Elastic Spin Relaxation Processes in Semiconductor Quantum Dots

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Abstract

Electron spin decoherence caused by elastic spin-phonon processes is investigated comprehensively in a zero-dimensional environment. Specifically, a theoretical treatment is developed for the processes associated with the fluctuations in the phonon potential as well as in the electron procession frequency through the spin-orbit and hyperfine interactions in the semiconductor quantum dots. The analysis identifies the conditions (magnetic field, temperature, etc.) in which the elastic spin-phonon processes can dominate over the inelastic counterparts with the electron spin-flip transitions. Particularly, the calculation results illustrate the potential significance of an elastic decoherence mechanism originating from the intervalley transitions in semiconductor quantum dots with multiple equivalent energy minima (e.g., the X valleys in SiGe). The role of lattice anharmonicity and phonon decay in spin relaxation is also examined along with that of the local effective field fluctuations caused by the stochastic electronic transitions between the orbital states. Numerical estimations are provided for typical GaAs and Si-based quantum dots.

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I. INTRODUCTION

Electron spin relaxation in semiconductor quantum dots (QDs) has received much attention as they are a natural candidate for the qubit in quantum computing (QC) (see Refs. 1 and 2 for a brief review). Particularly, a significant effort has been devoted to investigate the spin transition probabilities associated with the spin-flip interactions, for example, the so-called longitudinal spin relaxation ($T_1$) (i.e., energy relaxation) induced via the inelastic process of phonon absorption/emission by an electron spin localized in a QD. Although generally an importance source of decoherence, the spin-flip events can sometimes be efficiently suppressed by an appropriate choice of external conditions including the magnetic field strength or direction. In this case, alternative mechanisms (such as the elastic spin-phonon scattering that does not assume energy exchange between the electron Zeeman energy and thermal bath) can be primarily responsible for decoherence. So far, however, the elastic processes have received relatively little attention. This disparity may be due, in part, to the mathematical simplicity in the treatment of the former case (e.g., calculations based on the Fermi’s golden rule), while the elastic processes of spin decoherence need more sophisticated approaches. At the same time, recent proliferation of the literature on electron spin relaxation has exacerbated the confusion on this complicated subject matter. Frequently, an individual study addresses only a specific aspect of spin decoherence or dephasing without elucidating its place in the general theory or the interrelationships with other relevant works. Consequently, one faces difficulties each time in determining which existing mechanisms, processes, or theories are relevant for spin relaxation under the given external conditions and specific QD properties.

The purpose of the present study is to systematically identify and analyze the crucial transversal ($T_2$) spin relaxation processes in the context of qubit dephasing in the electron spin based QC. Specifically, the role of spin-phonon interaction on the elastic decoherence processes is examined theoretically from the perspective of the fluctuations in the effective magnetic field. The calculation identifies the conditions in which the elastic spin-lattice processes can dominate over the inelastic counterparts. The analysis is applicable to the electron spin in both group IV and III-V QDs. Note that the related issues from the spintronics point of view were surveyed in Refs. 3 and 4.

The rest of this paper is organized as follows. Section II provides a systematic classifica-
tion of most, if not all, of the processes identified so far that lead to a type of electron spin relaxation (i.e., dephasing, decoherence or energy relaxation). Then, the discussion focuses on those caused by the spin-phonon interaction and describes different manifestations of the spin-phonon interaction in a manner consistent to both elastic and inelastic processes. Subsequently, the elastic decoherence processes that warrant further analysis are recognized and their potential significance outlined in the context of QC. Section III summarizes a theoretical framework necessary for the mathematical description of electron spin relaxation in terms of quantum kinetic equation.

Each of the following three sections describes a particular elastic process that can be dominant under a certain condition. In Sec. IV, we analyze the effects of spin precession fluctuations due to the irregular phonon phase disturbances originating from phonon relaxation via the lattice anharmonicity or imperfections. Such a situation applies not only to real (thermally activated) phonons but also to zero-point phonons that the classical picture cannot address. Section V considers spin phase diffusion due to the modulations in the longitudinal g-factor and hyperfine interaction through the phonon-assisted transitions between the lowest electronic states (without spin-flip). Section VI concerns electron spin in a semiconductor QD with multiple equivalent minima (e.g., Si$_{1-x}$Ge$_x$). As the degeneracy is lifted due to structural inversion asymmetry, phonon-mediated transitions between the "valley-split" states can be allowed when the local crystal imperfections are present. This process can be very significant since the energy separation of the valley-split states is generally comparable to the thermal energy in the QC operation. Then, the discussion on the comparative importance of different processes in the parameter space is provided in Sec. VII, followed by a brief conclusion at the end.

II. SPIN RELAXATION PROCESSES AND MECHANISMS

For a systematic analysis, Table I shows simple classification of the relevant spin relaxation processes identified so far. The term ”dephasing” (with the characteristic time $T^*_2$) concerns phase relaxation of a QC system consisting of multiple qubits, while ”decoherence” ($T_2$) is for a single qubit (i.e., single electron spin in a QD). Hence, decoherence contributes to dephasing ($T_2 \geq T^*_2$). The ”random local fields” category denotes the processes that lead to phase diffusion of the multi-qubit system without causing decoherence in the individual
qubits. The decoherence or transversal relaxation has contributions from both the elastic and inelastic processes. The latter involves spin flip transitions leading to the energy (or longitudinal) relaxation of the spin states (i.e., $T_1$). Of the spin decoherence processes, those marked by the # sign in the table are induced by the spin-phonon interaction. The listed $T_2$ processes correspond to the uncontrollable interactions with the environment that assumes to be in a thermodynamic equilibrium at temperature $T$. Inaccuracies in the parameters of the control pulses during QC operations as well as uncontrollable perturbations introduced with these pulses are beyond the current description. Table II summarizes various manifestations of the spin-phonon mechanism in a manner consistent to both elastic and inelastic processes. A brief description on each category is given below.

A. Dephasing in random fields

The fastest relaxation in a quantum computer consisting of a large number of qubits $[N \sim 1000$ (see Ref. 2)] is the dephasing process associated with transversal magnetization loss with the characteristic time $T_2^*$. Clearly, the electrons experience a dispersion of spin precession in the presence of a random effective magnetic field. Even when the magnetic field can be considered a constant spatially and temporally for a given qubit within the time scale of QC operation, its random variation across the ensemble of qubits can induce dephasing of the system. Although some of its effect may be potentially mitigated by QC algorithms,\textsuperscript{2,5} the absence of this additional complexity provides an obvious advantage.

One reason for the dispersion in the precession frequency is the $g$-tensor variation over the $N$ qubits. Consequently, even in a very pure crystal, the unavoidable fluctuation in the QD sizes results in the $g$-factor dispersion $\Delta g$, which subsequently induces dephasing with the rate of $T_{g^*}^{-1} = \Delta g \mu_B B$ (where $\mu_B$ is the Bohr magneton and $B$ the external magnetic field). This mechanism of spin phase relaxation has not yet been investigated in the zero-dimensional (0D) structures. A related case of 2D hole localization in a fluctuating potential was studied in Refs. 6 and 7. In addition, the randomness of spin-orbital coupling in a quantum well due to the fluctuation of dopant concentration was considered in Ref. 8.

