Enhancement of the Spin Accumulation at the Interface between a Spin-Polarized Tunnel Junction and a Semiconductor

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We report on spin injection experiments at a $\mathrm{Co/Al_2O_3/GaAs}$ interface with electrical detection. The application of a transverse magnetic field induces a large voltage drop ΔV at the interface as high as 1.2 mV for a current density of $0.34\ \mathrm{nA}\cdot\mu\mathrm{m}^{-2}$. This represents a dramatic increase of the spin accumulation signal, well above the theoretical predictions for spin injection through a ferromagnet/semiconductor interface. Such an enhancement is consistent with a sequential tunneling process via localized states located in the vicinity of the $\mathrm{Al_2O_3/GaAs}$ interface. For spin-polarized carriers these states act as an accumulation layer where the spin lifetime is large. A model taking into account the spin lifetime and the escape tunneling time for carriers traveling back into the ferromagnetic contact reproduces accurately the experimental results.

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The electron's spin degree of freedom is expected to provide additional functionalities in future electronic devices [1,2]. The ability to inject spins into a semiconductor (SC) by electrical means along with the possibility to convert a spin accumulation into an electrical signal is a prerequisite for spin transport and spin manipulation. In this context, lateral structures for nonlocal bipolar spin detection operations are of particular interest [3] in the aim to add a gate control. Several approaches have been proposed to inject spin currents into a semiconductor (SC), using either ferromagnetic (FM) metals [4] or diluted magnetic semiconductors [5,6] or by taking advantage of the spin-orbit coupling through spin Hall effect experiments [7]. Even though experimental evidences of spin injection into GaAs [8–10] or silicon [11] from spin-polarized tunnel junctions have recently been reported, the amplitudes of the electrical signals related to the spin accumulation remain quite modest. In this letter, we report on the dramatic enhancement of the spin accumulation at the interface between a $\mathrm{Co(FM)/Al_2O_3}$ insulating (I) tunnel junction and a *n*-type GaAs lateral channel (FM/I/SC interface). The spin accumulation amplitude is well above the predictions of the standard theory of spin injection in the diffusive regime for a single FM/SC interface [12,13]. We show, through the bias dependence of the electrical Hanle signal, that an additional mechanism in the spin injection process has to be taken into account to explain our measurements. It involves an intermediate tunneling step into some localized states located in the region close to the I/SC interface. Both spin lifetime of the carriers and average back tunneling time towards the FM emitter are determined experimentally.

The samples were grown by molecular beam epitaxy on a semi-insulating GaAs (001) substrate. A Si-doped *n*-GaAs channel ($n = 5\times10^{18}\ \mathrm{cm}^{-3}$, resistivity $\rho = 11\Omega\cdot\mu\mathrm{m}$, thickness $w = 50$ nm) was grown on the top of an undoped GaAs buffer, and followed by a 15 nm thick n^+-GaAs layer ($n^+ = 2\times10^{19}\ \mathrm{cm}^{-3}$) in order to minimize the depletion region. The samples were then capped with amorphous As and air-transferred to another chamber. The As desorption was performed under RHEED until a clean (2×4)-GaAs(001) reconstruction was obtained. After cooling down to room temperature, the samples were transferred *in situ* to a sputtering chamber where a 1.5 nm thick Al layer was deposited and etched under 10^{-2} mbar of O_2 plasma. A 15 nm thick Co layer was then deposited and capped with 15 nm Au. The structure was processed into 200 μm wide and 2 mm long channels characterized by a resistance of 2.2 kΩ using standard contact optical lithography. Each channel was contacted with several electrodes of size ranging from $6\times196\ \mu\mathrm{m}^2$ to $30\times196\ \mu\mathrm{m}^2$ (W is the electrode width). The long sides were aligned along the in-plane easy axis of Co. In order to prevent any channel damage, only the Au layer was dry etched, while selective wet etching was used for the Co and $\mathrm{Al_2O_3}$ layers. In a last step, remote ohmic contacts were formed.

