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Spin density matrix of a two-electron system. II. Application to a system of two quantum dots

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This work is a sequel to our preceding work [Phys. Rev. B **77**, 045319 (2008)]. Here we compare *pure-spin* and *pseudospin* dynamics using as an example a system of two quantum dots, a pair of localized conduction-band electrons in an *n*-doped GaAs semiconductor. Pure-spin dynamics is obtained by tracing out the orbital degrees of freedom, whereas pseudospin dynamics retains (as is conventional) an implicit coordinate dependence. We show that magnetic field inhomogeneity and spin-orbit interaction result in a nonunitary evolution in pure-spin dynamics, whereas these interactions contribute to the effective pseudospin Hamiltonian via terms that are asymmetric in spin permutations, in particular, the Dzyaloshinskii-Moriya (DM) spin-orbit interaction. We numerically investigate the nonunitary effects in the dynamics of the triplet-state population, purity, and Lamb energy shift as a function of interdot distance and magnetic field difference $\Delta\vec{B}$. The spin-orbit interaction is found to produce effects of roughly 4 orders of magnitude smaller than those due to $\Delta\vec{B}$ in the pure-spin model. We estimate the spin-orbit interaction magnitude in the DM-interaction term. Our estimate gives a smaller value than that recently obtained by Kavokin [Phys. Rev. B **64**, 075305 (2001)], who did not include double-occupancy effects. We show that a necessary and sufficient condition for obtaining a universal set of quantum logic gates, involving only two spins, in both pure-spin and pseudospin models is that the magnetic field inhomogeneity $\Delta\vec{B}$ and the Heisenberg interaction are both nonvanishing. We also briefly analyze pure-spin dynamics in the electron on liquid helium system recently proposed by Lyon [Phys. Rev. A **74**, 052338 (2006)].

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I. INTRODUCTION

The spin degree of freedom of a localized particle, e.g., an electron or nucleus, is a popular carrier of quantum information. It serves as a qubit which can be manipulated in order to accomplish a computational task. The spin of electrons localized in quantum dots (QDs) or by donor atoms has been the subject of extensive recent studies.[1–15]

Consider two electrons trapped in two sites, A and B, e.g., two QDs each containing one electron. The two-electron system is fully described by the total wave function $|\Psi_{\text{tot}}\rangle$, which depends on the electrons' coordinates $\vec{r}$ and spin variables σ. The two-electron spin density matrix, obtained by tracing out the orbital degrees of freedom, $\rho=\text{Tr}_{\vec{r}}|\Psi_{\text{tot}}\rangle\langle\Psi_{\text{tot}}|$, fully describes the spin dynamics as long as one cannot or does not wish to apply measurements that can separate or localize electrons spatially; the only observable is then the electron spin, $s^{\alpha}=\frac{1}{2}\sigma_{\alpha}$, where σ_{α} are the Pauli spin one-half matrices with $\alpha=x,y,z$. Since the spin system is not closed—there is a coupling to the electrons' spatial degrees of freedom—we observe open system effects, i.e., the spin dynamics becomes, in general, nonunitary. We refer to this dynamics as *pure-spin* dynamics.

In contrast, *pseudospin* dynamics is the standard case where the electron spin observable is not free from coordinate dependence but includes information about the electron's localization orbital. In the pseudospin case, one defines the electron spin operator as a bilinear combination of electron annihilation and creation Fermi operators, c_{As}, $c_{As}^{\dagger}$, in a localized orbital ϕ_A (s is a spin index and A is the QD index): $s_A^{\alpha}=\frac{1}{2}\Sigma_{ss'=1}^{2}c_{As}^{\dagger}(\sigma_{\alpha})_{ss'}c_{As'}$, $\alpha=x,y,z$. Then, the operators $\{s_A^{\alpha}\}_{\alpha}$ obey the usual $su(2)$ commutation rules.

This paper is the sequel to our work in Ref. 16 (henceforth "Part I"), where we derived an operator-sum representation as well as a master equation in the Lindblad and time-convolutionless forms for the spin density matrix of a two-electron system. In this sequel, we focus on a detailed comparison of pure-spin and pseudospin dynamics. We are interested, in particular, in how nonunitary effects in pure-spin dynamics are translated into the corresponding unitary ones in pseudospin dynamics and vice versa. We show that as long as there is no magnetic field inhomogeneity, the pure-spin dynamics is unitary, but in the presence of magnetic field inhomogeneity, this dynamics is nonunitary.

The paper is organized as follows. We begin in Sec. II by highlighting the differences and relationship between pseudospin and pure-spin models. Section III provides a concrete illustration in terms of a system of two QDs trapping one electron each. In it, we examine the role of the different interactions in both pseudospin and pure-spin dynamics. We first derive the coordinate part of the Hamiltonian (Sec. III A) and the form of the dipolar interaction (Sec. III B). In Secs. III C and III D, respectively, we then present calculations illustrating effects due to both external magnetic field inhomogeneity and the spin-orbit interaction in the pure-spin model. In Secs. III E and III F, we discuss universal quantum gates in both pseudospin and pure-spin models. Section III G presents our estimates for spin-orbit interaction effects in the pseudospin model and compares these estimates to the results of Ref. 17. We conclude in Sec. V.

Atomic units, $\hbar = e = m_e = 1$, $1/c \simeq 1/137$, are used throughout the paper unless stated otherwise.

II. PSEUDOSPIN VERSUS PURE-SPIN APPROACHES

In this section, we discuss the relation between the present approach based on the spin density matrix and the pseudospin effective Hamiltonian approach. The latter is usually developed as a low-energy mapping within the Hubbard model Hamiltonian of interacting electrons.[9,11,18–23] We do not follow the Hubbard model since it is highly simplified and neglects many interactions that we would like to keep here. For Hubbard model analyses in the quantum computation context, see, e.g., Ref. 9.

A. Pseudospin effective Hamiltonian

In order to keep the present treatment as simple as possible, we restrict ourselves to the two-orbital approximation used in Part I; inclusion of excited-state orbitals is straightforward. Consider the four single-occupancy basis states $\{\Phi_{s1}, \Phi_{ti}, i=1,2,3\}$, where Φ_{s1} is a singlet wave function with two electrons localized on different QDs, A and B, while Φ_{ti} are the corresponding triplet wave functions. The two double-occupancy states $\{\Phi_{s2}, \Phi_{s3}\}$ describe two electrons in a singlet state, localized on the same QD, A or B. The total wave function $\Psi_{\text{tot}}(t)$ in this basis set takes the form

$$\Psi_{\text{tot}}(t) = \sum_{i=1}^{3} [a_{si}(t)\Phi_{si} + a_{ti}(t)\Phi_{ti}],$$

where the complex amplitudes $\{a_{si}(t), a_{ti}(t)\}$ define, respectively, the singlet- and triplet-state populations. In the total Hilbert space, the state is defined by 11 real parameters (12 real parameters defining $\{a_{si}(t), a_{ti}(t)\}$ minus a normalization condition). The unitary evolution in the total Hilbert space is described by

$$\begin{pmatrix} |a_s(t)\rangle \\ |a_t(t)\rangle \end{pmatrix} = \exp(-iHt)\begin{pmatrix} |a_s(0)\rangle \\ |a_t(0)\rangle \end{pmatrix},$$

where H is the total two-electron system Hamiltonian.

Since these basis states are orthonormal, projection operators into the corresponding subspaces can be written as

$$P = |\Phi_{s1}\rangle\langle\Phi_{s1}| + \sum_{i=1}^{3} |\Phi_{ti}\rangle\langle\Phi_{ti}|,$$

$$Q = |\Phi_{s2}\rangle\langle\Phi_{s2}| + |\Phi_{s3}\rangle\langle\Phi_{s3}|, \tag{1}$$

where Q projects onto the double-occupancy states. Then, using the method of projection operators, one obtains the Schrödinger (eigenvalue) equation projected into the P subspace,

$$[\mathcal{H}_{\text{eff}}(E) - E]P\Psi = 0, \tag{2}$$

where

$$\mathcal{H}_{\text{eff}}(E) = PHP + PHQ\frac{1}{E - QHQ}QHP. \tag{3}$$

Observe that Eq. (2) is *exact* but nonlinear and has six solutions.

Due to interelectron repulsion, the double-occupancy states are usually much more energetic than the singly occupied ones if the electrons are well localized in QDs. We consider the low-energy physics described by Eq. (2), where the total energy E is near the energies of singly occupied states. In general, $\mathcal{H}_{\text{eff}}$ is not a Hamiltonian since it is a function of the energy E. However, if the energy gap between the P and Q states is large enough, one can expand and approximate

$$\begin{aligned}\mathcal{H}_{\text{eff}}(E) &= \mathcal{H}_{\text{eff}}(\bar{E}) + \sum_{n=1}^{\infty} PHQ\frac{(\bar{E}-E)^n}{(\bar{E}-QHQ)^{n+1}}QHP \\ &= \mathcal{H}_{\text{eff}}(\bar{E}) + \mathcal{H}_{\text{eff}}^{(1)}(\bar{E})(\bar{E}-E) + O[(\bar{E}-E)^2] \\ &\approx \mathcal{H}_{\text{eff}}(\bar{E}) + \mathcal{H}_{\text{eff}}^{(1)}(\bar{E})(\bar{E}-E),\end{aligned} \tag{4}$$

where $\bar{E}$ is the average energy in the P subspace and $\mathcal{H}_{\text{eff}}^{(1)}(\bar{E}) = PHQ(\bar{E}-QHQ)^{-2}QHP$. Keeping terms up to the first order in Eq. (4), the nonlinear Eq. (2) can be reduced to a generalized linear equation problem,

$$\{\mathcal{H}_{\text{eff}}(\bar{E}) + \mathcal{H}_{\text{eff}}^{(1)}(\bar{E})\bar{E} - [1 + \mathcal{H}_{\text{eff}}^{(1)}(\bar{E})]E\}P\Psi = 0. \tag{5}$$

Solving Eq. (5), we obtain four low-energy solutions; the two high-energy, double-occupancy solutions are lost in this approximation. Therefore, in the low-energy, pseudospin approximation, the state is described by seven real parameters.

In the following, we assume $\mathcal{H}_{\text{eff}}^{(1)}(\bar{E}) \equiv 0$ for simplicity. The effective Hamiltonian Eq. (4) can be recast into a pseudospin form. Using Eq. (1), we have

$$\begin{aligned}\mathcal{H}_{\text{eff}} &= \mathcal{H}^{ss}|\Phi_{s1}\rangle\langle\Phi_{s1}| + \sum_{i,j=1}^{3} \mathcal{H}_{ij}^{tt}|\Phi_{ti}\rangle\langle\Phi_{tj}| \\ &\quad + \sum_{i=1}^{3} (\mathcal{H}_i^{st}|\Phi_{s1}\rangle\langle\Phi_{ti}| + \mathcal{H}_i^{ts}|\Phi_{ti}\rangle\langle\Phi_{s1}|),\end{aligned} \tag{6}$$

where

$$\mathcal{H}^{ss} = \langle\Phi_{s1}|\mathcal{H}_{\text{eff}}(\bar{E})|\Phi_{s1}\rangle, \quad \mathcal{H}_{ij}^{tt} = \langle\Phi_{ti}|\mathcal{H}_{\text{eff}}(\bar{E})|\Phi_{tj}\rangle,$$

$$\mathcal{H}_i^{st} = \langle\Phi_{s1}|\mathcal{H}_{\text{eff}}(\bar{E})|\Phi_{ti}\rangle, \quad \mathcal{H}_i^{ts} = (\mathcal{H}_i^{st})^*. \tag{7}$$

In the second quantization representation, the P-subspace basis vectors take the form

$$|\Phi_{s1}\rangle = \tfrac{1}{\sqrt{2}}(c_{A\uparrow}^\dagger c_{B\downarrow}^\dagger - c_{A\downarrow}^\dagger c_{B\uparrow}^\dagger)|0\rangle = \tfrac{1}{\sqrt{2}}(|\uparrow\rangle_A \otimes |\downarrow\rangle_B - |\downarrow\rangle_A \otimes |\uparrow\rangle_B),$$

$$|\Phi_{t1}\rangle = c_{A\uparrow}^\dagger c_{B\uparrow}^\dagger|0\rangle = |\uparrow\rangle_A \otimes |\uparrow\rangle_B,$$

$$|\Phi_{t2}\rangle = \tfrac{1}{\sqrt{2}}(c_{A\uparrow}^\dagger c_{B\downarrow}^\dagger + c_{A\downarrow}^\dagger c_{B\uparrow}^\dagger)|0\rangle = \tfrac{1}{\sqrt{2}}(|\uparrow\rangle_A \otimes |\downarrow\rangle_B + |\downarrow\rangle_A \otimes |\uparrow\rangle_B),$$

$$|\Phi_{t3}\rangle = c_{A\downarrow}^\dagger c_{B\downarrow}^\dagger|0\rangle = |\downarrow\rangle_A \otimes |\downarrow\rangle_B, \tag{8}$$

where we introduced pseudospin states $|s\rangle_\alpha$, $s=\uparrow,\downarrow$, $\alpha=A,B$, localized near the A and B sites (the term pseudospin emphasizes the fact that these are not really spin states since they depend on the electron orbital degrees of freedom). Equation (8) establishes a one-to-one correspondence between four basis states $\{\Phi_{s1},\Phi_{ti},i=1,2,3\}$ and four tensor-product pseudospin states $|s\rangle_\alpha\otimes|s'\rangle_\beta$, where $s,s'=\uparrow,\downarrow$, $\alpha,\beta=A,B$. Then, relabeling the pseudospin states as $|0,1\rangle=|\uparrow,\downarrow\rangle$ and introducing the pseudospin Pauli and identity operators,

$$\sigma_x=|0\rangle\langle 1|+|1\rangle\langle 0|,$$

$$\sigma_y=-i(|0\rangle\langle 1|-|1\rangle\langle 0|),$$

$$\sigma_z=|0\rangle\langle 0|-|1\rangle\langle 1|,$$

$$I=|0\rangle\langle 0|+|1\rangle\langle 1|, \tag{9}$$

where we temporarily dropped the subscripts A and B, one easily finds that

$$|\Phi_{s1}\rangle\langle\Phi_{s1}|=\mathbf{S},\quad |\Phi_{ti}\rangle\langle\Phi_{tj}|=\mathbf{T}_{ij},$$

$$|\Phi_{s1}\rangle\langle\Phi_{ti}|=\mathbf{K}_i. \tag{10}$$

Here the pseudospin operators $\mathbf{S}$ and $\mathbf{T}_{ij}$ are defined as

$$\mathbf{S}=\tfrac{1}{4}I-\vec{s}_A\cdot\vec{s}_B,\quad \mathbf{T}_{11}=\tfrac{1}{4}I+\tfrac{1}{2}S_z+s_{Az}s_{Bz},$$

$$\mathbf{T}_{22}=\tfrac{1}{4}I+s_{Ax}s_{Bx}+s_{Ay}s_{By}-s_{Az}s_{Bz},$$

$$\mathbf{T}_{33}=\tfrac{1}{4}I-\tfrac{1}{2}S_z+s_{Az}s_{Bz},$$

$$\mathbf{T}_{12}=\tfrac{1}{\sqrt{2}}\left[\tfrac{1}{2}S_+ +J_s\right],\quad \mathbf{T}_{23}=\tfrac{1}{\sqrt{2}}\left[\tfrac{1}{2}S_+ -J_s\right],$$

$$\mathbf{T}_{13}=s_{Ax}s_{Bx}-s_{Ay}s_{By}+i(s_{Ax}s_{By}+s_{Bx}s_{Ay}),$$

$$\mathbf{T}=\tfrac{3}{4}I+\vec{s}_A\cdot\vec{s}_B,\quad \mathbf{T}_{21}=\mathbf{T}_{12}^\dagger,$$

$$\mathbf{T}_{31}=\mathbf{T}_{13}^\dagger,\quad \mathbf{T}_{32}=\mathbf{T}_{23}^\dagger, \tag{11}$$

where

$$J_s=s_{Az}s_{Bx}+s_{Ax}s_{Bz}+i(s_{Az}s_{By}+s_{Ay}s_{Bz}),$$

$$S_\pm=S_x\pm iS_y,\quad \vec{S}=\vec{s}_A+\vec{s}_B,$$

and $\mathbf{K}$ is defined as

$$\mathbf{K}_1=-\frac{i}{2\sqrt{2}}\{(\vec{J}_{as})_x-i(\vec{J}_{as})_y\},\quad \mathbf{K}_2=\frac{i}{2}(\vec{J}_{as})_z,$$

$$\mathbf{K}_3=\frac{i}{2\sqrt{2}}\{(\vec{J}_{as})_x+i(\vec{J}_{as})_y\}, \tag{12}$$

where $\vec{J}_{as}=[\vec{s}_B-\vec{s}_A\times\vec{S}]$. In fact, Eqs. (11) and (12) can be obtained from the corresponding ones in Part I if the pure-spin operators $\vec{s}_{1,2}$ are replaced, respectively, by the pseudospin ones, $\vec{s}_{A,B}$. We reproduce these formulas here in order to make the presentation as self-contained as possible.

