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Angaben zur Veröffentlichung / Publication details:

Häusler, Wolfgang, and Peter Hänggi. 2006. "Spin conversion rates due to dipolar interactions in monoisotopic quantum dots at vanishing spin-orbit coupling." *Physical Review B* 73 (12): 125329. https://doi.org/10.1103/physrevb.73.125329.

Spin conversion rates due to dipolar interactions in monoisotopic quantum dots at vanishing spin-orbit coupling

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(Received 16 August 2005; revised manuscript received 31 January 2006; published 22 March 2006)

Dipolar interaction between the magnetic moments of electrons is studied as a source for electron spin decay in quantum dots or arrays of quantum dots. This magnetic interaction will govern spin decay, after other sources, such as the coupling to nuclear spins or spin-orbit coupling, have been eliminated by a suitable sample design. Electron-electron (Coulomb) interactions, important for magnetic properties, are included. Decomposing the dipolar operator according to the symmetric group of electron permutations allows one to deduce vanishing decay channels as a function of electron number and spatial symmetries of the quantum dot(s). Moreover, we incorporate the possibility of rapid phonon-induced spin-conserving transitions which crucially affect the temperature dependence of spin decay rates. An interesting result is that a sharp increase of the spin decay rate occurs already at relatively low temperatures.

DOI: 10.1103/PhysRevB.73.125329 PACS number(s): 73.21.La, 72.25.Rb, 75.75.+a, 85.35.Be

I. INTRODUCTION

One of the proposals to realize qubits for quantum computing[1] uses the electron's spin in semiconducting solid state nanostructures.[2,3] This approach could benefit from traditional electronics device experience and allows for straightforward scalability. Recent efforts have succeeded to demonstrate controlled preparation and detection of single[4] and of pairs[5] of electron spins in quantum dots. Aiming for long running coherent computations, however, solid state based devices somewhat suffer from relatively short spin decoherence and relaxation times as a drawback, for example, compared to nuclear spin-based qubits.[6] Exponential decay of the upper Zeeman level population has been observed in GaAs quantum dots[4] over times not exceeding 10^{-3} s. Therefore, it is important to know (and, if possible, to control) any kind of mechanism causing spin relaxation in solids, and particularly in semiconductors. Previous theoretical work has valued several contributions. They can be subdivided into two classes regarding the magnetic mechanism to mix spin states as a source for spin decay: (i) spin-orbit coupling,[7–10] also, recently, in its interplay with the electron-electron interaction,[11,12] or (ii) coupling to nuclear spins. The latter can act through the spin-flip Overhauser effect by hyperfine interactions.[13–16] In principle, both magnetic sources for spin mixing can be eliminated by a proper device design and by the choice of the semiconductor material. Nonvanishing spin-orbit splitting can have several causes in semiconductors which we briefly address: *p*-type bands, i.e., usually valence bands, may split by spin-orbit effects, arising near the nuclei. Secondly, the lack of spatial inversion symmetry produces spin splitting even of *s*-type bands, either by the Dresselhaus mechanism arising in the absence of crystallographic centrosymmetry as in zinc blende or by wurtzite structures. The latter particularly refers to all III–V semiconductors with GaAs being the most striking example. Also devices lacking structural inversion symmetry, e.g., near surfaces or in asymmetric quantum wells, producing internal electric fields show spin splitting due to the Rashba mechanism. Fortunately, this latter spin-orbit source may be suppressed by fine tuning suitable gate voltages.[17] The goal to avoid spin-orbit coupling effects therefore suggests using spins of conduction band electrons in Si or in Ge in carefully symmetrically prepared structures. Also the attempt to avoid coupling to nuclear spins favors the use of Si or Ge: their natural isotopic mixture contains nuclear spin $I=0$ to more than 95% (Si) or more than 92% (Ge), respectively.[18]

With this work we consider the effect of dipolar interactions between the magnetic moments of electron spins which, for fundamental reasons, *cannot* be removed by design. While considerably weaker[19] than the above quoted mechanisms, this interaction unavoidably causes spin relaxation and, in the absence of other magnetic interactions, combined with the never vanishing electron-phonon coupling,[20] will set the ultimate limit for long time quantum computations using electron spins,[3] even in optimally designed structures. We study transitions between energy levels differing in their *total* spins[21] and disregard here effects associated with transitions between Zeeman levels (which conserve the symmetry of many-body electron levels, see below) at finite magnetizations when an external magnetic field is applied. After introducing the model in Sec. II we reveal circumstances of particular spin stability with respect to dipolar interactions (Sec. III A), also regarding excited (many electron) states in Sec. III D, depending on the electron number and on the symmetry of the single or the ensemble of quantum dots.

We explicitly include Coulomb interactions[11,12] due to their importance for magnetic properties. For example, they can cause total ground state spins greater[22–25] than $S=0$ or

$S=\frac{1}{2}$, as expected for even or odd numbers N of noninteracting electrons. Here, we take the electrons to be confined inside one quantum dot or in different quantum dots. As a complementary approach to the noninteracting or weakly interacting regime we focus on strong Coulomb interactions at low electron densities, where pocket states[26–28] offer a reliable description of many-body states localized by Coulomb repulsion, even in single quantum dots.[23] Pocket states are briefly reviewed in Sec. III C for the present purpose to determine matrix elements of the dipolar interaction in Sec. III D.

Pursuing most of the foregoing theoretical work on electron spin decay in quantum dots we consider in Sec. IV A phonons (which themselves cannot change spin states) to provide the transition energy between discrete dot levels. Contrary to extended bulk situations, this transition energy is much bigger than mere magnetic energies which is is one reason for the relative stability of quantum dot compared to bulk electron states, in accordance with experimental fact.[29] Spin-changing transitions due to the combined action of dipolar energy and phonons are discussed in Sec. IV B. Generalizing previous results, we account for rapid *spin-conserving* excitations of the electron system induced by phonons that occur already at relatively low temperatures; these transitions turn out to govern predominantly the temperature dependence of spin decay times, discussed in Sec. IV C. Finally, we summarize and value our findings in Sec. V.

II. MODEL

Specifically, we consider the N-electron system

$$H_0 = \sum_{i=1}^{N} \left(\frac{\mathbf{p}_i^2}{2m^*} + v(\mathbf{r}_i) \right) + \frac{1}{2} \sum_{i \neq j} w(|\mathbf{r}_i - \mathbf{r}_j|) \tag{1}$$

confined by the potential $v(\mathbf{r})$ which is supposed to describe a single quantum dot or more complex situations of many quantum dots, such as for example N quantum dots each containing a single electron. To be realistic, particularly regarding magnetic properties, we include interactions between electrons $w(r)=\frac{e^2}{\kappa} r^{-1}$, depending on the static dielectric constant κ of the host material; Coulomb interactions are always considerably stronger than dipolar energies. Moderate screening, not reducing the interaction range to values smaller than the electron separation, will not affect qualitatively our results. In Eq. (1), $\mathbf{p}_i$ and $\mathbf{r}_i$ are d-component momentum and position vectors, depending on the dimensionality d of the quantum dot wave functions (in heterostructures, $d=2$); m^* is the band electron mass.

Notice that at strong Coulomb repulsion, which is the focus of this work, N electrons Wigner crystallized[23,30] in a single quantum dot become in their theoretical treatment at low energies very similar to the case of N electrons localized in separate quantum dots. The essential physics of both situations is captured by an antiferromagnetic Heisenberg lattice model.[26,31] Eigenstates of H_0 exhibit well-defined spins S and can be classified according to the eigenvalues of the z-component $\hat{S}_z$ and the square $\hat{\mathbf{S}}^2$ of the total spin operator $\hat{\mathbf{S}}=\Sigma_{i=1}^{N}\hat{\mathbf{S}}_i$ yielding eigenvalues S_z and $S(S+1)$, respectively. With $SU(2)$ symmetry in spin space, Zeeman multiplets $-S \leqslant S_z \leqslant +S$ are degenerate. We index eigenstates $|\phi_n\rangle$ and eigenvalues E_n of H_0 by n, taken to incorporate the values of S and S_z. Transitions between $|\phi_n\rangle$ and $|\phi_{n'}\rangle$ may or may not change S. The present work focuses on inelastic transitions that change the *total* spin values $S \to S'$ rather than on transitions within a Zeeman multiplet. As already mentioned, many-electron ground states $|\phi_{n=0}\rangle$ may exhibit total spin values $S_0 > \frac{1}{2}$ as a result of electron-electron interactions.[22–25]

III. TRANSITION MATRIX ELEMENTS

In order to satisfy the Pauli principle an N-fermion state $\langle \mathbf{r}_1, s_1, \ldots, \mathbf{r}_N, s_N | \phi_n \rangle$ must belong to the $A_2 \equiv [1^N]$ representation of the symmetric (permutational) group S_N with respect to permutations $p \in S_N$ of the particle enumeration, $\{1, \ldots, N\} \to \{p(1), \ldots, p(N)\}$, see Ref. 32. When permuting only spin coordinates $\{s_1, \ldots, s_N\} \to \{s_{p(1)}, \ldots, s_{p(N)}\}$ the state $|\phi_n\rangle$ transforms according to the irreducible representation (partition) $\Gamma=[N/2+S, N/2-S]$ of S_N for spin-$\frac{1}{2}$ fermions[27,28] at given $S=\{{}^{0}_{1/2}\}, \ldots, N/2$ for $\{{}^{\text{even}}_{\text{odd}}\}N$. Correspondingly, when permuting only positions $\{\mathbf{r}_1, \ldots, \mathbf{r}_N\} \to \{\mathbf{r}_{p(1)}, \ldots, \mathbf{r}_{p(N)}\}$, $|\phi_n\rangle$ transforms according to $\bar{\Gamma}=[2^{N/2-S}, 1^{2S}]$ (with $\Gamma \times \bar{\Gamma}$ containing the A_2 representation). We notice that total spin-changing transitions require altering the wave function's symmetry, which necessitates operators acting simultaneously in position and in spin space (by contrast, transitions within a Zeeman multiplet leave unaltered the symmetries of wave functions).