The tunneling *I–V* curve acquired in a 3-points configuration [Fig. 1(a)] at the $\mathrm{Co/Al_2O_3}/n^+$-GaAs interface for a $15\ \mu\mathrm{m}\times196\ \mu\mathrm{m}$ contact is shown in Fig. 1(b). The behavior is nonlinear and quasisymmetric with respect to zero bias. This highlights the dominant role of the $\mathrm{Al_2O_3}$ barrier in the tunneling transmission. Moreover, we compare the $\mathrm{Al_2O_3}$ tunnel resistance, $R_b^* \approx 10^9\ \Omega\cdot\mu\mathrm{m}^2$ at low bias with the range of interface resistances calculated by Fert *et al.* [13] for efficient spin injection and detection. First R_b^* exceeds the minimum interface resistance needed for the injection of a spin-polarized current into the GaAs channel, i.e., $R_b^* \gg r_1 = (\rho\ell_{sf})W/w \approx 10^3\ \Omega\cdot\mu\mathrm{m}^2$ for a spin diffusion length ℓ_{sf} of 1 μm (ρ is the resistivity of the GaAs and W and w are geometrical parameters that can

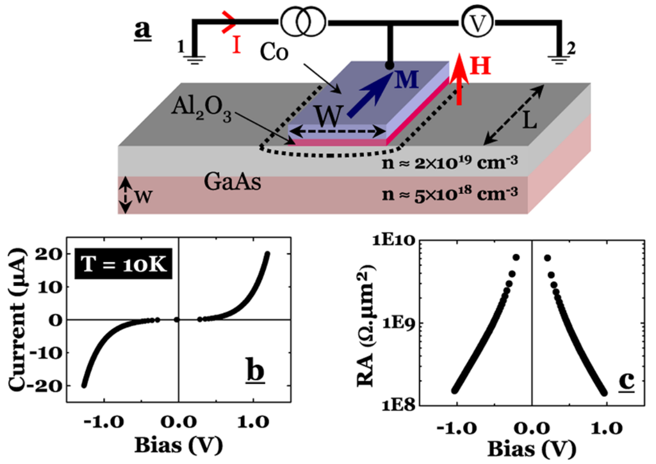

FIG. 1 (color online). (a) Diagram of the structure described in the text. The region within the dashed line represents the depletion layer. (b) Current vs bias curve of a typical $15 \times 196\ \mu\mathrm{m}^2$ Co/Al_2O_3 interface. (c) *RA* product vs bias measured at the same contact.

be seen in Fig. 1). However, even if the channel length L between two spin-polarized contacts was 10 times smaller than ℓ_{sf}, a significant spin-valve signal (in both local and nonlocal geometry) could be obtained only for interfaces' resistances much smaller than $r_1 \ell_{sf}/L \approx 10^4\ \Omega \cdot \mu\mathrm{m}^2$ in the highest limit [13]. R_b^* is thus too large to observe a two-interface spin-valve-like signal as confirmed by our experiments. Therefore we focused on the measurement of the single-interface signal related to spin accumulation under the injection contact. This can be done by using the Hanle effect [8] in the 3-points geometry of Fig. 1(a). A large enough transverse magnetic field suppresses the spin accumulation. The resulting voltage drop is proportional to the drop of spin accumulation. Equivalently, it can be viewed as the reduction of the current flowing back as the electron bath becomes fully depolarized.

The voltage drop as a function of the magnetic field is plotted in Fig. 2 for a current density $j \simeq -0.34\ \mathrm{nA} \cdot \mu\mathrm{m}^{-2}$ ($I = -1\ \mu\mathrm{A}$, contact size of $15 \times 196\ \mu\mathrm{m}^2$ and electrons flowing from the Co/Al_2O_3 contact into the GaAs channel). A significant potential drop $\Delta V \approx 1.2$ mV occurs when the magnetic field increases from zero to about 3 kOe. This quantity is related to the spin accumulation $\Delta\mu$ at the interface by $e\Delta V = \gamma \Delta\mu/2$, where $\gamma \simeq 0.4$ is the spin transmission coefficient through the tunnel barrier in the Co/Al_2O_3 system [14,15]. This corresponds to a spin resistance $R_s = \Delta V/I \simeq 1.2\ \mathrm{k}\Omega$, or equivalently a spin resistance-area product $R_s \cdot A \simeq 3.6\ \mathrm{M}\Omega \cdot \mu\mathrm{m}^2$. This value is well above the spin resistance expected from the theory of spin injection in the diffusive regime applied at a single FM/SC interface, which would give $r_1 = (\rho \ell_{sf}) W/w \approx 10^3\ \Omega \cdot \mu\mathrm{m}^2$ [13].