As is seen from Eqs. (11) and (12), the first line of Eq. (6) is symmetric with respect to spin permutations $[A\leftrightarrow B]$, while the second one is asymmetric representing, in particular, the Dzyaloshinskii-Moriya (DM)-type interaction term.[24,25] Notice that these asymmetric (in spin permutations) terms cancel out of unitary spin dynamics after averaging over orbital degrees of freedom, as demonstrated in Part I. However, they do not completely disappear but rather are converted into the corresponding nonunitary terms plus the *Lamb shift* term as will be seen in the next subsection. From the symmetric part of the Hamiltonian Eq. (6), using Eq. (11), one can derive the isotropic Heisenberg exchange interaction term,

$$\mathcal{H}_H=J_H\vec{s}_A\cdot\vec{s}_B, \tag{13}$$

where the Heisenberg exchange interaction constant $J_H=\frac{1}{3}\Sigma_i\mathcal{H}_{ii}^{tt}-\mathcal{H}^{ss}$; in contrast, as was demonstrated in Part I, the Heisenberg interaction term does not affect the unitary evolution of the spin density matrix except for the Lamb energy shift. In Sec. III C, we demonstrate numerically the effects of the Heisenberg interaction on both the Lamb energy shift and the nonunitary part of the spin density matrix evolution.

Observe that the asymmetric part of the Hamiltonian Eq. (6) is proportional to the singlet-triplet subspace interaction matrix $\mathcal{H}_i^{st}$, which is responsible for the coupling between singlet and triplet states. As will be demonstrated in Sec. III, the nonzero coupling between these states is due to $\vec{B}$-field spatial inhomogeneity (i.e., it cannot arise due to the homogeneous component of the external magnetic field), as well as due to the spin-orbit interaction.

B. Spin density matrix

In Part I, we derived the Lindblad-type master equation for the spin density matrix,

$$\frac{\partial\rho(t)}{\partial t}=-i[\tilde{\mathbf{H}}_\alpha^{tt},\rho(t)]+\mathcal{L}_\alpha[\rho(t)],$$

$$\tilde{\mathbf{H}}_\alpha^{tt}=\sum_{ij}\left(H^{tt}+\frac{1}{2}P_\alpha\right)_{ij}\mathbf{T}_{ij}=\mathbf{H}^{tt}+\frac{1}{2}\mathbf{P}_\alpha,$$

$$\mathcal{L}_\alpha[\rho(t)]=\frac{1}{2}\sum_{ij}(\chi_\alpha)_{ij}([\mathbf{K}_i,\rho(t)\mathbf{K}_j^\dagger]+[\mathbf{K}_i\rho(t),\mathbf{K}_j^\dagger]), \tag{14}$$

where the first and second terms describe, respectively, unitary and nonunitary contributions to the evolution. $\tilde{\mathbf{H}}_\alpha^{tt}$ is an effective pure-spin Hamiltonian which includes the Lamb shift term, $\frac{1}{2}\mathbf{P}_\alpha$; the pure-spin operators $\mathbf{T}_{ij}$ and $\mathbf{K}_i$ are defined by Eqs. (11) and (12) where $\vec{s}_{A,B}\rightarrow\vec{s}_{1,2}$. The index $\alpha=\{s,t,m\}$ specifies which initial state $\rho(0)$, singlet s, triplet t, or a mixed one m, is taken.

As mentioned in Part I, all the matrix functions in Eq. (14) as well as the pseudospin Hamiltonian Eq. (6) are expressible in terms of H matrix elements,

$$H = \begin{pmatrix} H^{ss} & H^{st} \\ H^{ts} & H^{tt} \end{pmatrix},$$

$$H_{ij}^{\alpha\beta} = \langle \Phi_{\alpha i} | H | \Phi_{\beta j} \rangle, \quad \alpha,\beta = s,t, \quad i,j = 1,2,3. \tag{15}$$

In the following example, we consider the triplet case for which we have

$$\chi_\alpha^T = i(Q_\alpha - Q_\alpha^\dagger),$$

$$P_\alpha = Q_\alpha + Q_\alpha^\dagger, \tag{16}$$

where

$$Q_\alpha = \sum_k \exp(-i\varepsilon_k t) H^{ts} [|e_{sk}\rangle\langle e_{sk}| R_\alpha(0) + |e_{sk}\rangle\langle e_{tk}|] \times \left(\sum_k \exp(-i\varepsilon_k t) [|e_{tk}\rangle\langle e_{sk}| R_\alpha(0) + |e_{tk}\rangle\langle e_{tk}|] \right)^{-1}. \tag{17}$$

Here, $R_\alpha(0)$ is a correlation matrix, which establishes an initial correlation between the singlet and triplet amplitudes,

$$a_s(0) = R_\alpha(0) a_t(0) \tag{18}$$

[in the triplet case, we have $R_{\alpha=t}(0) \equiv 0$; in the mixed case, where both $a_s(0) \neq 0$ and $a_t(0) \neq 0$, $R_{\alpha=m}(0) \neq 0$; for the singlet case, see Part I] and ε_k, $|e_{sk}\rangle$, and $|e_{tk}\rangle$ are solutions to the eigenvalue problem,

$$\begin{pmatrix} H^{ss} & H^{st} \\ H^{ts} & H^{tt} \end{pmatrix} \begin{pmatrix} |e_{sk}\rangle \\ |e_{tk}\rangle \end{pmatrix} = \varepsilon_k \begin{pmatrix} |e_{sk}\rangle \\ |e_{tk}\rangle \end{pmatrix}, \quad k = 1, \ldots, 6. \tag{19}$$

III. EXAMPLE: SYSTEM OF TWO QUANTUM DOTS

In this section, we investigate the role of different interactions in the calculation of the H matrix. Let us consider a system of two electrons trapped at sites $\vec{r}_A$ and $\vec{r}_B$ ($\vec{r}_{A,B}$ are radius vectors of the centers of QDs in the $z=0$ plane) created by a system of charged electrodes in a semiconductor heterostructure so that the electrons are confined in the $z=0$ plane or a system of localized conduction-band electrons in n-doped GaAs, as in our calculation example. The heterostructure trapping potential,

$$V_{\text{tr}}(z,\vec{r}) = V_\perp(z) + V_A(\vec{r}) + V_B(\vec{r}), \tag{20}$$

is separable in the in-plane and out-of-plane directions; $V_\perp(z)$ and $V_{A,B}(\vec{r})$ are the trapping potentials in the z direction and in the $z=0$ plane around $\vec{r}_{A,B}$, respectively. If the electron system is placed in a constant magnetic field $\vec{B}_0$ directed along the z axis (with vector potential $\vec{A}_0 = \frac{1}{2}[\vec{r} \times \vec{B}_0]$), then the in-plane motion, in a superposition of the in-plane confining oscillatory potential and a perpendicular magnetic field, is described by the Fock-Darwin (FD) states.[26] Approximating the confining potential by a quadratic one,

$$V_{A,B}(\vec{r}) \approx \tfrac{1}{2}\omega_{A,B}^2 (\vec{r} - \vec{r}_{A,B})^2, \tag{21}$$

we can take as basis "atomic" orbitals the ground-state functions

$$\phi_{A,B}(z,\vec{r}) = \varphi_0(z) R_{A,B}^{\text{FD}}(|\vec{r} - \vec{r}_{A,B}|), \tag{22}$$

where the out-of-plane motion in the z direction is "frozen" in the ground state $\varphi_0(z)$ in the potential $V_\perp(z)$, and the ground FD state is

$$R_{A,B}^{\text{FD}} = \frac{1}{\sqrt{2\pi} l_{A,B}} \exp\left(-\frac{r^2}{4 l_{A,B}^2} \right),$$

$$l_{A,B} = \frac{l_c}{\sqrt[4]{1 + 4\omega_{A,B}^2/\omega_c^2}}, \quad l_c = \sqrt{\frac{c}{B_0}}. \tag{23}$$

Here, $l_{A,B}$ is the effective length scale, equal to the magnetic length l_c in the absence of the confining potential, $\omega_{A,B} \equiv 0$; $\omega_c = B_0/c$ is the cyclotron frequency.

The orbitals [Eq. (22)] must be orthogonalized. One way to do this is a simple Gram-Schmidt orthogonalization procedure:

$$\tilde{\phi}_A = \phi_A,$$

$$\tilde{\phi}_B = \frac{1}{\sqrt{1 - S^2}} (\phi_B - S\phi_A), \tag{24}$$

where the overlap matrix element $S_{AB} = S_{BA} = S = \langle \phi_A | \phi_B \rangle$ can be calculated analytically,

$$S = \frac{2 l_A l_B}{l_A^2 + l_B^2} \exp\left(-\frac{r_{AB}^2}{4(l_A^2 + l_B^2)} \right). \tag{25}$$

For appropriate values of system parameters such as the interdot distance r_{AB} and the external magnetic field B_0, the overlap becomes exponentially small.

The other more symmetric way is to make a transition to the "molecular" or two-centered orbitals by prediagonalizing the coordinate part of Pauli's nonrelativistic Hamiltonian $\hat{h}_c$, which describes the electron's motion in a superposition of the trapping potential and magnetic fields:

$$\tilde{\phi}_A = c_{AA}\phi_A + c_{AB}\phi_B,$$

$$\tilde{\phi}_B = c_{BA}\phi_A + c_{BB}\phi_B,$$

$$\langle \tilde{\phi}_i | \tilde{\phi}_j \rangle = \delta_{ij}, \quad i,j = A,B,$$

$$\langle \tilde{\phi}_i | \hat{h}_c | \tilde{\phi}_j \rangle = \varepsilon_i \delta_{ij}, \quad i,j = A,B. \tag{26}$$

The two-state eigenvalue problem Eq. (26) is solved analytically in terms of atomic orbital matrix elements: $h_{ij} = \langle \phi_i | \hat{h}_c | \phi_j \rangle$.

In general, given the molecular Eq. (26) or "half-molecular" Eq. (24) basis choices, one cannot ascribe a spin to a particular QD, since an electron in a molecular orbital belongs to both QDs.

The total Hamiltonian contains both coordinate and spin-dependent terms. First, we consider the coordinate part of the Hamiltonian in the $\tilde{\phi}_{A,B}$ basis set.

A. Coordinate part of the Hamiltonian

In view of the orthogonality of the singlet and triplet spin wave functions, the spin-independent part of the Hamiltonian does not contribute to the singlet-triplet coupling, $H_c^{st}=H_c^{ts}=0$, whereas for the singlet-singlet and triplet-triplet Hamiltonians, we get

$$H_c^{ss}=\{H_{cij}^{ss}\}_{i,j=1}^{3},\quad H_{cij}^{ss}=H_{cji}^{ss*}$$

$$H_{c11}^{ss}=\tilde{h}_{AA}+\tilde{h}_{BB}+\tilde{v}_{ee}(AB;AB)+\tilde{v}_{ee}(AB;BA),$$

$$H_{c12}^{ss}=\sqrt{2}[\tilde{h}_{BA}+\tilde{v}_{ee}(AB;AA)],$$

$$H_{c13}^{ss}=\sqrt{2}[\tilde{h}_{AB}+\tilde{v}_{ee}(AB;BB)],$$

$$H_{c22}^{ss}=2\tilde{h}_{AA}+\tilde{v}_{ee}(AA;AA),\quad H_{c23}^{ss}=\tilde{v}_{ee}(AA;BB),$$

$$H_{c33}^{ss}=2\tilde{h}_{BB}+\tilde{v}_{ee}(BB;BB); \tag{27}$$

where

$$H_c^{tt}=\varepsilon_t I,$$

$$\varepsilon_t=\tilde{h}_{AA}+\tilde{h}_{BB}+\tilde{v}_{ee}(AB;AB)-\tilde{v}_{ee}(AB;BA), \tag{28}$$

where

$$\tilde{h}_{ij}=\langle\tilde{\phi}_i|\hat{h}_c|\tilde{\phi}_j\rangle,\quad i,j=A,B,$$

$$\tilde{v}_{ee}(ij;kl)=\langle\tilde{\phi}_i(1)\tilde{\phi}_j(2)|\frac{1}{\varepsilon r_{12}}|\tilde{\phi}_k(1)\tilde{\phi}_l(2)\rangle,$$

$$i,j,k,l=A,B, \tag{29}$$

with $\tilde{h}_{ij}=\varepsilon_i\delta_{ij}$ for molecular orbitals and $\tilde{v}_{ee}$ being the interelectron electrostatic interaction matrix elements. The matrix H_c^{ss} is diagonally dominated if the overlap $S\ll 1$; H_{c11}^{ss} is the singlet energy of the singly occupied state, whereas H_{c22}^{ss} and H_{c33}^{ss} are energies of doubly occupied states if one neglects the coupling between single- and double-occupancy states. Observe that the Heisenberg constant $J_H=\varepsilon_t-\varepsilon_s$, where ε_s is the lowest eigenvalue of the matrix H_c^{ss}. The matrix elements $\tilde{h}_{ij}$ and $\tilde{v}_{ee}(ij;kl)$ can be trivially expressed in terms of the corresponding matrix elements h_{ij} and $v_{ee}(ij;kl)$, where the orthonormalized states $\tilde{\phi}_i$ are replaced by ϕ_i using the relations in Eq. (24) or (26).

