Non-monotonic spin relaxation and decoherence in graphene quantum dots with spin-orbit interactions

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We investigate the spin relaxation and decoherence in a single-electron graphene quantum dot with Rashba and intrinsic spin-orbit interactions. We derive an effective spin-phonon Hamiltonian via the Schrieffer-Wolff transformation in order to calculate the spin relaxation time $T_1$ and decoherence time $T_2$ within the framework of the Bloch-Redfield theory. In this model, the emergence of a non-monotonic dependence of $T_1$ on the external magnetic field is attributed to the Rashba spin-orbit coupling-induced anticrossing of opposite spin states. A rapid decrease of $T_1$ occurs when the spin and orbital relaxation rates become comparable in the vicinity of the spin-mixing energy-level anticrossing. By contrast, the intrinsic spin-orbit interaction leads to a monotonic magnetic field dependence of the spin relaxation rate which is caused solely by the direct spin-phonon coupling mechanism. Within our model, we demonstrate that the decoherence time $T_2$ is dominated by relaxation processes for the electron-phonon coupling mechanisms in graphene up to leading order in the spin-orbit interaction. Moreover, we show that the energy anticrossing also leads to a vanishing pure spin dephasing rate for these states for a super-Ohmic bath.

I. INTRODUCTION

Carbon-based materials such as graphene and carbon nanotubes are of recognized importance for their potential spintronic and quantum computation applications. Notably, single-layer graphene, a one-carbon-atom-thick layer arranged in a honeycomb crystal lattice, has attracted much interest in the last decade due to its unique electronic properties. The electron spin degree of freedom in graphene quantum dots makes them promising candidates for universal scalable quantum computing, which would rely on spin relaxation and decoherence times much longer than the gate operation times. Graphene has a relatively weak hyperfine interaction and spin-orbit (SO) couplings. A graphene sheet is composed naturally of 99% of $^{12}$C with nuclear spin 0, and of 1% $^{13}$C with nuclear spin 1/2, leading us to long dephasing times in carbon-based quantum dots due to a weak hyperfine interaction. Thus graphene emerges as a good candidate to host a spin qubit, in contrast to GaAs quantum dots, whose spin dynamics is strongly modified by the nuclear spin bath. Moreover, the weak SO couplings in graphene generates a spin-splitting on the order of tens of $\mu$eV due to the low atomic weight of carbon atoms. Long spin relaxation times are expected since the mechanisms that enable relaxation channels arise as a combined effect of non-piezo-electric electron-phonon interaction and weak SO coupling.

Despite the lack of measurements of the spin relaxation and dephasing times in graphene quantum dots, experimental results have already been reported in a two-electron $^{13}$C nanotube double quantum dot that has been isotopically-enriched. These results showed a non-monotonic magnetic field dependence of the spin relaxation time near the energy anticrossing. In this case, the spin relaxation minimum is related to the coupling between electron spin in the quantum dot and the nanotube deformation potential.

In this paper, we derive a spin-phonon Hamiltonian using the Schrieffer-Wolff transformation for all mechanisms of electron-phonon and spin-orbit interactions. This effective Hamiltonian captures the combined effect of the SO interaction and electron-phonon-induced potential fluctuations. Within the Bloch-Redfield theory, we find that a non-monotonic behavior of the spin relaxation time occurs as a function of the external magnetic field around the spin mixing energy-level anticrossing by the Rashba SO coupling in combination with the deformation potential and bond-length change electron-phonon mechanisms. We predict that the minimum of the spin relaxation time $T_1$ could be experimentally observed in graphene quantum dots. This energy anticrossing takes place between the first two excited energy levels at the accidental degeneracy for a certain value $B^*$ of the external magnetic field. We treat the accidental degeneracy mixed by the Rashba SO coupling using degenerate-state per-

Figure 1: Schematic of a gate-tunable circular graphene quantum dot setup. An homogeneous magnetic field is applied perpendicularly to the gapped graphene sheet. A metallic gate put on top of the graphene defines the confinement potential for a single-electron. Figure not drawn to scale.

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turbation theory. $T_1$ strongly increases at the energy anticrossing, reaching the same order as the orbital relaxation time $\tau_{\text{orb}}$. In contrast with carbon nanotubes, the intrinsic SO does not couple these states due to the selection rules in a circular quantum dot, exhibiting a monotonic magnetic field dependence of $T_1$ due to direct spin-phonon coupling (deflection coupling mechanism). We also demonstrate that pure spin dephasing rates vanish in the leading order of the electron-phonon interaction and SO interactions causing a decoherence dominated by relaxation processes, i.e., $T_2 = 2T_1$. Moreover, we find a vanishing spin dephasing rate for a super-Ohmic bath as a general property of the energy anticrossing spectrum.

