Singlet-triplet dephasing in asymmetric quantum dot molecules

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We discuss pure dephasing of singlet-triplet superpositions in two-electron double quantum dots due to elastic phonon scattering. We generalize our previous results to a system built of two non-identical dots. We show that the asymmetry must be very strong in order to considerably affect the dephasing rate.

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The Hamiltonian of the system is then

\[ H = H_{\text{DQD}} + H_{\text{ph}} + H_{\text{int}}. \]

The first term describes the electrons and has the form

\[ H_{\text{DQD}} = \Delta \sum_s \left( a_{sL}^\dagger a_{sL} - a_{sR}^\dagger a_{sR} \right) - t_1 \sum_s \left( a_{sL}^\dagger a_{sR} + \text{h.c.} \right) + \frac{1}{2} \sum_{s,s', i,j,k,l} V_{ijkl} a_{is}^\dagger a_{js} a_{ks'}^\dagger a_{ls}, \]

respectively, and \( s = \uparrow, \downarrow \) labeling the spin orientation. The first term in Eq. (1) accounts for the energy difference between single-electron states in the two dots. The second term represents single-particle inter-dot tunneling. The third term describes the Coulomb interaction, with \( V_{ijkl} = V_{ijlk} = V_{kijl} = V_{lkji} \) (the wave functions may be chosen such that the matrix elements are real).

The Hamiltonian of the phonon reservoir is given by

\[ H_{\text{ph}} = \sum_{\lambda, k} \hbar \omega_{k, \lambda} b_{k, \lambda}^\dagger b_{k, \lambda}, \]

where \( b_{k, \lambda}^\dagger, b_{k, \lambda} \) are annihilation and creation operators for a phonon from a branch \( \lambda \) with a wave vector \( k \) and \( \hbar \omega_{k, \lambda} \) are the corresponding energies. The electron-phonon interaction is described by

\[ H_{\text{int}} = \sum_{s,i} \sum_{k,\lambda} F^{(\lambda)}(k) a_{is}^\dagger a_{is}(b_{k, \lambda} + b_{k, \lambda}^\dagger), \]

where \( F^{(\lambda)}(k) \) are coupling constants. We use the usual coupling constants for confined charges (see Refs. [2] [3] for explicit expressions).

The Hamiltonian \( H_{\text{DQD}} \) can be diagonalized in a simple way. The resulting singlet eigenstates can be written in the form

\[ |S_0\rangle = \cos \frac{\phi}{2} \left[ \cos \frac{\theta}{2}(1,1)S - \sin \frac{\theta}{2}(+)(S) \right] - \sin \frac{\phi}{2} (-)(S) \]

\[ |S_1\rangle = -\sin \frac{\phi}{2} \left[ \cos \frac{\theta}{2}(1,1)S - \sin \frac{\theta}{2}(+)(S) \right] + \cos \frac{\phi}{2} (-)(S) \]

\[ |S_+\rangle = \sin \frac{\theta}{2}(1,1)S + \cos \frac{\theta}{2}(+)(S), \]

where \(|(\pm)(S)\rangle = |(2,0)(S) \pm |(0,2)(S)\rangle|/\sqrt{2} \) and \(|(m,n)(S)\rangle \) denotes the configuration with \( m \) electrons in the left dot and \( n \) electrons in the right one. The corresponding energies are denoted by \( E_0, E_1, E_+ \).

Here \( \theta \) describes mixing of the \(|(1,1)(S)\rangle \) and \(|(+)(S)\rangle \) configurations due to tunneling and Coulomb interactions. In the present work we fix \( \theta = 0.5 \), which corresponds to \( t_1/(E_1-E_0) = 0.2 \).