B. Dipole spin-spin interaction

In the total spin representation, the dipole spin-spin interaction can be rewritten as[27]

$$V_{\text{dip}}=\frac{1.45}{2}\ \text{meV}\left[\frac{S^2r_{12}^2-3(\vec{S}\cdot\vec{r}_{12})^2}{r_{12}^5}-\frac{8\pi}{3}\left(S^2-\frac{3}{2}\right)\delta(\vec{r}_{12})\right]. \tag{30}$$

Since $\vec{S}|\chi_s\rangle=0$ and $f_t(\vec{r}_1=\vec{r}_2)=0$, where $|\chi_s\rangle$ and f_t are singlet-state spin and triplet-state coordinate wave functions, we have $H_{\text{dip}}^{st}=H_{\text{dip}}^{ts}=0$ and a nonzero contribution to H_{dip}^{ss} comes only from the contact term:

$$(H_{\text{dip}}^{ss})_{ij}=[1.45(2\pi)]\ \text{meV}\langle f_{si}|\delta(\vec{r}_{12})|f_{sj}\rangle$$

$$=1.09\Lambda\ \text{meV}\begin{pmatrix} d_{11} & d_{12} & d_{13}\\ d_{12} & d_{22} & \frac{1}{2}d_{11}\\ d_{13} & \frac{1}{2}d_{11} & d_{33}\end{pmatrix}, \tag{31}$$

where Λ is an effective constant of the interaction that confines electrons in the z plane and

$$d_{11}=2\langle\tilde{\phi}_A^2|\tilde{\phi}_B^2\rangle=\frac{1}{l_A^2+l_B^2}\exp\left(-\frac{r_{AB}^2}{2(l_A^2+l_B^2)}\right),$$

$$d_{12}=\sqrt{2}\langle\tilde{\phi}_A^3|\tilde{\phi}_B\rangle=\sqrt{2}\frac{l_B/l_A}{3l_B^2+l_A^2}\exp\left(-\frac{3}{4}\frac{r_{AB}^2}{3l_B^2+l_A^2}\right),$$

$$d_{13}=\sqrt{2}\langle\tilde{\phi}_A|\tilde{\phi}_B^3\rangle=\sqrt{2}\frac{l_A/l_B}{3l_A^2+l_B^2}\exp\left(-\frac{3}{4}\frac{r_{AB}^2}{3l_A^2+l_B^2}\right),$$

$$d_{22}=\langle\tilde{\phi}_A^2|\tilde{\phi}_A^2\rangle=\frac{1}{4l_A^2},\quad d_{33}=\langle\tilde{\phi}_B^2|\tilde{\phi}_B^2\rangle=\frac{1}{4l_B^2}. \tag{32}$$

The magnetic dipole contribution to the triplet-triplet interaction Hamiltonian can be written as

$$H_{\text{dip}}^{tt}=0.36\ \text{meV}\begin{pmatrix} \bar{t}_0 & -\frac{3}{\sqrt{2}}\bar{t}_1^* & -3\bar{t}_2^*\\ -\frac{3}{\sqrt{2}}\bar{t}_1 & -2\bar{t}_0 & \frac{3}{\sqrt{2}}\bar{t}_1^*\\ -3\bar{t}_2 & \frac{3}{\sqrt{2}}\bar{t}_1 & \bar{t}_0\end{pmatrix}, \tag{33}$$

where t_i, $i=0,1,2$, are dipole tensor operators,

$$t_0=\frac{1-3\cos^2\theta_{12}}{r_{12}^3},$$

$$t_1=\frac{\sin 2\theta_{12}\exp(i\varphi_{12})}{r_{12}^3},$$

$$t_2=\frac{\sin^2\theta_{12}\exp(2i\varphi_{12})}{r_{12}^3}, \tag{34}$$

with $(r_{12},\theta_{12},\varphi_{12})$ being spherical coordinates of the interelectron radius vector $\vec{r}_{12}=\vec{r}_1-\vec{r}_2$; the bar over t_i denotes averaging over the triplet coordinate wave function:

$$\bar{t}_i=\int\int d^3\vec{r}_1d^3\vec{r}_2|f_t(\vec{r}_1,\vec{r}_2)|^2t_i(\vec{r}_1,\vec{r}_2). \tag{35}$$

Taking into account the fact that the electrons are exponentially localized at sites $\vec{r}_A$ and $\vec{r}_B$ in the f_t state, a good approximation to $\bar{t}_i$ is to approximate the function t_i by a constant value at those points where $f_t(\vec{r}_1,\vec{r}_2)$ is localized, thus obtaining the estimate

$$H_{\mathrm{dip}}^{tt} = \frac{0.36}{r_{AB}^3}\ \mathrm{meV}\begin{pmatrix} 1 & 0 & -3 \\ 0 & -2 & 0 \\ -3 & 0 & 1 \end{pmatrix}. \tag{36}$$

In order to further improve the estimate, the function $t_i(\vec{r}_1, \vec{r}_2)$ can be expanded in a Taylor series around the localization points and the remaining integrals in the expansion terms be calculated analytically. From Eq. (36), we find the estimate $H_{\mathrm{dip}}^{tt} \sim (0.36/r_{AB}^3)$ meV $\approx 5.0 \times 10^{-8}$ meV at $r_{AB} = 100$ Å.

C. $\vec{B}$-field interaction in the pure-spin model

For the magnetic field, one gets

$$H^{tt}(\vec{B}) = \begin{pmatrix} B_{avz} & B_{av}^- & 0 \\ B_{av}^+ & 0 & B_{av}^- \\ 0 & B_{av}^+ & -B_{avz} \end{pmatrix}, \tag{37}$$

where

$$\vec{B}_{av} = \tfrac{1}{2}(\langle \tilde{\phi}_A | \vec{B} | \tilde{\phi}_A \rangle + \langle \tilde{\phi}_B | \vec{B} | \tilde{\phi}_B \rangle),$$

$$B_{av}^{\pm} = \tfrac{1}{\sqrt{2}}(B_{avx} \pm i B_{avy}). \tag{38}$$

Using Eqs. (11) and (37), one derives the Zeeman interaction Hamiltonian of the total spin $\vec{S}$ with the magnetic field $\vec{B}_{av}$:

$$\mathbf{H}^{tt}(\vec{B}) = \sum_{ij=1}^{3} H_{ij}^{tt}(\vec{B}) \mathbf{T}_{ij} = \vec{B}_{av} \cdot \vec{S}. \tag{39}$$

Similarly, for the singlet-triplet matrix, we have

$$H^{st}(\vec{B}) = \begin{pmatrix} \frac{1}{2\sqrt{2}}\Delta B^+ & -\frac{1}{2}\Delta B_z & -\frac{1}{2\sqrt{2}}\Delta B^- \\ \frac{1}{2}\delta B^+ & -\frac{1}{\sqrt{2}}\delta B_z & -\frac{1}{2}\delta B^- \\ -\frac{1}{2}\delta B^{-*} & \frac{1}{\sqrt{2}}\delta B_z^* & \frac{1}{2}\delta B^{+*} \end{pmatrix}, \tag{40}$$

where

$$\Delta\vec{B} = \langle \tilde{\phi}_B | \vec{B} | \tilde{\phi}_B \rangle - \langle \tilde{\phi}_A | \vec{B} | \tilde{\phi}_A \rangle,$$

$$\Delta B^{\pm} = \Delta B_x \pm i \Delta B_y,$$

$$\delta\vec{B} = \langle \tilde{\phi}_A | \vec{B} | \tilde{\phi}_B \rangle, \quad \delta B^{\pm} = \delta B_x \pm i\, \delta B_y. \tag{41}$$

If the $\vec{B}$ field is homogeneous, from Eq. (41), we obtain $\Delta\vec{B} = \delta\vec{B} = 0$ and $H^{st}(\vec{B}) = 0$. In this case, the spin dynamics is unitary and is described by the Zeeman Hamiltonian $\mathbf{H}^{tt}(\vec{B})$ [Eq. (39)]; the spin-spin dipole interaction H_{dip}^{tt} is too small and can usually be safely neglected.

Let us consider modifications due the to $\vec{B}$-field inhomogeneity in the pure-spin model. Neglecting contributions from the double-occupancy states within the first-order perturbation approximation in the singlet-triplet interaction H^{st}, we find the following for the nonunitary term in Eq. (14),

$$\mathcal{L}_t = \frac{1}{2}\sum_{ij} (\chi_t)_{ij}([\mathbf{K}_i, \rho \mathbf{K}_j^\dagger] + [\mathbf{K}_i \rho, \mathbf{K}_j^\dagger])$$

$$= \frac{\sin(J_H t)}{J_H}([\mathbf{K}, \rho \mathbf{K}^\dagger] + [\mathbf{K}\rho, \mathbf{K}^\dagger]), \tag{42}$$

where

$$\mathbf{K} = \sum_i H_{1i}^{st}(\vec{B}) \mathbf{K}_i = -\frac{i}{4}\Delta\vec{B} \cdot \vec{J}_{\mathrm{as}} \tag{43}$$

and

$$\vec{J}_{\mathrm{as}} = [\vec{s}_2 - \vec{s}_1 \times \vec{S}] = 2[\vec{s}_2 \times \vec{s}_1] + i(\vec{s}_2 - \vec{s}_1) \tag{44}$$

is an asymmetric spin operator containing both linear and bilinear parts. Observe that $\mathcal{L}_t = 0$ at the "swap" times $t_n = \pi n / J_H$, $n = 0, 1, \ldots$.

Similarly, for the Lamb shift in Eq. (14), we have

$$\mathbf{L}_t = \frac{1}{2}\sum_{ij} (P_t)_{ij} \mathbf{K}_i^\dagger \mathbf{K}_j = \frac{1 - \cos(J_H t)}{J_H} \mathbf{K}^\dagger \mathbf{K}. \tag{45}$$

Observe that $\mathcal{L}_t$ and $\mathbf{L}_t$ are quadratic in the difference field $\Delta\vec{B}$. Besides, notice that the magnetic field due to spin-orbit coupling does not contribute to the difference field $\Delta\vec{B}_{\mathrm{so}} = 0$ but contributes to the $\delta\vec{B}_{\mathrm{so}}$ field that is present in the coupling between the triplet states and the double-occupancy, singlet states in Eq. (40). If the external magnetic field $\vec{B}_{\mathrm{ex}}$ is homogeneous, then the singlet-triplet state coupling comes only from the spin-orbit interaction. Since the double-occupancy states should be involved in the dynamics in order to obtain nonzero spin-orbit interaction effects, these effects are expected to be especially small, proportional to δB_{so}^2, in the pure-spin model. An estimate of these spin-orbit effects will be given in a numerical example in the next section.

Clearly, there is an important qualitative difference between pure-spin and pseudospin models. In the former, the singlet-triplet state coupling is a second order effect, while in the latter, this coupling is of first order in H^{st} [cf. Eqs. (42) and (6)]. Thus, in pure-spin models, effects due to $\vec{B}$-field inhomogeneity should be especially (quadratically) small as compared to the corresponding pseudospin model effects. In case of negligible $\vec{B}$-field inhomogeneity, as follows from Eq. (40), the pure-spin dynamics is unitary and is governed by the spin Hamiltonian $\mathbf{H}^{tt}$.

Let us now consider a simple numerical example for the nonunitary effects due to the difference field $\Delta\vec{B}$ for an electron localized on a donor impurity in an n-doped GaAs semiconductor. To simplify numerics, we assume that $H^{ss} = \mathrm{diag}(\varepsilon_{s1}, \varepsilon_{s2}, \varepsilon_{s3})$ is diagonal and the singlet-triplet coupling field $\Delta\vec{B}$ has only a nonzero z component, ΔB_z. Then, the corresponding eigenvalue problem [Eq. (19)] can be reduced to a biquadratic polynomial equation which could, in principle, be solved exactly. If we neglect the exponentially small coupling field δB_z, proportional to the overlap S, the biquadratic equation reduces to a quadratic one and we find the following for the nonunitary term:

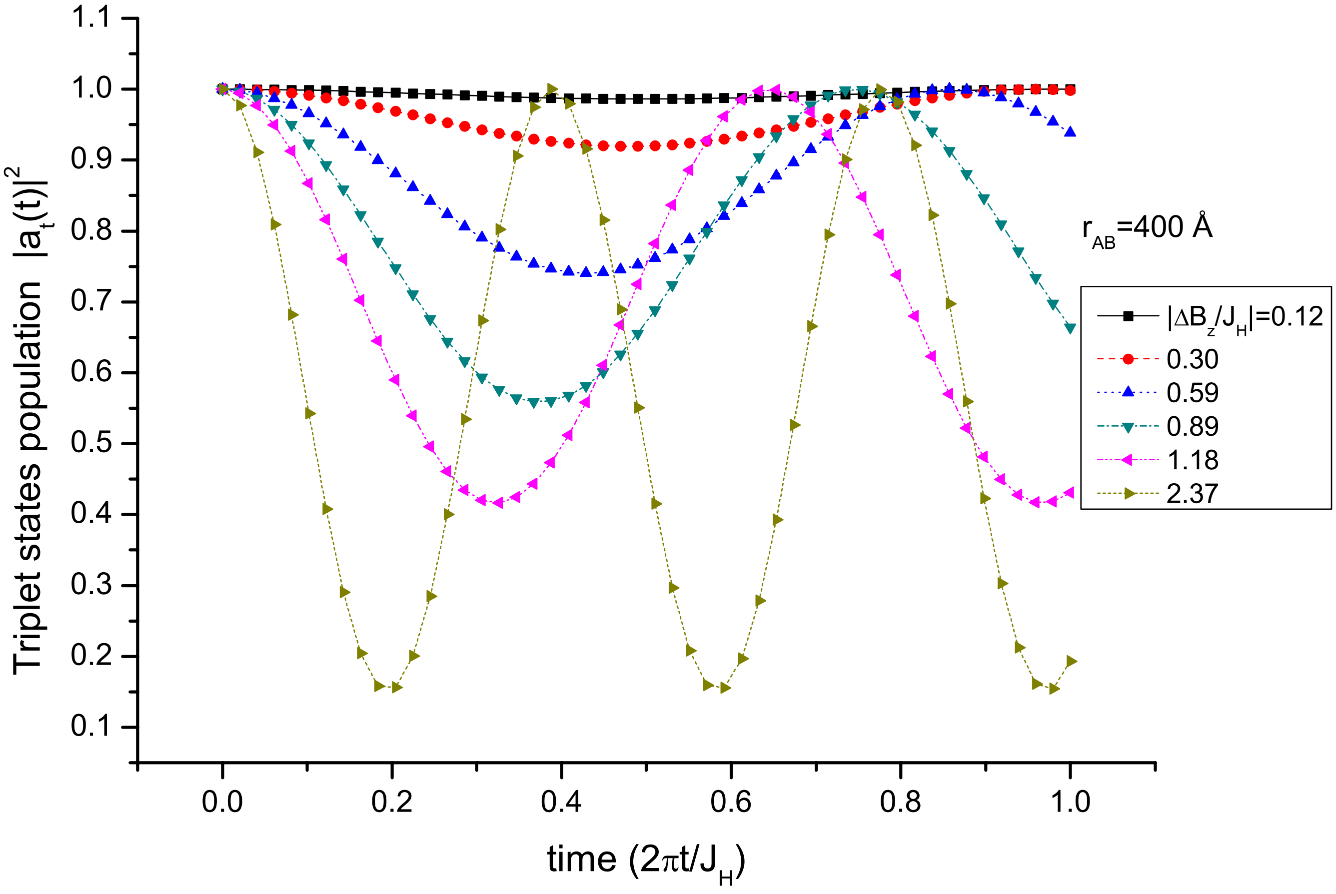

FIG. 1. (Color online) The triplet-state population of electrons in shallow QD centers in GaAs as a function of time at different magnetic field differences ΔB_z=0.01, 0.025, 0.05, 0.075, 0.1, 0.2 T, normalized to the Heisenberg exchange J_H constant. Initially, the system is in the triplet state $|S=1,M_S=0\rangle$. The distance between QDs is 400 Å.

$$\mathcal{L}_t = \frac{\omega \sin \omega t}{J_H^2 + \frac{1}{2}\Delta B_z^2(1+\cos \omega t)}([\mathbf{K},\rho\mathbf{K}^\dagger]+[\mathbf{K}\rho,\mathbf{K}^\dagger]),$$

$$\mathbf{K} = -\frac{i}{4}\Delta B_z \cdot J_{asz} \qquad (46)$$

and the Lamb shift:

$$\mathbf{L}_t = \frac{J_H(1-\cos \omega t)}{J_H^2 + \frac{1}{2}\Delta B_z^2(1+\cos \omega t)}\mathbf{K}^\dagger\mathbf{K}, \qquad (47)$$

where $\omega=\sqrt{J_H^2+\Delta B_z^2}$. In the limit of small magnetic field inhomogeneity, $|\Delta B_z/J_H| \ll 1$, $\omega \to |J_H|$ and Eqs. (46) and (47) go over into Eqs. (42) and (45), respectively. Equation (47) describes the Lamb energy shift of the triplet state $|S=1,M_S=0\rangle$ due to the coupling between singlet and triplet states induced by the magnetic field inhomogeneity ΔB_z. At the magnetic field geometry we have chosen, there is no coupling between $|S=1,M_S=\pm 1\rangle$ and $|S=1,M_S=0\rangle$ states.