This paper is organized as follows: In Sec. [III] we introduce the model to describe a circular graphene quantum dot. In Sec. [III] we derive the effective spin-phonon Hamiltonian. In Sec. [IV] we present a calculation of the spin relaxation time $T_1$ within the Bloch-Redfield theory. In Sec. [V] we discuss the vanishing spin dephasing rate within our model. Finally, we summarize our results and draw our conclusions in Sec. [VI].

II. THE MODEL

In this section, we introduce the model for a circular and gate-tunable graphene quantum dot. Within our model, we consider a gapped graphene taking into account electron-phonon coupling mechanisms and spin-orbit interactions. We also analyze the energy spectrum of the quantum dot and its energy-level degeneracy. The degenerate levels are mixed by the Rashba SO coupling, and the energy crossings are removed using the standard degenerate perturbation theory.

A. Graphene quantum dots

The low-energy effective Hamiltonian for graphene is analogous to the two-dimensional massless Dirac equation. The characteristic linear dispersion for massless fermions occurs at the two non-equivalent points $K$ and $K'$ (valleys), in the honeycomb lattice Brillouin zone. The graphene energy bands in the vicinity of these high-symmetry points constitute a solid-state realization of relativistic quantum mechanics. However, confining electrons in graphene quantum dots is a difficult task, since the particles tend to escape from the electrostatic confinement potential due to Klein tunneling. This problem can be overcome by putting graphene on top of a substrate, such as SiC[18] and BN[19,20] that induces a non-equivalent potential for each atom of the two carbon sublattices and adds a mass term to the Hamiltonian[21]. The sublattice A(B) will feel a potential parametrized by $+(-)\Delta$ which breaks inversion symmetry, opening a gap $2\Delta$ in the electron-hole energy spectrum. Combined with the mass term, an external magnetic field $B$ is necessary to break the time-reversal symmetry and lift the valley degeneracy. Thus it is reasonable to confine a single electron in a quantum dot with the restriction of its being localized in a single valley.

Consider then, a circular and gate-tunable graphene quantum dot at the two non-equivalent points $K$ and $K'$ of its being localized in a single valley.

and the electron-phonon interaction described by the following low-energy Hamiltonian for the $K$ valley[22]

$$\mathcal{H} = \mathcal{H}_d + \mathcal{H}_Z + \mathcal{H}_{SO} + \mathcal{H}_{ph} + \mathcal{H}_{\text{e-ph}}.$$  \hspace{1cm} (1)

with the quantum dot Hamiltonian $\mathcal{H}_d$ and the Zeeman term $\mathcal{H}_Z$, respectively, given by

$$\mathcal{H}_d = \hbar v_F \mathbf{p} \cdot \mathbf{e} + U(r) + \Delta \sigma_z, \quad \mathcal{H}_Z = \frac{1}{2} \gamma \mu_B B \cdot \mathbf{s},$$  \hspace{1cm} (2)

where $\mathbf{p} = \mathbf{p} - e \mathbf{A}$ is the canonical momentum. The vector potential is chosen such that $\mathbf{B} = \nabla \times \mathbf{A} = (0, 0, B)$, i.e., perpendicular to the graphene sheet. Here, $v_F = 10^5$ m/s is the Fermi velocity, $U(r) = U_0 \Theta(r - R)$ is the circular-shaped electrostatic potential, with $\Theta(x) = 1$ for $x \geq 0$ and $\Theta(x) = 0$ for $x < 0$. The operator $\mathbf{s}$ acts on the pseudospin subspace (A,B sublattices), while $\mathbf{e}$ acts on the real spin. Both operators $\sigma$ and $\mathbf{s}$ are represented by Pauli matrices.

The SO Hamiltonian for the $K$ valley reads[23]

$$\mathcal{H}_{SO} = \mathcal{H}_d + \mathcal{H}_R = \lambda_d \sigma_z s_z + \lambda_B (\sigma_x s_y - \sigma_y s_x),$$  \hspace{1cm} (3)

where $\mathcal{H}_d$ and $\mathcal{H}_R$ denote the intrinsic and Rashba SO effective Hamiltonians[23,24] respectively. The intrinsic SO coupling originates from the local atomic SO interaction. At first, only the contribution from the $\sigma - \pi$ orbital coupling was considered, resulting in a second-order term to the intrinsic SO coupling strength $\lambda_d[25]$. However, some $d$ orbitals hybridize with $p_z$ forming a $\pi$-band that gives a first-order contribution which plays a major role in the spin-orbit-induced gap[26]. The Rashba SO coupling, also called the extrinsic contribution, arises when an electric field is applied perpendicular to the graphene sheet. The major contribution of the SO coupling $\lambda_R$ comes from the $\sigma - \pi$ hybridization[27] in contrast with the intrinsic case. The Rashba SO could also be enhanced by curvature effects in the graphene sheet[28].