A. Dipolar energy

Here, we investigate the dipolar interaction H^D between electrons. As seen in Eq. (3) below, it contains products of position and spin operators and, indeed, mixes spin states. However, it is by far too weak to provide the energy separating quantum dot eigenlevels. Focusing on Si, we consider in Sec. IV acoustical deformation potential phonons[33] to supply the necessary transition energy. Unaided electron-phonon coupling, though, does not mix spin states and thus leaves spins unaltered. Eventually, it turns out that dipolar interaction, as a result of its smallness, causes considerably smaller transition rates at low temperatures than, for instance, nuclear spin-induced spin mixing.[13]

The operator of the dipolar energy

$$H^D = \frac{1}{2} \sum_{i \neq j} H_{ij}^D \tag{2}$$

is, as required for identical particles, invariant with respect to permuting the electron enumeration; however, H^D can be decomposed into parts that are *not* invariant under permuting coordinates $\mathbf{r}_i$ or spins $\hat{\mathbf{S}}_i$ separately. Let us first recap the interaction between a pair of magnetic moments

$$H_{ij}^{D} = \frac{\gamma^2}{r_{ij}^5}[r_{ij}^2 \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j - 3(\mathbf{r}_{ij} \cdot \hat{\mathbf{S}}_i)(\mathbf{r}_{ij} \cdot \hat{\mathbf{S}}_j)], \tag{3}$$

where $\gamma = ge\hbar/2mc$ (c is the velocity of light and the g factor for dot carriers which even in few electron quantum dots is found to take basically bulk values).[5,34] Its Heisenberg-like first part is manifestly $SU(2)$-invariant in spin space and commutes with $\hat{\mathbf{S}}^2$. This part neither changes S nor S_z and just renormalizes the energies slightly. It therefore can be ignored in view of the smallness of dipolar energies compared to the dot level separations. In Eq. (3) we abbreviate $\mathbf{r}_{ij} := \mathbf{r}_i - \mathbf{r}_j$ and $r_{ij} := |\mathbf{r}_{ij}|$. The second part of H_{ij}^D can be decomposed as

$$\frac{1}{r_{ij}^5}(\mathbf{r}_{ij} \cdot \hat{\mathbf{S}}_i)(\mathbf{r}_{ij} \cdot \hat{\mathbf{S}}_j) = [H_{ij}^{(0)} + H_{ij}^{(1)} + H_{ij}^{(2)}], \tag{4}$$

where the three terms

$$H_{ij}^{(0)} = \frac{|\varrho_{ij}|^2}{4}(\hat{S}_{+i}\hat{S}_{-j} + \hat{S}_{-i}\hat{S}_{+j}) + \zeta_{ij}^2 \hat{S}_{zi}\hat{S}_{zj}, \tag{5}$$

$$H_{ij}^{(1)} = \frac{\zeta_{ij}}{2}[\varrho_{-ij}(\hat{S}_{+i}\hat{S}_{zj} + \hat{S}_{zi}\hat{S}_{+j}) + \varrho_{+ij}(\hat{S}_{-i}\hat{S}_{zj} + \hat{S}_{zi}\hat{S}_{-j})], \tag{6}$$

$$H_{ij}^{(2)} = \frac{\varrho_{+ij}^2}{4}\hat{S}_{-i}\hat{S}_{-j} + \frac{\varrho_{-ij}^2}{4}\hat{S}_{+i}\hat{S}_{+j}, \tag{7}$$

are responsible to alter symmetries and spins after carrying out summation over ($i \neq j$). In Eqs. (5)–(7) they change S_z by 0, ±1, and ±2, respectively. $\hat{S}_{\pm} := \hat{S}_x \pm i\hat{S}_y$ denote usual rising or lowering operators in spin space, $\varrho_{\pm ij} := (x_{ij} \pm iy_{ij})/r_{ij}^{5/2}$ is a complex (angular momentum generating) coordinate in the plane perpendicular to the axes of spin quantization, taken as the z axes, and $\zeta_{ij} := z_{ij}/r_{ij}^{5/2}$.

The spin-changing part Eqs. (5)–(7) of H^D can now further be decomposed according to partitions Γ of the symmetric group S_N,

$$\sum_{i \neq j}(H_{ij}^{(0)} + H_{ij}^{(1)} + H_{ij}^{(2)}) = H^{\Gamma=[N]} + H^{\Gamma=[N-1,1]} + H^{\Gamma=[N-2,2]}. \tag{8}$$

This latter representation is particularly useful to deduce nonzero transition matrix elements between quantum dot eigenstates of different *total* spins. No other partitions occur since H_{ij}^D transforms as a product of two vector operators in position as well as in spin space [cf. Eq. (3)], i.e., as a tensor of rank two. In Eq. (8) $H^{\Gamma=[N-1,1]}$ changes the total spin S of dot by ±1 and $H^{\Gamma=[N-2,2]}$ by ±2, where the latter occurs only for $N \geqslant 4$ while the former already for $N \geqslant 3$. H^D cannot achieve spin changes by more than ±2. For example, we can conclude already at this stage that the rate for *direct* transitions of an excited $S=3$ quantum dot state into the (assumed) $S=0$ singlet ground state will be of the order $\mathcal{O}[(H^D)^4]$ and therefore will be very small. All properly symmetrized operators $H^{\Gamma=[N-1,1]}$ and $H^{\Gamma=[N-2,2]}$ for $N=3$ and $N=4$ are listed in Appendix A.

Note, that the property of $H_{ij}^{(0-2)}$ to change S_z by $0, \pm 1, \pm 2$, respectively, is unrelated to their respective capability to change S. In the absence of further symmetries of the quantum dot shape, all three operators $H_{ij}^{(0-2)}$ contain both, $H^{\Gamma=[N-1,1]}$ and $H^{\Gamma=[N-2,2]}$. In cases of frozen electron motion in z direction, as it applies to quantum dots (or arrays of quantum dots) fabricated on the basis of semiconducting heterostructures,[35] all of the above contributions involving ζ_{ij} vanish. Then $H^{(0)}$ simplifies and $H^{(1)}$ vanishes entirely, so that S_z can either remain unaltered (through $H^{(0)}$) or change by ±2 (through $H^{(2)}$).

B. Two electrons

Let us first focus on two electrons, i.e., $N=2$. This is relevant, for example, for double dots containing one electron on either side to realize the basic entity of coupled qubits.[3,36] As already mentioned in the previous section, non-A_1 symmetric partitions Eq. (8) of H^D occur only for $N \geqslant 3$. Further, H^D does not contain the A_2 partition for any N. Therefore, spin conversion transitions from a triplet excited state into the singlet ground state[37] will never be mediated by H^D. In the related physics context of nuclear spin conversion of H_2 molecules[38] the stability of orthohydrogen (even over weeks) is traced back[39] to parity symmetry of both, the molecule and the magnetic dipolar interactions between the two protons (of actually close proximity which enhances dipolar forces) to prevent the transition from the odd-parity ortho $S=1$ state into the (by 80 Kelvin lower) even-parity para $S=0$ ground state. In this case, spins refer to the protons. In the context of quantum dots we can generalize this finding: Irrespective of the shape of the quantum dot confining potential and of the functional form of the electron-electron interaction $w(\mathbf{r}_1 - \mathbf{r}_2)$, the dipolar interaction will not change (triplet or singlet) spin states as a result of permutational symmetry and quantum mechanical particle identity of $N=2$ electrons. This statement is not restricted to the lowest (golden rule) order $\mathcal{O}[(H^D)^2]$ but even holds true to any order of H^D. As one neat corollary we conclude that two electrons in square-shaped quantum dots (in the absence of other magnetic mechanisms) will stay in their respective spin states. This supports a corresponding proposal for quantum computations based on superpositions of states where the two electrons occupy either of the two equivalent electrostatic energy minimum positions at diagonally opposite corners in a square.[40]

C. Strong interaction, pocket states

In case of more than two electrons we focus on strong Coulomb forces at low carrier densities, i.e., at large values of the electron gas parameter $r_s \gg 1$. Then, the kinetic energy is small and the electron system lowers its energy by Wigner localizing[30] the charge density near electrostatically favorable places. Precursors of Wigner crystallization have been found already at $r_s \approx 4$ in two-dimensional quantum dots.[23] A similar localization of charge density arises when the external confining potential separates the electrons, such as in the case of N quantum dots, each containing a single electron. In either case, at strong Coulomb interactions, eigenstates $|\phi_n\rangle$

of H_0 Eq. (1) are well described by pocket states,[26–28] which exploit the electron localization. They allow to estimate the spin-dependent low energy spectrum to exponential accuracy with increasing r_s, or with increasing dot separation.