In Figs. 1–3, we show the results of calculations for the triplet-state population, purity, and Lamb shift energy, respectively, as a function of time at a fixed interdot separation (r_{AB}=400 Å) and different ΔB_z. For the Heisenberg interaction constant J_H, we used an asymptotically correct expression[28–30] obtained for hydrogenlike centers in GaAs (note that our $J_H=\varepsilon_t-\varepsilon_s$ is related to the exchange integral J in Ref. 28 via $J_H=-2J$). Initially, the system is assumed to be in the $|S=1,M_S=0\rangle$ state. As can be seen from Fig. 1, there is a redistribution between singlet- and triplet-state populations due to the singlet-triplet subspace coupling. At $|\Delta B_z/J_H| \lesssim 0.1$, the probability of redistribution is negligible and the time evolution is basically unitary. With increasing ΔB_z, this probability redistribution is seen to be more pronounced, time evolution becomes nonunitary (Fig. 2) and $|a_t(t)|^2$ can drop to the value J_H^2/ω^2 at $t=\pi n/\omega, n=1,3,\ldots$. Observe that the nonunitary dynamics reveals repetitions in time, and at moments of maximal (minimal) singlet-triplet state probability redistribution, we find maximal (minimal) Lamb energy shifts (Fig. 3). Thus, the nonunitary effects observed are not irreversible and they do not result in a real decoherence process. We do not have a real, external, and infinite "bath" in our two-electron model, coupling to which would result in irreversible decoherence effects in the spin system. In Figs. 4 and 5, we demonstrate the dependence of triplet-state population and Lamb energy shifts on the interdot distance r_{AB} at a fixed ΔB_z=0.05 T.

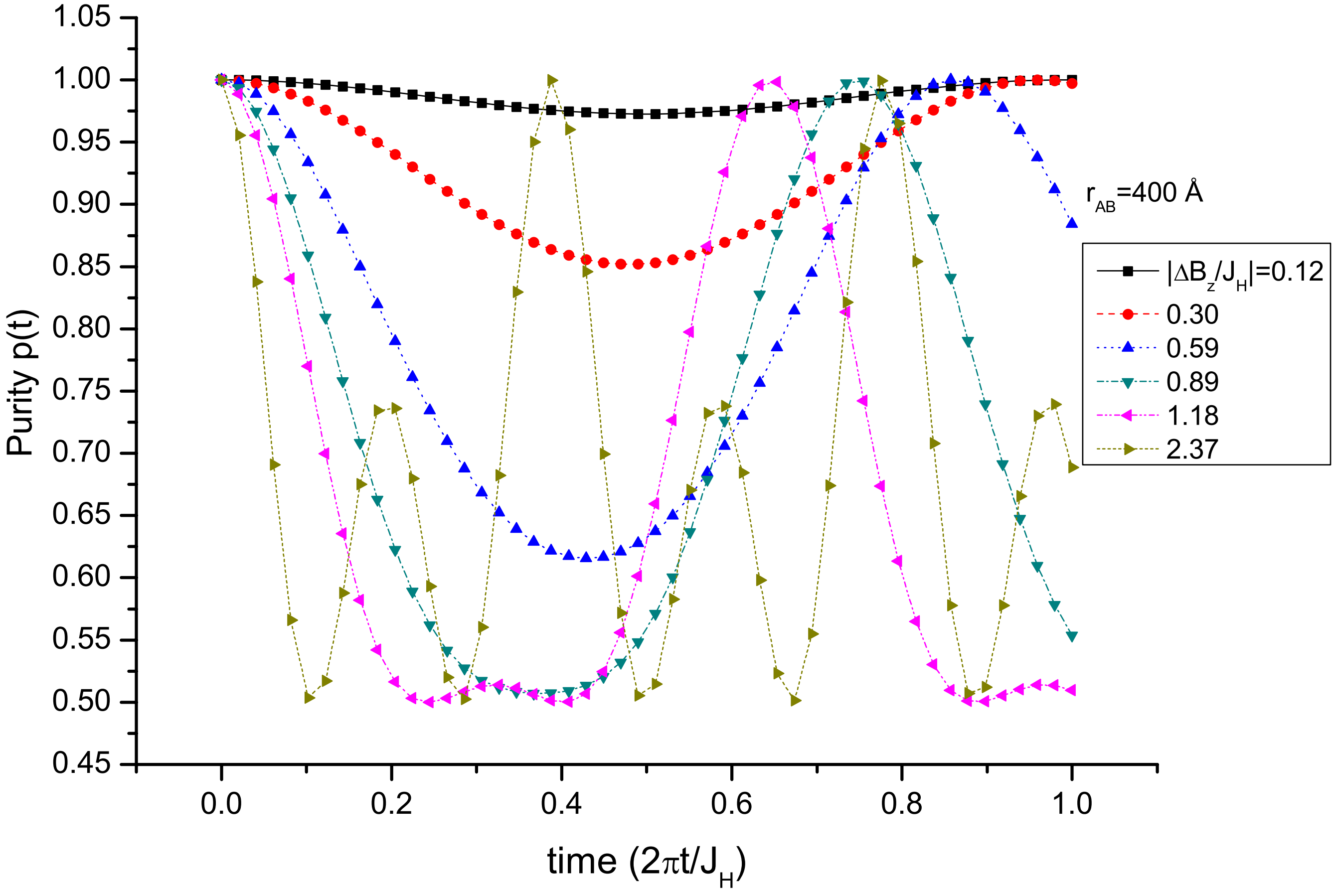

FIG. 2. (Color online) The purity $p(t) = \mathrm{Tr}\,\rho^2(t)$ for the same parameters as in Fig. 1.

D. Spin-orbit interaction in pure-spin model

In this subsection, we estimate the nonunitary effects in the pure-spin model due to spin-orbit interaction. For simplicity, we assume that the external magnetic field is homogeneous and directed along the z axis, with B_{oz} being its z component. Since $\delta\vec{B}_{so}$ is a pure imaginary field (its components are matrix elements between the real states $\tilde{\phi}_A$ and $\tilde{\phi}_B$ of an odd vector function of the momentum operator, both in vacuum and in the bulk of semiconductors that lack inversion symmetry, Dresselhaus fields,[31] as well as in heterostructure zinc blendes, Rashba fields[32]) we have

$$(\delta B_{so}^{\pm})^{*} = -\,\delta B_{so}^{\mp}, \qquad \delta B_{soz}^{*} = -\,\delta B_{soz}. \tag{48}$$

Using these relationships, the singlet-triplet spin-orbit coupling can be written as

$$H^{st}(\vec{B}_{so}) = \begin{pmatrix} 0 & 0 & 0 \\ \frac{1}{2}\,\delta B_{so}^{+} & -\frac{1}{\sqrt{2}}\,\delta B_{soz} & -\frac{1}{2}\,\delta B_{so}^{-} \\ \frac{1}{2}\,\delta B_{so}^{+} & -\frac{1}{\sqrt{2}}\,\delta B_{soz} & -\frac{1}{2}\,\delta B_{so}^{-} \end{pmatrix}. \tag{49}$$

The couplings between double-occupancy, singlet and triplet states are seen to be the same. We assume that $H^{ss} = \mathrm{diag}(\varepsilon_s, \varepsilon_{do}, \varepsilon_{do})$, where ε_s and ε_{do} are the singlet- and double-occupancy state energies, and $H^{tt} = \mathrm{diag}(\varepsilon_{t+}, \varepsilon_t, \varepsilon_{t-})$, where ε_t is a triplet-state energy and $\varepsilon_{t\pm} = \varepsilon_t \pm B_{0z}$. Within these approximations, the 6×6 eigenvalue problem Eq. (19) is then reduced to computing the roots of the biquadratic equation[33]

$$E^4 + a_3 E^3 + a_2 E^2 + a_1 E + a_0 = 0, \tag{50}$$

where

$$a_3 = -\sum_{i=1}^{4} \varepsilon_i, \quad a_2 = \sum_{i \neq j} \varepsilon_i \varepsilon_j - \sum_{\alpha = x,y,z} |\delta B_{so\alpha}|^2,$$

$$a_1 = -\sum_{i \neq j \neq k} \varepsilon_i \varepsilon_j \varepsilon_k + \left(|\delta B_{soz}|^2 + \frac{1}{2}[|\delta B_{sox}|^2 + |\delta B_{soy}|^2] \right) \times (\varepsilon_2 + \varepsilon_4) + (|\delta B_{sox}|^2 + |\delta B_{soy}|^2)\varepsilon_3,$$

$$a_0 = \prod_{i=1}^{4} \varepsilon_i - |\delta B_{soz}|^2 \varepsilon_2 \varepsilon_4 - \frac{1}{2}[|\delta B_{sox}|^2 + |\delta B_{soy}|^2]\varepsilon_3(\varepsilon_2 + \varepsilon_4),$$

$$\varepsilon_1 = \varepsilon_s, \quad \varepsilon_2 = \varepsilon_{t+}, \quad \varepsilon_3 = \varepsilon_t, \quad \varepsilon_4 = \varepsilon_{t-}.$$

For hydrogenlike centers, one can estimate the energies ε_{do} and ε_t as follows. The ground energy of two well separated hydrogen atoms is $E_{2\mathrm{H}} \approx -27.2$ eV. Using the scaling factor $K_{\mathrm{GaAs}} = m^{*}/\varepsilon^2 \approx 4.6 \times 10^{-4}$ for GaAs, one can estimate $\varepsilon_t \approx K_{\mathrm{GaAs}} E_{2\mathrm{H}} = -12.6$ meV. ε_{do} is located higher than ε_t due

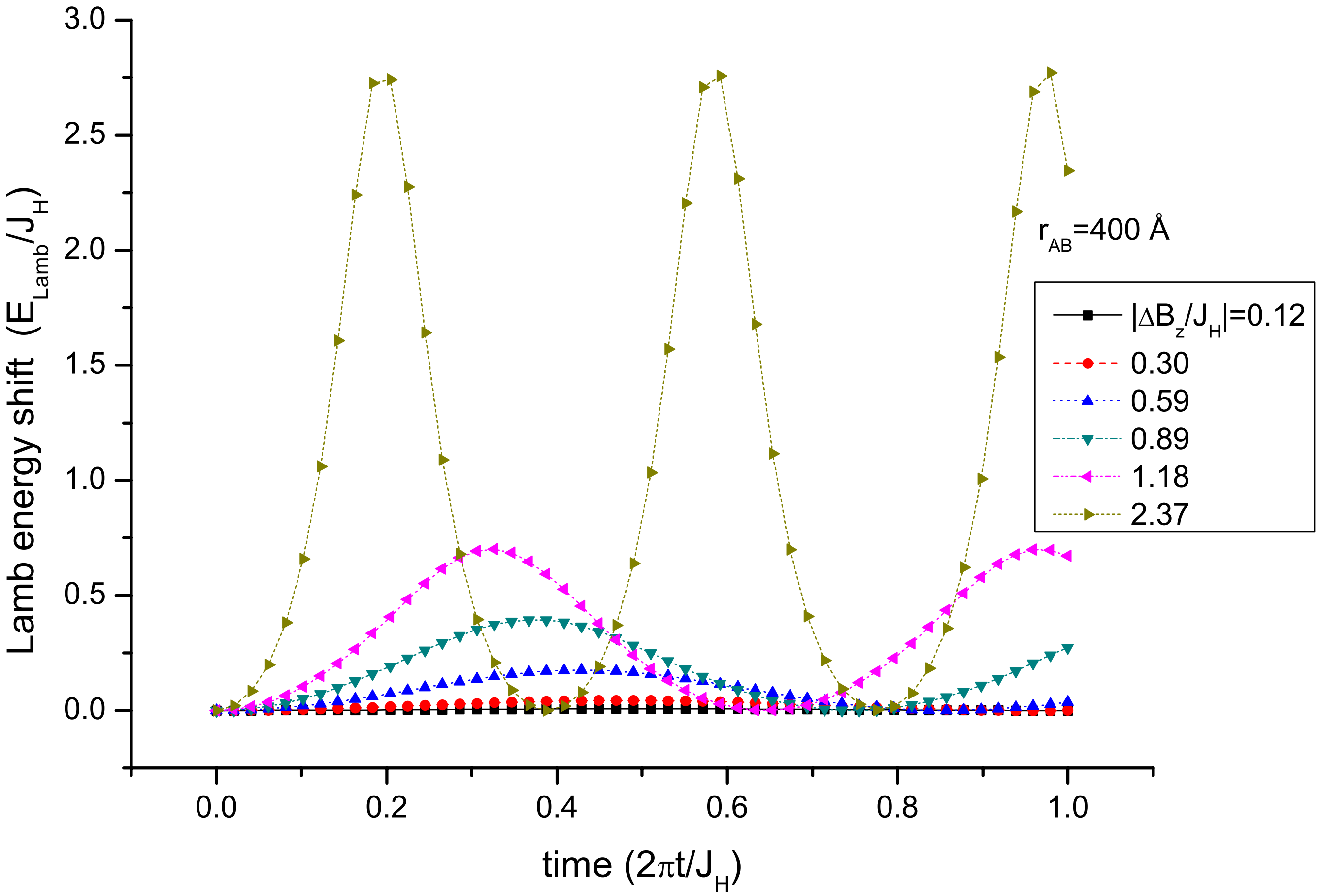

FIG. 3. (Color online) The Lamb shift energy as a function of time for the same parameters as in Fig. 1.

to mainly interelectron repulsion $\tilde{v}_{ee}(AA;AA)$ so that $\varepsilon_{do}-\varepsilon_t=\tilde{v}_{ee}(AA;AA)\approx 12.6$ meV.

If $\delta B_{so\alpha}\equiv 0$, $\alpha=x,y,z$, the roots of Eq. (50) E_i are equal to ε_i, $i=1,\ldots,4$. The two other roots are $E_5=\varepsilon_{do}$ and $E_6=\varepsilon_s$. The corresponding eigenvectors are

$$|e_k\rangle = d_k\left(0,1,1,-\frac{\delta B_{so}^-}{E_k-\varepsilon_2},\frac{\sqrt{2}\,\delta B_{soz}}{E_k-\varepsilon_3},\frac{\delta B_{so}^+}{E_k-\varepsilon_4}\right)^T,$$

$$k=1,\ldots,4,$$

$$|e_5\rangle=\tfrac{1}{\sqrt{2}}(0,1,-1,0,0,0)^T,$$

$$|e_6\rangle=(1,0,0,0,0,0)^T, \tag{51}$$

where

$$d_k=\left(2+[|\delta B_{sox}|^2+|\delta B_{soy}|^2]\left(\frac{1}{(E_k-\varepsilon_2)^2}+\frac{1}{(E_k-\varepsilon_4)^2}\right)+\frac{2|\delta B_{soz}|^2}{(E_k-\varepsilon_3)^2}\right)^{-1/2}.$$

Notice that the above formulas are not valid in the degenerate case: $B_{0z}=0$ and $\varepsilon_2=\varepsilon_3=\varepsilon_4=\varepsilon_t$. In this case, the biquadratic Eq. (50) reduces to two quadratic ones, two roots of which are degenerate, $E_1=E_2=\varepsilon_t$. Formally, one gets singularities in Eq. (51) at $E_1=E_2=\varepsilon_t$. Therefore, the simpler, degenerate case should be analyzed separately and the corresponding formulas (not shown here) can be derived.