The free phonon Hamiltonian is given by

$$\mathcal{H}_{ph} = \sum_{q,\mu} \hbar \omega_{q\mu} b^\dagger_{q\mu} b_{q\mu}$$  \hspace{1cm} (4)

with the dispersion relation $\omega_{q\mu} = s_\mu |q|m$, where $s_\mu$ is the sound velocity and $m = 1, 2$ depending on the type of phonon branch.

Finally, we have the electron-phonon interaction $\mathcal{H}_{\text{e-ph}}$. We consider long-wavelength acoustic phonons represented by two main mechanisms: the deformation potential and the bond-length change mechanism[29]. The former is an effective potential generated by static distortions of the lattice. It is represented in the sublattice space as a diagonal energy shift in the bond structure. The latter are off-diagonal terms due to modifications of the bond-length between neighboring carbon atoms, which causes changes in the hopping amplitude. The electron-phonon interaction in the sublattice space is given by[29]

$$\mathcal{H}_{\text{e-ph}} = \sum_{q,\mu} \frac{q}{\sqrt{4 \pi \omega_{q\mu}}} \left( \begin{array}{c} g_1 a_1 \\ g_2 a_2 \\ g_1 a_1 \end{array} \right) \left( e^{i q \mathbf{B} \cdot \mathbf{r}} b_{q\mu}^+ - e^{-i q \mathbf{B} \cdot \mathbf{r}} b_{q\mu} \right).$$  \hspace{1cm} (5)
where $g_1$ and $g_2$ are the deformation potential and bond-length change coupling constants. Here, $A$ is the area of the graphene layer and $\rho$ is the mass area density. The constants $a_1$, $a_2$ and the sound velocities $s_{LA}$, $s_{TA}$ for the longitudinal-acoustic ($\mu = LA$) and transverse-acoustic ($\mu = TA$) modes are given in Table [I]. Both phonon branches have a linear dispersion relation given by $\omega_{\mu \mathbf{k}} = s_{\mu} |q|$. Optical phonons are not taken into account in this work, since their energies do not match the Zeeman splitting for typical laboratory fields. The out-of-plane phonons ($\mu = ZA$) will be discussed further below. Notice that the electron-phonon interaction is spin independent and can only cause a spin relaxation when assisted by the SO interaction.

In the following subsection, we analyze the bare quantum dot spectrum and perform a perturbation theory calculation for degenerate levels treating the SO Hamiltonian as a perturbative term.

### B. Degenerate state perturbation theory

In order to calculate $T_1$ and $T_2$, we use the quantum dot eigenstates perturbed by the SO interaction. Before doing so, we have to get rid of the degeneracies in the quantum dot spectrum by applying degenerate state perturbation theory. This procedure makes it clearer to define which states constitute our spin qubit and where the spin relaxation occurs.

Due to the selection rules for the matrix elements of the SO interaction $\mathcal{H}_D$ only the Rashba SO term couples states from the degenerate subspace. Thus we intend to find a linear combination of eigenstates from the degenerate subspace of the quantum dot such that these states are not coupled by the Rashba SO Hamiltonian $\mathcal{H}_R$.

Consider then, first the bare quantum dot Hamiltonian in the $K$ valley $\mathcal{H}_D$, with $\mathcal{H}_D [j, \nu, s] = E_{j,\nu}[j, \nu, s]$ and the quantum dot wave function $\chi_{j,\nu, s}(r, \phi)$:

$$\chi_{j,\nu, s}(r, \phi) = \psi_{j,\nu, s}(r, \phi) = e^{i(j-1/2)\phi} \left( \chi_A \chi_B^* e^{i\delta} \right). \quad (6)$$

The spinor components $\chi_{A,B}(r)$ are proportional to the confluent hypergeometric functions and are described by the set $j, \nu, s$, where we introduce the angular $(j = \pm 1/2, \pm 3/2, \ldots)$, radial ($\nu = 1, 2, 3, \ldots$) and spin $s = \uparrow, \downarrow$ quantum numbers.

Matching the spinors at $r = R$ results in a transcendental equation for the eigenvalues $E_{j,\nu}$ which can be obtained numerically. Since we are going to calculate the spin relaxation rates due to transitions between the lowest three energy levels of the quantum dot, we restrict ourselves to the analysis of the subspace $\{|+1/2, 1, \downarrow\}, |1/2, 1, \uparrow\}, |−1/2, 1, \downarrow\}|$. Including the Zeeman spin-splitting, it leads to a crossing of the energy levels $E_{1/2,1,\uparrow}$ and $E_{−1/2,1,\downarrow}$, at $B = B^*$ (solid lines in the inset). The spin relaxation rate takes place between the states $|\gamma_0\rangle$ and $|\gamma_1\rangle$ ($\Gamma_1 = \Gamma_{\gamma_0\gamma_1}$) before the anticrossing, and between the states $|\gamma_0\rangle$ and $|\gamma_2\rangle$ ($\Gamma_2 = \Gamma_{\gamma_0\gamma_2}$) after the anticrossing. Inset: Blowup of the energy levels in the vicinity of the crossing region.

