Phonon-assisted carrier tunneling in coupled quantum dot systems with hyperfine-induced spin flip

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We calculate the rates of phonon-assisted hyperfine spin flips during electron and hole tunneling in a QDM. We show that the hyperfine process dominates over the spin-orbit-induced spin relaxation in magnetic fields up to almost 1 T for electron, while for holes this cross-over takes place at fields below 0.01 T.

I. INTRODUCTION

The main source of dephasing of electron spins in self-assembled quantum dots (QDs) is their hyperfine (hf) interaction with the nuclear spins [1–5]. These processes are, however, inefficient in generating electron spin-flip-flop process due to large mismatch between electronic and nuclear Zeeman energies. They are suppressed even more for holes that interact with nuclei via dipole interactions that are weaker than the contact interaction of electrons [6–8]. In fact, for a purely p-type valence band the transverse coupling to carriers, that enables spin-flip processes, could only result from weak band mixing effects [6–8]. Recent experiments show that the main source of such coupling is the atomic d-shell admixture to valence band states [9]. In view of the Zeeman energy mismatch, spin-orbit (SO) coupling dominates over such simple spin-flip processes in spin relaxation within a Zeeman doublet already at moderate magnetic fields [9, 10].

In quantum dot molecules (QDMs), built of two coupled QDs, even in the s shell, spin relaxation can not only take place between states within one Zeeman doublet but can also accompany charge relaxation (dissipative tunneling) between the QDs. Reliable description of processes involving tunneling requires reasonable knowledge of wave functions, which is achievable with the k·p method in the envelope function approximation [11]. The k·p method allows one also to describe all kind of phonon-assisted processes, including those involving spin-orbit-induced spin flips [12]. Recently, we have also combined the k·p model with the hyperfine Hamiltonian and provided a description of hyperfine couplings based on multi-band wave functions [13].

In this paper we calculate the rates of phonon-assisted hyperfine spin flip-flops during electron and hole relaxation between the two branches of the s shell in a QDM (corresponding to states localized in two different QDs if the system is away from the resonance). We compare the result with the spin-orbit-induced phonon-assisted spin-flip tunneling and show that the hyperfine process dominates for fields below approximately 1 T for electrons, while for holes it becomes important only for fields below 0.01 T.

II. MODEL

We consider two coupled, vertically stacked self-assembled InGaAs quantum dots. The dots are placed on the wetting layers of 0.565 nm thickness and In0.4Ga0.6As composition. The shape of each dot is defined by the wetting layers of 0.4 nm. Both dots have a trumpet-shape 565 nm thickness and In0.4Ga0.6As composition[15], where position dependent In content is

FIG. 1. (Color online) Material distribution in the system (a) and scheme of the lowest energy levels (b).
given by

\[ C(r) = C_b + (C_t - C_b) \exp \left( \frac{-\sqrt{x^2 + y^2} \exp(-z/z_c)}{r_c} \right), \]

where \( C_t = 0.5, \ C_b = 0.3 \) are related to the composition in the top/bottom region of given QD, \( z_c = 1.2 \) nm and \( r_c = 0.6 \) nm. To simulate material intermixing, the dots are processed by Gaussian blur with the standard deviation of 0.6 nm.

The static strain related to the lattice mismatch is accounted for within continuous elasticity approach[10]. The strain-induced piezoelectric potential is calculated, where we account for the polarization up to the second order in strain tensor elements[17] and parameters are taken from[18]. The wave functions for the four lowest electron and hole states (corresponding to the two QDs and two spin orientations) are obtained using the eight band \( k \cdot p \) theory in the envelope function approximation. The full Hamiltonian, the material parameters and implementation is widely described in the Appendix of Ref.[19].