Let us now find the spin-orbit field,

$$\begin{aligned}\delta\vec{B}_{so}&=\langle\tilde{\phi}_A|\vec{B}_{so}(\vec{p})|\tilde{\phi}_B\rangle\approx\langle\phi_A|\vec{B}_{so}(\vec{p})|\phi_B\rangle\\&=\int d\vec{r}\phi_A(|\vec{r}-\vec{R}|)\vec{B}_{so}(-i\nabla_{\vec{r}})\phi_B(r)\\&=\vec{B}_{so}(-i\nabla_{\vec{R}})\int d\vec{r}\phi_A(|\vec{r}-\vec{R}|)\phi_B(r)=\vec{B}_{so}(-i\nabla_{\vec{R}})S(R),\end{aligned}\tag{52}$$

where $\vec{B}_{so}(\vec{p})$ is an odd function of the momentum operator $\vec{p}=-i\nabla_{\vec{r}}$ and $\vec{R}=\vec{r}_{AB}$. In particular, in zinc-blende semiconductors such as GaAs, $\vec{B}_{so}$ is cubic in the components of $\vec{p}$:[31,34]

$$B_{so\alpha}=A_{so}p_\alpha(p_\beta^2-p_\gamma^2),$$

$$\alpha,\beta,\gamma=\{\text{cyclic permutations of } x,y,z\},$$

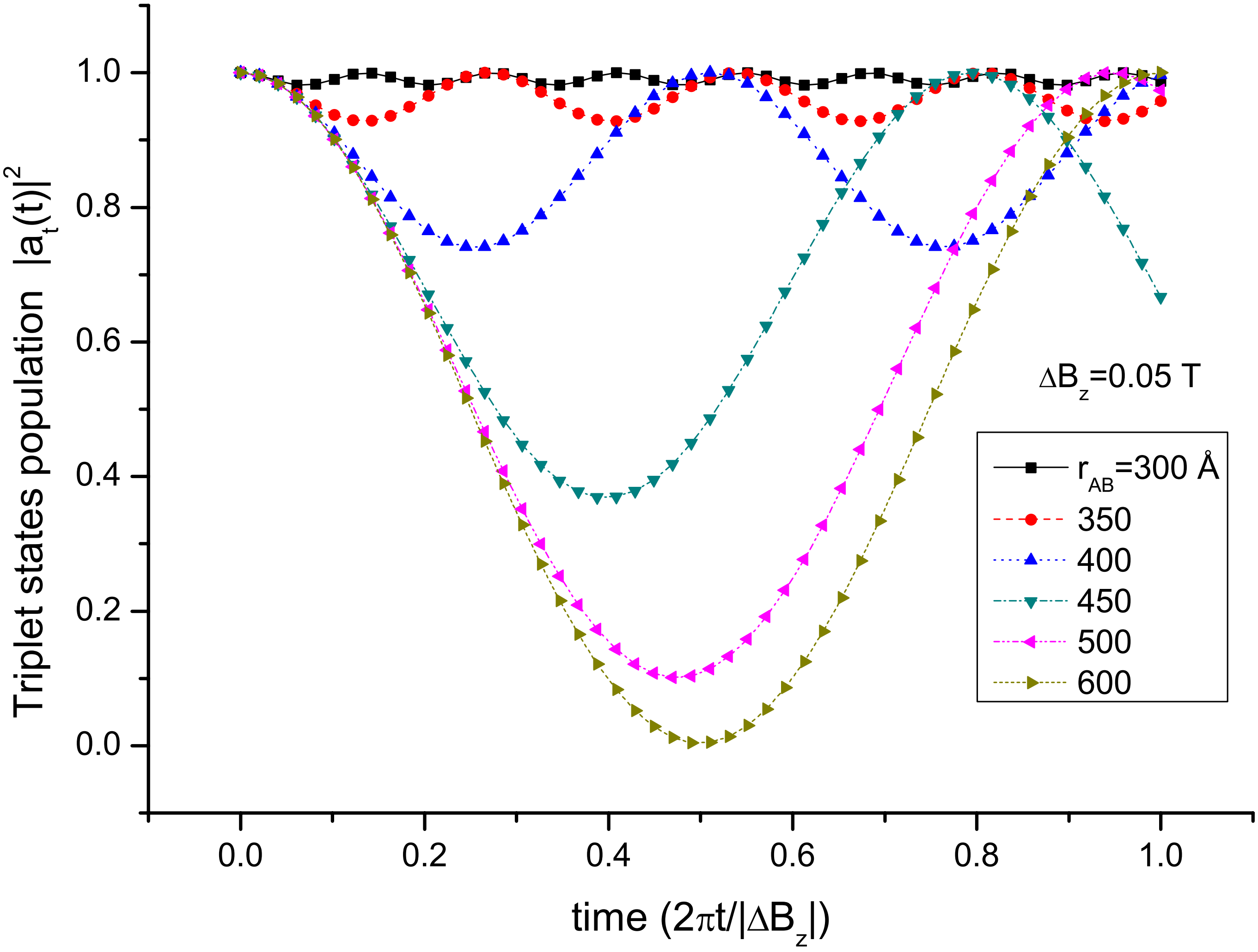

FIG. 4. (Color online) Triplet-state population dependence on interdot separations r_{AB}=300, 350, 400, 450, 500, and 600 Å at a fixed ΔB_z=0.05 T.

$$A_{so} = \alpha_{so}(m^* \sqrt{2m^* E_g})^{-1}, \tag{53}$$

where m^* is the effective mass of the electron, E_g is the band gap ($m^* \approx 0.072$, $E_g \approx 1.43$ eV for GaAs), and p_x, p_y, p_z are components of the momentum along the cubic axes [100], [010], and [001], respectively. The dimensionless coefficient α_{so}=0.07 for GaAs. From Eqs. (52) and (53) we obtain

$$B_{so\alpha} = iA_{so}\frac{R_\alpha(R_\beta^2 - R_\gamma^2)}{R^3}\left\{S'''(R) - \frac{3}{R}S''(R) + \frac{3}{R^2}S'(R)\right\}. \tag{54}$$

The overlap integral[35] $S(r=R/a_B)=(1+r+r^2/3)\exp(-r)$ for hydrogenlike centers ($a_B \approx 92$ Å for GaAs) and Eq. (54) reduces to

$$B_{sox} = i(0.83 \text{ meV})\sin\theta\cos\varphi(\sin^2\theta\sin^2\varphi - \cos^2\theta) \times\left(-\frac{1}{3}\right)r^2\exp(-r), \tag{55}$$

where (R,θ,φ) are spherical coordinates of the vector $\vec{R}$, with other components being obtained by cyclic interchange of indices.

In Figs. 6 and 7, we display the time dependence of the triplet-state population and the purity, which is induced by the spin-orbit interaction [Eq. (55)] at a fixed orientation ($\theta = \pi/4, \varphi = \pi/3$) and different r_{AB} in the range of 200–500 Å. Observe that the maximal redistribution of singlet-triplet probability occurs at $2\pi t/|\varepsilon_{do}-\varepsilon_t|=0.5$ and the spin-orbit interaction effect diminishes as r_{AB} increases. The maximal singlet-state probability achieved at r_{AB}=200 Å is seen to be quite small, $\sim 10^{-5}$. As compared to the nonunitary effects induced by $\vec{B}$-field inhomogeneity, the spin-orbit effects are on average 4 orders of magnitude smaller. The angular dependence of the population of triplet states on the interdot radius-vector orientation at a fixed r_{AB}=200 Å is illustrated in Fig. 8.

E. $\vec{B}$-field interaction in the pseudospin model

Using Eqs. (37) and (40), the effective Hamiltonian matrix Eq. (6) in the basis $\{\Phi_{s1}, \Phi_{ti}, i=1,2,3\}$ can be rewritten as

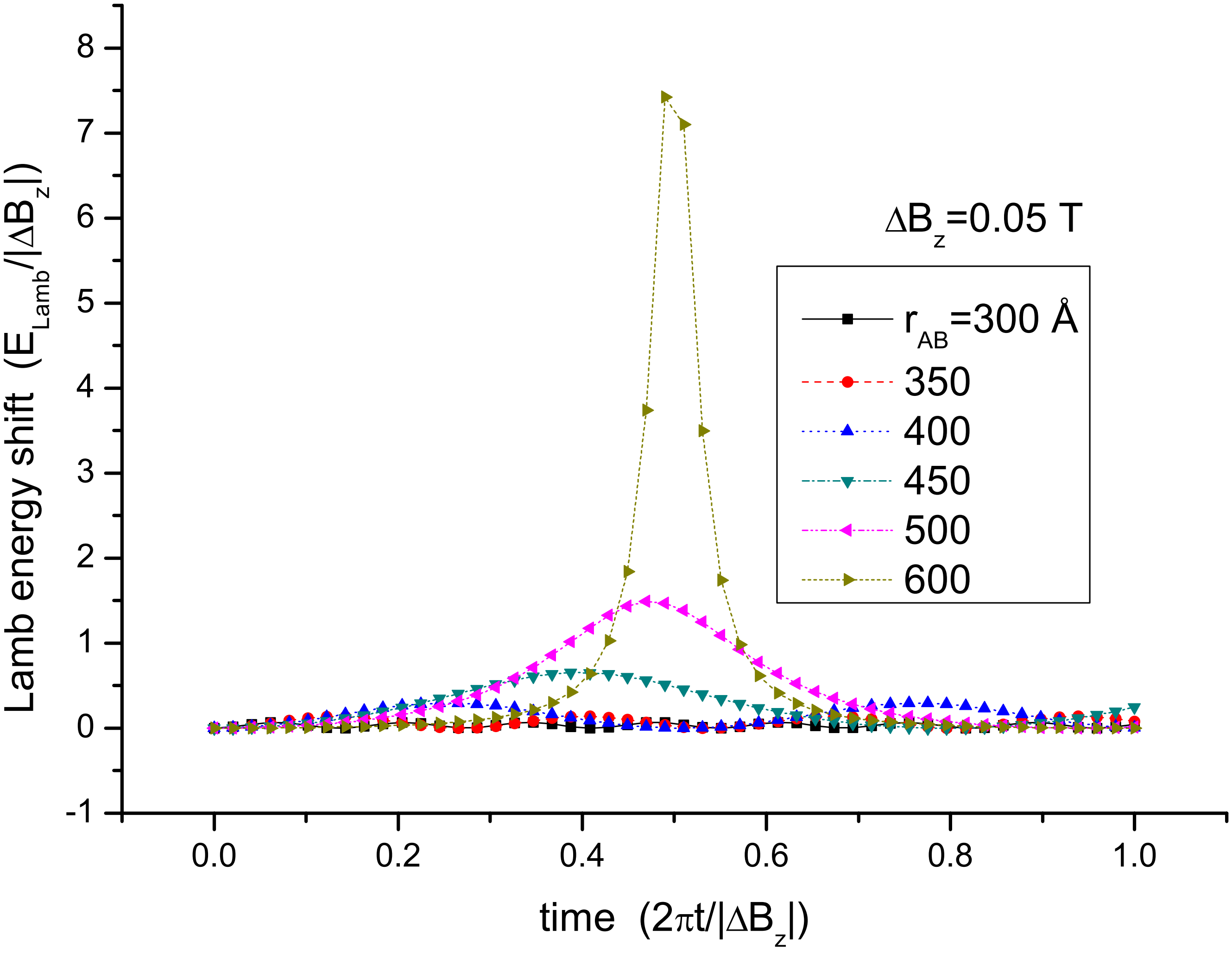

FIG. 5. (Color online) Lamb energy shift dependence on interdot distances r_{AB}=300, 350, 400, 450, 500, and 600 Å at a fixed ΔB_z =0.05 T.

$$\mathcal{H}_{\text{eff}} = \begin{pmatrix} \varepsilon_s & \frac{1}{2\sqrt{2}}\Delta B^+ & -\frac{1}{2}\Delta B_z & -\frac{1}{2\sqrt{2}}\Delta B^- \\ \frac{1}{2\sqrt{2}}\Delta B^- & \varepsilon_t + B_{avz} & B^-_{av} & 0 \\ -\frac{1}{2}\Delta B_z & B^+_{av} & \varepsilon_t & B^-_{av} \\ -\frac{1}{2\sqrt{2}}\Delta B^+ & 0 & B^+_{av} & \varepsilon_t - B_{avz} \end{pmatrix}, \tag{56}$$

where, for simplicity, we neglected contributions from the double-occupancy states [the resolvent term in Eq. (3)]. Alternatively, in the pseudospin representation, we get

$$\mathcal{H}_{\text{eff}} = \left(\frac{1}{4}\varepsilon_s + \frac{3}{4}\varepsilon_s\right)I + J_H\vec{s}_A \cdot \vec{s}_B + \vec{B}_{av} \cdot (\vec{s}_A + \vec{s}_B) + \frac{\Delta\vec{B}}{2} \cdot (\vec{s}_A - \vec{s}_B) = \left(\frac{1}{4}\varepsilon_s + \frac{3}{4}\varepsilon_t\right)I + J_H\vec{s}_A \cdot \vec{s}_B + \vec{B}_A \cdot \vec{s}_A + \vec{B}_B \cdot \vec{s}_A, \tag{57}$$

where $\vec{B}_A = \vec{B}_{av} + \Delta\vec{B}/2$ and $\vec{B}_B = \vec{B}_{av} - \Delta\vec{B}/2$ are the local magnetic fields at sites A and B, respectively. The term $J_H\vec{s}_A \cdot \vec{s}_B$ is the familiar Heisenberg interaction. In matrix form, Eq. (57) can be rewritten as

$$\mathcal{H}_{\text{eff}} = \begin{pmatrix} \varepsilon_1 & \frac{1}{2}B^-_A & 0 & 0 \\ B^+_A & \varepsilon_2 & \frac{1}{2}J_H & 0 \\ 0 & \frac{1}{2}J_H & \varepsilon_3 & \frac{1}{2}B^-_B \\ 0 & 0 & \frac{1}{2}B^+_B & \varepsilon_4 \end{pmatrix},$$

$$\varepsilon_1 = \varepsilon_t + \tfrac{1}{2}B_{Az}, \quad \varepsilon_2 = \varepsilon_t - \tfrac{1}{2}(J_H + B_{Az}),$$

$$\varepsilon_3 = \varepsilon_t - \tfrac{1}{2}(J_H - B_{Bz}), \quad \varepsilon_4 = \varepsilon_t - \tfrac{1}{2}B_{Bz}, \tag{58}$$

where $B^{\pm}_{A,B} = B_{A,Bx} \pm iB_{A,By}$.