Table I: Electron-phonon constants and sound velocities for longitudinal (LA) and transverse (TA) acoustic phonons. The phonon emission angle is denoted by $\phi_y$.

|          | $a_1$ | $a_2$ | $s_j (10^7 \text{ m/s})$ |
|----------|-------|-------|--------------------------|
| LA       | $\ell$ | $\ell e^{i\phi_y}$ | 1.95*                   |
| TA       | 0     | $\ell e^{i\phi_y}$ | 1.22*                   |

*From Ref. [31]
with the associated first-order eigenvalues
\[
E_{\gamma} = E_{1/2, 1} - \frac{\hbar \omega_{2}}{2}, \quad \epsilon_+ = \epsilon = \sqrt{\epsilon^2 + |\Delta_{SO}|^2},
\] (8)
plotted in Fig. 2. We define \( \epsilon_+ = (E_{1/2, 1} + E_{-1/2, 1})/2 \) and \( \epsilon_- = (E_{1/2, 1} - E_{-1/2, 1} + \hbar \omega_{2})/2 \). The Rashba SO interaction \( \gamma \) is defined as
\[
\gamma = g_{\text{R}} B = g_{\text{R}} B_0.
\]

For the Rashba SO coupling, we have to consider the new basis \( |\gamma_1, \gamma_2\rangle \) calculated in Sec. II B using perturbation theory for the degenerate levels. As explained in Sec. II B, we are interested in transitions from the excited states \( |\gamma_2\rangle \) to the ground state \( |\gamma_0\rangle \). In this case, we calculate the matrix element of the effective spin-phonon Hamiltonian \( \langle \gamma_0 | H_{\text{R-ph}} | \gamma_2 \rangle \), where \( \gamma_2 = \gamma_1, \gamma_2 \).

\[
\langle \gamma_0 | H_{\text{R-ph}} | \gamma_2 \rangle = \langle \gamma_0 | H_{\text{R-ph}} | \gamma_1 \rangle + \sum_{n,s \neq \gamma_2} \frac{\Omega_1(n, \gamma_1, \gamma_2)}{E_{\gamma_0} - E_n} + \sum_{n,s \neq \gamma_1} \frac{\Omega_2(n, \gamma_1, \gamma_2)}{E_{\gamma_2} - E_n},
\] (10)

where the degenerate subspace is given by \( \Delta = \{|1/2, 1, \uparrow\}, |1/2, 1, \downarrow\} \). Here, we have defined the product of the matrix elements as
\[
\Omega_1(n, \gamma_1, \gamma_2) = \langle \gamma_0 | H_{\text{R}} | n, s \rangle \langle n, s | H_{\text{R-ph}} | \gamma_2 \rangle, \quad \Omega_2(n, \gamma_1, \gamma_2) = \langle \gamma_0 | H_{\text{R}} | n, s \rangle \langle n, s | H_{\text{R-ph}} | \gamma_1 \rangle.
\] (11)

The matrix elements of the Rashba SO coupling give the selection rule \( |j - j'| = 0 \). These transitions are compatible with the selection rules of the electron-phonon interaction mechanisms depending on the order of the dipole expansion considered in the term \( \epsilon_{\text{eq}} \) in this instance, the selection rules match \( |j - j'| = 1 \) for the first order and zero order of the dipole expansion of the deformation potential (LA) and bond-length change (LA, TA), respectively.

For the intrinsic SO, the matrix element of the spin-phonon Hamiltonian is given by
\[
\langle n_0, \downarrow | H_{\text{R-ph}} | n_0, \uparrow \rangle = \langle n_0, \downarrow | S, H_{\text{R-ph}} | n_0, \uparrow \rangle, \quad \text{with the ground state set of angular and radial quantum numbers } n_0 = (1/2, 1), \text{ since } H_{\text{R}} \text{ does not connect the quantum states related with the crossed energy levels. Explicitly, we have}
\]
\[
\langle n_0, \downarrow | H_{\text{R-ph}} | n_0, \uparrow \rangle \propto \sum_{n' \neq n_0} \delta_{j, j'} \left( N_{AA}^{n_0} - N_{BB}^{n_0} \right),
\] (13)

where \( N_{AA}^{n_0} = \int dr r \chi^{\dagger}_{n_0}(r) \chi_{n_0}(r) \) and \( N_{BB}^{n_0} = \int dr r \chi^{\dagger}_{n_0}(r) \chi_{n_0}(r) \).