Similarly, electron interaction with nuclear spins can induce the spin phase diffusion due to the random distribution of the nuclear spin states and their spatial locations.\textsuperscript{9,10,11} In principle, replacement of the magnetic isotopes by nonmagnetic counterparts ($I = 0$) can
remove or suppress this source of relaxation.\textsuperscript{12} However, it may not be a practical option as a significant level of purification is required to be effective; the dephasing rate depends only weakly on the nuclear spin concentration $n_I$ (i.e., $\propto \sqrt{n_I}$). As indicated in Table I, the random fields of magnetic impurities also contribute to the dephasing process. The last one listed in this category, the magneto-dipole interaction between the qubits, plays a specific role since it can be incorporated (thus, eliminated) to the QC algorithm under certain conditions.\textsuperscript{2,13,14}

**B. Inelastic processes**

Among the inelastic processes in Table I, the direct spin-flip via one phonon emission/absorption (see Fig. 1) is the most studied due to its simplicity in concept as well as the importance at low temperatures.\textsuperscript{15,16,17,18,19,20} The rate of this process is proportional to the phonon density of states $g(\omega_q)[\sim \omega_q^2]$ at the resonant phonon energy $\hbar \omega_q = g \mu_B B$ ($\equiv h\omega$) as well as the square of the spin-phonon matrix element ($\sim \omega_q B^2$). As a result, a strong magnetic field dependence ($\sim B^5$) is expected for the direct spin-flip. Thus, at low magnetic fields, the effect of this process can be significantly reduced. When $k_B T < \hbar \omega$, the influence of the temperature can be ignored as the phonon emission determines the relaxation rate.

The Orbach process\textsuperscript{21,22} considers the spin-flip events through successive transitions between the ground and excited orbital states separated by an energy $\delta_0$. For example, an electron first makes a spin-flip transition to the excited state with opposite spin by absorbing a phonon with energy $\delta_0 \mp \hbar \omega$, followed by the return to the ground state through emission of a phonon with energy $\delta_0$. An alternative channel is to make a transition to the excited state without spin flip (i.e., $\delta_0$) and, then, return to the ground state with opposite spin (i.e., $\delta_0 \mp \hbar \omega$). Thus, the net effect is the electron spin flip with the energy of $\hbar \omega$ (i.e., the Zeeman splitting) transferred to the thermal bath. Under certain conditions, the corresponding matrix elements can exceed those for the direct spin-flip transitions between the Kramers doublet.\textsuperscript{23} It should be stressed that the Orbach process assumes two one-phonon transitions in tandem. Hence, the first transition with the energy threshold of $\delta_0 \mp \hbar \omega$ or $\delta_0$ provides the primary temperature dependence along with a weak influence of the magnetic field.
The two-phonon process concerns the longitudinal spin relaxation via two-phonon absorption, emission or inelastic phonon scattering (Raman process). It can be further divided into three groups; the direct spin–two-phonon transitions due to the nonlinearity of the spin-phonon Hamiltonian\(^{24}\) [Fig. 2(a)], the virtual transitions mediated by one-phonon interactions through the excited states\(^{15,17}\) [Fig 2(b)], and the anharmonic Raman process\(^{25,26}\) [Fig. 2(c)]. In the case of virtual transitions, the integration over phonons includes the poles at the phonon energy \(\hbar \omega_q \sim \delta_0\) if \(\delta_0 < k_B \Theta\), where \(\Theta\) is the Debye temperature. The treatment of this singularity (the so-called phonon resonant fluorescence) beyond the conventional perturbation theory\(^{27}\) involves essentially the finite lifetime \(\tau_R\) of the excited states that exactly reproduces the relaxation rate of the Orbach process\(^{21,28}\). This coincidence/similarity is lifted (i.e. poles can be ignored as compared with Orbach process) when the finite phonon lifetime \(\tau_q\) is considered since \(\tau_R^{-1} \ll \tau_q^{-1}\). As the processes involving the inelastic phonon scattering show a strong dependence on temperature \((T^7 - T^{11})\),\(^{15,17}\) it has been mostly ignored in the consideration of QC that operates at very low temperatures. However, a recent analysis\(^{12}\) suggests its potential significance in the low magnetic field regime due to the weak dependence on the field strength \((B^2)\).

The process of directly transferring the electron Zeeman energy to the nuclear dipole-dipole reservoir is another possible channel for longitudinal relaxation.\(^{29}\) This, however, can be effective only in the external magnetic fields comparable or weaker than the local nuclear fields, which are generally sufficiently small. In addition to those listed in Table I, the simultaneous flip-flop of several spins generally entails energy exchange with the phonon or spin-spin reservoir. Much less attention has been paid to such processes in the literature.\(^{30,31,32}\)

### C. Elastic processes

The elastic processes can be imagined as a result of uncontrollable fluctuations of Zeeman frequency or its equivalent, i.e., the fluctuations of longitudinal magnetic field that conserve the electron spin projection on the quantization axis. As summarized in Table I, there are at least four different manifestations of this process. Of these, the first one listed (“anharmonic vibration”) represents the case that the irregular phonon phase disturbances originating from the phonon relaxation induces a net effect of electron spin phase decay via the spin-
phonon interaction. If the phonons are treated as harmonic oscillators without damping, the longitudinal component of the ”effective” interaction field takes the form of a harmonic perturbation and does not cause alteration in the electron spin phase.\(^{33}\) One exception may be the small spin phase decoherence ($\sim 10^{-9}$) that can be acquired under the additional assumption of identical phase for all the harmonic phonon modes at the initial instant.\(^{34}\) However, a different situation can be realized when the phonon oscillation is interrupted by a series of random phase disturbances as schematically illustrated in Fig. 3. The reason of such phonon phase steps can be lattice anharmonicity, phonon scattering at the impurities or lattice defects, etc. These irregular phonon phase perturbations affect the electron spin precession resulting in its phase relaxation.\(^{33}\)

The fluctuations in the local effective magnetic field (and, thus, spin decoherence) can also result from the transitions between the electronic (orbital) states with different Zeeman frequencies. As the electron experiences the stochastic transitions between these orbital states without spin flip, electron spin phase can change due to the finite and random lifetime in the states with different precession frequencies (Fig. 4).\(^{35}\) It should be emphasized that although the transitions are mediated by phonons, the energy exchange between the Zeeman and phonon reservoirs does not take place in contrast to the Orbach process; i.e., this case undoubtedly belongs to the elastic processes.

Electron spin can accumulate the phase shift through random thermal changes in the nuclear spin distribution as well. Although a quantum state of individual nuclear spin generally has a relatively long lifetime, a large ensemble of nuclear spins (or spin flip transitions) produces a rapidly evolving effective field distribution. Its significance can be gauged in terms of electron resonant frequency diffusion (i.e., the so-called spectral diffusion) in the spin-echo experiments.\(^{12,36,37,38,39}\) A peculiar property of this process is the cubic exponential decay of spin-echo signal $\sim \exp[-t^3/T_M^3]$, where the memory time $T_M$ can be calculated in the model of uncorrelated nuclear spin flips\(^{38}\) or in the model taking into account the flip-flop processes for nuclear spin pairs.\(^{39}\) Note that the spectral diffusion process is probably irrelevant to QC where only the initial phase decay of $\sim 10^{-4} - 10^{-6}$ matters for fault tolerant operation. In the corresponding short time frame, the processes with a linear exponential decay prevail as demonstrated in Ref. \cite{12}. Decoherence can also be attributed to isolated (i.e. non-interacting) nuclear spins.\(^{40}\) This is because each nuclear spin precesses around the electron hyperfine field, which is inhomogeneous over the QD volume. Such uncorrelated
nuclear spin precession, in turn, leads to the change in the strength of nuclear hyperfine interaction field affecting the electron spin and its phase. The efficiency of this process, however, is much weaker than that of spectral diffusion when the inter-nuclear interaction is taken into account.

The elastic processes in Table I include the one mediated by fluctuations of Berry geometric phase. The efficiency of this process compared to the others is not clearly understood yet. In addition, an elastic phonon scattering on a group of spins can cause them out of phase. However, this process is less relevant for single qubit decoherence.