In the Wigner crystal state, electrons vibrate about electrostatic energy minimum positions. Linearizing the (Coulomb and external) forces yields the plasmon spectrum of the confined N-electron system. Energy level separations $\omega_{\mathrm{pl}}^2 \sim \omega_0^2 + A r_s^{-3}$ can be estimated from the dynamical matrix with a prefactor A depending on N and on the dot layout; ω_0 is the confining frequency of the quantum dot(s). Due to the electron spin each plasmon level is 2^N-fold degenerate. Quantum corrections (partly) split this degeneracy into sublevels, with all exhibiting well-defined total spins $S = \{{0 \atop 1/2}\}, \ldots, N/2$ for $\{{\text{even} \atop \text{odd}}\}$ N [of $(2S+1)$-fold Zeeman degeneracy, by spin-rotation invariance], according to Sec. II. The ground state (in more than one spatial dimension) need not be of minimal spin $S_0=0$ or $S_0=\frac{1}{2}$.[22,24,25,27] A given spin S may appear more than once in such a spin-split plasmon level; examples of spectra are discussed in Refs. 26–28. The splitting arises due to permutational electron exchanges by quantum mechanical tunneling through the electrostatic barrier consisting of the v term plus the w term in Eq. (1). In the simplest case there are $N!$ different, but all energetically precisely equivalent, possibilities to arrange the localized electrons; this defines $1 \leqslant p \leqslant N!$ pocket states $|p\rangle$. The width of each pocket state corresponds to plasmonic zero point oscillations and scales roughly as $\omega_{\mathrm{pl}}^{-1/2}$ in Nd-dimensional configuration space (d being the spatial dimensionality of the quantum dot, often[35] $d=2$ but also $d=1$ is realized, for example in rods of carbon nanotubes[41]). The energy scale Δ for spin splittings of plasmon levels through quantum mechanical electron exchanges is tuned by the magnitude of overlap integrals $\langle p'|H_0|p\rangle$ between two different arrangements p and p'. This latter quantity can be estimated semiclassically[27,28,31] to read $\Delta \sim \langle p'|H_0|p\rangle \sim \omega_{\mathrm{pl}} \exp(-\sqrt{r_s})$ so that $\Delta/\omega_{\mathrm{pl}} \ll 1$. Numerically obtained quantum dot spectra[23,27,42] indeed nicely follow this behavior. For example, it exhibits the predicted[24] crossover into a spin polarized $S=\frac{3}{2}$ ground state in a spherical two-dimensional quantum dot containing $N=3$ electrons at sufficiently low electron density.[23,25] As a result, all eigenstates

$$|\phi_n\rangle = \frac{1}{N_n} \sum_p c_{np} |p\rangle \tag{9}$$

belonging to the plasmon ground multiplet can approximately be expressed through the set $\{|p\rangle\}$. The (real) coefficients c_{np}, appearing in Eq. (9), ensue from the irreducible representation Γ of the permutational group S_N, according to the wave functions symmetry which at the same time fixes the total spin S of $|\phi_n\rangle$; $N_n = \sqrt{\Sigma_{pp'} c_{np} c_{np'} \langle p'|p\rangle}$ ensures normalization, $\langle \phi_n | \phi_n \rangle = 1$.

D. Dipolar matrix elements

Pocket states allow to conveniently estimate the matrix elements $\langle \phi_n | H^D | \phi_{n'} \rangle$ of the dipolar energy since, to leading order, electron positions may be taken as being well localized, δ-function such as, for ϱ_{ij} and ζ_{ij} in Eqs. (5)–(7) or in the already symmetrized expressions in Appendix A. This leads to a finite lattice spin problem. Having constructed symmetrized spin states, the matrix elements of H^D for $N > 4$ follow straightforwardly from Eq. (8).

We demonstrate our approach for the particularly symmetric cases of $N=3$ and $N=4$ electrons occupying equilateral electrostatic equilibrium positions, as in a two-dimensional spherical quantum dot,[23,24] in triangularly or square-shaped quantum dots,[35] or in equilateral triangular or square arrangements of single electron quantum dots. We assume frozen motion in z direction, as in heterostructures, so that terms involving ζ_{ij} or ζ_{ij}^2 are irrelevant in Eqs. (5)–(7). Symmetrized, nontrivial spin states of minimal S_z components are presented in Table I.

1. $N=3$

In this case $\Sigma_{i \neq j} \varrho_{ij}^2 = 0$ and, in Appendix A, we replace $|\varrho_{ij}|^2$ by r^2 for $N=3$ where r is the mean interelectron separation. Then, the only nonvanishing term $H^{(2)[2,1]}$ takes the value

$$\sum_{i \neq j} H_{ij}^{(2)} = \frac{r^2}{4} \sum_{i \neq j} e^{i2\vartheta_{ij}} \hat{S}_{-i} \hat{S}_{-j} + \text{H.c.}$$

where $\vartheta_{ij} = 0, \frac{2\pi}{3}, \frac{-2\pi}{3}$ is the azimuthal angle of $\mathbf{r}_{ij}$.

Thus, H^D necessarily changes S_z by ± 2 and has nonvanishing matrix elements only between the A and the E states $\{S=\frac{3}{2}, S_z=\frac{3}{2}\} \leftrightarrow \{S=\frac{1}{2}, S_z=-\frac{1}{2}\}$ and $\{S=\frac{3}{2}, S_z=-\frac{3}{2}\} \leftrightarrow \{S=\frac{1}{2}, S_z=+\frac{1}{2}\}$ of Table I. Their value emerges as

$$\langle \phi_{\{A, S_z=\pm 3/2\}} | H^D | \phi_{\{E_{\mathrm{a,b}}, S_z=\mp 1/2\}} \rangle = -\frac{3\sqrt{3}}{4} \frac{\gamma^2}{r^3}.$$

In particular, this means that the not-Zeeman-aligned states of $S=\frac{3}{2}$ with $S_z=\pm\frac{1}{2}$ remain unaffected from dipolar decay.

2. $N=4$

For a square arrangement of $N=4$ electrons two distances occur: r along one edge and $\sqrt{2}r$ across the diagonal. Inspecting all the terms $H^{(0)[3,1]}, \ldots, H^{(2)[2,2]}$ for $N=4$ in Appendix A reveals that only contributions $\frac{1}{16}\Sigma_{i \neq j}[\varrho_{+ij}^2 \pm \varrho_{+\bar{i}\,\bar{j}}^2] \times [\hat{S}_{-i}\hat{S}_{-j} \pm \hat{S}_{-\bar{i}}\hat{S}_{-\bar{j}}] + \text{H.c.}$ remain nonvanishing; here $(\bar{i}, \bar{j})$ take the two values out of 1,...,4 that are both different from (i,j). Again, S_z has to change by ± 2. The term $\sim[\varrho_{+ij}^2 + \varrho_{+\bar{i}\,\bar{j}}^2]$ vanishes across the diagonal of the square, while $[\varrho_{+ij}^2 - \varrho_{+\bar{i}\,\bar{j}}^2] = 0$ along any edge. An evaluation yields the nonzero matrix elements between symmetrized spin states of Table I, i.e.,

$$\langle \phi_{\{A, S_z=\pm 2\}} | H^D | \phi_{\{T_z, S_z=0\}} \rangle = -\frac{3}{\sqrt{2} \cdot 32} \frac{\gamma^2}{r^3},$$

$$\langle \phi_{\{A, S_z=\pm 1\}} | H^D | \phi_{\{T_z, S_z=\mp 1\}} \rangle = -\frac{3}{64} \frac{\gamma^2}{r^3},$$

TABLE I. Symmetrized, nontrivial spin states of minimal S_z component for $N=3$ and $N=4$. Spin states of larger $|S_z|$ are obtained easily.