Thus \( H_{\text{R-ph}} \) can be rewritten as proportional to \( \{j, \sigma | j', \sigma'\} \) with \( j \neq j' \) which is identically zero. Consequently, the admixture mechanism due to the intrinsic SO does not contribute to the spin relaxation and dephasing process within our model.

In addition to the admixture mechanism, the spin relaxation can also take place due to the direct coupling of spin and local...
The system and the bath become correlated via the interaction mechanisms and SO interactions via the mapping, characterized by the Redfield tensor. First, we introduce the Bloch-Redfield theory. The spin-phonon terms presented here will be used to calculate the spin relaxation and dephasing rates in the following sections.

IV. SPIN RELAXATION RATES

In this section, we calculate the spin relaxation time using the effective spin-phonon Hamiltonian derived in the previous section. First, we introduce the Bloch-Redfield theory, which allows us to derive the general expression for the spin relaxation and decoherence times.

Consider a general Hamiltonian given by $H = H_s + H_B + H_{SB}$, where $H_s$ describes the system, $H_B$ a reservoir in thermal equilibrium (bath) and $H_{SB}$ describes the interaction between them. This general Hamiltonian is analogous to the one derived in Sec. 5 for all electron-phonon mechanisms and SO interactions via the mapping, $H_s \rightarrow H_s + H_E$, $H_B \rightarrow H_{ph}$ and $H_{SB} \rightarrow H_{s-ph}$. The system and the bath are uncorrelated initially, i.e., their spin matrices $\rho$ can be represented as $\rho(0) = \rho_s(0)\rho_B(0)$. Nevertheless, as time goes by, the system and the bath become correlated via the interaction term $H_{s-ph}$. This system dynamics is described by an equation of motion for the density matrix in the interaction picture $\hat{\rho}(t) = e^{-i\hat{H}_{s-ph}\hat{t}}\hat{\rho}(0)e^{i\hat{H}_{s-ph}\hat{t}}$ with the bath variables traced out $\hat{\rho}_s = \text{Tr}_B[\hat{\rho}]$ as

$$\frac{d}{dt}\hat{\rho}_s(t) = -\frac{i}{\hbar}\int_0^t\text{Tr}_B\left[\hat{H}_{s-ph}(t')\hat{\rho}_s(t')\hat{\rho}_s(0)\right]dt'$$

(15)

This equation of motion for the reduced density matrix is called the Nakajima-Zwanzig equation. If we assume that the coupling system-bath can be further simplified by neglecting terms up to $O(H_{s-ph}^2)$ in Eq. (15), which is equivalent to approximating the density matrix in the integral as $\rho(t) = \rho(0)e^{-i\hat{H}_{s-ph}\hat{t}}$. Considering a phonon bath, we assume that the time evolution of the $\rho_S(t)$ depends only on its present value and not on its past state (Markov approximation), i.e., $\hat{\rho}(t') \rightarrow \hat{\rho}(t)$ in the integral of Eq. (15). Taking the matrix elements of Eq. (15) between the eigenstates of $\hat{H}_s$, we have

$$\frac{d}{dt}\hat{\rho}_{mn}(t) = -\frac{i}{\hbar}\omega_{mn}\rho_{mn} - \sum_{kl}R_{mnkl}\rho_{kl}(t)$$

(16)

where $\rho_{mn} = \langle m|\rho|n \rangle$ and $\omega_{mn} = \epsilon_m - \epsilon_n$. The term $R_{mnkl}$ is the Redfield tensor

$$R_{mnkl} = \delta_{mn}\sum_i \Gamma^+_{ir} + \delta_{nk}\sum_i \Gamma^-_{im} - \Gamma^-_{mn} - \Gamma^+_{nk}$$

(17)

where $\Gamma^-_{mn} = \int_0^\infty dt e^{-i\omega_{mn}t}\langle m|\hat{H}_{s-ph}|n \rangle\langle n|\hat{H}_{s-ph}(t)|m \rangle$, with $\Gamma^+_{mn} = \left\langle \Gamma_{mn} \right\rangle$. Here, the overbar denotes the average over a phonon bath in thermal equilibrium at temperature $T$. Using Eq. (16) in the secular approximation where $R_{mnkl}$ is approximately given by a diagonal tensor and $\langle dS_z/dt \rangle = \text{Tr}[\langle d\rho/dt \rangle dt]$, we can derive the differential equation describing the time evolution of the average values of the spin components, also known as Bloch equations. The solution for the $S_z$ component with a magnetic field applied along the same direction is $S_z(t) = S_z^0 - S_z(t)e^{-i\mu B t}$, where $S_z^0$ is the equilibrium spin polarization (ensemble of spin-down electrons) and $S_z(t)$ is the initial non-equilibrium spin alignment considered in the problem (ensemble of spin-up electrons).