D. Mechanisms of spin-phonon interaction

Table II summarizes the microscopic mechanisms of spin-phonon interaction that give rise to the elastic and inelastic spin relaxation processes (marked by \# in Table I). Despite the great multiplicity of the mechanisms considered in the literature, they can be classified into three groups following the global characteristics; i.e., the mechanisms resulting from the (i) spin-orbit, (ii) hyperfine, and (iii) spin-spin interactions. In turn, these interactions manifest themselves in a number of different ways, all of which can contribute to spin phase and longitudinal relaxation.

Taking into account that interactions (i), (ii) and (iii) mentioned above possess, generally speaking, non-zero matrix elements between the states appurtenant to the Zeeman doublet, their modulation by phonon vibrations can be expressed as a Hamiltonian realizing the spin-phonon interaction (listed as "Direct" in Table II). Mathematically, if a phonon mediates an electronic potential \( V_{ph}(r) \), the spin-orbit interaction is changed by the term \( H_{so-ph} = \frac{\hbar}{2m^2c^2}|\nabla V_{ph} \times p|s \) ("SOI modulation" mechanism). At the same time, the shift \( \Delta R_j \) of an atomic position from the site \( R_j \) due to the lattice vibration (i.e., phonons) also changes the Hamiltonian for the hyperfine interaction \( H_{hf}(R_j) = \sum_j \mathbf{s} \cdot \hat{a}_{hf}(r - R_j)I \) by \( H_{hf-ph} = H_{hf}(R_j + \Delta R_j) - H_{hf}(R_j) \) ("HFI modulation" mechanism); here, \( \hat{a}_{hf}(r - R_j) \) is the hyperfine interaction tensor. The case of spin-orbit interaction adapted to the QDs was considered in Ref. 15. Direct phonon fluctuations of hyperfine interaction were studied for a donor in silicon, \( F \)-centers in alkali halides, and adapted to QDs.

In a similar manner, one can introduce the spin-phonon interaction mediated by the spin-spin interactions \( H_{ss-ph} \). In this case, one can further separate the dipole-dipole and
exchange interactions. Obviously, these mechanisms depend on the inter-spin distances. Phonon modulation of dipole-dipole interaction ("Waller" mechanism) was considered in Ref. 46; the role of exchange interaction including the Dzialoshinski-Moria terms\textsuperscript{47,48} (that can be important in crystals with no inversion symmetry\textsuperscript{49}) was analyzed in Ref. 30.

The direct mechanisms considered above are not the only manifestation of the spin-phonon interactions. In the strict sense, the Kramers doublets actually represent non-multiplicative functions of spin and space variables due to the the spin-orbit or hyperfine interactions (i.e., mixing of orbital and spin states; "Admixture" in Table II). This makes a spin-independent phonon potential capable of evoking the transitions between the Kramers doublet. Such a mechanism becomes apparent in the case of hole spin relaxation in a QD, where the spin-orbit interaction imposes a linear combination of different spin and orbital states for the basis of valence band\textsuperscript{50,51}. Mathematically, the mechanism constitutes only the non-zero matrix elements that can be symbolically transformed to the form

\begin{equation}
\sum_{\{e\}} \left[ \frac{\langle g \uparrow | V_{ph}(r) | e \uparrow \rangle \langle e \uparrow | H_{mix} | g \downarrow \rangle}{E_g - E_e} + \frac{\langle g \uparrow | H_{mix} | e \downarrow \rangle \langle e \downarrow | V_{ph}(r) | g \downarrow \rangle}{E_g - E_e} \right],
\end{equation}

if a perturbation theory is applicable for the system with the electron energies $E_g$ and $E_e$ in the ground ($|g\rangle$) and excited ($|e\rangle$) states; \(\uparrow\) and \(\downarrow\) denote different spin states. Equation (1) assumes that the diagonal part of the Hamiltonian $H_{mix}$ enters into $E_g$ and $E_e$, while the off-diagonal element mixes the ground and excited states with opposite spins.

In the second-order perturbation theory, Eq. (1) represents the spin-phonon interaction as the process of virtual transition to the spin-flip excited state due to $H_{mix}$ followed by a transition to the ground state without spin flip via the spin-independent electron-phonon interaction, or vice versa. In the case of electron in a QD, this mechanism can possess an advantage over the direct spin-phonon interaction due to the relatively strong spin coupling between the non-Kramers (i.e., orbital) states. A distinctive feature of such indirect spin-phonon interactions is their proportionality to the matrix elements of electron-phonon interaction between the ground and excited states. Consequently, these mechanisms can be further classified based on the types of electron-phonon interactions as well as the potentials that cause effective mixture of the ground and excited states.

The mechanism caused by simultaneous manifestation of spin-orbit and spin-independent electron-phonon interactions was initially developed by Kronig\textsuperscript{52} and Van Vleck\textsuperscript{53} for magnetic ions. Adaptation of this "Kronig-Van Vleck" mechanism to the conduction electrons
in a QD can be conveniently achieved by extracting the spin-orbit coupling in the form of the Rashba and Dresselhause terms (which efficiently mix the QD ground and excited states).\textsuperscript{54,55} Then, the phonon potential of various origins such as the deformation potential, piezoelectric potential,\textsuperscript{15,18,19,20} or the ripple effect at the QD interface\textsuperscript{56} gives rise to the effective spin-phonon coupling in the second order. The spin-orbit coupling in the valence band is generally much stronger; hence, holes are less attractive as a qubit.\textsuperscript{50,51}

When $H_{\text{mix}}$ is due to the hyperfine interaction, the spin flip can be considered in a manner similar to that with the spin-orbit interaction as discussed in Refs.\textsuperscript{57,45}; an isomorphous case of a diluted magnetic semiconductor QD was also investigated.\textsuperscript{58} It seems that this admixture spin-phonon interaction is more effective than the direct counterpart (i.e., phonon modulation of the hyperfine interaction). As to the spin-spin interactions (both dipole-dipole and exchange), they can also induce the mixing of spin and orbital states similarly; however, such a treatment has not yet been developed to the best of our knowledge.

In addition to the direct and admixture mechanisms discussed above, one can also consider "indirect" manifestation of spin dependent interactions. This group of mechanisms accounts for the random fluctuations in the effective field through the phonon-mediated stochastic transitions between different orbital states. As the characteristics of $g$-tensor, hyperfine field, or spin-spin interactions vary with the orbital states, electrons experience uncontrollable changes in these parameters while undergoing spin-independent phonon scattering between them. Such fluctuations cause the transversal relaxation of electron spin\textsuperscript{35} along with a potential contribution to the longitudinal relaxation. However, the latter (i.e., spin-flip) is expected to be small due to the negligibly small influence on the spectral density at typical Zeeman frequencies.

Finally, there is a higher-order phonon-mediated mechanism that can affect the spin relaxation. Although the phonon potential $V_{\text{ph}}(r)$ cannot influence spin precession directly, its incorporation with the electron potential $V_0(r)$, which contributes to the $g$-tensor in a different manner,\textsuperscript{59,60} results in a specific spin-phonon interaction $H_{g-\text{ph}} = \sum (\delta g_{ij}/\delta V_0) V_{\text{ph}}(r) \mu_B B_i s_j$, where $s_j$ are the components of electron spin operator. This mechanism was analyzed in a QD with respect to the Orbach spin-flip processes\textsuperscript{15} as well as in a bulk semiconductor\textsuperscript{61}.