The left inset of Fig. 2 shows the voltage drop or Hanle signal vs the applied bias for two different contact sizes. At small bias the curves exhibit a linear increase with the injected current. They reach a maximum of 1.2 mV at −430 mV and 0.4 mV at +500 mV before decreasing in a nonsymmetric way at higher (absolute) bias. More quantitative information can be extracted from the relative variation of the Hanle signal $\Delta V/V$ (magnetoresistance, MR) with the applied bias V as shown in Fig. 3(a). The most striking feature is a nonlinear and asymmetric dependence with respect to zero bias which we attribute to the effect of the depletion layer. The MR admits a single maximum at $V = -430$ mV ($j = -0.34\ \mathrm{nA} \cdot \mu\mathrm{m}^{-2}$) followed by a rapid decrease of the signal, which disappears for $V < -0.85$ V. Reversing the electron flow at positive bias results in the gradual reduction of the spin accumulation signal down to zero for bias larger than +1.1 V.

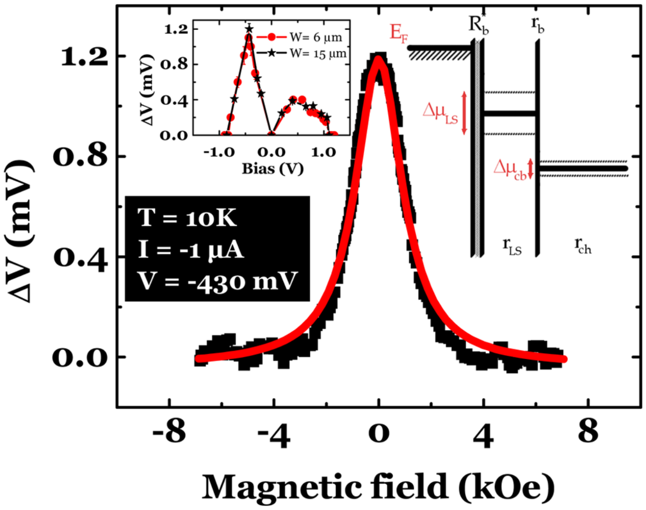

FIG. 2 (color online). Voltage drop ΔV vs transverse magnetic field measured at 10 K on a typical contact (size $15 \times 196\ \mu\mathrm{m}^2$, bias of $V = -430$ mV and current $I = -1\ \mu\mathrm{A}$). We have substracted a small positive quadratic background to the bare signal corresponding to the intrinsic tunneling anisotropic magnetoresistance of the Al_2O_3 barrier. The red line corresponds to a typical fit obtained with Eq. (3) for $\tau_{sf}^* = 230$ ps and $\tau_n = 15$ ns. Left inset: bias dependence of ΔV for 2 typical Co/Al_2O_3 electrodes (width 6 and 15 μm and similar *RA*). Right inset: sketch of the model described in the discussion.

We ascribe our results to a sequential tunneling process through localized states (LS), e.g ionized Si donors within the depletion layer or surface states at the I/SC interface. These LS act as a confining layer which enhances the spin accumulation in the vicinity of the $Al_2O_3/GaAs$ interface reducing the spin accumulation in the n-GaAs channel. By describing the spin injection with an intermediate stage in the LS (see right inset of Fig. 2) using standard drift-diffusion equations, one finds the following expressions for the spin accumulation in the LS and in the n-GaAs at the injection interface:

$$\Delta\mu_{LS} \simeq 2e\gamma j \frac{r_{LS}(r_{ch} + r_b)}{r_{LS} + r_{ch} + r_b}, \qquad \Delta\mu_{ch} \simeq 2e\gamma j \frac{r_{LS} r_{ch}}{r_{LS} + r_{ch} + r_b}. \tag{1}$$