N	S	S_z	index	
3	$\frac{3}{2}$	$\frac{1}{2}$	A	$\frac{1}{\sqrt{3}}(\lvert\uparrow\uparrow\downarrow\rangle+\lvert\uparrow\downarrow\uparrow\rangle+\lvert\downarrow\uparrow\uparrow\rangle)$
3	$\frac{1}{2}$	$\frac{1}{2}$	E_a	$\frac{1}{\sqrt{3}}(\lvert\uparrow\uparrow\downarrow\rangle+e^{i2\pi/3}\lvert\uparrow\downarrow\uparrow\rangle+e^{-i2\pi/3}\lvert\downarrow\uparrow\uparrow\rangle)$
3	$\frac{1}{2}$	$\frac{1}{2}$	E_b	$\frac{1}{\sqrt{3}}(\lvert\uparrow\uparrow\downarrow\rangle+e^{-i2\pi/3}\lvert\uparrow\downarrow\uparrow\rangle+e^{i2\pi/3}\lvert\downarrow\uparrow\uparrow\rangle)$
4	2	0	A	$\frac{1}{\sqrt{6}}(\lvert\uparrow\uparrow\downarrow\downarrow\rangle+\lvert\downarrow\uparrow\uparrow\downarrow\rangle+\lvert\downarrow\downarrow\uparrow\uparrow\rangle+\lvert\uparrow\downarrow\downarrow\uparrow\rangle+\lvert\uparrow\downarrow\uparrow\downarrow\rangle+\lvert\downarrow\uparrow\downarrow\uparrow\rangle)$
4	1	0	T_x	$\frac{1}{2}(\lvert\uparrow\uparrow\downarrow\downarrow\rangle-i\lvert\downarrow\uparrow\uparrow\downarrow\rangle-\lvert\downarrow\downarrow\uparrow\uparrow\rangle+i\lvert\uparrow\downarrow\downarrow\uparrow\rangle)$
4	1	0	T_y	$\frac{1}{2}(\lvert\uparrow\uparrow\downarrow\downarrow\rangle+i\lvert\downarrow\uparrow\uparrow\downarrow\rangle-\lvert\downarrow\downarrow\uparrow\uparrow\rangle-i\lvert\uparrow\downarrow\downarrow\uparrow\rangle)$
4	1	0	T_z	$\frac{1}{\sqrt{2}}(\lvert\uparrow\downarrow\uparrow\downarrow\rangle-\lvert\downarrow\uparrow\downarrow\uparrow\rangle)$
4	0	0	E_1	$\frac{1}{2}(\lvert\uparrow\uparrow\downarrow\downarrow\rangle-\lvert\downarrow\uparrow\uparrow\downarrow\rangle+\lvert\downarrow\downarrow\uparrow\uparrow\rangle-\lvert\uparrow\downarrow\downarrow\uparrow\rangle)$
4	0	0	E_2	$\frac{1}{\sqrt{8}}(\lvert\uparrow\uparrow\downarrow\downarrow\rangle+\lvert\downarrow\uparrow\uparrow\downarrow\rangle+\lvert\downarrow\downarrow\uparrow\uparrow\rangle+\lvert\uparrow\downarrow\downarrow\uparrow\rangle-2\lvert\uparrow\downarrow\uparrow\downarrow\rangle-2\lvert\downarrow\uparrow\downarrow\uparrow\rangle)$

$$\langle\phi_{\{A,S_z=\pm2\}}|H^D|\phi_{\{E_1,S_z=0\}}\rangle=-\frac{3}{32}\frac{\gamma^2}{r^3}.$$

All other matrix elements vanish identically. In particular, states of symmetries T_x, T_y, E_2, and A with $S_z=0$ do not exhibit dipolar decay.

E. Mixed spin states

Nonvanishing elements of H^D slightly mix eigenstates $|\phi_n\rangle$ of H_0. Here we are interested in admixtures to spin states S,

$$|\psi_{n_S}\rangle=|\phi_{n_S}\rangle+\sum_{S'\neq S}\sum_{n_{S'}}\frac{\langle\phi_{n_{S'}}|H^D|\phi_{n_S}\rangle}{E_{n_S}-E_{n_{S'}}}|\phi_{n_{S'}}\rangle, \tag{10}$$

arising from other spins $S'\neq S$. In Eq. (10) we have disregarded the very unlikely case of accidental degeneracy between eigenlevels of H_0 [cf. Eq. (1)] of different spins.[43]

Eventually, this mixing will cause spin-changing transitions and thus spin relaxation. We disregard dipolar admixtures from other states $|\phi_{n'_S}\rangle$ of the same spin in Eq. (10) as those occur much more efficiently by phonons; see the subsequent section. To this end, we take $\{|\psi_{n_S}\rangle\}$ as exact eigenstates of H_0+H^D. From now on we denote by n_S the subset of n values enumerating eigenstates of H_0 that belong to the definite spin S.

IV. SPIN RELAXATION RATES

Typically, the electron-phonon interaction $H^{el\text{-}ph}$ establishes thermal equilibrium between electron and lattice reservoirs on short time scales compared to the times on which spin-changing transitions occur. This is so because the latter cannot be achieved directly by $H^{el\text{-}ph}$ (cf. Sec. IV A), so that equilibrium will be established rapidly only among dot levels of given total spins. This suggests to divide the total Hilbert space,

$$\mathcal{H}=\bigoplus_S\mathcal{H}_S,$$

of *coupled* electron-phonon states into orthogonal subspaces $\mathcal{H}_S$, labeled according to the *electron* spin S. Transitions among subspaces occur only slowly by the action of H^D while thermal equilibrium resides within each of the subspaces after much shorter times $\tau^{el\text{-}ph}$ at the lattice temperature $(k_B\beta)^{-1}$. Consider a certain electronic spin state S', as it may have been prepared, for example, using electronic transport techniques.[2,44,45] Then, the rate

$$R_{S\leftarrow S'}=\left.\frac{d}{dt}\langle P_S(t)\rangle_{S'}\right|_{t\gtrsim\tau^{el\text{-}ph}} \tag{11}$$

for its decay into a particular spin $S\neq S'$ is given as the temporal increase of the spin S-population $\langle P_S(t)\rangle_{S'}$, assuming an initial (i.e., after intra-$\mathcal{H}_{S'}$ equilibration has taken place) S' thermal equilibrium state,

$$P_{S'}e^{-\beta H}P_{S'}/\mathrm{Tr}\{P_{S'}e^{-\beta H}P_{S'}\}.$$

Here, $P_S=\Sigma_{n_S}|\phi_{n_S}\rangle\langle\phi_{n_S}|\otimes\mathbb{1}^{ph}$ projects onto $\mathcal{H}_S$, $\mathbb{1}^{ph}$ denotes a unit operator on the phonon space. Transition rates $R_{S\leftarrow S'}$

observe the detailed balance condition, ensuring one vanishing (stationary) eigenvalue of the matrix $M_{SS'}=R_{S'\leftarrow S}-\tau_{S'}^{-1}\delta_{SS'}$, which governs the rate dynamics. In the present context we are primarily interested in the total decay rate of the initial spin S' population; i.e.,

$$\tau_{S'}^{-1}=\sum_{S\neq S'}R_{S\leftarrow S'}. \tag{12}$$

In Eq. (11) the time evolution refers to the Hamiltonian $H=H_0+H^D+H^{ph}+H^{el\text{-}ph}$ where the electron-phonon interaction $H^{el\text{-}ph}$ will be discussed next. This approach, in principle, accounts for rapid thermalizing spin-conserving multiphonon transitions within subspaces $\mathcal{H}_S$.

A. Coupling to phonons

Electron-phonon coupling in semiconductors has been studied intensively in the 1950s and 1960s. For homopolar semiconductors, such as Si or Ge, deformation potential coupling[46] has been established. It can be expressed as[47]

$$H^{el\text{-}ph}=\sum_{\mathbf{q}}g_{\mathbf{q}}\rho(\mathbf{q})(b_{\mathbf{q}}+b_{-\mathbf{q}}^{+}), \tag{13}$$

where we have suppressed the phonon branch index. Considerably below room temperature, pertinent to possible quantum computing, optical phonons don't contribute so that $b_{\mathbf{q}}^{+}$ in Eq. (13) is meant to create a longitudinal acoustical phonon of momentum $\mathbf{q}$. For excitations of the electronic system most relevant are phonon wavelengths $2\pi c_s/\omega_{\mathrm{pl}}=50$ nm or $2\pi c_s/\Delta=500$ nm, assuming[35] $\omega_{\mathrm{pl}}\simeq 3$ meV and $\Delta\simeq 0.3$ meV, respectively; cf. Sec. III C for the definitions of the energies Δ and ω_{pl}. At these wavelengths, intravalley scattering dominates. Its strength,

$$g_{\mathbf{q}}^2=\frac{E_2^2}{2\rho_{\mathrm{M}}V_{c_s}}|\mathbf{q}|, \tag{14}$$

mainly is regulated by the deformation potential constant E_2 for longitudinal coupling which takes values of about[47,48] 10 eV in Si. Further, $g_{\mathbf{q}}^2$ depends on the mass density ρ_{M}, the normalization volume V for the phonon modes, and on the sound velocity c_s. In Eq. (13) the operator

$$\rho(\mathbf{q})=N\sum_{n_{S'}n_S'}\rho_{n_Sn_S'}(\mathbf{q})|\phi_{n_S'}\rangle\langle\phi_{n_S}| \tag{15}$$

of the total electron density may excite the correlated electron system at nonzero $\mathbf{q}$, though at conserved total spin S (and conserved z-component S_z). It can be decomposed into the basis $\{|\phi_n\rangle\}$, where $|\phi_n\rangle$ and $|\phi_{n'}\rangle$ have same spin S, i.e.,

$$\rho_{n_Sn_S'}(\mathbf{q})=\int d\mathbf{r}e^{i\mathbf{q}\mathbf{r}}\int d\mathbf{r}_2\ldots d\mathbf{r}_N\langle\mathbf{r},\mathbf{r}_2,\ldots,\mathbf{r}_N|\phi_{n_S}\rangle \times\langle\phi_{n_S'}|\mathbf{r},\mathbf{r}_2,\ldots,\mathbf{r}_N\rangle. \tag{16}$$