In summarizing the possible mechanisms of spin-phonon interaction, it must be pointed out that all of them make *correlated* contributions to the processes marked \# in Table I.
For example, the direct spin-flip process results from the phonon-induced fluctuation of the effective magnetic field (with the resonant frequency $\hbar \omega$) that is realized by additive contributions from the spin-phonon mechanisms listed in Table II (i.e., the spin-orbit, hyperfine, and spin-spin interactions). Consequently, the corresponding matrix elements of individual mechanisms must be summed before the probability of any process is calculated. Although the importance of interference for certain mechanisms was already discussed in Refs. 62 and 18, it requires further theoretical scrutiny. However, the anticipated difficulty may be circumvented for reasonably weak magnetic fields if the spin-phonon Hamiltonian $H_{\text{s-ph}}$ is considered in terms of a phenomenological tensor $A$ following the analysis of Ref. 63. This parameter $A$ is designed to represent the net/total contribution of the spin-orbit mechanisms and can be found experimentally from the dependence of electron paramagnetic resonance signal on the applied stress.

As for the contributions of different processes (Table I) to spin relaxation, they do not interfere and, thus, can be considered independently. As mentioned earlier, the present paper investigate dominant elastic spin relaxation processes via the spin-phonon mechanisms, particularly those originated from the spin-orbit and hyperfine interactions.

### III. BASIC EQUATION FOR ELECTRON SPIN EVOLUTION

With the mechanisms responsible for the spin-phonon interaction $H_{\text{SL}}$ ascertained in Table II, the evolution of electron spin can be described according to the rubrics under "Decoherence ($T_2$)" in Table I. In contrast to the calculation of $H_{\text{SL}}$ that requires individualized treatments for each mechanisms, most processes, elastic and inelastic, marked by # in Table I (with an exception of "geometric phase" process) can be approached by a general formalism based on kinetic equations. For simplicity, the derivation of these equations is restricted to the first Born approximation in the present consideration. As such, the two-phonon processes, which appear on the second order of $H_{\text{SL}}$ [Fig. 2(b)], are not included.

There are several comparable approaches to describe the evolution of spin $S = 1/2$ interacting with a thermal bath. For the purposes of decoherence evaluation in the QDs, it is desirable to apply a system of equations directly for the mean spin components (rather than for the density matrix components), which analyze their temporal progression on the Bloch sphere from an initial state to that corresponding to the thermal equilibrium of the
system. At the same time, it is desirable to express the relaxation coefficients in terms of the spectral density of the thermal bath operators that allows application of the formalism well-developed in the case of thermodynamic equilibrium. Finally, the equations must correctly account for the anisotropy of spin interaction with the thermal bath. Such equations were studied previously in the cases of specific symmetry properties of the correlation functions. This section generalizes and adopts these equations to the problem of spin decoherence.

It is convenient to represent the Hamiltonian of one electron system in terms of the effective spin operator $s$ for the Kramers doublet $\{g\} = \{|g\rangle, |\tilde{g}\rangle\}$ of the ground electronic state. The linear magnetic splitting of Kramers doublet $\{g\}$ is described by an effective Hamiltonian in the common form $H_S = g_{\mu\nu}\mu_B B_\mu s_\nu$, where the $g$-tensor $g_{\mu\nu}$ is introduced. Similarly, the electron spin interaction $H_{SL}$ with the thermal bath (of Hamiltonian $H_L$) is expressible as a linear form of the spin operators, $H_{SL} = \Omega^{(g)} s$ if only the states $\{g\}$ are involved in the relaxation. Actually, $H_{SL}$ also describes the transitions between different Kramers doublets. If these transitions from the ground $\{g\}$ to excited $\{e\}$ states are virtual (as is the case at sufficiently low temperatures $kT << \delta_0$, where $\delta_0$ is the energy separation between the $\{g\}$ and $\{e\}$ states), reduction of the Hamiltonian $H_{SL}$ to the basis $\{g\}$ of spin operators $s$ poses no specific problem. For example, the off-diagonal elements of $H_{SL}$ between $\{g\}$ and $\{e\}$ states can be eliminated through the procedure of canonical transformation. Hence, the evolution of the spin system at low temperatures is fully described by an effective $2 \times 2$ Hamiltonian,

$$H = H_S + H_L + H_{SL}. \quad (2)$$

This Hamiltonian can also be applicable to the cases other than at low temperatures. Note that the problems involving a group of Kramers doublets (e.g., $\{n\} = \{g\}, \{e\}$) sometimes allow description by an effective Hamiltonian $H_S^{\text{eff}}$ with $\langle \{n\}|H_S^{\text{eff}}|\{n'\}\rangle = \delta_{n,n'}(E_n + g_{\mu\nu}^{(n)}\mu_B B_\mu s_\nu)$ and $\langle \{n\}|H_{SL}|\{n'\}\rangle = (\hat{\Omega})_{n,n'} s$; here, the matrix of orbital energy $\delta_{n,n'} E_n$ can alternatively be considered a part of $H_L$ as well. Hence, this case leaves room for the Hamiltonian in the form of Eq. (2) even at elevated temperatures. Some examples will be discussed later.

For a detailed description, we start with the kinetic equation for the spin density matrix

$$\hat{\sigma} = \text{Tr}_L \rho. \quad (3)$$

Here $\rho$ obeys the Liouville equation $i\dot{\rho} = [H, \rho]$, the trace is taken over all variables of the
system except the given electron spin, and $\hbar$ and $k_B$ are set to 1 in this section. Assuming that the thermal bath is in a thermodynamic equilibrium, one can introduce its statistical operator

$$f = \frac{\exp(-\beta H_L)}{\text{Tr}_L \exp(-\beta H_L)}; \quad \beta = \frac{1}{T}. \quad (4)$$

Following the approach developed in Ref. 65 with a projection operator method (see Ref. 66), the kinetic equation of operator $\hat{\sigma}$ in the Born approximation is obtained as

$$\frac{d\hat{\sigma}(t)}{dt} = -i[H_S, \hat{\sigma}(t)] - \int_0^t \text{Tr}_L[V(t, t'), [V(t, t'), f\hat{\sigma}(t)]]dt', \quad (5)$$

where $[A, B] = AB - BA$. Equation (5) implies renormalization of the spin-bath interaction $V = \langle \Omega^{(g)} - \langle \Omega^{(g)} \rangle \rangle s$, so that the averaging over the bath $\langle \ldots \rangle = \text{Tr}_L f \ldots$ results in $\langle V \rangle = 0$ while $H_S = H_S^{(g)} + \langle \Omega^{(g)} \rangle s$. The last term in Eq. (5) resembles a collision integral for the common kinetic equation, where the possible influence of alternating magnetic fields is ignored. In the case of time-independent $H_S$, the operators on the right-hand side of Eq. (5) take the forms

$$V(t, t') = \exp\{-iH_S (t - t')\} V(t') \exp\{iH_S (t - t')\}, \quad (7)$$

$$V(t) = \exp(iH_L t)V \exp(-iH_L t). \quad (8)$$

Although the integrand in Eq. (5) can be calculated for arbitrary coordinates, its subsequent application is not very convenient. Without the loss of generality, the $z$ axis can be chosen along the quantization direction that reduces the spin Hamiltonian to

$$H_S = \omega s_z. \quad (9)$$

Note that the $z$ axis, generally speaking, does not coincide with the direction of the external magnetic field if the $g$-tensor reflects the low symmetry of a QD and $\omega = \sqrt{\sum_i \left( g_i \mu_B B_i + \langle \Omega_i^{(g)} \rangle \right)^2} \equiv g_{\text{eff}} \mu_B B$. Here $g_i$ are the principal values of $g$-tensor while $B_i$ and $\Omega_i^{(g)}$ are the vector components along the main axes of $g$-tensor. By adopting the specified coordinate system, Eqs. (7) and (8) can be easily calculated with $\Omega_{\mu} = \Omega_{\mu}^{(g)} - \langle \Omega_{\mu}^{(g)} \rangle$,

$$V(t, t') = \Omega_x(t')(s_x \cos u + s_y \sin u) + \Omega_y(t')(s_y \cos u - s_x \sin u) + \Omega_z(t')s_z, \quad (10)$$