Explicitly, the spin relaxation rate is given by

$$\Gamma_1 = \frac{1}{T_1} = 2\pi \frac{\langle |\gamma_\uparrow|\hat{H}_{s-ph}|\gamma_\uparrow \rangle^2}{\hbar\omega_{\gamma_\uparrow}\coth\left(\frac{\hbar\omega_{\gamma_\uparrow}}{2k_B T}\right)}$$

(18)

Equation (18) can be simplified to

$$\frac{1}{T_1} = 2\pi \frac{2}{\hbar}\sum_q \left\langle |\gamma_q|\hat{H}_{s-ph}|\gamma_q \rangle^2 \delta(\hbar\omega_{\gamma_q} - \hbar\omega_q)\coth\left(\frac{\hbar\omega_{\gamma_q}}{2k_B T}\right)\right.$$  

(19)

The spin relaxation rate is then calculated combining Eqs. (19) and (10). The contribution due to the deformation potential (LA) combined with the Rashba SO coupling is given by

$$\Gamma_1^{\gamma_\uparrow LA} = \frac{\pi}{2} \frac{\hbar^2}{\hbar^2 s_L^2} \int_0^{2\pi} d\phi \left[\frac{E}{\hbar s_L^2}\right]_\gamma^2 \frac{2}{\hbar^2 s_L^2}$$

(20)

And those due to the bond-length change mechanism for $\mu = LA, TA$.

$$\Gamma^{\gamma_\uparrow LA, TA} = 2\pi \frac{\hbar^2}{\hbar^2 s_L^2} \int_0^{2\pi} d\phi \left[\frac{E}{\hbar s_L^2}\right]_\gamma^2 \frac{2}{\hbar^2 s_L^2}$$

(21)

where we imply summation over the repeated index $i = 1, 2, 3$. In the above we have defined

$$\Lambda_{\gamma}^i(A, g, \delta) = \gamma^i \left(1, 2, \downarrow | A_{1, g, \delta} - 1, 2, \uparrow \right) \rho_k$$

(22)
\[ \Lambda_2^a(A_{\text{eff}}, \epsilon) = \sum_{n=1/2,1} \lambda_2^a(1/2, 1, 1/2|A_{\text{eff}}, \epsilon| n, \downarrow) \times \langle n| H_R|1/2, 1, \uparrow \rangle \sigma_z, \]

\[ \Lambda_2^s(A_{\text{eff}}, \epsilon) = \sum_{n=1/2,1} \lambda_2^s(1/2, 1, 1|H_B| n, \uparrow) \times \langle n|H_{\text{eff}}|1/2, 1, \uparrow \rangle \sigma_y, \]

where \( A_{\text{eff}} = a_1 I_{2x2}, A_{\text{eff}} = g_2 (\sigma_x a_2^s + \sigma_y a_2^s), \) with \( \sigma_+ = (\sigma_x \pm i \sigma_y)/2. \) Their respective matrix elements are given by

\[ \langle n|A_{\text{eff}}|n'\rangle = M_{nn'} \left( \delta_{j,j'+1} e^{-i\delta \gamma} + \delta_{j,j'-1} e^{i\delta \gamma} \right), \]

with \( M_{nn'} = \int dr r^2 \left( \lambda_2^a \chi_0^A \chi_0^B + \lambda_2^s \chi_0^A \chi_0^B \right), \)

\[ \langle n|A_{\text{eff}}|n'\rangle = (g_2 a_1^s \delta_{j,j'+1} N_{n,n'}^B + g_2 a_2^s \delta_{j,j'-1} N_{n,n'}^B), \]

where \( N_{n,n'}^B = \int dr r^2 \chi_0^A(r) \chi_0^B(r). \) Here, \( \rho_1 = -\sin(\theta/2), \sigma_{y1} = \cos(\theta/2) \) and \( \rho_2 = \cos(\theta/2), \sigma_{y2} = \sin(\theta/2). \) The energy-dependent denominators are given by \( \lambda_2^a = 1, \lambda_2^s = 1/E_k - \Delta_n + g_2 \mu_B/2, \lambda_2^z = 1/E_{n/2,1} - \Delta_n - g_2 \mu_B/2. \)

As stated in Sec. IIIB, the energy relaxation accompanied by a spin-flip transition occurs between the states \( |\gamma_0\rangle \) and \(|\gamma_1\rangle\) before the energy anticrossing \( \Gamma_{11}^e = \Gamma_{\gamma_0 \rightarrow \gamma_1} \) and between the states \( |\gamma_0\rangle \) and \( |\gamma_2\rangle \) after the energy anticrossing \( \Gamma_{11}^e = \Gamma_{\gamma_0 \rightarrow \gamma_2} \), for all electron-phonon mechanisms \( \Gamma_{11}^e = \Gamma_{11}^{\text{LA}} + \Gamma_{11}^{\text{SA}} + \Gamma_{11}^{\text{TA}} \).