At small $q\equiv|\mathbf{q}|$ these coefficients are expanded, $\rho_{nn'}(\mathbf{q})=\delta_{nn'}+\alpha(q\ell)^{\nu}$ where, to lowest nonvanishing order, $\nu=1$ unless the electron charge density distribution of the quantum dot or of the ensemble of quantum dots is parity symmetric, in which case $\nu=2$. The quantity ℓ either equals the typical distance between electrons if n and n' belong to the same plasmon multiplet, or $\ell\simeq(m\omega_{\mathrm{pl}})^{-1/2}$ for n and n' from different plasmon multiplets. The magnitude of α can be estimated by inserting Eq. (9) into Eq. (16) and using, for convenience, the density distribution $\rho_{nn'}(\mathbf{r})$ in real space. This reveals that α is proportional to the maximum overlap between unequal pocket states, i.e., $\max_{p,p';p\neq p'}\langle p|p'\rangle$, a quantity which, in turn, is proportional[27] to $\Delta/\omega_{\mathrm{pl}}$.

B. Transition rates

We are now in the position to calculate the phonon-mediated transition rate Eq. (11) as a result of spin mixing; see Eq. (10). Assuming a not too strong electron-phonon coupling, use of standard time-dependent perturbation theory with respect to $H^{el\text{-}ph}$, as explicated in Appendix B, yields to leading order the rate $R_{S\leftarrow S'}$, cf. Eq. (11), reading

$$R_{S\leftarrow S'}=\frac{2\pi}{Z_{S'}}\sum_{n_Sn_{S'}}e^{-\beta E_{n_{S'}}}J_{n_{S'}n_S}(|E_{n_{S'}}-E_{n_S}|)[n(|E_{n_{S'}}-E_{n_S}|)+\Theta(E_{n_{S'}}-E_{n_S})], \tag{17}$$

where we have defined the (temperature-independent) coupled density of phonon states for transitions between spins S and S', respectively, as

$$J_{n_{S'}n_S}(\omega)=\sum_{\mathbf{q}}g_{\mathbf{q}}^2\delta(\omega-|\mathbf{q}|c_s)\times\left|\sum_{n_S'}\rho_{n_Sn_S'}(\mathbf{q})\frac{\langle\phi_{n_S'}|H^D|\phi_{n_{S'}}\rangle}{E_{n_{S'}}-E_{n_S'}}-\sum_{n_{S'}'}\rho_{n_{S'}'n_{S'}}(\mathbf{q})\frac{\langle\phi_{n_S}|H^D|\phi_{n_{S'}'}\rangle}{E_{n_{S'}'}-E_{n_S}}\right|^2. \tag{18}$$

In Eq. (17) $Z_S=\Sigma_{n_S}e^{-\beta E_{n_S}}$ denotes the partition function inside the subspace $\mathcal{H}_S$, $n(\omega)=(e^{\beta\omega}-1)^{-1}$ the Bose function, and $\Theta(x)$ the Heaviside step function. Straightforwardly, higher order terms regarding $H^{el\text{-}ph}$ can also be considered for $R_{S\leftarrow S'}$, although the corresponding explicit expressions are rather lengthy. In Eq. (10) we have assumed that phonon states and energy eigenvalues remain unaffected by the weak dipolar mixing.

At low temperatures, $T\ll\Delta/k_{\mathrm{B}}$ compared to the typical distance Δ between dot levels of same or of different total spins inside the lowest plasmon multiplet, the Bose factor $n(|\Delta|)\ll 1$ is small and only the ground level n_{0S} will be occupied within each subspace $\mathcal{H}_S$. In this temperature regime thermalization into the global ground state $n=0_{S_0}$ of spin S_0 will take place exclusively through the direct process by emission of a resonant phonon of energy Δ so that the coupled density of states $J_{n_{0S}0_{S_0}}(\Delta)$ controls the relaxation rate $R_{S_0\leftarrow S}$. Still, a summation over excited levels $n_{S_0}'>0$ and $n_{0S}'>n_{0S}$ appears in Eq. (18), as the lowest terms $n_{S_0}'=0$ and $n_{0S}'=n_{0S}$ cancel exactly. In Sec. IV A it has been estimated

that nondiagonal coefficients $\rho_{nn'}(\mathbf{q}) \sim (\Delta/\omega_{\rm pl})(r\Delta/c_{\rm s})^{\nu}$ for low energy and long wavelength transitions; here r denotes the distance between electrons and $\nu=1$ or $\nu=2$ in the absence or presence of parity symmetry. For the electron-phonon coupling Eq. (14), this results in a spin-transition rate at zero temperature through the direct process, reading

$$R_{S_0\leftarrow S} = 2\pi J_{n_{0S}0_{S_0}}(\Delta), \tag{19}$$

with

$$J_{n_{0S}0_{S_0}}(\Delta) = \frac{E_2^2\gamma^4}{c_{\rm s}^7\rho_{\rm M}\pi^2\omega_{\rm pl}^2} N^2 n_{\rm a}^2 \Delta^5,$$

unless this transition is not suppressed entirely for cases discussed in Sec. III D. In Eq. (19) we have assumed for simplicity that level separations $E_{n'_{S_0}} - E_{0_{S_0}} \sim \Delta$ and $E_{n'_S} - E_{n_{0S}} \sim \Delta$ both are of the order[49] Δ. Also, we have inserted the areal density $n_{\rm a}=r^{-2}$ of electrons, focusing on the measured quantity in two-dimensional samples.

In Si the rate Eq. (19) appears to be very small at zero temperature, $\sim 10^{-7}$ s^{-1} for three electrons at densities corresponding to $r_{\rm s}=1$, and considering a quantum dot[35] of $\omega_{\rm pl}=3$ meV and $\Delta=0.3$ meV. However, this number strongly varies with parameters, as seen in Eq. (19). Parity symmetric quantum dots (where $\nu=2$) would suppress this decay rate even further at small transition energies due to $J_{n0}(\Delta) \sim \Delta^7$ in this case. These values are, of course, considerable smaller than the decay rates estimated from spin-orbit effects,[7–9] if present.

They are also smaller than the rates estimated from the hyperfine interaction with nuclei of nonzero spin.[13,14,16] Particularly in Ref. 13 a hybrid mechanism is considered which is closely related to the one presented here in combining the electron-phonon coupling with a spin-mixing interaction. Transitions between total spin $S=1$ and $S=0$ of a two-electron quantum dot are investigated. The low temperature rate has been estimated[13] to $\sim 10^{-2}$ s^{-1} s for similar quantum dot parameters as above, assuming a two-dimensional dot fabricated on the basis of heterostructures. This rate is proportional to the number $N_{\rm n}$ of nonvanishing nuclear spins covered by the electron wave function. In GaAs almost every nucleus has spin $I=\frac{3}{2}$. It is instructive to determine from this result[13] a critical concentration $\tilde{C}_{\rm n}$ of ^{29}Si nuclei in silicon, the only ones of nonvanishing spin $I=\frac{1}{2}$, beyond which the here-described dipolar mechanism should prevail over the spin decay via nuclear spins. For a quantum dot of the same excitation energy, the electron wave function in natural silicon covers only about $N_{\rm n}^{\rm Si} \approx 10^3$ of the ^{29}Si nuclei while[14] in GaAs $N_{\rm n}^{\rm GaAs} \approx 10^5$. Two further important differences between Si and GaAs have to be taken into account. Firstly, the type of electron-phonon coupling which is piezoelastic in GaAs while we have deformation potential coupling in Si. Accidentally, for the here-considered quantum dot parameters (and assuming again heterostructures and now laterally parabolic confining potential) $J_{nn'}(\omega)$ in Si is only by 0.6 smaller than in GaAs. Secondly, the nuclear spin $I^{\rm Si}=\frac{1}{2}$ of ^{29}Si as compared to $I^{\rm GaAs}=\frac{3}{2}$ in GaAs which reduces the coupling by $I^{\rm Si}(I^{\rm Si}+1)/I^{\rm GaAs}(I^{\rm GaAs}+1)=\frac{1}{5}$. This yields $\tilde{C}_{\rm n}^{\rm Si} \approx 2\times 10^{-4}$ nm^{-3} (note that $C_{\rm n}^{\rm Si} \approx 2.5\times 10^{-3}$ nm^{-3} has been reported[50] experimentally). This value is less stringent than the isotopic purification required for the quantum computer,[6] based on the nuclear spins of ^{31}P donors, where $C_{\rm n}$ should be smaller than $N^{-1}10^{-4}$ nm^{-3} in Si with N being the number of qubits.