$$V(t) = \Omega_x(t)s_x + \Omega_y(t)s_y + \Omega_z(t)s_z, \quad (11)$$
where \( u = \omega(t - t') \) and \( \Omega_{\mu}(t) \) is defined similar to Eq. (8). One can see that the collision integral can be expressed in terms of double-time correlation functions

\[
\langle \Omega_{\mu}(t), \Omega_{\nu}(t') \rangle = \text{Tr}_L \{ e^{-\beta H_L} \Omega_{\mu}(t), \Omega_{\nu}(t') \} \tag{12}
\]

with an evident property \( \langle \Omega_{\mu}(t), \Omega_{\nu}(t') \rangle = \langle \Omega_{\mu}(t - t'), \Omega_{\nu}(0) \rangle = \langle \Omega_{\mu}(0), \Omega_{\nu}(t' - t) \rangle \). We consider that these correlation functions reduce to zero as soon as \( |t - t'| \) exceeds the correlation time \( \tau_c \). Thus, two regimes of spin evolution must be distinguished (Ref. 68). In the first regime for short times \( t \ll \tau_c \), the relaxation coefficients in Eq. (5) are proportional to \( t \). This leads to a quadratic-on-\( t \) deviation from the initial states of the system that reproduces the very general results obtained previously in various analyses (see Refs. 69 and 70). Such a non-linearity results in only a marginal effect of spin transition from the ground state at sufficiently short times (Zeno effect 71). Consequently, we focus on the second, opposite case of reasonably long time \( t \gg \tau_c \), when the upper limit of the integral in Eq. (5) can tend to infinity, \( t \to \infty \). In this case, the relaxation coefficients are independent of time and Eq. (5) describes the Markovian process with exponential evolution of \( \hat{\sigma}(t) \).

To proceed further, it is convenient to introduce the Fourier transformation of the correlation functions [Eq. (12)],

\[
\gamma_{\mu\nu}(\omega) \equiv \langle \Omega_{\mu}(\tau), \Omega_{\nu}(0) \rangle_{\omega} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle \Omega_{\mu}(\tau), \Omega_{\nu}(0) \rangle e^{i\omega \tau} d\tau. \tag{13}
\]

Considering the functional form of Eq. (5), another useful relation is

\[
\int_{0}^{\infty} \langle \Omega_{\mu}(\tau), \Omega_{\nu}(0) \rangle e^{i\omega \tau} d\tau = \pi \gamma_{\mu\nu}(\omega) + iP \int_{-\infty}^{\infty} \frac{\gamma_{\mu\nu}(\omega')}{\omega - \omega'} d\omega', \tag{14}
\]

where we use the symbolic identity \( \int_{0}^{\theta} e^{i\omega \tau} d\tau = \pi \delta(\omega) + iP \frac{1}{\omega} \). \( P \) denotes the principal value of the integral. Finally, let us express the density matrix \( \hat{\sigma}(t) \) in terms of mean values \( \langle s_x \rangle \equiv X, \langle s_y \rangle \equiv Y, \) and \( \langle s_z \rangle \equiv Z, \)

\[
\hat{\sigma}(t) = \frac{1}{2} \left[ \hat{1} + 4 (X s_x + Y s_y + Z s_z) \right], \tag{15}
\]

so that \( \text{Tr}_s \hat{\sigma}(t) s_{\mu} = \langle s_{\mu} \rangle \), where \( \hat{\sigma}(t) \) is taken from Eq. (15); \( \hat{1} \) is the \( 2 \times 2 \) identity matrix and the trace \( \text{Tr}_s \) is taken over spin states. Multiplying Eq. (5) sequentially by \( s_x, s_y \) and \( s_z \) from the right and taking \( \text{Tr}_s \), one can obtain kinetic equations for \( S = \{X, Y, Z\} \), which define the spin vector evolution on the Bloch sphere in the most general form

\[
\frac{d}{dt} S = \mathcal{H} \times S - \Gamma (S - S_0). \tag{16}
\]
In this equation, the 3×3 matrix \( \Gamma \) represents a rather complicated expression, \( \mathcal{J} = \{0, 0, \omega\} \), \( S_0 = \{0, 0, Z_0\} \), and \( Z_0 = -\frac{1}{2} \tanh \frac{\beta \omega}{2} \) is a static spin polarization due to the spin splitting \( \omega \).

The matrix \( \Gamma \) can be simplified notably if one takes into account the symmetry properties of the Fourier transformation of correlation functions [Eq. (13)]:

\[
\langle \Omega_\mu(\tau), \Omega_\nu \rangle_\omega = e^{\beta \omega} \langle \Omega_\nu(\tau), \Omega_\mu \rangle_\omega = e^{\beta \omega} \langle \Omega_\nu, \Omega_\mu(\tau) \rangle_\omega.
\]

At the same time, only the symmetrical part \( \Gamma = ||(\Gamma_{\mu \nu} + \Gamma_{\nu \mu})/2|| \) is relevant to spin relaxation. This is because the asymmetrical part \( \Delta \Gamma \) \( (\Gamma = \Gamma + \Delta \Gamma) \) contributes to Eq. (16) as an additional effective magnetic field that cannot reduce the length of vector \( \Delta S = S - S_0 \):

\[
d\Delta S/dt = \frac{\Delta S}{dt} = \frac{\Delta S}{dt} \cdot [(\mathcal{J} - \mathcal{J}) \times \Delta S] - \frac{\Delta S}{dt} \cdot \Gamma \Delta S = -\frac{\Delta S}{dt} \Gamma \Delta S; \Delta \omega_x = -\Delta \gamma_{yz}, \text{ etc.}
\]

Subsequently, the contribution of \( \Delta \Gamma \) to spin relaxation is ignored. We also neglect in the calculation the second term on the right-hand side of Eq. (14) (proportional to \( iP \)) since it plays an insignificant role as well.\(^{65, 73, 74}\)

The final result of the symbolic computation for the matrix \( \Gamma \) reads

\[
\Gamma = \pi \begin{pmatrix}
\gamma_{zz}^0 + n(\gamma_{yy} + \gamma_{xy}) & -n\gamma_{xy} & -\frac{n}{2}(\gamma_{xx} - \gamma_{yz}) - \frac{1}{2}\gamma_{zz}^0 \\
-n\gamma_{xy} & \gamma_{zz}^0 + n(\gamma_{xx} + \gamma_{xy}) & -\frac{n}{2}(\gamma_{yz} - \gamma_{z}x) - \frac{1}{2}\gamma_{yz}^0 \\
-\frac{n}{2}(\gamma_{xx} - \gamma_{yz}) - \frac{1}{2}\gamma_{zz}^0 & -\frac{n}{2}(\gamma_{yz} - \gamma_{z}x) - \frac{1}{2}\gamma_{yz}^0 & n(\gamma_{xx} + \gamma_{yy}) + 2n\gamma_{xy}^0
\end{pmatrix},
\]

where \( n = n(\omega) = (1 + e^{-\beta \omega})/2 \), \( \gamma_{\mu \nu} = [\gamma_{\mu \nu}(\omega) + \gamma_{\nu \mu}(\omega)]/2 \), \( \gamma_{\mu \nu} = i[\gamma_{\mu \nu}(\omega) - \gamma_{\nu \mu}(\omega)]/2 \), and \( \gamma_{\mu \nu}^0 = \gamma_{\mu \nu}(0) \).