The hyperfine Hamiltonian for the interaction of the carrier with nuclear spins is

\[ H_{hf} = 3E_{hf} \sum_\alpha \zeta_\alpha \mathbf{A}(r - R_\alpha) \cdot \mathbf{I}_\alpha / \hbar, \]

where \( \alpha \) labels the ions, \( R_\alpha \) are their positions,

\[ E_{hf} = \frac{2\mu_0}{3\pi} \mu_B \mu_N a_B^{-3} = 0.5253 \text{ meV}, \]

\( \mu_B \) and \( \mu_N \) are Bohr and nuclear magnetons, respectively, \( a_B \) is the Bohr radius, \( \mu_0 \) is the vacuum permeability, \( I_\alpha \) is the nuclear spin, \( \zeta_\alpha \) defines the nuclear magnetic moment for a given nucleus via \( \mu_\alpha = \zeta_\alpha \mu_N I_\alpha \), and

\[ \mathbf{A}(r) = \frac{a_B^3}{4\hbar} \left[ \frac{8\pi}{3} \mathbf{S}(r) \cdot \mathbf{L} / \hbar^2 + \frac{3(\mathbf{r} \cdot \mathbf{S}) \mathbf{r} - \mathbf{S}}{r^3} \right]. \]

We consider the two most common In isotopes, two Ga isotopes and one As isotope with the angular momenta \( j = 9/2 \), \( 3/2 \), and \( 3/2 \), respectively. The full set of parameters describing the hyperfine coupling for an 8-component wave function as obtained in the \( k \cdot p \) method is given in Ref.[19]. The carrier Zeeman splitting is much larger than the nuclear one and we assume a temperature high enough to assume equal probability for any nuclear configuration at thermal equilibrium. The nuclear Zeeman energies are therefore neglected.

Coupling to phonons is described in the usual way... The phonon subsystem and the carrier-phonon interaction are described by the general Hamiltonian

\[ H_{ph} = \sum_{k, \lambda} \hbar \omega_{k, \lambda} b_{k, \lambda}^\dagger b_{k, \lambda} + \sum_{k, \lambda} \left\{ \Phi(r), e^{ik \cdot r} \right\} \left( b_{k, \lambda} + b_{k, \lambda}^\dagger \right), \]

where \( k \) and \( \lambda \) denote the wave vector and polarization of a phonon mode, respectively, \( b_{k, \lambda}, b_{k, \lambda}^\dagger \) are the corresponding annihilation and creation operators, and \( \Phi(r) \) is an \( 8 \times 8 \) matrix of operators in the coordinate representation, corresponding to the 8-band structure of the \( k \cdot p \) theory. The detailed description of spin-phonon Hamiltonian is given in Ref.[20].

III. PHONON-ASSISTED SPIN-FLIP TRANSITIONS

In this section we present the theory for the phonon-assisted tunneling of carriers between the ground state manifolds of the two QDs with a simultaneous spin flip-flop between the carrier and a nuclear spin. The 8-band \( k \cdot p \) theory treats the electron and hole states on equal footing (the latter upon a standard transition from the electron picture of the completely filled valence band to the hole picture) and we present our theory for single-particle states in a general form, without specifying the kind of the carrier. In the following, the term “spin” is used to identify one of the two the sub-bands of the conduction or heavy-hole valence band. We will focus on the transition from the

In order to find the rate for such a process we first calculate the hyperfine flip-flop correction to the system state. We denote the carrier state in the \( n \)th QD \((n = 1, 2)\) with the nominal spin orientation \( \sigma \) (resulting from the \( k \cdot p \) diagonalization) by \( |n\sigma\rangle \) and its energy by \( E_{\sigma}^{(n)} \). The state of the nuclei is labeled by \(|...m_\alpha,...\rangle\), where \( m_\alpha \) is the quantum number for the \( z \) projection of the respective nuclear spin. The states unperturbed by the hyperfine coupling are of a product form \(|n\sigma;...m_\alpha,...\rangle = |n\sigma\rangle \otimes |...m_\alpha,...\rangle\). In the lowest order of perturbation theory with respect to the hyperfine interaction the eigenstates of the system are then