C. Temperature dependence

Through the marked increase of $J_{n0}(\omega) \sim \omega^{4+2\nu}$ as a function of transition energy ω, relaxation can take advantage from spin-conserving thermal excursions to plasmonic excited levels and accomplish the spin transition at an elevated energy. In NMR theory this possibility is called the "Orbach process"[51] and shows up in a steeply increasing relaxation rate with temperature. Our formulation, Eq. (17), of the transition rate explicitly incorporates such thermal excursions. They turn out to influence considerably the temperature dependence of $R_{S_0\leftarrow S}$ which only at low temperatures follows the Bose behavior $\sim J_{n0}(\Delta)n(\Delta)$ of direct transitions. Already at temperatures not much exceeding Δ the severely stronger increase $\sim J_{n0}(\omega_{\rm pl})\exp(-\omega_{\rm pl}/k_{\rm B}T)$, following from Eq. (17), can easily enhance the transition rate by three orders of magnitudes, depending on system parameters. A similarly pronounced increase of (nuclear) spin relaxation rates has been discussed in detail[52] in the context of quantum rotating molecules: substantial increases in spin-changing transition rates by more than six orders of magnitudes are depicted in Fig. 3(b) of Ref. 52. Again, the stronger increasing density of coupled phonon states in quantum dots of parity symmetry should lead to even more pronounced temperature sensitivity.

V. RESUME

We have investigated dipolar interactions between the magnetic moments of electrons confined to one or to several quantum dots and studied the rate of inelastic total spin-changing transitions. As compared to the coupling to nuclear spins[13–16,53] and to spin-orbit induced decay,[8–12] dipolar spin decay turns out as much weaker. However, either of the earlier studied mechanisms can, at least in principle, be eliminated by a suitable sample design. It is therefore possible that the dipolar interactions between electronic spin moments, together with the coupling to lattice modes,[20] as discussed in the present work, will ultimately limit long-time quantum computations, even when devices become optimally designed. Experimentally, the dipolar mechanism should show up most directly by observing at low temperatures the dependence on the electron density; cf. Eq. (19).

Upon generalizing previous approaches we incorporate here electron-electron interactions[11,12] in the quantum dot(s) which, additionally, are important for magnetic features.[22–25,31] For example, ground state spins may exceed the values $S_0=0$ or $S_0=\frac{1}{2}$ expected for noninteracting electrons. We focus on the limit of strong interactions, where electronic many-body wave functions can be described as "pocket states"[26–28] and where the spectrum exhibits spin-split plasmon multiplets. The dipolar interaction is decomposed according to the symmetric group and nonvanishing

matrix elements are determined in their dependence on spatial parts of the collective electron wave functions. As we have shown, particular spatial symmetries of the quantum dot(s) can reduce the number of nonzero elements.

One important result is the stability of $N=2$ electron spins. Irrespective of the dimensionality, the shape of the quantum dot(s), or of the electron-electron interaction strength, the decay of triplet states into the singlet ground state is always suppressed. Any dipolar decay channel will require participation of further electrons. This is important, for example, for two coupled qubits, the basic (gate) element for quantum information processing.

Also at larger electron numbers, nonground state spins can be stable with respect to dipolar interactions. We have discussed the case of $N=3$ electrons on an equilateral triangle. Here, $S_z=\pm\frac{1}{2}$ states of the $S=\frac{3}{2}$ submanifold prove robust against decay into any $S=\frac{1}{2}$ state. Only spin-polarized $S_z=\pm\frac{3}{2}$ states decay into the $S=\frac{1}{2}$ submanifold. Further, $N=4$ electrons on a square-shaped quantum dot exhibit robust $S=1$ states of T_x and T_y symmetry, and $S=0$ states of E_2 symmetry.

Owing to the smallness of magnetic and, in particular, dipolar energies, compared to dot level separations, the energy accompanied with an actual spin transition has to be provided by the reservoir of lattice vibrations. As in previous approaches[7–10,13] we have considered the coupling to acoustic phonons. Parity symmetric dots are weaker coupled to phonons, which further suppresses spin decay in this case. Additionally, we have accounted for rapid thermal excursions of the system *within* electron-phonon subspaces of given (many-electron) dot spins. This enables one to deduce the dependence of spin relaxation over a wider range of temperatures as compared to the resonant direct process. As a result we found a very striking increase of the spin decay rate. This rate grows with temperature considerably steeper than the naively expected proportionality to the Bose function describing direct processes: It occurs already at temperatures that barely exceed the energy difference between the lowest levels of different spins, but is still considerably smaller than the energy for plasmon excitations. Although we find amazingly stable spin configurations at low temperatures, this marked temperature sensitivity restricts the operation temperatures of quantum computing dots (unless quantum computation can be confined to the stable spin configurations) to values that are not exceeding much the lowest level separations.

Because the same phonon energy reservoir is considered in previous work[7–10,13,14] for spin decay, a similar scenario regarding the dot symmetries and the spin-conserving phonon induced excursions should apply also to magnetic mechanisms of the spin-orbit or of the hyperfine type. We expect therefore a similarly striking temperature sensitivity as obtained here for these mechanisms as well.

ACKNOWLEDGMENTS

This work has been supported by the DFG Collaborative Research Center SFB 631: "Solid State Based Quantum Information Processing: Physical Concepts and Materials Aspects," project A5.

APPENDIX A: SYMMETRIZED OPERATORS INDUCING SPIN-CHANGING TRANSITIONS FOR $N=3$ AND $N=4$ ELECTRONS

The following explicit form of dipolar operators Eq. (8) contain nonvanishing elements for $N=3$,

$$H^{(0)[2,1]}=\sum_{i\neq j}H_{ij}^{(0)}-\frac{1}{6}\left\{\left[\frac{1}{4}\sum_{i\neq j}|\varrho_{ij}|^2\right]\left[\sum_{i\neq j}(\hat{S}_{+i}\hat{S}_{-j}+\hat{S}_{-i}\hat{S}_{+j})\right]+\left[\sum_{i\neq j}\zeta_{ij}^2\right]\left[\sum_{i\neq j}\hat{S}_{zi}\hat{S}_{zj}\right]\right\},$$

$$H^{(1)[2,1]}=\sum_{i\neq j}H_{ij}^{(1)}-\frac{1}{12}\left\{\left[\sum_{i\neq j}\zeta_{ij}\varrho_{-ij}\right]\left[\sum_{i\neq j}(\hat{S}_{+i}\hat{S}_{zj}+\hat{S}_{zi}\hat{S}_{+j})\right]+\left[\sum_{i\neq j}\zeta_{ij}\varrho_{+ij}\right]\left[\sum_{i\neq j}(\hat{S}_{-i}\hat{S}_{zj}+\hat{S}_{zi}\hat{S}_{-j})\right]\right\},$$

$$H^{(2)[2,1]}=\sum_{i\neq j}H_{ij}^{(2)}-\frac{1}{24}\left\{\left[\sum_{i\neq j}\varrho_{+ij}^2\right]\left[\sum_{i\neq j}\hat{S}_{-i}\hat{S}_{-j}\right]+\left[\sum_{i\neq j}\varrho_{-ij}^2\right]\times\left[\sum_{i\neq j}\hat{S}_{+i}\hat{S}_{+j}\right]\right\},$$

and for $N=4$,

$$H^{(0)[3,1]}=\frac{1}{4}\sum_{i\neq j}\left\{\left[\frac{|\varrho_{ij}|^2-|\varrho_{\bar{i}\bar{j}}|^2}{4}\right][(\hat{S}_{+i}\hat{S}_{-j}+\hat{S}_{-i}\hat{S}_{+j})-(\hat{S}_{+\bar{i}}\hat{S}_{-\bar{j}}+\hat{S}_{-\bar{i}}\hat{S}_{+\bar{j}})]+[\zeta_{ij}^2-\zeta_{\bar{i}\bar{j}}^2][\hat{S}_{zi}\hat{S}_{zj}-\hat{S}_{z\bar{i}}\hat{S}_{z\bar{j}}]\right\},$$

$$H^{(1)[3,1]}=\frac{1}{8}\sum_{i\neq j}\{[\zeta_{ij}\varrho_{-ij}-\zeta_{\bar{i}\bar{j}}\varrho_{-\bar{i}\bar{j}}][(\hat{S}_{+i}\hat{S}_{zj}+\hat{S}_{zi}\hat{S}_{+j})-(\hat{S}_{+\bar{i}}\hat{S}_{z\bar{j}}+\hat{S}_{z\bar{i}}\hat{S}_{+\bar{j}})]+[\zeta_{ij}\varrho_{+ij}-\zeta_{\bar{i}\bar{j}}\varrho_{+\bar{i}\bar{j}}]\times[(\hat{S}_{-i}\hat{S}_{zj}+\hat{S}_{zi}\hat{S}_{-j})-(\hat{S}_{-\bar{i}}\hat{S}_{z\bar{j}}+\hat{S}_{z\bar{i}}\hat{S}_{-\bar{j}})]\},$$