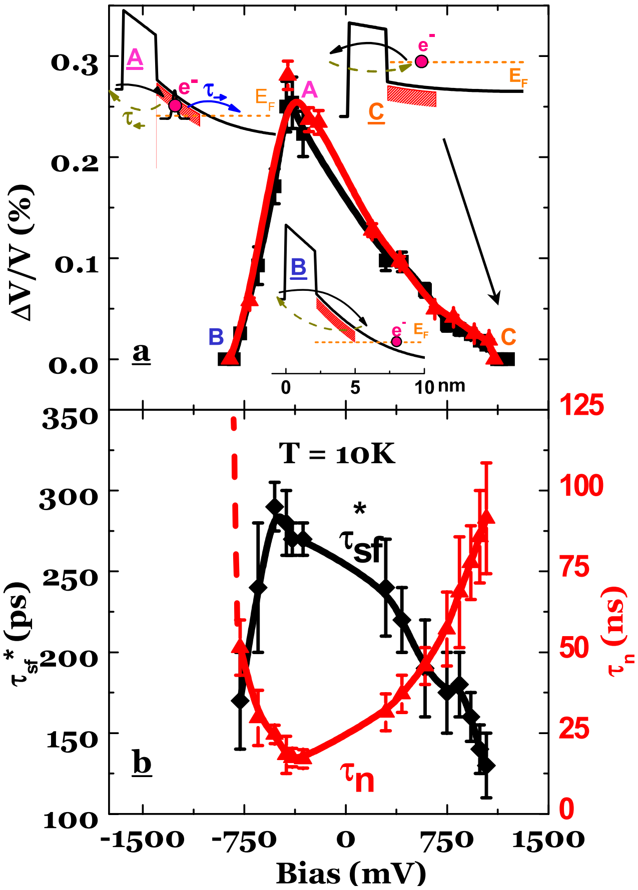

FIG. 3 (color online). Bias dependence of (a) the MR for 2 typical Co/Al$_2$O$_3$ interfaces with different sizes 6×196 μm^2 (squares, black) and 15×196 μm^2 (triangles, red) and (b) the spin lifetime τ^*_{sf} and tunneling escape time τ_n for the 6×196 μm^2 junction. The dashed red line is an extrapolation of the behavior of τ_n. Figure 3(a) insets: band bendings A, B, and C corresponding to different applied bias and described in text.

The corresponding MR that will be detected by the Hanle effect follows:

$$\frac{\Delta V}{V} \simeq \frac{\gamma^2}{1-\gamma^2}\left(\frac{r_{LS}}{R^*_b + r_b}\right)\frac{r_b + r_{ch}}{r_b + r_{LS} + r_{ch}}. \qquad (2)$$

In the above equation are included: (i) the large Al$_2$O$_3$ spin-dependent tunnel resistance used for spin injection namely R^*_b, (ii) a bias-dependent leakage resistance r_b at the interface between the LS and the n-GaAs channel, (iii) a spin-flip resistance associated with these LS, $r_{LS} = \tau^{LS}_{sf}/(e^2 \mathcal{N}^{LS}_{3-D} d_{LS})$, as well as (iv) the spin-flip resistance of the n-GaAs channel $r_{ch} \simeq \tau^{ch}_{sf}/(e^2 \mathcal{N}^{ch}_{3-D} d_{ch})$. The superscripts ch and LS hold for the n-GaAs channel and the localized state layer respectively τ^{α}_{sf}, $\mathcal{N}^{\alpha}_{3-D}$ and d_α are the spin lifetime, density of states per unit volume and thickness of each layer. By taking into account the relaxation volume including lateral spin flow, d_{ch} is found to be equal to $d_{ch} = w + 2\frac{w}{W}\ell^{ch}_{sf}$. d_{LS} is estimated to be of the order of a nm. Hereafter, we call $\mathcal{N}^{LS} = \mathcal{N}^{LS}_{3-D} d_{LS}$ and $\mathcal{N}^{ch} = \mathcal{N}^{ch}_{3-D} d_{ch}$ the two-dimensional density of states (2D DOS) integrated over the thickness of the LS layer and n-GaAs channel, respectively, with $\mathcal{N}^{ch} \gg \mathcal{N}^{LS}$ by at least 2 orders of magnitude. A large "leakage" tunnel resistance r_b leads to a large spin accumulation in the LS layer, $\Delta\mu_{LS} \simeq 2e\gamma j r_{LS}$. This is the situation that can be observed experimentally as soon as $r_{LS} \gg r_{ch} \approx 10^3\ \Omega \cdot \mu\text{m}^2$. At the same time it reduces the spin accumulation signal in the n-GaAs channel, $\Delta\mu_{ch} \ll 2e\gamma j r_{ch}$, compared to the case of a direct injection into the channel without sequential tunneling. Typical values of $\tau^{LS}_{sf} \simeq 1$ ns for LS in an impurity band [16] and $\mathcal{N}^{LS}_{3-D} = 10^{19}$ eV^{-1} cm^{-3} would give r_{LS} of the order of 1 M$\Omega \cdot \mu$m^2 and a MR $\frac{\Delta V}{V} \approx 0.1\%$ for $R^*_b \simeq 10^9 \Omega \cdot \mu\text{m}^2$, close to our experiments. This interpretation appears even more convincing when one converts each resistance into a corresponding characteristic time: $R^*_b = \tau^{LS}_{\leftarrow}/(e^2 \mathcal{N}^{LS}_{3-D} d_{LS})$ and $r_b = \tau^{LS}_{\rightarrow}/(e^2 \mathcal{N}^{LS}_{3-D} d_{LS})$. $\tau^{LS}_{\leftrightarrows}$ represent the mean escape (or leakage) tunneling times of carriers from a LS into the FM contact on the left ($\leftarrow$) or towards the GaAs channel on the right ($\rightarrow$). In the limit of a large tunneling resistance R^*_b (or a large $\tau^{LS}_{\leftarrow}$), the MR can then be rewritten:

$$\frac{\Delta V}{V} \simeq \frac{\gamma^2}{1-\gamma^2}\left(\frac{\tau^*_{sf}}{\tau_n}\right) \qquad (3)$$

with

$$\tau^*_{sf} = \tau^{LS}_{sf}\frac{\mathcal{N}^{ch}\tau_{\rightarrow} + (\mathcal{N}^{ch} + \mathcal{N}^{LS})\tau^{ch}_{sf}}{\mathcal{N}^{ch}(\tau_{\rightarrow} + \tau^{LS}_{sf}) + \mathcal{N}^{LS}\tau^{ch}_{sf}},$$
$$\tau_n = \left(1 + \frac{\mathcal{N}^{ch}\tau^{ch}_{sf}}{\mathcal{N}^{LS}\tau^{ch}_{sf} + \mathcal{N}^{ch}\tau^{LS}_{\rightarrow}}\right)\tau^{LS}_{\leftarrow}, \qquad (4)$$

τ^*_{sf} is an average spin lifetime in the SC (both n-GaAs channel and LS) and τ_n is the mean escape (reabsorbing) time of carriers towards the FM contact after having been injected into the SC. They depend explicitly on the characteristic leakage time $\tau^{LS}_{\rightarrow}$ between the LS and the GaAs reservoir. The prefactor appearing in the Eq. (4), $\mathcal{N} = 1 + \frac{\mathcal{N}^{ch}\tau^{ch}_{sf}}{\mathcal{N}^{LS}\tau^{ch}_{sf} + \mathcal{N}^{ch}\tau^{LS}_{\rightarrow}}$, stands for a renormalization factor relative to the localized state 2D DOS, $\mathcal{N}^{LS}$. That way, $\mathcal{N} \times \mathcal{N}^{LS}$ appears to be the true 2D DOS seen by an electron in the SC before its spin flips. In Hanle experiments τ^*_{sf} has to be replaced in the Eq. (3) by $\tau^*_{sf}(H) = \tau^*_{sf}/[1 + (\omega_H \tau^*_{sf})^2]$ to take into account the decoherence induced by the applied magnetic field H. We used a decoherence convolution function [14] $f = 1/[1 + (\omega_H \tau^*_{sf})^2]$, where $\omega_H = \frac{g\mu_B B}{\hbar}$ is the Larmor frequency, μ_B the Bohr magneton, and g is the Landé factor of GaAs.

The $\Delta V/V$ vs H curves can be fitted using the Eq. (3) with $\tau^*_{sf} = \tau^*_{sf}(H)$ to extract τ^*_{sf} and τ_n for each bias [Fig. 3(b)]. They are determined independently since τ^*_{sf} is responsible for the full width at half maximum of the Lorentzian curve whereas its amplitude is given by τ^*_{sf}/τ_n.