\[ |\Psi_{n\sigma;...m_\alpha,...}\rangle = |n\sigma;...m_\alpha,...\rangle + \sum_\alpha c^{(n\sigma)}_\alpha ||\sigma\rangle;...m_\alpha + 1,...\rangle \]

\[ + \sum_\alpha c^{(n\sigma)}_{\alpha} ||\sigma\rangle;...m_\alpha - 1,...\rangle, \]

where \( \sigma \) denotes inverted spin and the nuclear configuration on the right-hand side is the same as the one on the left-hand side except for the one explicitly given modified spin. The coefficients of the perturbative correction are

\[ c^{(n\sigma)}_{\alpha} = \frac{\langle n\sigma;...m_\alpha + 1,...|H_{hf}|n\sigma;...m_\alpha,...\rangle}{\hbar \omega_{\sigma\sigma'}} \]

\[ = \frac{3E_{hf}(\zeta_\alpha)}{2\hbar \omega_{\sigma\sigma'}} \sqrt{j(j + 1) - m_\alpha (m_\alpha \pm 1)} \langle n\sigma|A_{\mp}|n\sigma\rangle, \]

where \( \omega_{\sigma\sigma'} = (E_{\sigma}^{(n)} - E_{\sigma'}^{(n)})/\hbar \).

From the Fermi golden rule, the probability of phonon-assisted transition from the state with spin \( \sigma \) in QD1 to the state with spin \( \sigma' \) in QD2 with a change of the nuclear
configuration from \{m_\alpha\} to \{m'_\alpha\} is
\begin{align*}
\Gamma_{\sigma \rightarrow \sigma'}^{\{m_\alpha\} \rightarrow \{m'_\alpha\}} &= \frac{2\pi}{\hbar} n_B(\omega_{\sigma \sigma'})^2 \left| \sum_{k, \lambda} \delta(\hbar \omega_{k, \lambda} - |\omega_{\sigma \sigma'}^{(12)}|) \right| \times \left| \langle \Psi_{2\sigma'; 3\sigma'; \ldots, \sigma' n_\sigma} | \{ \Phi(r), e^{ik \cdot r} \} | \Psi_{1\sigma; 3\sigma; \ldots, m_\alpha} \rangle \right|^2.
\end{align*}

Substituting the perturbation expansion from Eq. (3), taking into account the obvious fact that the phonon interaction conserves nuclear spins, and denoting
\begin{equation}
F_{\sigma' \sigma}(k) = \langle 2\sigma' | \{ \Phi(r), e^{ik \cdot r} \} | 1\sigma \rangle
\end{equation}

one finds
\begin{align}
\langle \Psi_{2\sigma'; \ldots, m_\alpha} | \{ \Phi(r), e^{ik \cdot r} \} | \Psi_{1\sigma; \ldots, m_\alpha} \rangle &= \sum_{\alpha, \pm} c^{(12)}_{\alpha, \pm} \langle m_\alpha | m_\alpha \pm 1 \rangle \ldots
+ \sum_{\alpha, \pm} c^{(12)*)}_{\alpha, \pm} \langle m'_\alpha | m_\alpha \pm 1 \rangle \ldots
+ \ldots.
\end{align}

Since the multi-band carrier wave functions are dominated by one leading component (determining the nominal ‘spin’ of the state), the couplings \(F_{\sigma' \sigma}(k)\) are large for \(\sigma = \sigma'\) and much smaller otherwise, when it relies only on spin-orbit couplings manifested by band mixing in the \(k \cdot p\) wave functions. The hyperfine admixture amplitudes \(c^{(12)}_{\alpha, \pm}\) are small, as well. Therefore, in Eq. (5) we kept only the contributions in the leading order in the spin-orbit or hyperfine couplings, neglecting those relying on both these weak couplings simultaneously. Furthermore, for a nominally spin-conserving process (\(\sigma = \sigma'\), the transition amplitude is by far dominated by the first contribution \(F_{\sigma \sigma}(k)\), which determines the spin-conserving phonon-assisted tunneling rate
\begin{equation}
\Gamma_{\sigma \rightarrow \sigma} = 2\pi R_{\sigma \sigma \sigma}(\omega_{\sigma \sigma}^{(12)}),
\end{equation}
where we define the spectral densities for the phonon bath as
\begin{equation}
R_{\sigma_1 \sigma_2 \sigma_3}(\omega) = \frac{1}{\hbar^2} |n_B(\omega) + 1| \times \sum_{k, \lambda} F_{\sigma_1 \sigma_2}(k) F_{\sigma_2 \sigma_3}(k) \delta(\hbar \omega_{k, \lambda} - |\omega|).
\end{equation}