$$H^{(2)[3,1]}=\frac{1}{16}\sum_{i\neq j}\{[\varrho_{+ij}^2-\varrho_{+\bar{i}\bar{j}}^2][\hat{S}_{-i}\hat{S}_{-j}-\hat{S}_{-\bar{i}}\hat{S}_{-\bar{j}}]+[\varrho_{-ij}^2-\varrho_{-\bar{i}\bar{j}}^2]\times[\hat{S}_{+i}\hat{S}_{+j}-\hat{S}_{+\bar{i}}\hat{S}_{+\bar{j}}]\},$$

$$H^{(0)[2,2]}=\frac{1}{4}\sum_{i\neq j}\left\{\left[\frac{|\varrho_{ij}|^2+|\varrho_{\bar{i}\bar{j}}|^2}{4}\right][(\hat{S}_{+i}\hat{S}_{-j}+\hat{S}_{-i}\hat{S}_{+j})+(\hat{S}_{+\bar{i}}\hat{S}_{-\bar{j}}+\hat{S}_{-\bar{i}}\hat{S}_{+\bar{j}})]+[\zeta_{ij}^2+\zeta_{\bar{i}\bar{j}}^2][\hat{S}_{zi}\hat{S}_{zj}+\hat{S}_{z\bar{i}}\hat{S}_{z\bar{j}}]\right\}-\frac{1}{12}\left\{\left[\frac{1}{4}\sum_{i\neq j}|\varrho_{ij}|^2\right]\left[\sum_{i\neq j}(\hat{S}_{+i}\hat{S}_{-j}+\hat{S}_{-i}\hat{S}_{+j})\right]+\left[\sum_{i\neq j}\zeta_{ij}^2\right]\left[\sum_{i\neq j}\hat{S}_{zi}\hat{S}_{zj}\right]\right\},$$

$$H^{(1)[2,2]} = \frac{1}{8}\sum_{i\neq j}\{[\zeta_{ij}\varrho_{-ij} + \zeta_{\bar{i}\,\bar{j}}\varrho_{-\bar{i}\,\bar{j}}][(\hat{S}_{+i}\hat{S}_{zj} + \hat{S}_{zi}\hat{S}_{+j}) + (\hat{S}_{+\bar{i}}\hat{S}_{z\bar{j}} + \hat{S}_{z\bar{i}}\hat{S}_{+\bar{j}})] + [\zeta_{ij}\varrho_{+ij} + \zeta_{\bar{i}\,\bar{j}}\varrho_{+\bar{i}\,\bar{j}}][(\hat{S}_{-i}\hat{S}_{zj} + \hat{S}_{zi}\hat{S}_{-j}) + (\hat{S}_{-\bar{i}}\hat{S}_{z\bar{j}} + \hat{S}_{z\bar{i}}\hat{S}_{-\bar{j}})]\} - \frac{1}{24}\left\{\left[\sum_{i\neq j}\zeta_{ij}\varrho_{-ij}\right]\left[\sum_{i\neq j}(\hat{S}_{+i}\hat{S}_{zj} + \hat{S}_{zi}\hat{S}_{+j})\right] + \left[\sum_{i\neq j}\zeta_{ij}\varrho_{+ij}\right]\left[\sum_{i\neq j}(\hat{S}_{-i}\hat{S}_{zj} + \hat{S}_{zi}\hat{S}_{-j})\right]\right\},$$

$$H^{(2)[2,2]} = \frac{1}{16}\sum_{i\neq j}\{[\varrho^2_{+ij} + \varrho^2_{+\bar{i}\,\bar{j}}][\hat{S}_{-i}\hat{S}_{-j} + \hat{S}_{-\bar{i}}\hat{S}_{-\bar{j}}] + [\varrho^2_{-ij} + \varrho^2_{-\bar{i}\,\bar{j}}]\times[\hat{S}_{+i}\hat{S}_{+j} + \hat{S}_{+\bar{i}}\hat{S}_{+\bar{j}}]\} - \frac{1}{48}\left\{\left[\sum_{i\neq j}\varrho^2_{+ij}\right]\left[\sum_{i\neq j}\hat{S}_{-i}\hat{S}_{-j}\right] + \left[\sum_{i\neq j}\varrho^2_{-ij}\right]\left[\sum_{i\neq j}\hat{S}_{+i}\hat{S}_{+j}\right]\right\}.$$

In the above expression, $(\bar{i},\bar{j})$ take the two values out of 1,…,4 that are both different from (i,j).

APPENDIX B: DERIVATION OF EQ. (17)

In a perturbative expansion with respect to $H^{el\text{-}ph}$ of either of the two time evolution operators appearing in Eq. (11) we write

$$e^{-i\tilde{H}t} = e^{-i\tilde{H}_0 t}\left[\mathbb{1}^{ph} - i\int_0^t dt' e^{i\tilde{H}_0 t'}H^{el\text{-}ph}e^{-i\tilde{H}_0 t'} - \int_0^t dt' \times \int_0^{t'} dt'' e^{i\tilde{H}_0 t'}H^{el\text{-}ph}e^{-i\tilde{H}_0(t'-t'')}H^{el\text{-}ph}e^{-i\tilde{H}_0 t''} + \ldots\right]. \tag{B1}$$

Here, $\tilde{H}_0 = H_0 + H^D + H^{ph}$ with H_0 defined in Eq. (1) and the eigenstates of $H_0 + H^D$ taken according to Eq. (10). To second (i.e., lowest nonvanishing) order in $H^{el\text{-}ph}$ only two of the second terms in the square bracket of (B1) contribute to Eq. (11), yielding with (13),

$$R_{S\leftarrow S'} = \frac{1}{Z_{S'}}\sum_{\mathbf{q}} g^2_{\mathbf{q}}\sum_{n_S n_{S'}} e^{-\beta E_{n_{S'}}}\frac{d}{dt}\int_0^t dt'\int_0^t dt'' i\langle\phi_{n_{S'}}|e^{i(H_0+H^D)t'} \times\rho(\mathbf{q})e^{-i(H_0+H^D)t'}|\phi_{n_S}\rangle(-i)\langle\phi_{n_S}|e^{i(H_0+H^D)t''} \times\rho(\mathbf{q})e^{-i(H_0+H^D)t''}|\phi_{n_{S'}}\rangle[\bar{n}_{\mathbf{q}}e^{ic_s|\mathbf{q}|(t'-t'')} + (\bar{n}_{\mathbf{q}}+1)e^{-ic_s|\mathbf{q}|(t'-t'')}].$$

Here, the Bose factors $\bar{n}_{\mathbf{q}} = (e^{\beta c_s|q|}-1)^{-1}$ result after thermal averaging over phonon modes. Inserting now eigenstates $|\psi_n\rangle$ of (H_0+H^D) for the $|\phi_n\rangle$, according to (10), and carrying out the long-time limit $t\to\infty$, yields for the decay rate

$$R_{S\leftarrow S'} = \frac{2\pi}{Z_{S'}}\sum_{\mathbf{q}} g^2_{\mathbf{q}}\sum_{n_S n_{S'}} e^{-\beta E_{n_{S'}}} \times\left[i\left(\langle\psi_{n_{S'}}| - \sum_{n_{S''}}\frac{\langle\phi_{n_{S'}}|H^D|\phi_{n_{S''}}\rangle}{E_{n_{S'}}-E_{n_{S''}}}\langle\psi_{n_{S''}}|\right)\rho(\mathbf{q}) \times\left(|\psi_{n_S}\rangle - \sum_{n_{S''}}\frac{\langle\phi_{n_{S''}}|H^D|\phi_{n_S}\rangle}{E_{n_S}-E_{n_{S''}}}|\psi_{n_{S''}}\rangle\right)\right] \times\left[-i\left(\langle\psi_{n_S}| - \sum_{n_{S''}}\frac{\langle\phi_{n_S}|H^D|\phi_{n_{S''}}\rangle}{E_{n_S}-E_{n_{S''}}}\langle\psi_{n_{S''}}|\right)\rho(\mathbf{q}) \times\left(|\psi_{n_{S'}}\rangle - \sum_{n_{S''}}\frac{\langle\phi_{n_{S''}}|H^D|\phi_{n_{S'}}\rangle}{E_{n_{S'}}-E_{n_{S''}}}|\psi_{n_{S''}}\rangle\right)\right] \times[\bar{n}_{\mathbf{q}}\delta(E_{n_{S'}}-E_{n_S}+c_s|\mathbf{q}|) + (\bar{n}_{\mathbf{q}}+1) \times\delta(E_{n_{S'}}-E_{n_S}-c_s|\mathbf{q}|)],$$

which is readily brought into the form (17) with (18), after employing (15) and using the orthogonality of spin states. Further calculational details can be found in Ref. 52.