The Hamiltonian Eq. (56) generates a unitary evolution,

$$U_{\text{eff}}(t) = \exp(-it\mathcal{H}_{\text{eff}}), \tag{59}$$

in $\mathbb{C}^4$. At a fixed set of parameters ε_s, ε_t, $\vec{B}_A$, and $\vec{B}_B$, the propagator $U_{\text{eff}}(t)$ does not provide a universal set of unitary gates in $\mathbb{C}^4$. Any unitary transformation $U \in U(4)$ can be rep-

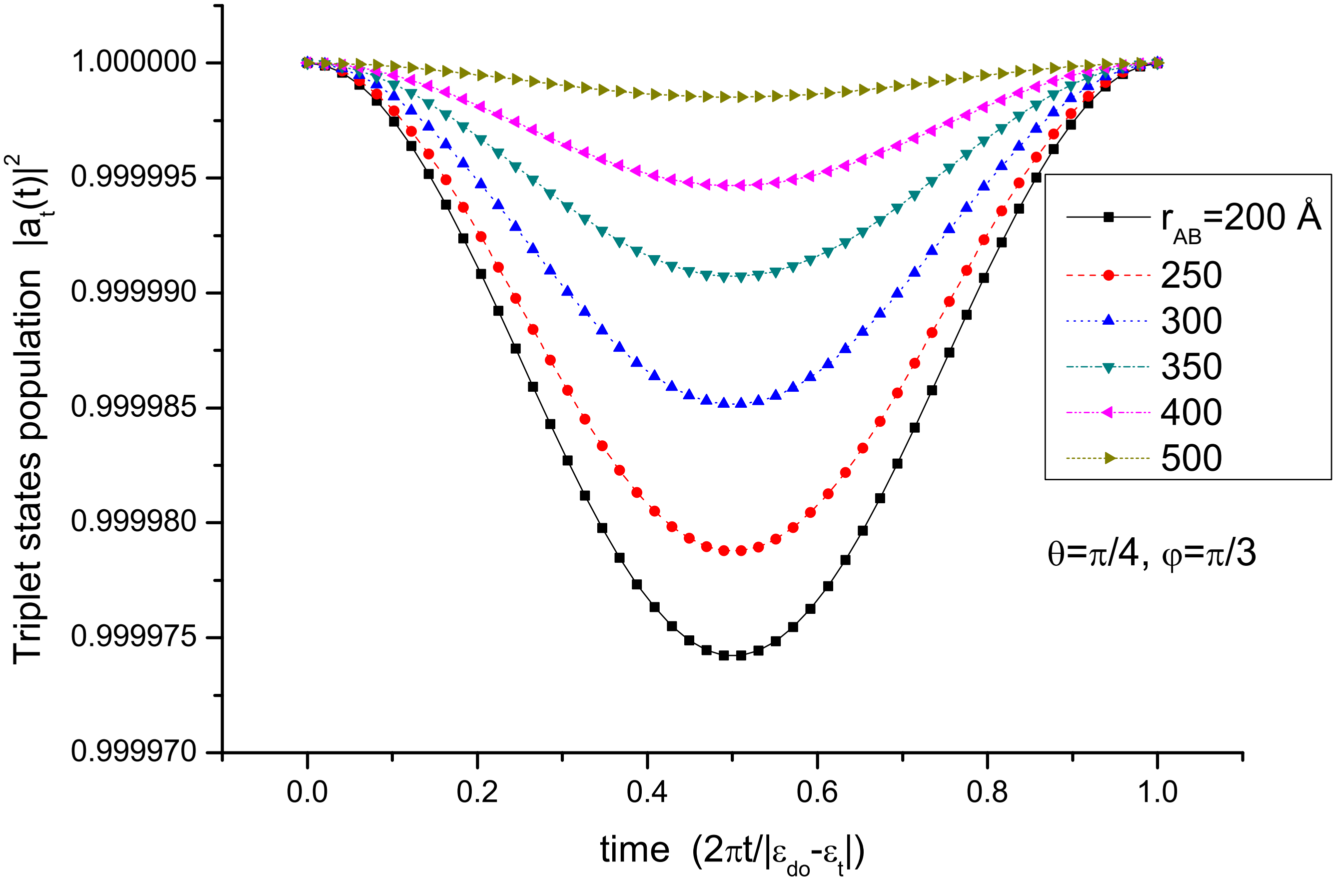

FIG. 6. (Color online) Triplet-state population dependence on spin-orbit interaction as a function of time at different interdot separations r_{AB} and fixed orientation of the interdot radius vector $\vec{r}_{AB}$ $(\theta=\pi/4,\varphi=\pi/3)$ (see text). The initial triplet-state population is taken to be equal. The external magnetic field $B_{0z}=0.5$ T.

resented as a product of a phase factor $\exp(i\alpha)$, where α is a real parameter, and a unitary transformation $U_S\in SU(4)$. Any transformation U_S is determined by $M=4^2-1=15$ independent real parameters $(\theta_1,\ldots,\theta_{15})$ so that

$$U_S(\theta_1,\ldots,\theta_M)=\exp\left(-i\sum_{i=1}^{M}\theta_iF_i\right), \qquad (60)$$

where the set of generators $\{F_i\}$ is an orthonormalized traceless, Hermitian matrix set that forms a Lie algebra $su(4)$ [F_i form a complete basis in a real M-dimensional vector space; they are analogs of Pauli matrices, σ_α, $\alpha=x,y,z$, in $su(2)$, see, e.g., Ref. 36]. From the representation Eq. (60), it follows immediately that $U_{\text{eff},S}(t)=\exp[-it(\frac{1}{4}\varepsilon_s+\frac{3}{4}\varepsilon_t)]U_{\text{eff}}(t)$ cannot match an arbitrary U_S because the number of independent parameters in Eq. (56) is at most 8—fewer than the number of θ_i. This can also be understood from the fact that the form of the Hamiltonian matrix [Eq. (56)] is not generic. In particular, the matrix is sparse, i.e., the entries (2,4) and (4,2) are zeros.

However, compositions of unitary transformation Eq. (59) taken at different sets of parameters can provide a universal set of unitary gates in $\mathbb{C}^4$. A well-known example of universal gates is provided by the Heisenberg interaction (at $J_H\neq 0$) with single-spin addressing (at $\Delta\vec{B}\neq 0$).[1]

From Eq. (56), it follows that a necessary and sufficient condition for obtaining a universal set of gates on two spins is to have an inhomogeneity in the magnetic field $\Delta\vec{B}\neq 0$ (the source of inhomogeneity can be different; it can be either strongly localized magnetic fields or g-factor engineered) and the Heisenberg interaction, $J_H\neq 0$. The reason is that when $\Delta\vec{B}=0$, the Hamiltonian Eq. (56) and the corresponding unitary transformations take a block-diagonal form, with singlet-triplet entries being zeros, while when $J_H=0$, the Hamiltonian form Eq. (58) will have zero off-diagonal block matrices. Clearly, even a composition of such unitary transformations taken at different sets of parameters, either $\varepsilon_s,\varepsilon_t,\vec{B}_A=\vec{B}_B$ or $\varepsilon_s=\varepsilon_t,\vec{B}_A,\vec{B}_B$, will be in a block-diagonal form and it cannot reproduce an arbitrary unitary transformation. Note that when one allows for encoding a qubit into three or more spins, the Heisenberg interaction alone is universal in the pseudospin model,[37,38] and Heisenberg along with an inhomogeneous magnetic field is universal for an encoding of a single qubit into a pair of spins.[39]

Moreover, it should be noted that in the homogeneous magnetic field case, unitary transformations restricted to the triplet subspace will not provide a universal set of gates. To

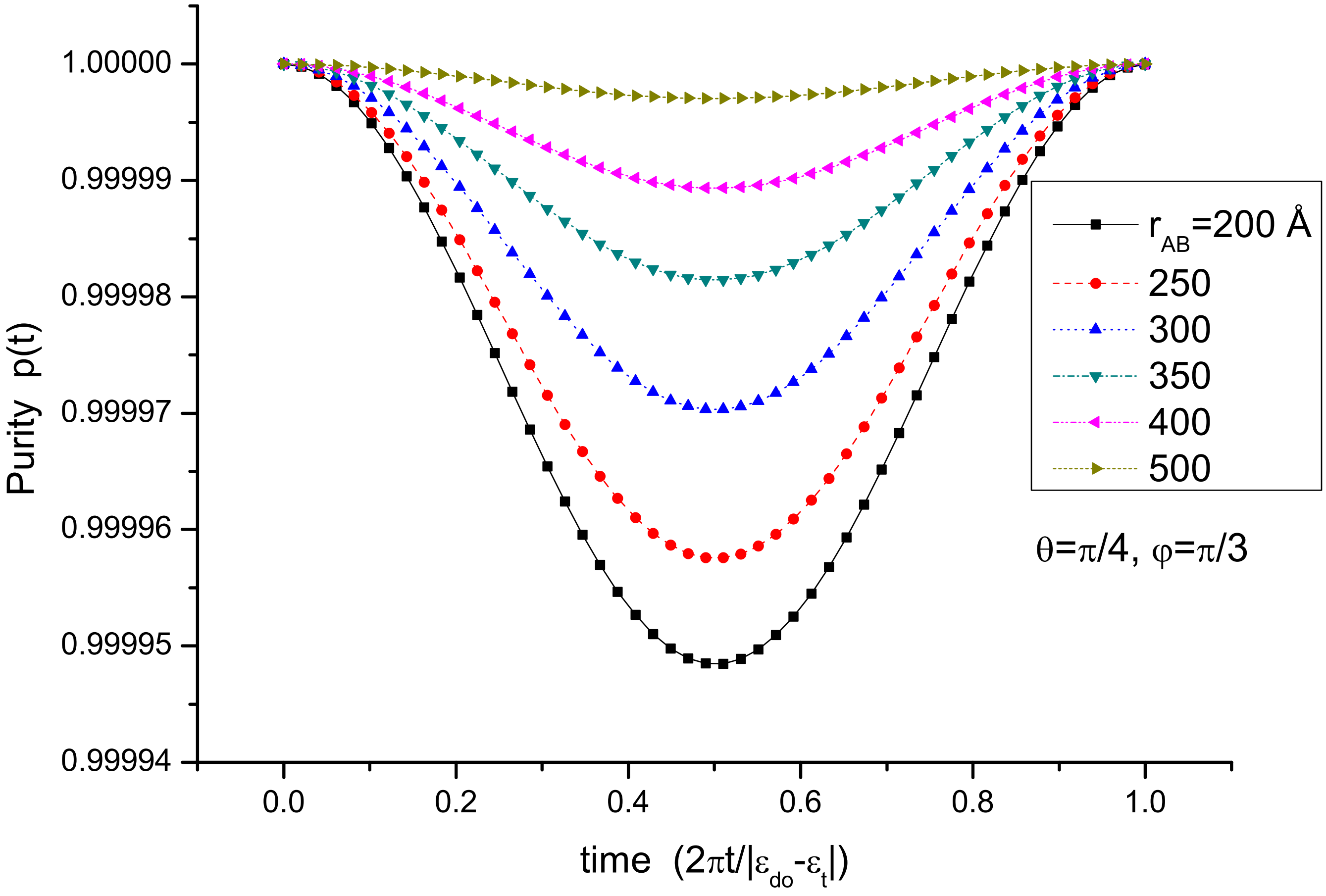

FIG. 7. (Color online) The purity in the presence of spin-orbit coupling. All parameters are the same as in Fig. 6.

prove this statement, let us consider a composition of two unitary transformations in the triplet subspace:

$$
\begin{aligned}
&\exp[-it_1H^{tt}(\vec{B}_1)]\exp[-it_2H^{tt}(\vec{B}_2)]\\
&\quad=\exp[-i(t_1+t_2)H^{tt}_{\mathrm{eff}}]\\
&\quad=\exp\Bigg\{-i(t_1+t_2)\varepsilon_t-it_1H^{tt}(\vec{B}_1)-it_2H^{tt}(\vec{B}_2)\\
&\qquad-\frac{t_1t_2}{2}[H^{tt}(\vec{B}_1),H^{tt}(\vec{B}_2)]+\cdots\Bigg\},
\end{aligned}
\tag{61}
$$

where on the right-hand side, we used the Campbell-Hausdorff formula.[40] From Eq. (37), one obtains

$$
[H^{tt}(\vec{B}_1),H^{tt}(\vec{B}_2)]=i\begin{pmatrix}2\mathcal{B}_z & \mathcal{B}^- & 0\\ \mathcal{B}^+ & 0 & \mathcal{B}^-\\ 0 & \mathcal{B}^+ & -2\mathcal{B}_z\end{pmatrix},
\tag{62}
$$

where $\vec{\mathcal{B}}=[\vec{B}_1\times\vec{B}_2]$ and $\mathcal{B}^{\pm}=\vec{\mathcal{B}}_x\pm i\vec{\mathcal{B}}_y$. Since the higher-order terms in the Campbell-Hausdorff formula consist of nested commutators between $H^{tt}(\vec{B}_1)$ and $H^{tt}(\vec{B}_2)$, we find that the effective Hamiltonian H^{tt}_{eff} corresponding to the product of two unitary transformations will still have a sparse form, with the (1,3) and (3,1) entries being zeros.

F. Is it possible to obtain a universal set of gates in the pure-spin model?

Simultaneously, we have just proved that in the pure-spin model, in the case of a homogeneous magnetic field, unitary transformations in the triplet subspace will not provide a universal set of gates. On the other hand, at $\Delta\vec{B}\neq 0$, we have already shown that the evolution of the spin density matrix is nonunitary. Let us assume that we have a nonunitary gate $\mathcal{L}_1$ so that $\rho(t_1)=\mathcal{L}_1(t_1)\rho(0)$. How could one define a composition of two nonunitary gates, $\mathcal{L}_2\mathcal{L}_1$? In order to do this unambiguously, $\mathcal{L}_2$ should obey a compatibility condition with the initial state (because a nonunitary $\mathcal{L}$ gate is not totally independent of the initial state, it includes some sort of correlation information encoded in the initial state), that is, a correlation $R_m(t_1)$ established between $a_s(t_1)$ and $a_t(t_1)$ amplitudes at $t=t_1$ should be included in the definition of the corresponding dynamics generator operators in $\mathcal{L}_2$. Equation (17), where the left-hand side and H^{ts} should be replaced by $R_m(t_1)$ and the identity matrix, respectively, provides a relationship between $R_m(t_1)$ and $R_m(0)$. If the correlation between the amplitudes at $t=0$ and $t=t_1$ is the same, $R_m(t_1)=R_m(0)$, then we obviously have $\mathcal{L}_1=\mathcal{L}_2=\mathcal{L}$ and $\mathcal{L}_2(t_2)\mathcal{L}_1(t_1)=\mathcal{L}(t_1+t_2)$.

