivity effect in chiral assemblies of π -conjugated polymers. *Nat. Mater.* **2024**, *23*, 782–789.
 - 105 Lee, J. W.; Oh, Y. W.; Park, S. Y.; Figueroa, A. I.; van der Laan, G.; Go, G.; Lee, K. J.; Park, B. G. Enhanced spin-orbit torque by engineering Pt resistivity in Pt/Co/AlO_x structures. *Phys. Rev. B* **2017**, *96*, 064405.
 - 106 Paul, B.; Ghosh, T. K. Understanding spin Hall effect in two-dimensional Fermionic systems with generic spin-orbit interaction in III-V heterostructures. *Phys. Lett. A* **2015**, *379*, 728–731.
 - 107 Neese, F. Prediction of electron paramagnetic resonance *g* values using coupled perturbed Hartree-Fock and Kohn-Sham theory. *J. Chem. Phys.* **2001**, *115*, 11080–11096.
 - 108 Yang, X.; Guo, A.; Yang, J.; Chen, J.; Meng, K.; Hu, S.; Duan, R.; Zhu, M.; Shi, W.; Qin, Y.; Zhang, R.; Yang, H.; Li, J.; Guo, L.; Sun, X.; Liu, Y.; Guo, Y. Halogenated-edge polymeric semiconductor for efficient spin transport. *Nat. Commun.* **2024**, *15*, 8368.
 - 109 Yu, Z. G. Spin-orbit coupling and its effects in organic solids. *Phys. Rev. B* **2012**, *85*, 115201.
 - 110 Yu, Z. G. Spin-orbit coupling, spin relaxation, and spin diffusion in organic solids. *Phys. Rev. Lett.* **2011**, *106*, 106602.
 - 111 Dediú, V.; Murgia, M.; Maticotta, F. C.; Taliani, C.; Barbanera, S. Room temperature spin polarized injection in organic semiconductor. *Solid State Commun.* **2002**, *122*, 181–184.
 - 112 Liang, S.; Geng, R.; Yang, B.; Zhao, W.; Subedi, R. C.; Li, X.; Han, X.; Nguyen, T. D. Curvature-enhanced spin-orbit coupling and spinterface effect in fullerene-based spin valves. *Sci. Rep.* **2016**, *6*, 19461.
 - 113 Nguyen, T. D.; Wang, F.; Li, X.-G.; Ehrenfreund, E.; Vardeny, Z. V. Spin diffusion in fullerene-based devices: morphology effect. *Phys. Rev. B* **2013**, *87*, 075205.
 - 114 Wilson, J. S.; Dhoot, A. S.; Seeley, A. J. A. B.; Khan, M. S.; Köhler, A.; Friend, R. H. Spin-dependent exciton formation in π -conjugated compounds. *Nature* **2001**, *413*, 828–831.
 - 115 Sheng, C. X.; Singh, S.; Gambetta, A.; Drori, T.; Tong, M.; Tretiak, S.; Vardeny, Z. V. Ultrafast intersystem-crossing in platinum containing π -conjugated polymers with tunable spin-orbit coupling. *Sci. Rep.* **2013**, *3*, 2653.
 - 116 Yu, Z. G.; Ding, F.; Wang, H. Hyperfine interaction and its effects on spin dynamics in organic solids. *Phys. Rev. B* **2013**, *87*, 205446.
 - 117 Dyakonov, V.; Rösler, G.; Schwoerer, M.; Frankevich, E. L. Evidence for triplet interchain polaron pairs and their transformations in polyphenylenevinylene. *Phys. Rev. B* **1997**, *56*, 3852–3862.
 - 118 Ding, S.; Tian, Y.; Li, Y.; Zhang, H.; Zhou, K.; Liu, J.; Qin, L.; Zhang, X.; Qiu, X.; Dong, H.; Zhu, D.; Hu, W. Organic single-crystal spintronics: magnetoresistance devices with high magnetic-field sensitivity. *ACS Nano* **2019**, *13*, 9491–9497.
 - 119 Nguyen, T. D.; Hukic-Markosian, G.; Wang, F.; Wojcik, L.; Li, X.-G.; Ehrenfreund, E.; Vardeny, Z. V. Isotope effect in spin response of π -conjugated polymer films and devices. *Nat. Mater.* **2010**, *9*, 345–352.