The angle \( \phi \) accounts for state mixing due to system asymmetry. The latter may be due to a difference be-
tween the energies of doubly occupied configurations in the two dots or to asymmetry of off-diagonal Coulomb elements. Accordingly, we define two parameters

\[
\Delta = 2\Delta_c + \frac{V_{RRR} - V_{LLL}}{2},
\]

\[
\eta = \frac{V_{RRR} + V_{LLL} - V_{LRR} - V_{RLL}}{2}.
\]

Neglecting the exchange terms compared to direct Coulomb energies, one finds \(\sin \phi \approx \sin(\theta/2)\Delta - \cos(\theta/2)\eta/(E_1 - E_0)\).

The carrier-phonon coupling for the two-electron system is written in the eigenbasis of \(H_{\text{DQD}}\),

\[
H_{\text{int}} = \sum_{\alpha,\beta, k, \lambda} F^{(\lambda)}_{\alpha\beta}(k)|\alpha\rangle\langle\beta|\left(b_{k,\lambda} + b^\dagger_{-k,\lambda}\right),
\]

with \(\alpha, \beta = S_0, S_1, S_+\). Then, the phonon spectral densities involving the low-energy state \(|S_0\rangle\) are found,

\[
R_{\alpha\beta}(\omega) = \sum_{k, \lambda} F^{(\lambda)}_{\alpha\beta}(k)F^{(\lambda)\ast}_{\alpha\beta}(k)|n(\omega) + 1|\delta(|\omega| - \omega_k),
\]

where \(\alpha, \beta = S_1, S_+\) and \(n(\omega)\) is the Bose distribution. In contrast to the previously studied symmetric case, two channels of phonon scattering are now present, corresponding to the two different excited states \((S_1 \text{ and } S_+)\) through which the phonons can scatter. The interference of different scattering paths is reflected by the presence of the whole family of spectral densities out of which only one, \(R_{S_0S_0}(\omega)\) survives for \(\phi \to 0\) and reduces to the spectral density found in Ref. [2].

Following the method worked out in our previous paper [2], we calculate the dephasing rate using the 4th order time-convolutionless equation for the evolution of the density matrix describing the state of the two-electron system. From this, we extract the Markov limit, which yields the dephasing rate due to two-phonon (scattering) processes. Similarly as in the symmetric case [2], this process dominates over the one-phonon real transitions at low temperatures. The resulting dephasing rate is given by

\[
\gamma = \pi \mathcal{P} \sum_{\alpha, \beta} \int_{-\infty}^{\infty} d\omega \left[ \frac{R_{\alpha\beta}(\omega)R_{\alpha\beta}(-\omega)}{(\omega + \omega_\alpha)(\omega - \omega_\beta)} + \frac{R_{\alpha\beta}(\omega)R_{\alpha\beta}(-\omega) - R_{\alpha\beta}(\omega_\alpha)R_{\alpha\beta}(-\omega_\beta)}{(\omega - \omega_\alpha)(\omega - \omega_\beta)} \right],
\]

where \(\omega_\alpha = (E_\alpha - E_0)/\hbar\) and \(\mathcal{P}\) denotes the Cauchy principal value. The above equation generalizes the previously found formula to the asymmetric case.

In the calculations, we set the energy differences \(E_1 - E_0\) = 0.9 meV, \(E_+ - E_0\) = 1.0 meV, the inter-dot distance \(D = 300\) nm, and use the material parameters as in Ref. [2].

The pure dephasing rates resulting from the scattering process are shown in Figs. 1(a,c) as a function of \(\Delta\) for a few values of \(\eta\) and in Fig. 1(b,d) as a function of \(\eta\) for a few values of \(\Delta\). In all the cases, the dephasing time varies from about 1 \(\mu\)s to several \(\mu\)s. As long as the asymmetry is small, the dephasing rate is rather insensitive to \(\eta\). Only for strong asymmetry, the dephasing rate grows considerably. One should keep in mind that the largest values of \(\eta\) shown in Fig. 1 are rather unrealistic since this parameter describes the variation of exchange-like Coulomb terms which are themselves small. In a real structure, \(\eta\) is likely to remain within a tenth of meV. It is clear from Fig. 1 that in this parameter range the dephasing rate is almost completely insensitive to \(\eta\), as well as to \(\Delta\).

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