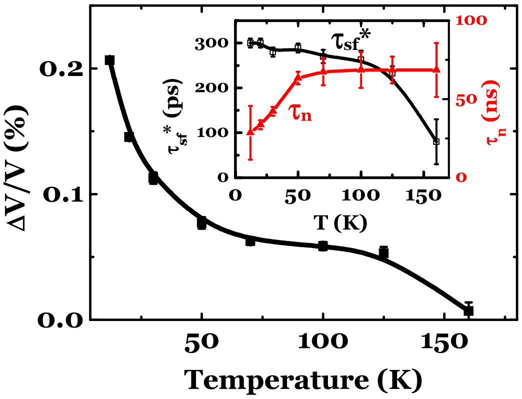

FIG. 4 (color online). Temperature dependence of the MR ($\Delta V/V$) acquired at $V = -0.3$ V on a $30 \times 196\ \mu\text{m}^2$ contact. The *RA* product is *1.5* times larger than the 2 junctions studied in Fig. 3(a). Inset: τ_{sf}^* and τ_n as a function of temperature and as determined by fitting the $\Delta V/V$ vs H curves by Eq. (3).

We now discuss three distinct injection regimes: low bias injection (A), large reverse polarization (B) and large direct polarization (C), as illustrated in Fig. 3(a) from 1D Poisson simulations [17]. At low negative bias (point A in Fig. 3(a): $V = -0.43$ V), the depletion region extends inside the GaAs resulting in a longer escape tunneling time $\tau_{\rightarrow}^{LS}$ into the GaAs channel. As long as $\tau_{sf}^{LS} \ll \tau_{\rightarrow}^{LS}$, the spin lifetime is now the LS spin lifetime, $\tau_{sf}^* = \tau_{sf}^{LS} \approx$ 300 ps [16]. The escape time $\tau_n = \tau_{\leftarrow}^{LS}$, measured to be less than 15 ns, is reduced by localization effects. It results from both combined effects in a dramatic increase of the MR. At large negative bias of $V = -0.85$ V [point B in Fig. 3(a)] the conduction band is pulled well below the FM contact Fermi level. Electrons are directly injected into the conduction band and taken away from the interface by the strong electric field which prevents carriers from flowing back: τ_n becomes infinite. Simultaneously, the spin lifetime is shortened to some tens of ps since it corresponds now to the spin lifetime in the n-GaAs channel [Fig. 3(b)] $\tau_{sf}^* = \tau_{sf}^{ch} < 100$ ps. This relaxation time is in agreement with reported value $\tau_{sf}^{ch} \approx 50$ ps for a similar doping level [18,19]. As a consequence the MR falls down. For direct polarization ($V > 0$ V), the depletion layer is reduced, resulting in the diminution of the carrier escape time towards the conduction band, $\tau_{\rightarrow}^{LS}$. The localization effects related to the LS play now a minor role. The extreme case (point C in Fig. 3(a): $V \simeq +1.1$ V) corresponds to a direct tunneling from the GaAs channel into the FM contact. The spin lifetime is again the n-GaAs spin lifetime $\tau_{sf}^* = \tau_{sf}^{ch}$. The escape time τ_n increases by 1 order of magnitude from 10 ns to about 100 ns at such a voltage. According to Eq. (4) the origin of this enhancement can be explained by the increase of the prefactor $\mathcal{N}$ when $\tau_{\rightarrow}^{LS}$ decreases. Another way to assess the spin injection mechanism is to study the MR temperature dependence (Fig. 4). The spin lifetime and escape time towards the FM contact were measured as a function of the temperature (inset of Fig. 4) for the bias $V = -0.3$ V. Thermal activation favors carrier escape towards the GaAs channel (shorter $\tau_{\rightarrow}^{LS}$) leading to a reduced spin lifetime of carriers, down to less than 100 ps at 150 K and to a longer tunneling escape time towards the FM contact [Eq. (4)].

In conclusion, we have succeeded in detecting a spin accumulation by electrical means in FM/SC contacts. The accumulation is large, $\Delta\mu \simeq 6$ meV ($\gamma = 0.4$) and exhibits an unusual bias dependence. This behavior can be explained by a sequential spin injection process into localized states nearby the interface. Such a spin injection process in several stages could also be effective in other experiments of spin injection into semiconductors. An analytical model was developed which expresses the dependence of the MR as a function of two characteristic times: the spin-lifetime τ_{sf} and carrier backward tunneling escape time τ_n. This opens new routes to extract the spin lifetime in assembled nano-objects by electrical means (e.g., SC quantum dots) rather than by optical measurements [20].

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