In the total Hilbert space, the state is defined by 11 real parameters. While in the reduced description the spin density

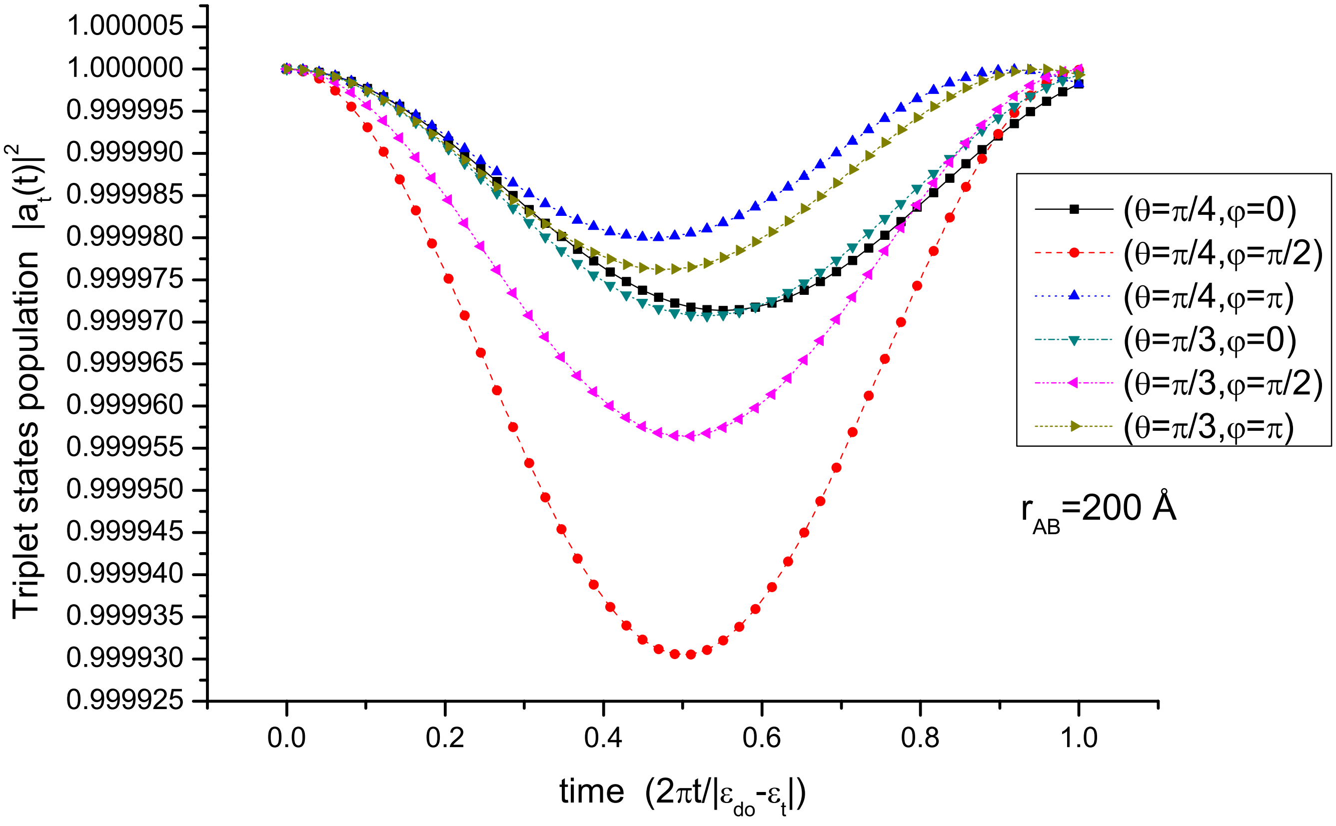

FIG. 8. (Color online) Effect of spin-orbit angular anisotropy on triplet-state population. All parameters are the same as in Fig. 6.

matrix is defined by five real parameters (for more on the spin density matrix parametrization in terms of a, see Sec. V). Fixing a correlation in the initial state, $a_s(0)=R_m(0)a_t(0)$, we have three complex equations between the amplitudes $a_s(0)$ and $a_t(0)$, which define a five-dimensional real manifold embedded into the total Hilbert space. Using these equations, we can separate six extra real degrees of freedom that we have in the total state description from those in the spin density matrix description. However, these extra degrees of freedom are not eliminated in the spin density description; they are included in the form of correlation matrix $R_\alpha(0)$, $\alpha=\{s,t,m\}$. It was shown in Part I that Eq. (14) provides an *exact* description of quantum evolution in the spin density space. Therefore, as long as we have a universal set of unitary gates in the total Hilbert space, this set of gates will be translated into the corresponding universal set of nonunitary gates generated by Eq. (14) because no information is lost in our "reduced" spin density matrix description.

G. Spin-orbit interaction in the pseudospin model

Let us consider spin-orbit effects, which are proportional to δB_{so}, in the pseudospin model. From Eq. (6), we obtain

$$\mathcal{H}_{so}(\delta\vec{B}_{so}) = \sum_{kk'=2,3} H^{ss}_{1k}\left(\frac{1}{\bar{E}I - H^{ss}}\right)^{-1}_{kk'} F_{k'}(\delta\vec{B}_{so}), \qquad (63)$$

where

$$F_k(\delta\vec{B}_{so}) = \sum_{i=1}^{3}\{H^{st}_{ki}(\delta\vec{B}_{so})\mathbf{K}_i + [H^{st}_{ki}(\delta\vec{B}_{so})]^*\mathbf{K}_i^\dagger\}. \qquad (64)$$

It follows from Eq. (49) that $H^{st}_{2i}=H^{st}_{3i}$ and $F_2=F_3$ and Eq. (64) can be reduced to

$$F_{2,3}(\delta\vec{B}_{so}) = F(\delta\vec{B}_{so}) = i\sqrt{2}\,\delta\vec{B}_{so}[\vec{s}_A \times \vec{s}_B]. \qquad (65)$$

Then, Eq. (63) can be rewritten as

$$\mathcal{H}_{so}(\delta\vec{B}_{so}) = \mathcal{A}_{do}F(\delta\vec{B}_{so}), \qquad (66)$$

where the coefficient $\mathcal{A}_{do}$ is proportional to the ratios of the amplitudes of double-occupancy transitions (H^{ss}_{12} and H^{ss}_{13}) and the energies of interelectron interaction ($H^{ss}_{22}-\bar{E}$ and $H^{ss}_{33}-\bar{E}$) in doubly occupied QDs:

$$\begin{aligned}\mathcal{A}_{do} &= \sum_{kk'=2,3} H^{ss}_{1k}\left(\frac{1}{\bar{E}I - H^{ss}}\right)_{kk'}\\ &= \frac{1}{\Delta}[H^{ss}_{12}(\bar{E}-H^{ss}_{33}-H^{ss}_{32}) + H^{ss}_{12}(\bar{E}-H^{ss}_{22}-H^{ss}_{23})]\\ &\approx \frac{H^{ss}_{12}}{\bar{E}-H^{ss}_{22}} + \frac{H^{ss}_{13}}{\bar{E}-H^{ss}_{33}}.\end{aligned} \qquad (67)$$

Here, the determinant is

$$\Delta = (\bar{E} - H^{ss}_{22})(\bar{E} - H^{ss}_{33}) - |H^{ss}_{23}|^2. \tag{68}$$

The effective spin-orbit interaction Hamiltonian Eq. (66) is different from the corresponding one obtained by Kavokin.[17] In our derivation, the double-occupancy states are essential, whereas in Ref. 17, these states are totally neglected. We showed above that neglecting double-occupancy states results in zero spin-orbit coupling. The very physical picture put forward in Ref. 17 to support the derivation was based on the assumption that when one of the two electrons localized at centers A or B tunnels to the adjacent center (say, from A to B), it experiences the influence of the spin-orbit field resulting from the underbarrier motion of the electron. Neglecting double-occupancy states means that the second electron should simultaneously tunnel from B to A so that the two electrons can never be found in the same QD. Indeed, in Ref. 17, this simultaneous two-electron transition is described by the product of two matrix elements: the overlap S and $(\delta\vec{B}_{so})_\alpha$, $\alpha=x,y,z$ [$\mathcal{H}_{so}\sim S(\delta\vec{B}_{so}\cdot[\vec{s}_A\times\vec{s}_B])$, our $\delta\vec{B}_{so}$ is related to Kavokin's **b** field via $\delta\vec{B}_{so}=-i\mathbf{b}$, and the overlap via $S=\Omega$]. With the orthogonalized molecular-type, two-center orbitals, such a one step two-electron transition gives a zero contribution since the spin-orbit interaction is a one-electron operator and the overlap $\tilde{S}=\langle\tilde{\phi}_A|\tilde{\phi}_B\rangle=0$. Equation (63) describes the two-step mechanism: In the first step, the two-electron system makes a transition from the singly occupied state Φ_{s1} to the intermediate, double-occupancy states Φ_{s2} and Φ_{s3} due to the interelectron interaction ($H^{ss}_{1k},k=1,2$ terms). Then, in the second step, as a result of the spin-orbit interaction, the system makes transitions from Φ_{s2} and Φ_{s3} to Φ_{ti} triplet states (the H^{st}_{ki} terms).

Let us find an estimate for

$$\mathcal{A}_{do}(R) \approx -\frac{2\sqrt{2}}{|\varepsilon_{do}-\varepsilon_t|}\int\int d\vec{r}_1 d\vec{r}_2\, \phi^2(r_1) \times \phi(|\vec{r}_2-\vec{R}|)\frac{1}{\varepsilon|\vec{r}_2-\vec{r}_1|}\phi(r_2), \tag{69}$$

where the hydrogenlike orbital $\phi(r)=(\pi a_B^3)^{-1/2}\exp(-r/a_B)$. Since electron 1 in Eq. (69) is localized around the effective Bohr radius a_B, one can approximate

$$\frac{1}{|\vec{r}_2-\vec{r}_1|} \approx \frac{1}{|\vec{r}_2 - a_B\vec{r}_1/r_1|}. \tag{70}$$

Then, the remaining integrals can be calculated exactly and we obtain

$$\mathcal{A}_{do}(r=R/a_B) \approx -\frac{4\sqrt{2}}{er}\{4eF_1(r) - 2[F_1(r+1)+F_1(r-1)] - [F_2(r+1)-F_2(r-1)] + 2[F_0(r+1)-F_0(r-1)]\}, \tag{71}$$

where

$$F_0(x) = \frac{\text{sgn}(x)}{48}[15(1+|x|)+6|x|^2+|x|^3]\exp(-|x|),$$

$$F_1(x) = \tfrac{1}{48}[3|x|(1+|x|)+|x|^3]\exp(-|x|),$$

$$F_2(x) = \frac{\text{sgn}(x)}{2}(1+|x|)\exp(-|x|). \tag{72}$$

Here, we used the same estimate for $\varepsilon_{do}-\varepsilon_t$ as in Sec. III D.

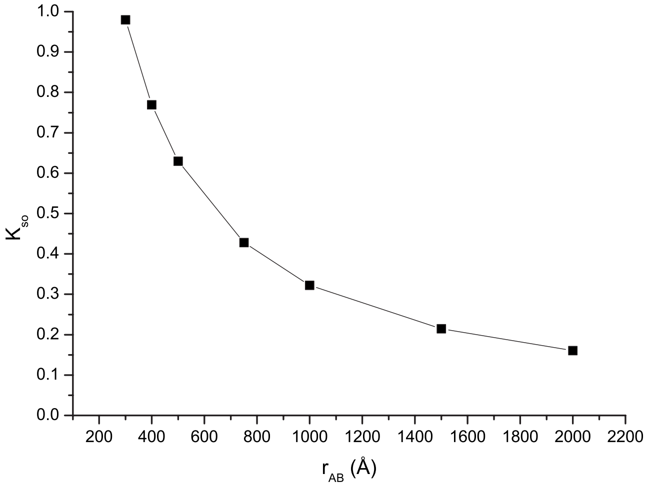

FIG. 9. Spin-orbit interaction reduction coefficient [Eq. (73)] as a function of interdot distance for GaAs.

In order to compare our calculations to Kavokin's result for GaAs, we have plotted in Fig. 9 the spin-orbit interaction reduction coefficient,

$$K_{so} = \frac{|\mathcal{A}_{do}|}{\sqrt{2}S}, \tag{73}$$

which is exactly the ratio of our and Kavokin's estimates as a function of interdot distance. As one can see, K_{so} decreases from 0.98 to 0.46 in the range $r_{AB}\sim 300-700$ Å. Interestingly, our results qualitatively agree with the results of Ref. 28, which were obtained in the region of interest [$r_{AB}\sim(3-7)a_B$], a reduction of about one-half relative to that of Refs. 17 and 41. According to Ref. 28, $\mathcal{H}_{so}\sim 2J\gamma_{\text{GK}}$, where J is the exchange integral calculated using the medium hyperplane method[29,30] and γ_{GK} is an angle of spin rotation due to spin-orbit interaction introduced by Gor'kov and Krotkov [$\gamma_{\text{GK}}\approx\frac{1}{2}\gamma_{\text{K}}$, γ_{K} being the corresponding angle of spin rotation introduced by Kavokin]. Note that Kavokin's γ_{K} and J are not independent parameters. In Ref. 17, γ_{K} was defined as $\Omega b/J$, so that their product $2J\gamma_{\text{K}}=2\Omega b$ does not depend explicitly on J.

IV. ELECTRONS ON LIQUID HELIUM

Recently, Lyon suggested that the spin of electrons floating on the surface of liquid helium (LHe) will make an excellent qubit.[42] Lyon's proposal, instead of using the spatial part of the electron wave function as a qubit as in the charge-based proposal,[43–46] takes advantage of the smaller vulnerability of the electron's spin to external magnetic perturbations and, as a consequence, a longer spin-coherence time. It also has the important advantage over semiconductor spin-based proposals (as first pointed out in the charge-based proposal[43–46]) that, with the electrons residing in the vacuum, several important sources of spin decoherence are eliminated so that the environment effects are highly suppressed (the spin-coherence time is estimated[42] to be $T_2 > 100$ s).

The geometry of the system with the electrons trapped at the LHe-vacuum interface (see Fig. 2 in Ref. 42) is conceptually similar to that of semiconductor heterostructures. The two electrons are trapped at sites $\vec{r}_A$ and $\vec{r}_B$ ($\vec{r}_{A,B}$ are radius vectors of the centers of quantum dots in the $z=0$ plane) by the two attractive centers created by two charged spherical electrodes located below the LHe surface at distance h and separated by the interdot distance $r_{AB}=|\vec{r}_A-\vec{r}_B|$:

$$V_{\mathrm{tr}}(z,\vec{r}) = -\frac{\Lambda}{z} + V_A(z,\vec{r}) + V_B(z,\vec{r}), \tag{74}$$

$$V_{A,B}(z,\vec{r}) = -\frac{Q_{A,B}}{\sqrt{(z+h)^2+(\vec{r}-\vec{r}_{A,B})^2}}, \tag{75}$$

where the first potential, $-\Lambda/z$, is due to attraction to the image charge induced by an electron in the LHe $[\Lambda=(\varepsilon-1)/[4(\varepsilon+1)]\approx 7\times10^{-3}$, with $\varepsilon\approx1.057$ being the dielectric constant of helium]. For purposes of interaction control, the $Q_{A,B}$ charges on the electrodes can be made variable in time. The electrons are prevented from penetrating into helium by a high potential barrier ($\sim$1 eV) at the helium surface, so that formally, one can set $V_{\mathrm{tr}}=\infty$ at $z<0$. The in-plane and out-of-plane motions of electrons in the potential of Eq. (74) are, in general, nonseparable. However, near the electrode's position $\vec{r}_{A,B}$, the potential of Eq. (75) is approximately separable in the z and $\vec{r}$ coordinates,

$$V_{A,B}(z,\vec{r}) \approx -\frac{Q_{A,B}}{h} + \mathcal{E}_{\perp A,B}z + \frac{1}{2}\omega_{A,B}^2(\vec{r}-\vec{r}_{A,B})^2, \tag{76}$$

where it is assumed that z and $|\vec{r}-\vec{r}_{A,B}|\ll h$ and $\mathcal{E}_{\perp A,B}=Q_{A,B}/h^2$ and $\omega_{A,B}=(Q_{A,B}/h^3)^{1/2}$. In the separable approximation of Eq. (76), the electron's motion in the z direction ($z>0$) is described by a one-dimensional (1D) Coulomb potential perturbed by a small Stark interaction and the in-plane motion by a two-dimensional (2D) oscillatory potential. We assume that the out-of-plane motion in the z direction is frozen in the ground state of a 1D Coulomb potential,

$$\varphi_0^C(z) = 2\sqrt{\Lambda}(\Lambda z)\exp(-\Lambda z), \tag{77}$$

and the in-plane motion, in a superposition of the in-plane confining oscillatory potential and, possibly, a perpendicular magnetic field, is described by the FD states of Eq. (23). Then, the calculation of h_{ij} and $v_{ee}(ij;kl)$ in Eqs. (27)–(29) in the chosen basis set is reduced to the 1D integrals

$$h_{AB} = \left[-\frac{\Lambda}{2} + \frac{1}{4l_A^2} - 2\Lambda Q_A g_c(\alpha,\beta,\lambda_A) - 2\Lambda Q_B g_c(\alpha,\beta,\lambda_B)\right] S_{AB},$$