If the correlation functions possess some supplementary symmetry, Eq. (18) allows further simplification. Assuming \( \gamma_{\mu \nu}(\omega) = \gamma_{\nu \mu}(\omega) \), one can find

\[
\Gamma = \pi \begin{pmatrix}
\gamma_{zz}^0 + n(\gamma_{yy} + \gamma_{xy}) & -n\gamma_{xy} & -\frac{n}{2}\gamma_{xx} - \frac{1}{2}\gamma_{zz}^0 \\
-n\gamma_{xy} & \gamma_{zz}^0 + n(\gamma_{xx} + \gamma_{xy}) & -\frac{n}{2}\gamma_{yz} - \frac{1}{2}\gamma_{yz}^0 \\
-\frac{n}{2}\gamma_{xx} - \frac{1}{2}\gamma_{zz}^0 & -\frac{n}{2}\gamma_{yz} - \frac{1}{2}\gamma_{yz}^0 & n(\gamma_{xx} + \gamma_{yy}) + 2n\gamma_{xy}^0
\end{pmatrix},
\]

which readily transforms to the relaxation matrix obtained in Ref. \(^{76}\) in the limit \( \omega \rightarrow 0 \).

In other case with \( \gamma_{\mu \nu}(\omega) = -\gamma_{\nu \mu}(\omega) \) for \( \mu \neq \nu \), Eq. (18) can be put in a simpler form

\[
\Gamma = \pi \begin{pmatrix}
\gamma_{zz}^0 + n(\gamma_{yy} + \gamma_{xy}) & 0 & \frac{n}{2}\gamma_{yz} \\
0 & \gamma_{zz}^0 + n(\gamma_{xx} + \gamma_{xy}) & \frac{n}{2}\gamma_{xx} \\
\frac{n}{2}\gamma_{yz} & \frac{n}{2}\gamma_{xz} & n(\gamma_{xx} + \gamma_{yy}) + 2n\gamma_{xy}^0
\end{pmatrix}.
\]
Note that Eqs. (16) and (18) are, generally speaking, derived for the case of anisotropic medium and arbitrary electron spin splitting \( \omega \). Equation (18) can be reduced to the common Bloch equations if one puts 
\[
T^{-1}_{1} = \pi n \left[ \gamma_{xx}(\omega) + \gamma_{yy}(\omega) \right], \\
T^{-1}_{2} = \pi \gamma_{zz}(0) + T^{-1}_{1}/2
\]
and omits the off-diagonal components of \( \Gamma \). However this reduction does not always happen, as it was shown in Ref. 64. The spectral properties of Eq. (17) with respect to the definition of \( n(\omega) \) reveal that the relaxation matrix given in Eq. (18) is an even function of electron spin splitting (i.e., \( \Gamma(\omega) = \Gamma(-\omega) \)), as it also follows from the time inversion symmetry. The kinetic equations described above do not assume any specific properties of the heat bath \( (H_L) \) nor the interaction mechanism \( (H_{SL}) \), and form the backbone of our theoretical approach. In the following, we show how they can be applied to the problem of QD spin decoherence, particularly the elastic processes described by \( \gamma_{zz}(0) \) in Eq. (18) corresponding to ”anharmonic vibration” and ”local field steps” in Table 1.

IV. ANHARMONIC VIBRATION

There are several manifestations of the elastic relaxation process termed ”anharmonic vibration”. To avoid the difficulties in determining adequate mechanisms of spin-phonon interaction and their interference, we consider this problem semi-phenomenologically by applying the parameters of the spin-deformation Hamiltonian, which can be extracted from the electron paramagnetic resonance (EPR) experiments on shallow donors in strained crystals or known dependencies of electron \( g \)-tensor shift in a low-symmetry potential. These parameters are mainly associated with a direct modulation of spin-orbit interaction as discussed above (see Table II). We also take into account the ”HFI modulation” mechanism when the spin-deformation Hamiltonian is not applicable. A brief preliminary account of this process was given earlier.

A. Qualitative analysis

Our analysis is based on the representation of spin-phonon interaction in terms of fluctuating effective magnetic field \( \Omega \) [in units of energy, see Eq. (15)] acting on the electron spin \( s \). This field is assumed to be composed of additive contributions \( \Omega_p \) from each phonon \( p = \{q, \kappa\} \) with a wave vector \( q \) and polarization \( \kappa \), i.e., \( \Omega = \Sigma_p \Omega_p \). For the moment, let
us focus on a single phonon contribution. Then, in the frame of reference rotating with a Zeeman frequency, the electron spin performs precession around the small $\Omega_p$, which oscillates with the phonon frequency $\omega_p$. No alteration in the electron spin phase occurs due to such a harmonic perturbation with a possible exception of spin phase shift $\Delta\phi_0$ acquired at the initial period of interaction $0 < t < 2\pi/\omega_p$ due to a random phonon phase $\theta_p$.28

A different situation can be realized when the phonon harmonic oscillation is interrupted and resumes at a series of instant times $t_{1i}$ and $t_{2i}$ ($i = 1, 2, ...$), respectively, as shown in Fig. 3. The reason of such phonon fluctuations can be lattice anharmonicity, phonon scattering at the impurities or lattice defects, etc. These irregular phonon perturbations affect the electron spin precession resulting in the phase shift $\Delta\phi_i$ at each interval of time $t_{2i} - t_{1i}$. Subsequently, the net effect of spin phase change $\phi_p(t)$ due to a phonon mode $p$ can be expressed as $\phi_p(t) = \Sigma_i \Delta\phi_i$, ($t_{2i} < t$).

Note that for a large number of small changes $\Delta\phi_i$, their total effect can be described by a diffusion equation. Its solution leads to an exponential decay of electron spin phase with a relaxation rate $T_p^{-1} = \frac{1}{2} \langle \Delta\phi_i^2 \rangle \tau_p^{-1}$, where $\tau_p$ is the mean time between sequential instants $t_{1i}$ (or $t_{2i}$).24 To estimate the spin phase change $\Delta\phi_i$ caused by a phonon perturbation during $t_{2i} - t_{1i}$, it is helpful to recognize that the oscillator does not change the spin phase during its full period $\Delta t_p = 2\pi/\omega_p$ as well as for any $n$ integer periods $n2\pi/\omega_p$. Hence, $\Delta\phi_i$ can be approximated as a spin rotation $\Omega_p \Delta t_p$ in an effective field $\Omega_p$, independently of the duration $t_{2i} - t_{1i}$. With the mean value $\langle \Delta\phi_i \rangle$ on the order of $\Omega_p/\omega_p$, one can expect $T_p^{-1} \sim \tau_p^{-1}\Omega_p^2/\omega_p^2$ for the phonon mode $p$ and $T_2^{-1} \sim \Sigma_p N_p \tau_p^{-1}\Omega_p^2/\omega_p^2$ when the contributions of all phonons (with the population factor $N_p$) are taken into account.

**B. Theoretical model**

The qualitative consideration provided above shows that electron spin phase relaxation can be strongly affected by the phonon phase damping of any origin such as phonon decay. Since this mechanism does not involve energy exchange, only the longitudinal (with respect to the external magnetic field $B$) component $\Omega_z$ of the effective fluctuating field is relevant to our case. Since the random single spin fluctuation associated with each phonon scattering is expected to be very small, we investigate the total result of these small fluctuations averaged over the time scale $\Delta t$ ($\tau_c \ll \Delta t \ll T_2$). Thus, the time evolution of the mean spin...
value $s$ can be described in terms of the quantum kinetic equation discussed in the previous section [Eqs. (16) and (19)]. The analysis of $T_{2,0}^{-1} = \pi \gamma^0_{zz}$ is the focus of the investigation as it determines the transversal relaxation stemming from the elastic processes.