 - 120 McCamey, D. R.; van Schooten, K. J.; Baker, W. J.; Lee, S.-Y.; Paik, S.-Y.; Lupton, J. M.; Boehme, C. Hyperfine-field-mediated spin beating in electrostatically bound charge carrier pairs. *Phys. Rev. Lett.* **2010**, *104*, 017601.
 - 121 Lunghi, A.; Sanvito, S. How do phonons relax molecular spins. *Sci. Adv.* **2019**, *5*, eaax7163.
 - 122 Teferi, M. Y.; Ogle, J.; Joshi, G.; Malissa, H.; Jamali, S.; Baird, D. L.; Lupton, J. M.; Brooks, L. W.; Boehme, C. Tuning effective hyperfine fields in PEDOT: PSS thin films by doping. *Phys. Rev. B* **2018**, *98*, 241201.
 - 123 Carey, R. L.; Ren, X.; Jacobs, I. E.; Elsner, J.; Schott, S.; Goldberg, E.; Wang, Z.; Blumberger, J.; Sirttinghaus, H. Long spin lifetimes of charge carriers in rubrene crystals due to fast transient-localization motion. *Nat. Commun.* **2025**, *16*, 7605.
 - 124 Elliott, R. J. Theory of the effect of spin-orbit coupling on magnetic resonance in some semiconductors. *Phys. Rev.* **1954**, *96*, 266–279.
 - 125 Yafet, Y. Calculation of the *g* factor of metallic sodium. *Phys. Rev.* **1952**, *85*, 478.
 - 126 Zheng, Y.; Wudl, F. Organic spin transporting materials: present and future. *J. Mater. Chem. A* **2014**, *2*, 48–57.
 - 127 Wang, Y.; Yao, J.; Ding, S.; Guo, S.; Cui, D.; Wang, X.; Yang, S.; Zhang, L.; Tian, X.; Wu, D.; Jin, C.; Li, R.; Hu, W. Spin injection and transport in single-crystalline organic spin valves based on TIPS-pentacene. *Sci. China Mater.* **2021**, *64*, 2795–2804.
 - 128 Tong, J.; Ruan, L.; Yao, X.; Qin, G.; Zhang, X. Defect states dependence of spin transport in iron phthalocyanine spin valves. *Phys. Rev. B* **2019**, *99*, 054406.
 - 129 Yang, X.; Guo, A.; Guo, L.; Liu, Y.; Sun, X.; Guo, Y. Organic semiconductors for room-temperature spin valves. *ACS Mater. Lett.* **2022**, *4*, 805–814.
 - 130 Gu, R. Y.; Xing, D. Y.; Dong, J. Spin-polarized tunneling between ferromagnetic films. *J. Appl. Phys.* **1996**, *80*, 7163–7165.
 - 131 Grünewald, M.; Wahler, M.; Schumann, F.; Michelfeit, M.; Gould, C.; Schmidt, R.; Würthner, F.; Schmidt, G.; Molenkamp, L. W. Tunneling anisotropic magnetoresistance in organic spin valves. *Phys. Rev. B* **2011**, *84*, 125208.
 - 132 Gobbi, M.; Golmar, F.; Llopis, R.; Casanova, F.; Hueso, L. E. Room-temperature spin transport in C₆₀-based spin valves. *Adv. Mater.* **2011**, *23*, 1609–1613.
 - 133 Butler, W. H.; Zhang, X. G.; Schulthess, T. C.; MacLaren, J. M. Spin-dependent tunneling conductance of Fe|MgO|Fe sandwiches. *Phys. Rev. B* **2001**, *63*, 054416.
 - 134 Xu, W.; Szulcowski, G. J.; LeClair, P.; Navarrete, I.; Schad, R.;

- Miao, G.; Guo, H.; Gupta, A. Tunneling magnetoresistance observed in $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ /organic molecule/Co junctions. *Appl. Phys. Lett.* **2007**, *90*, 072506.
- 135 Lu, Q.; Xie, S.; Qu, F. Hopping-dominated spin transport in unintentionally doped organic semiconductors. *J. Phys. Chem. Lett.* **2021**, *12*, 3540–3544.