The contribution from the out-of-plane flexural phonons via the deflection coupling mechanism, calculated using Eq. (19), combined with Eq. (15), is

\[ \Gamma_{11}^{ZA} = \frac{4\pi^2 \lambda_2^z}{\rho} \left( g_2 \mu_B B Q(B) \right)^{1/2} \left( \frac{-s_Z^A + Q(B)}{2\mu} \right)^{1/2} \times \left| \int dr r \left( |\chi_A^z|^2 - |\chi_B^z|^2 \right)^2 \right|, \]

where we define \( Q(B) = \sqrt{s_{ZA}^2 + 4\rho(g_2 \mu_B B)} \), with \( s_{ZA} = 0.25 \times 10^3 \text{m/s}. \) In the low magnetic field limit, the term \( \Gamma_{11}^{ZA} \) simplifies to

\[ \Gamma_{11}^{ZA} = \frac{4\pi^2 \lambda_2^z}{\rho} \left( g_2 \mu_B B \right)^{1/2} \times \left| \int dr r \left( |\chi_A^z|^2 - |\chi_B^z|^2 \right)^2 \right|. \]

The magnetic field dependence of the spin relaxation rate for each electron-phonon coupling mechanism can be understood using the spectral density of the system-bath interaction

\[ J_{\gamma_0 \gamma}(\omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t} \langle \gamma_0 | H_{\text{eff}} | \gamma \rangle \langle \gamma | H_{\text{eff}} | \gamma \rangle \langle \gamma | H_{\text{eff}} | \gamma \rangle (29) \]

Further simplifications in Eq. (18) allow us to find the following relation \( 1/T_1 \propto J_{\gamma_0 \gamma}(\omega_{\gamma_0 \gamma}) \), where \( \omega_{\gamma_0 \gamma} \propto \omega_2 \propto g_2 \mu_B B. \) In a general form, we have that \( 1/T_1 \propto \sum_q K_q |\gamma_0 \rangle e^{i\omega_T q |\gamma_0 \rangle} \langle \gamma_0 | H_{\text{SO}} | \gamma_0 \rangle \delta(\omega - \omega_{\gamma_0 \gamma}) \), where \( K_q = q/\sqrt{4q^2 - 4} \) since \( \mathcal{H}_{\text{eff}} \propto K_q e^{i\omega_T q}. \) Also, \( \sum_q \propto \int dq q^{-1}, \) where \( d = 2 \) is the dimensionality of graphene. Each SO coupling defines the selection rule for the quantum number \( j \) and consequently, the order of the dipole expansion as explained in Sec. III. We find that for the Rashba SO coupling, \( J_{\gamma_0 \gamma}(\omega_{\gamma_0 \gamma}) \propto \omega_2^j \) with \( s = 4 \) for the deformation potential (LA) and \( s = 2 \) for the bond-length change mechanism (LA, TA). Also, for the intrinsic SO, \( s \geq 2 \) for the direct spin-phonon coupling (ZA). Therefore the spectral density of the system-bath interaction is super-Ohmic (\( s > 1 \)) with a strong dependence with the bath frequency for all phonons considered in graphene.

V. SPIN DEPHASING RATES

Next we evaluate the spin dephasing rates for all the electron-phonon mechanisms introduced in Sec. III. With the Bloch-Redfield theory, we can also solve the Bloch equations for the spin components perpendicular to the magnetic field, which are given by \( \langle S_z(\gamma_0 \gamma) \rangle \times s_0 \gamma_0(\omega_2) e^{-i/T_2} \) and \( \langle S_y(\gamma_0 \gamma) \rangle \times s_0 \gamma_0(\omega_2) e^{-i/T_2} \), where \( s_0 \gamma_0 \) are the initial spin polarizations along the \( x, y \) directions. The decoherence time

Table II: Parameters for the numerical evaluation of the spin relaxation rates. The electron-phonon coupling constants for the deformation potential \( g_1 \) and for the bond-length change mechanism \( g_2 \) are obtained after the external electric field \( E \) and the intrinsic \( \lambda_i \) SO couplings. The graphene layer is characterized by its mass area density \( \rho \). The quantum dot parameters are its radius \( R \), potential height \( U_0 \) and the substrate-induced energy gap \( \Delta \). The system is assumed to be in thermal equilibrium with the bath at temperature \( T \).

| Parameter   | Value |
|-------------|-------|
| \( g_1 \)   | 30 eV  |
| \( g_2 \)   | 1.5 eV  |
| \( \lambda_0 \) | 11 \mu eV  |
| \( E \)     | 50 V/300 nm² |
| \( \lambda_i \) | 12 \mu eV  |
| \( \rho \)   | 7.5 \times 10^{-7} \text{ kg/m}² |
| \( R \)     | 35 nm   |
| \( U_0 \)   | 260 meV |
| \( T \)     | 100 mK  |
In the low-temperature limit, we find that \( \Gamma^+_{\gamma_\gamma} \) and the out-of-plane phonons \( Z_A \) (light, dot-dashed). In other words, there are no phonons available in leading order to contribute to the spectral density of the system-bath interaction as \( 1/T \). The absence of Van Vleck cancellation leads to a finite value for \( \Gamma_\phi \) at \( B = 0 \).