The $z$ component of the fluctuating field operator due to the phonon decay can be expressed in a form linear in the creation and annihilation operators $a_p^\dagger$ and $a_p$ of the phonon mode $p = \{q, \kappa\}$ ($q$ and $\kappa$ are the phonon wave vector and its polarization, $-p \equiv \{-q, \kappa\}$); i.e.,

$$\Omega_z = \sum_p V_p^z Q_p; \quad (21)$$

$$Q_p = a_p^\dagger - a_{-p}; \quad (22)$$

with a matrix element $V^z_p$ of the spin-phonon interaction, whose specific form will be discussed later. Skipping the detailed elaboration of the routine calculation of the phonon correlation functions, the final result for the spin relaxation rate associated with the elastic "anharmonic vibration" process is obtained as

$$T_{2,0}^{-1} = \sum_p |V^z_p|^2 (2N_p + 1) \frac{\tau_p^{-1}}{\omega_p^2}; \quad (23)$$

where $\tau_p$ is the phonon relaxation time (see Ref. 81 and the references therein) and the phonon population factor $N_p$ is given as $[\exp (\hbar \omega_p / k_B T) - 1]^{-1}$. This expression is in accordance with the qualitative analysis discussed in previous section with the exception of the second term in the factor $2N_p + 1$ (i.e., "1") that is attributed to the zero-point oscillations. It reflects the quantum effects, which could not be described in terms of classical approach but may be important at low temperatures.

To evaluate Eq. (23), one needs the knowledge of the relevant phonon mode $p$ (such as the dispersion relation $\omega_p$ and the relaxation time $\tau_p$) as well as its interaction with electron spins. Taking into account the conditions frequently adopted in QC, we assume that the radius of the electronic state $a_0$ (i.e., the QD size) is much larger than the lattice constant. This restricts the summation in Eq. (23) to long wavelength phonons as the matrix element $V^z_p$ falls off quickly for $q > \pi/a_0$. Accordingly, $\tau_p = \tau_p(q)$, which is a complex function of the temperature and phonon frequencies [see Eq. (6.2) in Ref. 82], can be considered in the long wavelength limit. Of a number of potential phonon decay mechanisms, only one term originating from the boundary scattering survives at sufficiently low temperatures $T \lesssim T_{bs}$.
\(T_{bs} \approx 10\ \text{K in the case of Ref. \[\text{82}\]}.\) Since this mechanism is insensitive to the temperature, it is adequate to assume a constant phonon relaxation time \(\tau_p \simeq \tau_{ph}\). The simple relaxation time approximation circumvents the difficult problem of the zone-edge phonons and the dependence \(\tau_p = \tau_\xi(q)\), which could be very specific for each particular sample.

C. Effect of acoustic modes

To evaluate Eq. (23) (i.e., \(|V_p^z|^2\)) for the acoustic phonons, we consider a phenomenological Hamiltonian of the spin-lattice interaction. It can be expressed in terms only of symmetry and does not depend on the specific mechanism:

\[
H_{SL}^{ac} = \sum_{ijkl} A_{ijkl} \mu_B B_i s_j \overline{u}_{kl} + \sum_{ijklm} A'_{ijklm} \mu_B B_i s_j \overline{w}_{klm}. \quad (24)
\]

Here \(\overline{u}_{kl}\) is the strain tensor \(u_{kl} = (\partial \Delta x_k / \partial x_l + \partial \Delta x_l / \partial x_k)/2\) averaged over the electron ground state \(|g\) (i.e., \(\overline{u}_{kl} = \langle g | u_{kl} | g \rangle\)), subscripts \(ijkl\) refer to the crystallographic axes, and the non-zero components of the tensor \(A_{ijkl}\) can be found from the symmetry of the QD.\([17,63]\)

The second term in Eq. (24) takes into account the effect of strain gradient \(w_{klm} = \partial u_{kl} / \partial x_m\), which will be neglected from consideration since it can be irrelevant to the elastic spin-phonon processes.\([17]\)

As a specific problem, we consider a lateral [001] QD with a \(z\)-directed magnetic field and a localized electron with the axial symmetry with respect to the \(z\) axis. This case can be applied to high-symmetry systems corresponding, for example, to shallow donors, etc. Now Eq. (24) can be reduced to the form \(H_{SL}^{ac} = \Omega s\) with \(\Omega_z = [(A_{33} - A_{31}) \overline{u}_{zz} + A_{31} \overline{\Delta}] \mu B\) and \(\Omega_x = \Omega_y = 0.\text{[17]}\) \(\Delta\) denotes the dilatation \(\Delta = \Sigma_i u_{ii}\) and the Voigt notation is adopted \((A_{33} = A_{zzzz}, A_{31} = A_{zzxx}, A_{66} = A_{xyxy})\). Then, as \(\Omega_z\) is expressed in terms of the creation and annihilation operators [Eq. (21)], the matrix element \(V_p^z\) is given as

\[
V_p^z = i \left(\frac{\hbar}{2 \rho V_0 \omega_p}\right)^{1/2} [(A_{33} - A_{31}) e_p^z q_z + \delta_{\xi, L} A_{31} q] \Phi(q) \mu_B B. \quad (25)
\]

Here \(\rho\) is the mass density of the crystal, \(q = \sqrt{q_x^2 + q_y^2 + q_z^2}\), \(V_0\) is the volume of the sample structure, \(e^p\) the polarization vector of the phonon \(p\), and \(\Phi(q) = \langle g | e^{i\mathbf{q} \cdot \mathbf{r}} | g \rangle\).

The spin-lattice relaxation rate in Eq. (23) can be calculated by treating the acoustic phonon modes based on the isotropic elastic continuum model with the longitudinal and transverse sound velocities \(v_L\) and \(v_T\). Then by taking advantage of the phonon relaxation
time approximation (in the long wavelength limit and at sufficiently low temperatures) as well as the axial symmetry for the localized electron [i.e., $\Phi (q) = \Phi (x, z)$ with $x = qa_0/2$, $z = qz/q$], one can obtain

$$T_{2,0}^{-1} = \tau_{ph}^{-1} \xi (B) \int_0^\infty x \sum_{\kappa=L,T} \frac{v_3^2}{\nu_{\kappa}^3} \coth \left( \frac{T_{eff}^\kappa x}{T} \right) F_{\kappa} (x) \, dx,$$

(26)

$$\xi (B) = \frac{(A_{33} - A_{31})^2 \mu_B^2 B^2}{2\pi^2 \hbar \nu_T^3 a_0^2},$$

$$F_{\kappa} (x) = \int_{-1}^1 P_{\kappa} (z) \left( z^2 + \zeta \right)^2 \Phi^2 (x, z) \, dz,$$

where we introduce $1/a_0^2$ and $a_0^2$ into $\xi (B)$ and integrant for convenience. In this equation, $P_L(z) = (z^2 + \zeta)^2$; $T_{L} (z) = z^2(1-z^2)$, $\tau_{ph}^{-1}$ is the average phonon relaxation rate as mentioned above, $T_{eff}^\kappa = \hbar \nu_{\kappa}/k_B a_0$ is the effective temperature, and $\zeta \equiv A_{31}/(A_{33} - A_{31}) = -1/3$ if one assumes that the strain induced part of the effective $g$-tensor $\tilde{g}_{ij} = \Sigma_{k,l} A_{ijkl} u_{kl}$ is characterized by zero trace, i.e., $A_{33} + 2A_{31} = 0$.

1. Spin decoherence of shallow donor

Let us evaluate spin relaxation of a shallow donor electron with an effective Bohr radius $a_B = a_0$ and $\Phi (x, z) = (1 + x^2)^{-2}$. The last factor determines the relevant phonons, which must be long wavelength modes with $q \lesssim 1/a_B$. Then, the integral in Eq. (26) has a simple analytical approximation,

$$T_{2,0}^{-1} = \frac{2\xi (B) \tau_{ph}^{-1}}{45} \left( \sqrt{1 + \frac{T^2}{T_{T}^2}} + \frac{2v_3^2}{3v_L^3} \sqrt{1 + \frac{T^2}{T_L^2}} \right),$$

(27)

where $T_L(T) = (16/15\pi) T_{eff}^L(T)$; this result is for the case of $B \parallel [001]$.