$$h_{AA} = -\frac{\Lambda}{2} + \frac{1}{4l_A^2} - 2\Lambda Q_A g_c(\alpha,\beta_A,0) - 2\Lambda Q_B g_c(\alpha,\beta_A,2\Lambda r_{AB}),$$

$$h_{BB} = -\frac{\Lambda}{2} + \frac{1}{4l_B^2} - 2\Lambda Q_A g_c(\alpha,\beta_B,2\Lambda r_{AB}) - 2\Lambda Q_B g_c(\alpha,\beta_B,0),$$

$$g_c(\alpha,\beta,\lambda) = \int_0^\infty dx J_0(\lambda x)\exp(-\alpha x-\beta x^2)/(x+1)^3,$$

$$\alpha = 2\Lambda h, \quad \beta = \frac{(2\Lambda l_A l_B)^2}{l_A^2+l_B^2},$$

$$\beta_{A,B} = 2(\Lambda l_{A,B})^2, \quad \lambda_{A,B} = \frac{2\Lambda l_{B,A}^2}{l_A^2+l_B^2} r_{AB}, \tag{78}$$

$$v_{ee}(AB;CD) = N_{ee} g_{ee}(a,b),$$

$$N_{ee} = \frac{\Lambda l_A l_B l_C l_D}{(l_A^2+l_C^2)(l_B^2+l_D^2)} \exp\left(-\frac{r_{AC}^2}{4(l_A^2+l_C^2)} - \frac{r_{BD}^2}{4(l_B^2+l_D^2)}\right),$$

$$g_{ee}(a,b) = \int_0^\infty dx J_0(bx)\exp(-ax^2)(3x^2+9x+8)(x+1)^{-3},$$

$$a = 4\Lambda^2\left(\frac{l_A^2 l_C^2}{l_A^2+l_C^2} + \frac{l_B^2 l_D^2}{l_B^2+l_D^2}\right),$$

$$b = 2\Lambda\left|\frac{l_C^2\vec{r}_A + l_A^2\vec{r}_C}{l_A^2+l_C^2} - \frac{l_D^2\vec{r}_B + l_B^2\vec{r}_D}{l_B^2+l_D^2}\right|, \tag{79}$$

where $i=A,B,C,D$ in the two-electron matrix elements denotes orbitals with the effective lengths l_i localized at $\vec{r}_i$, and $J_0(x)$ is the zeroth order Bessel function. From Eq. (79), one can obtain the following expression for the Heisenberg interaction constant:

$$J_H = -1.36\times10^4\Lambda g_{ee}(a,0)S^2 \text{ meV},$$

$$a = \frac{8\Lambda^2 l_A^2 l_B^2}{l_A^2+l_B^2}. \tag{80}$$

Note that J_H is proportional to the square of the overlap matrix element S [Eq. (25)], and the integral $g_{ee}(a,0)$ does not depend on the interdot distance r_{AB}. As a rough estimate, one can approximate the rational function in the integral $g_{ee}(a,0)$ by a constant 8, obtaining $g_{ee}(a,0)\approx 4\sqrt{\pi/a}$ as a result.

Figure 10 shows the magnitudes of the Heisenberg and dipole-dipole spin interaction, $|J_H|$ and J_{dip} from Eqs. (80)

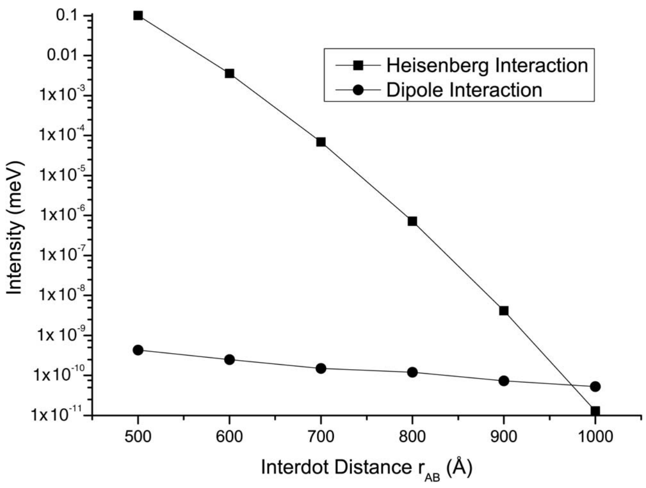

FIG. 10. The Heisenberg and dipole-dipole spin interaction magnitudes as a function of interdot distance. The distance from the electrodes to the LHe surface $h=800$ Å and the charges on the electrodes $Q_A=Q_B=1$ a.u. The magnetic field $\vec{B}_0=0$.

and (36) respectively, as a function of interdot distance. As estimated, the magnitude of the Heisenberg interaction is comparable to the weak dipole-dipole interaction at $r_{AB} \simeq 1000$ Å. However, we remark that the strong dependence of $S \sim \exp(-\alpha r_{AB}^2)$ (quadratic in the interdot distance) is due to the quadratic dependence on coordinates in the exponent of the corresponding oscillatory wave functions. Asymptotically, the confining potential in Eq. (75) behaves as a 2D Coulomb potential, so that one should expect a milder coordinate dependence, $S \sim \exp(-\alpha r_{AB})$, at large distances (assuming $B_0=0$) and the rough estimate of Eq. (80) provides a lower bound for the Heisenberg interaction strength.

Lyon suggested[42] using, instead of the exchange interaction, the magnetic dipole-dipole interaction between the spins in order to implement two-qubit gates, motivating this by the strong sensitivity of the exchange coupling to the parameters of the system and, hence, the corresponding difficulties with attempting to control this interaction. Our analysis confirms this, though, of course, it is not easy to control the dipole-dipole interaction either: Eq. (36) shows that this interaction depends on only one controllable parameter, the interdot distance (the g factor is a constant in vacuum).

Similarly to Figs. 1 and 2, Figs. 11 and 12 demonstrate nonunitary effects in the pure-spin model due to magnetic field inhomogeneity in the electrons-on-LHe system. The interdot distance shown is $r_{AB}=900$ Å. At this distance, the Heisenberg interaction still prevails over the dipole interaction by at least an order of magnitude. Again, the pattern seen in the singlet-triplet state population redistribution is clearly oscillatory.

V. DISCUSSION AND CONCLUSION

We have performed a comparative study of pure-spin and pseudospin dynamics for a system of two interacting electrons trapped in two QDs. We have shown that when there is negligible coupling between the spin and orbital degrees of freedom, which is the case of near $\vec{B}$-field homogeneity and negligible spin-orbit interaction, the system spin dynamics is unitary in both pure-spin and pseudospin models and is governed by the Zeeman interaction Hamiltonian of the total spin $\vec{S}$ ($S=1$) with the magnetic field $\vec{B}_{av}$. The singlet and triplet states are totally decoupled; the total spin is conserved. The spin system Hilbert space can be decomposed into two independent, singlet and triplet subspaces, the singlet spin states being magnetically inactive ($S=0$). Thus, the two-electron spin system restricted to the triplet subspace physically embodies a qutrit. The Heisenberg interaction operates differently in pure-spin and pseudospin models. If, for

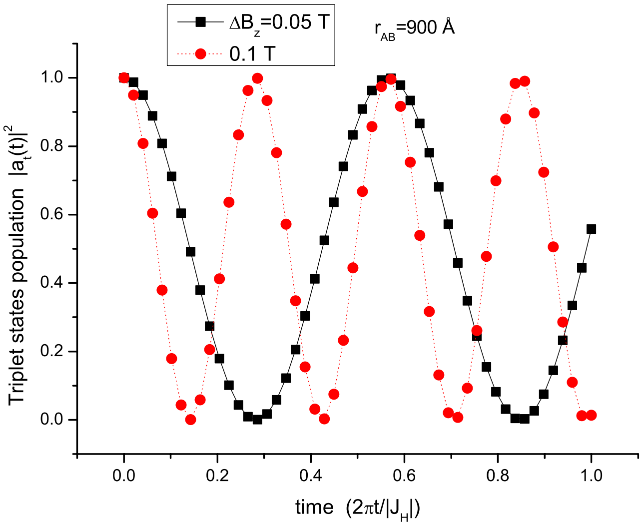

FIG. 11. (Color online) The triplet-state population of electrons trapped in QDs on a LHe surface as a function of time at different magnetic field differences $\Delta B_z=0.05$ and 0.1 T. Initially, the system is in the triplet state $|S=1, M_S=0\rangle$. The distance between QDs is 900 Å. All other parameters are the same as in Fig. 10.

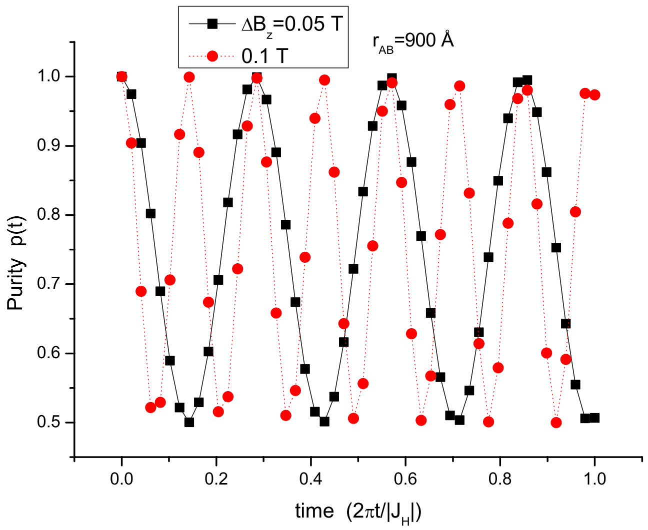

FIG. 12. (Color online) The purity $p(t)$ for the same parameters as in Fig. 11.

simplicity, we neglect double-occupancy states, the pseudospin state is totally defined by four complex amplitudes: $\{a_{s1}(0), a_{ti}(0), i=1,2,3\}$ in the basis $\{\Phi_{s1}, \Phi_{ti}\}$, so that the Heisenberg interaction results in a phase unitary transformation: $\{a_{s1}(t)=\exp(-i\varepsilon_s t)a_{s1}(0), a_{ti}(t)=\exp(-i\varepsilon_t t)a_{ti}(0)\}$. Since the spin density matrix $\rho(t)$ is a bilinear combination of a: $\rho_{tij}(t)=a_{ti}(t)a_{tj}^*(t)$, $\rho_s(t)=1-\Sigma_i\rho_{tii}(t)$, the ρ state will not be affected by the unitary transformation induced by the Heisenberg interaction.

We have also shown that unitary quantum gates realized in both spin models do not provide a universal set of gates under the condition $\Delta\vec{B}=0$. In order to obtain a universal set of gates, there should be both nonzero coupling between singlet and triplet states ($\Delta\vec{B}\neq 0$) and nonzero Heisenberg interaction ($J_H\neq 0$). Although at $\Delta\vec{B}\neq 0$ pure-spin dynamics becomes nonunitary, one can establish a relationship between unitary gates in pseudospin and the corresponding nonunitary gates in pure-spin dynamics, so that a universal set of quantum gates constructed within the pseudospin model will generate a universal set of nonunitary gates in pure-spin dynamics.

To demonstrate the nonunitary effects, which are proportional to the square of the magnetic field inhomogeneity, in pure-spin dynamics, we have calculated how singlet- and triplet-state populations, as well as the purity and the Lamb energy shift, are affected by $\Delta\vec{B}\neq 0$ and spin-orbit interaction in n-doped GaAs semiconductors. These effects are found to be strongly dependent on the ratio of $\vec{B}$-field inhomogeneity and the Heisenberg interaction constant, $|\Delta B_z/J_H|$. For example, the singlet-triplet state population redistribution is maximal at $t=\pi n/\omega$, where $\omega=\sqrt{J_H^2+\Delta B_z^2}$ and $n=1,3,\ldots$, and the singlet-state population can achieve the value $\Delta B_z^2/\omega^2$. Thus, we can conclude that the Heisenberg interaction, characterized by the interaction constant J_H, plays an essential role in producing nonunitary effects in pure-spin dynamics. Spin-orbit interaction effects are found to be roughly 4 orders of magnitude smaller as compared to those caused by $\vec{B}$-field inhomogeneity.

As shown in Figs. 1–8, there are clear oscillations in the pure-spin dynamics and the nonunitary behavior of the spin density matrix does not show the decaying pattern characteristic of a real decoherence process. This should be expected since the bath, the electron orbitals, in our spin model is not a real stochastic or infinite external bath, an interaction with which may result in irreversible decoherence. In essence, the spin dynamics is embedded in space and our bath is too small. The coordinate Hilbert space in the two-orbital ground-state approximation adopted in the present paper is represented by four two-electron coordinate basis wave functions. In principle, the coordinate bath can be large in a system where couplings between excited- and ground-state orbitals are not negligible. This is an interesting question for future investigation: how will couplings to excited orbitals affect the nonunitary spin dynamics? The other interesting generalization of the present model is inclusion of real environment effects, i.e., the real stochastic bath representing the interaction of electron spins with the semiconductor medium. We will consider these and other generalizations in future publications.

In the pseudospin model, where $\vec{B}$-field inhomogeneity results in first-order effects, we have estimated the contribution of the spin-orbit interaction to the effective pseudospin Hamiltonian, namely, the DM spin-orbit interaction term, and have suggested a two-step mechanism: coupling between the singly occupied singlet state and triplet states occurs via intermediate, double-occupancy states (direct coupling between these states turns out to be zero due to orthogonality of the orbitals involved in the transition). Our calculations predict a smaller magnitude of the spin-orbit interaction as compared to the estimates of Ref. 17 but are consistent with the results of Ref. 28.

In our second application, we demonstrated (in Figs. 11 and 12) nonunitary effects due to $\Delta\vec{B}\neq 0$ in a system of electrons trapped above a liquid helium surface, namely, the spin-based quantum computing proposal by Lyon.[42] A more thorough investigation of spin dynamics in this system is left for a future publication.

Although the ρ dynamics becomes nonunitary in general (at $\Delta\vec{B}\neq 0$), it is controllable by modulating the interaction parameters, J_H, $\vec{B}_{av}$, and $\Delta\vec{B}$. Since the nonunitarity comes from the magnetic field inhomogeneities, $\Delta\vec{B}$ and/or $\delta\vec{B}_{so}$, and since the ρ dynamics is quadratically protected from these fields, this might prove to be important in practical quantum computing as minimizing coupling between spin and orbital degrees of freedom *quadratically* improves the fidelity of unitary gates in the ρ-state space.

In conclusion, we note that the two-electron spin density matrix description advocated in this paper is expected to be useful when localized electrons (e.g., trapped in QDs) are not spatially resolved or resolvable. Formalization of this physical situation results in a requirement imposed on the observables accessible by a measurement, namely, that they should not depend on the electron's coordinates. This means that any measurable observable should be represented as a tensor product $\mathcal{O}=I_{\vec{r}}\otimes\mathcal{O}_S$, where $I_{\vec{r}}$ is the identity operator acting in the coordinate space and $\mathcal{O}_S$ is a pure (Hermitian) spin operator. Since any observable for a system of identical particles must be invariant under permutations of the particles, we also require that $\mathcal{O}_S$ should be symmetric with respect to electron permutations. The observable dynamics is then completely described by the spin density matrix ρ: $\bar{O}(t)=\langle\Psi_{\mathrm{tot}}(t)|\mathcal{O}|\Psi_{\mathrm{tot}}(t)\rangle=\mathrm{tr}[\mathcal{O}_S\rho(t)]$. As an example, we consider a nonlocal pure-spin operator $\mathcal{O}_S=\mathbf{S}=|\chi_s\rangle\langle\chi_s|=\frac{1}{4}I-\vec{s}_1\cdot\vec{s}_2$, in which case we obtain the singlet-state population, $\bar{O}(t)=\Sigma_{i=1}^3|a_{si}(t)|^2$.

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