For a spin-flip process, there are two mechanisms that may, in principle, yield comparable contributions: the spin-orbit channel entering via the first term and the hyperfine channel accounted for by the two other terms on the right-hand side of Eq. (7). The total spin-flip transition rate is then a sum of the spin-orbit rate,
\begin{equation}
\Gamma_{\sigma \rightarrow \sigma'}^{(so)} = 2\pi R_{\sigma \sigma' \sigma}(\omega_{\sigma \sigma}^{(12)}),
\end{equation}
and the hyperfine rates for transitions, summed up over final configurations of the nuclear bath, differing by one nuclear spin-flip from the initial one,
\begin{equation}
\Gamma_{\sigma \rightarrow \sigma'}^{(hf)}(\ldots m_\alpha \ldots) = \sum_{\alpha} \Gamma_{\sigma \rightarrow \sigma'}^{(hf), \alpha}(\ldots m_\alpha \ldots),
\end{equation}
where we explicitly noted the dependence on the initial configuration of the nuclear bath and
\begin{equation}
\Gamma_{\sigma \rightarrow \sigma'}^{(hf), \alpha}(\ldots m_\alpha \ldots) = 2\pi \sum_{a, \pi} \sum_{b, \pi} Q_{a \alpha b} R_{ab \alpha}(\omega_{\alpha}^{(12)}),
\end{equation}
with
\begin{align*}
Q_{a \sigma}^{(\alpha)} &= \left( \frac{3E_{hf} \Omega_{\alpha}}{2\hbar \omega_{\sigma}^{(12)}} \right)^2 \left\{ [j(j+1) - m_\alpha(m_\alpha-1)] |(2\sigma | A_- | 2\sigma')|^2 + [j(j+1) - m_\alpha(m_\alpha+1)] |(2\sigma | A_+ | 2\sigma')|^2 \right\},
\end{align*}
\begin{align*}
Q_{a \sigma}^{(\alpha)} &= \left( \frac{3E_{hf} \Omega_{\alpha}}{2\hbar \omega_{\sigma}^{(12)}} \right)^2 \left\{ [j(j+1) - m_\alpha(m_\alpha+1)] |(1\sigma | A_- | 1\sigma')|^2 + [j(j+1) - m_\alpha(m_\alpha-1)] |(1\sigma | A_+ | 1\sigma')|^2 \right\},
\end{align*}
\begin{align*}
Q_{a \sigma}^{(\alpha)} &= \left( \frac{3E_{hf} \Omega_{\alpha}}{4\hbar^2 \omega_{\sigma}^{(12)}} \right)^2 \left\{ [j(j+1) - m_\alpha(m_\alpha+1)] |(1\sigma | A_- | 1\sigma') (2\sigma | A_+ | 2\sigma') \right\} + [j(j+1) - m_\alpha(m_\alpha-1)] |(1\sigma | A_+ | 1\sigma') (2\sigma | A_- | 2\sigma'|) \right\}.
\end{align*}

Note that \(R_{\sigma \sigma \sigma}(\omega) = R_{\sigma \sigma \sigma}^{(so)}(\omega)\) so \(\Gamma_{\sigma \rightarrow \sigma'}^{(hf), \alpha}\) is real.