 - 136 Yoo, J. W.; Jang, H. W.; Prigodin, V. N.; Kao, C.; Eom, C. B.; Epstein, A. J. Giant magnetoresistance in ferromagnet/organic semiconductor/ferromagnet heterojunctions. *Phys. Rev. B* **2009**, *80*, 205207.
 - 137 Coropceanu, V.; Cornil, J.; da Silva Filho, D. A.; Olivier, Y.; Silbey, R.; Brédas, J.-L. Charge transport in organic semiconductors. *Chem. Rev.* **2007**, *107*, 926–952.
 - 138 Lin, R.; Wang, F.; Rybicki, J.; Wohlgenannt, M.; Hutchinson, K. A. Distinguishing between tunneling and injection regimes of ferromagnet/organic semiconductor/ferromagnet junctions. *Phys. Rev. B* **2010**, *81*, 195214.
 - 139 Sun, X.; Bedoya-Pinto, A.; Mao, Z.; Gobbi, M.; Yan, W.; Guo, Y.; Atxabal, A.; Llopis, R.; Yu, G.; Liu, Y.; Chuvilin, A.; Casanova, F.; Hueso, L. E. Active morphology control for concomitant long distance spin transport and photoresponse in a single organic device. *Adv. Mater.* **2016**, *28*, 2609–2615.
 - 140 Massé, A.; Friederich, P.; Symalla, F.; Liu, F.; Meded, V.; Coehoorn, R.; Wenzel, W.; Bobbert, P. A. Effects of energy correlations and superexchange on charge transport and exciton formation in amorphous molecular semiconductors: an *ab initio* study. *Phys. Rev. B* **2017**, *95*, 115204.
 - 141 Abdalla, H.; Zuo, G.; Kemerink, M. Range and energetics of charge hopping in organic semiconductors. *Phys. Rev. B* **2017**, *96*, 241202.
 - 142 Hu, B.; Wu, Y. Tuning magnetoresistance between positive and negative values in organic semiconductors. *Nat. Mater.* **2007**, *6*, 985–991.
 - 143 Nguyen, T. D.; Ehrenfreund, E.; Vardeny, Z. V. Spin-polarized light-emitting diode based on an organic bipolar spin valve. *Science* **2012**, *337*, 204–209.
 - 144 Prieto-Ruiz, J. P.; Miralles, S. G.; Prima-García, H.; López-Muñoz, A.; Riminucci, A.; Graziosi, P.; Aeschlimann, M.; Cinchetti, M.; Dediu, V. A.; Coronado, E. Enhancing light emission in interface engineered spin-OLEDs through spin-polarized injection at high voltages. *Adv. Mater.* **2019**, *31*, 1806817.
 - 145 Bairagi, K.; Romero, D. G.; Calavalle, F.; Catalano, S.; Zuccatti, E.; Llopis, R.; Casanova, F.; Hueso, L. E. Room-temperature operation of a p-type molecular spin photovoltaic device on a transparent substrate. *Adv. Mater.* **2020**, *32*, 1906908.
 - 146 Yu, Z. G. Impurity-band transport in organic spin valves. *Nat. Commun.* **2014**, *5*, 4842.
 - 147 Droghetti, A.; Sanvito, S. Impurity band magnetism in organic semiconductors. *Phys. Rev. B* **2019**, *99*, 094413.
 - 148 Arnold, T.; Atxabal, A.; Parui, S.; Hueso, L. E.; Ortmann, F. Hot electrons and hot spins at metal–organic interfaces. *Adv. Funct. Mater.* **2018**, *28*, 1706105.
 - 149 Meng, K.; Zheng, R.; Gu, X.; Zhang, R.; Guo, L.; Qin, Y.; Yang, T.; Li, M.; Hu, S.; Zhang, C.; Wu, M.; Guo, A.; Yang, X.; Zhang, J.; Sun, X. In-device ballistic-electron-emission spectroscopy for accurately *in situ* mapping energy level alignment at metal–organic semiconductors interface. *Adv. Mater.* **2025**, *37*, 2412758.
 - 150 Yang, T.; Qin, Y.; Gu, X.; Sun, X. Molecular design for enhanced spin transport in molecular semiconductors. *Nano Res.* **2023**, *16*, 13457–13473.