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[1] *Semiconductor Spintronics and Quantum Computing*, edited by D. D. Awschalom, D. Loss, and N. Samarth (Springer, New York, 2002).

[2] J. M. Elzerman, R. Hanson, L. H. Willems van Beveren, B. Witkamp, J. S. Greidanus, R. N. Schouten, S. De Franceschi, S. Tarucha, L. M. K. Vandersypen, and L. P. Kouwenhoven, in *Quantum Dots: a Doorway to Nanoscale Physics*, Series: Lecture Notes in Physics, Vol. 667, edited by W. D. Heiss (Springer, Berlin, 2005).

[3] D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).

[4] J. M. Elzerman, R. Hanson, L. H. Willems van Beveren, B. Witkamp, L. M. K. Vandersypen, and L. P. Kouwenhoven, Nature **430**, 431 (2004).

[5] R. Hanson, L. M. K. Vandersypen, L. H. Willems van Beveren, J. M. Elzerman, I. T. Vink, and L. P. Kouwenhoven, Phys. Rev. B **70**, 241304 (2004).

[6] B. E. Kane, Nature **393**, 133 (1998).

[7] A. V. Khaetskii and Y. V. Nazarov, Phys. Rev. B **61**, 12639 (2000).

[8] A. V. Khaetskii and Y. V. Nazarov, Phys. Rev. B **64**, 125316 (2001).

[9] L. M. Woods, T. L. Reinecke, and R. Kotlyar, Phys. Rev. B **69**, 125330 (2004).

[10] V. N. Golovach, A. Khaetskii, and D. Loss, Phys. Rev. Lett. **93**, 016601 (2004).

[11] K. V. Kavokin, Phys. Rev. B **64**, 075305 (2001).

[12] An interesting new mechanism for spin decay, arising from the interplay between Coulomb interaction and spin-orbit coupling, has been detected recently by S. C. Badescu, Y. B. Lyanda-

Geller, and T. L. Reinecke, Phys. Rev. B **72**, 161304(R) (2005). This contribution is proportional to the energy of the split-off band. It requires structural but not crystallographic inversion asymmetry.

[13] S. I. Erlingsson, Y. V. Nazarov, and V. I. Fal'ko, Phys. Rev. B **64**, 195306(R) (2001).

[14] S. I. Erlingsson and Y. V. Nazarov, Phys. Rev. B **66**, 155327 (2002).

[15] A. V. Khaetskii, D. Loss, and L. Glazman, Phys. Rev. Lett. **88**, 186802 (2002); Phys. Rev. B **67**, 195329 (2003).

[16] V. A. Abalmassov and F. Marquardt, Phys. Rev. B **70**, 075313 (2004).

[17] J. Nitta, T. Akazaki, H. Takayanagi, and T. Enoki, Phys. Rev. Lett. **78**, 1335 (1997); G. Engels, J. Lange, T. Schäpers, and H. Lüth, Phys. Rev. B **55**, R1958 (1997); T. Schäpers, G. Engels, J. Lange, T. Klocke, M. Hollfelder, and H. Lüth, J. Appl. Phys. **83**, 4324 (1998); C.-M. Hu, J. Nitta, T. Akazaki, H. Takayanagai, J. Osaka, P. Pfeffer, and W. Zawadzki, Phys. Rev. B **60**, 7736 (1999); D. Grundler, Phys. Rev. Lett. **84**, 6074 (2000); T. Matsuyama, R. Kürsten, C. Meißner, and U. Merkt, Phys. Rev. B **61**, 15588 (2000); K. M. Fonseca Romero, S. Kohler, and P. Hänggi, Chem. Phys. **296**, 307 (2004); C. Schierholz, T. Matsuyama, U. Merkt, and G. Meier, Phys. Rev. B **70**, 233311 (2004).

[18] H. Frauenfelder and E. M. Henley, *Subatomic Physics* (Prentice-Hall Inc. Englewood Cliffs, 1974).

[19] Already in E. Abrahams, Phys. Rev. **107**, 491 (1957) first estimates were published on the magnitude of electronic dipolar interactions.

[20] Note that the dipolar interaction alone, in the absence of phonons, would slightly mix spins but would not cause spin decay; cf. Ref. 53.

[21] Quantum computing employs entanglement of different total spin states, such as the singlet and the triplet state in the simplest situation of two electron spins, at Zeeman energies that are comparable to the level separations between correlated electron energies.

[22] For an overview, see S. M. Reimann and M. Manninen, Rev. Mod. Phys. **74**, 1283 (2002).

[23] R. Egger, W. Häusler, C. H. Mak, and H. Grabert, Phys. Rev. Lett. **82**, 3320 (1999); Phys. Rev. Lett. **83**, 462(E) (1999).

[24] W. Häusler, Europhys. Lett. **49**, 231 (2000).

[25] S. A. Mikhailov, Phys. Rev. B **65**, 115312 (2002).

[26] W. Häusler, Adv. Solid State Phys. **34**, 171 (1994).

[27] W. Häusler, Z. Phys. B: Condens. Matter **99**, 551 (1996).

[28] W. Häusler, Ann. Phys. **5**, 401 (1996).

[29] For example, when comparing Ref. 4 with optical data taken from bulk semiconductors, cf. D. Hägele, M. Oestreich, W. W. Rühle, N. Nestle, and K. Eberl, Appl. Phys. Lett. **73**, 1580 (1998), or J. M. Kikkawa and D. D. Awschalom, Nature **397**, 139 (1999).

[30] K. Jauregui, W. Häusler, and B. Kramer, Europhys. Lett. **24**, 581 (1993).

[31] J. H. Jefferson and W. Häusler, Phys. Rev. B **54**, 4936 (1996).

[32] M. Hamermesh, *Group Theory and its Applications to Physical Problems* (Dover Publications, New York, 1989).

[33] J. Bardeen and W. Shockley, Phys. Rev. **80**, 72 (1950).

[34] L. P. Rokhinson, L. J. Guo, S. Y. Chou, and D. C. Tsui, Phys. Rev. B **63**, 035321 (2001).

[35] L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, Rep. Prog. Phys. **64**, 701 (2001); and references therein.

[36] M. Thorwart and P. Hänggi, Phys. Rev. A **65**, 012309 (2002).

[37] By use of time reversal invariance, it can be shown that the ground state of two spin-$\frac{1}{2}$, fermions is $S=0$; cf. E. Lieb and D. Mattis, Phys. Rev. **125**, 164 (1962).

[38] K. F. Bonhoeffer and P. Harteck, Berichte der Berliner Akademie, p. 103 (1929).

[39] I. F. Silvera, Rev. Mod. Phys. **52**, 393 (1980).

[40] J. H. Jefferson, M. Fearn, D. L. J. Tipton, and T. P. Spiller, Phys. Rev. A **66**, 042328 (2002); note that the physical basis for this work was established in Refs. 27 and 28.

[41] P. Jarillo-Herrero, S. Sapmaz, C. Dekker, L. P. Kouwenhoven, and H. S. J. van der Zant, Nature **429**, 389 (2004).

[42] C. E. Creffield, W. Häusler, J. H. Jefferson, and S. Sarkar, Phys. Rev. B **59**, 10719 (1999).

[43] Exact degeneracy would entail straightforward degenerate perturbative treatment (which poses no conceptual difficulty when defining new "effective spins"); this case is, however, beyond the scope of the present work.

[44] D. Weinmann, W. Häusler, and B. Kramer, Phys. Rev. Lett. **74**, 984 (1995).

[45] D. Weinmann, W. Häusler, and B. Kramer, Ann. Phys. **5**, 652 (1996).

[46] J. Bardeen and W. Shockley, Phys. Rev. **80**, 72 (1950).

[47] L. Pintschovius, J. A. Vergés, and M. Cardona, Phys. Rev. B **26**, 5658 (1982).

[48] *Numerical data and Function Relationships in Science and Technology*, Vol. 17 of Landolt-Börnstein Series, Group IV, Part A, edited by O. Madelung (Springer, Berlin, 1982).

[49] This is reasonable for $N>4$. Only for smaller electron numbers the first excited level of given spin may be already a member of the first excited plasmon multiplet; whence in this case $E_{n'_S} - E_{n_{0S}} \sim \omega_{\mathrm{pl}}$; cf. W. Häusler and B. Kramer, Phys. Rev. B **47**, 16353 (1993) and Ref. 26.

[50] A. M. Tyryshkin, S. A. Lyon, A. V. Astashkin, and A. M. Raitsimring, Phys. Rev. B **68**, 193207 (2003).

[51] C. P. Slichter, *Principles of Magnetic Resonance* (Springer, Berlin, 1980).

[52] W. Häusler, Z. Phys. B: Condens. Matter **81**, 265 (1990).

[53] Spin decay by coupling to the bath of dipolarly broadened nuclei has been considered by R. de Sousa and S. Das Sarma, Phys. Rev. B **68**, 115322 (2003).