Since the shape of the wave function for a given spatial state in a given QD very weakly depends on the spin orientation, all the spectral densities in Eq. (11) are almost identical upon an appropriate choice of the arbitrary phases. This allows one to write
For a transition between the Zeeman states in a single QD the distinction between \textquotedblleft 1\textquotedblright{} and \textquotedblleft 2\textquotedblright{} disappears, the two matrix elements of $A_\pm$ become identical and the transition rate is suppressed by destructive interference. In contrast, in the DQD system the states are spatially separated each is effectively coupled to at most to one carrier state (the one localized in the same QD as the ion) and only one of the two interfering amplitudes can be large.

For unpolarized nuclei the physically meaningful rate is obtained by averaging Eq. (10) over all the initial configurations of the nuclear spins and summing up over all nuclear spin flips,

$$\Gamma_{\sigma\rightarrow\sigma}^{(hf)} = 2\pi \sum_{a=\sigma,\bar{\sigma}} \sum_{b=\sigma,\bar{\sigma}} \bar{Q}_{ab} R_{abbb}(\omega^{(12)}_\alpha), \quad (12)$$

Since $\langle j+1 - m_\alpha (m_\alpha - m_\beta) \rangle = 2(j+1)/3$ one finds

$$\bar{Q}_{\sigma\sigma}^{(\alpha)} = \sum_{\alpha} \left(3E_{hf}\zeta_\alpha \over 2\hbar\omega^{(22)}_\alpha \right)^2 \frac{2j(j+1)}{3} \times \left( |\langle 2\sigma | A_- | 2\sigma \rangle |^2 + |\langle 2\sigma | A_+ | 2\sigma \rangle |^2 \right) \quad (13a)$$

$$Q_{\sigma\sigma}^{(\alpha)} = \sum_{\alpha} \left(3E_{hf}\zeta_\alpha \over 2\hbar\omega^{(11)}_\alpha \right)^2 \frac{2j(j+1)}{3} \times \left( |\langle 1\sigma | A_- | 1\sigma \rangle |^2 + |\langle 1\sigma | A_+ | 1\sigma \rangle |^2 \right) \quad (13b)$$

$$\bar{Q}_{\sigma\sigma}^{(\alpha)} = Q_{\sigma\sigma}^{(\alpha)} = \sum_{\alpha} \left(3E_{hf}\zeta_\alpha \over 4\hbar^2\omega^{(11)}_\alpha \omega^{(22)}_\alpha \right)^2 \frac{2j(j+1)}{3} \times \left( |\langle 1\sigma | A_- | 1\sigma \rangle \langle 2\sigma | A_+ | 2\sigma \rangle \right) \quad (13c)$$

\[ \text{wave functions according to the definitions in Eq. (4) and Eq. (8).} \]

In Fig. 2 we show the spin-flip tunneling rates for the electron in presence of magnetic field $B$ and electric field $F$ (both oriented in $z$ direction). It is clearly seen, that all of the rates oscillate with the electric field. This is related to the interference effects of phonon emission in growth direction. The period of such oscillations depends on the speed of sound and the distance between the dots [21]. At weak magnetic fields (we fixed $B = 0.1 \text{T}$) the hyperfine channel dominates over the spin-orbit one, and the resulting spin-flip transition becomes faster. On the other hand, for moderate and strong values of magnetic field
the ratio \( \Gamma_{(so)} / \Gamma_{(hf)} \) exhibits pronounced maxima.

The results for the hole are presented in Fig. 4. In that case, the hyperfine-induced spin flip is much weaker compared to the electron one, as expected, while the spin-orbit-induced process is a few times more effective. As a result, the cross-over between the predominantly spin-orbit and predominantly hyperfine processes takes place already at magnetic fields below 0.01 T.

V. CONCLUSIONS

We have calculated the rates of phonon-assisted hyperfine spin flips during electron and hole tunneling in a QDM. We have shown that the hyperfine process dominates over the spin-orbit-induced spin relaxation in magnetic fields up to almost 1 T for electron, while for holes this cross-over takes place at much smaller fields.

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