The dephasing time can also be rewritten in terms of the spectral density of the system-bath interaction as \( 1/T_\phi \propto \lim_{\omega \to 0} J(\omega) \langle \omega \rangle \propto \lim_{\omega \to 0} J(\omega)/\omega \).

As we have shown in Sec. III the spectral function for all electron-phonon coupling mechanisms considered in this work are super-Ohmic. Thus the spin dephasing vanishes in all cases, since \( 1/T_\phi \propto \lim_{\omega \to 0} \omega^s/\omega \to 0 \), with \( s > 1 \). In other words, there are no phonons available in leading order to cause dephasing in graphene quantum dots. The decoherence time \( T_2 \) is determined only by the relaxation contribution, i.e., \( T_2 = 2T_1 \). Notice that this relation is no longer necessarily true considering two-phonon processes since the combination of emission and absorption energies can fulfill the energy conservation requirement.

Additionally, the spin dephasing rate could also vanish at the energy anticrossing for a super-Ohmic bath. Within the subspace spanned by the states \([|+1/2, 0, \uparrow\rangle, |-1/2, 0, \downarrow\rangle\), the Hamiltonian \( \mathcal{H} \) can be rewritten as \( \mathcal{H}_\phi = \Delta_x(B) \tau_x + \Delta_x(B) \tau_x \), where \( \tau_x \) denote a Pauli matrix and \( \Delta_x = (E_{\uparrow} + E_{\downarrow})/2 \). This magnetic field can be divided into two contributions \( B = B_0 + \delta B(t) \): an external source \( B_0 \) and an internal contribution \( \delta B(t) \) due to the bath. For small fluctuation of \( \delta B(t) \), the \( \mathcal{H}_\phi \) is approximately given by

\[
\mathcal{H}_\phi = (\Delta_x(B_0) + \partial B \Delta_x(B_0) \delta B(t)) \tau_x,
\]

where we have not included the term proportional to \( \Delta_x \parallel \omega \) since it does not cause spin dephasing. Calculating the spin dephasing rate within the Bloch-Redfield theory using Eq. (30) we find that

\[
\frac{1}{T_\phi} = \left( \frac{2}{\hbar} \partial B \Delta_x(B_0) \right)^2 \lim_{\omega \to 0} \int_0^\infty dt e^{-i\omega t} \langle \delta B(t) \delta B(t') \rangle ,
\]

where \( \langle A(t) \rangle \) is the thermal equilibrium expectation value of the operator \( A(t) \) on the bath. Therefore the spin dephasing rate goes to zero at the energy anticrossing, since \( \partial B \Delta_x(B_0) \to 0 \). This condition is valid under the assumption that the thermal average of the fluctuating magnetic field does not diverge.

Following the result given by Eq. (31), the spin dephasing rate still vanishes as long as the spectral density of the system-bath interaction is super-Ohmic, i.e., \( J(\omega) \propto \omega^s \), with \( s > 1 \).

VI. CONCLUSION

In summary, we find a minimum in the spin relaxation time as a function of the magnetic field that is induced by the Rashba SO coupling and is controllable by an external electric field. In larger quantum dots, the intrinsic SO dominates the spin relaxation over the Rashba SO contribution at low magnetic fields. As the magnetic field increases, the extrinsic contribution takes over, generating a non-monotonic behaviour of \( T_1 \) due to the Rashba SO interaction-induced level anticrossing. We have also analyzed the spectral density of the system-bath interaction for the first-order electron-phonon interaction and we have identified a vanishing contribution to the energy-conserving dephasing process. Therefore the phonon-induced pure spin dephasing rate is of the same order of magnitude as the spin relaxation rate, i.e., \( T_2 = 2T_1 \), in the leading order of the electron-phonon interaction. Other mechanisms such as nuclear spins from the \( ^{13}C \) atoms and charge noise combined with SO interaction could lead to a non-vanishing spin dephasing rate. Nevertheless, these mechanism are expected to be weak in graphene. Moreover, we have shown that any super-Ohmic bath has a vanishing spin dephasing rate at the energy anticrossing.

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