As an example, we consider a Phosphorus shallow donor in Si with $a_B = 1.8$ nm. The phonon relaxation time can be extracted from the low temperature measurements of Si thermal resistivity in terms of the theory developed in Refs. 84 and 81 ($\tau_{ph} = 2.4 \times 10^{-8}$ s). The spin-phonon coupling constants were estimated in the works of Refs. 85 and 86. However, we believe that direct determination of coupling constants by means of EPR measurements of Si:P under an applied stress gives more reliable data. A corresponding experiment was performed in Ref. 87, where the constant $A_{66} = 0.44$ was found. Similarly,
our estimation obtains $A_{33} = 0.31$ and $A_{31} = -0.155$ that gives $T_{2,0}^{-1} = 1.3 \times 10^{-4}$ s$^{-1}$ at the magnetic field of 1 T and low temperatures $T \ll T_{L(T)} \simeq 10$ K. In another important case of a Si shallow donor in Al$_{0.4}$Ga$_{0.6}$As, the data on EPR under a uniaxial stress provide rather strong spin-phonon constants of $A_{33} = 19.6$ and $A_{31} = -9.8$. This gives the estimation $T_{2,0}^{-1} = 6.1 \times 10^{-2}$ s$^{-1}$ and $6.1 \times 10^{-4}$ s$^{-1}$ for the magnetic fields of 1 T and 0.1 T, respectively, at $T = 4$ K under the assumption that phonon lifetimes are identical in these crystals.

2. Spin decoherence in a QD

Corresponding calculations can be performed for an electron localized in a QD of $L_{xy} = 2a_0$ in the lateral width and $L_z = \epsilon L_{xy}$ in the thickness. Under the condition $\epsilon \lesssim 0.1$, an approximate formula takes the form

$$T_{2,0}^{-1} = \xi(B)\tau_{ph}^{-1} \left( \sum_{i=L,T} b_i \sqrt{v_i^2 + d_i^2 T_i^2 / T_i^2} \right), \quad (28)$$

where the fitting coefficients are $b_T = 1$, $b_L = v_T^3 / v_L^3$, $v_T = 0.33 - 1.27 \epsilon^2$, $d_T = 0.35 - 0.395 \epsilon^2$, $v_L = 0.97 - 28.5 \epsilon^2$, and $d_L = 0.40 - 3.76 \epsilon^2$.

To examine the importance of spin phase relaxation caused by the phonon decay, the result of Eq. (28) is compared with the admixture spin-flip process in a GaAs QD with $L_z = 3$ nm and $L_{xy} = 25$ nm, assuming $\tau_{ph} = 2.4 \times 10^{-8}$ s and $A_{33} = 19.6$. For the relatively strong magnetic field of 1 T and $T = 4$ K, the present elastic process and the spin-flip give $T_{2,0}^{-1} \approx 0.1$ s$^{-1}$ and $\frac{1}{2} T_1^{-1} = T_{2,0}^{-1} = 10$ s$^{-1}$, respectively, while for $B = 0.1$ T both predict almost the same rate of $\approx 10^{-3}$ s$^{-1}$. At lower magnetic fields, the elastic spin relaxation prevails. The Si QD gives a qualitatively similar (but smaller) result; its numerical estimation will be discussed in Sec. VII.

D. Role of optical phonons

The optical phonons possess high energies compared to the typical energy scale (such as the thermal energy $k_B T$) and, thus, are generally ignored in the problem of QC based on QD electron spins. However, this is not the case in the elastic process under consideration since the uncontrolled variation of spin phase can happen without the presence of thermal phonons as shown in Eq. (23) at $N_p \to 0$. Zero-point optical vibrations, which survive even
at very low temperatures $T \ll \hbar \omega_{op}$ ($\omega_{op}$ is the optical phonon frequency), contribute to spin decoherence as it happens with acoustic modes in the case $T \ll T_{L,T}$ [Eq. (28)]. The potential significance of the optical modes stems from their relatively large contributions at small wave vectors $q$ ($|V_z^p|^2 \to \text{constant}$ as $q \to 0$) as well as the substantially shorter phonon lifetime. In contrast, $|V_z^p|^2 \to q$ for acoustic phonons in the long wavelength limit.

For a detailed analysis, consider a QD made of a polar crystal. The longitudinal-optical phonons induce the electron potential

$$\phi_{op} = \sqrt{\frac{2\pi \hbar \omega_{op}}{V_0 \kappa}} \sum_q \frac{\exp(iqr)}{q} (a_q^\dagger - a_{-q}),$$

(29)

where $1/\tilde{\kappa} = 1/\kappa_\infty - 1/\kappa_0$; $\kappa_\infty$ and $\kappa_0$ are the corresponding dielectric constants. Assume that the QD possesses an asymmetrical potential in the growth direction $z$ due to an effective electric field $F$ imposed along this axis (e.g., the gate bias). Since $F$ influences the electron $g$-factor, its superposition with $\phi_{op}$ results in a spin-phonon interaction $H_{SL}^{op} = \mu_B B (dg/dF) d\phi_{op}/dz$ that is linear in the phonon operator. After substitution of Eq. (29), this expression reduces to the form of Eq. (21) with

$$|V_z^p|^2 = \left(\frac{\mu_B B}{dg/dF}\right)^2 \frac{2\pi \hbar \omega_{op} q^2}{q^2} \Phi^2(q).$$

(30)

Finally, the spin relaxation rate is obtained as:

$$T_{2,0}^{-1} = \tau_{op}^{-1} \xi_{op}(B),$$

(31)

$$\xi_{op}(B) = \frac{2}{\sqrt{\pi \tilde{\kappa} \hbar \omega_{op} V_{QD}}} \left(\frac{dg}{df} \mu_B B\right)^2,$$

(32)

where $V_{QD}$ is the QD volume ($= L_{xy}^2 L_z$).

We perform numerical estimation for a GaAs QD of $L_{xy} = 50 \text{ nm}$ and $L_w = 5 \text{ nm}$. Here, evaluating the QD potential asymmetry a propri provides the most significant challenge. Following the calculations performed for Ga$_{0.47}$In$_{0.53}$As/InP quantum wells, a typical value for $\frac{dg}{df}$ seems to be approximately 0.085 nm/mV at $F > 10 \text{ mV/nm}$ with $B$ directed along the growth axis, while for the perpendicular direction $\frac{dg}{df} \leq 0.035 \text{ nm/mV}$. As the effect of optical phonons quenches in the case of a symmetrical potential ($\frac{dg}{df} = 0$ with $F = 0$), only the upper limit of this contribution can be estimated, i.e., $\xi_{op}(1T) \leq 7 \times 10^{-6}$. Taking into account that $\tau_{op}^{-1} \simeq 10^{11} \text{ s}^{-1}$ one can find $T_{2,0}^{-1} \leq C_{op} B^2$ where $C_{op} = 7 \times 10^5 \text{ s}^{-1} \text{T}^{-2}$. This estimation shows that the transversal relaxation via zero-point optical oscillations can be a very effective decoherence mechanism in asymmetrical structures.
E. Effects of hyperfine interaction and two-phonon processes

Let us consider the elastic one-phonon process associated with the modulation of hyperfine interaction as well as the two-phonon process. Since these processes result in sufficiently slow/long spin relaxation, we shall restrict our consideration to a brief discussion.

The Hamiltonian of hyperfine interaction with the nuclear spin $I_j$ located at site $j$ with the position $\mathbf{r}_j$ takes the form

$$H_{hf} = a_{hf} \sum_j |\psi_g(\mathbf{r}_j)|^2 I_j s, \quad (33)$$

where $\psi_g(\mathbf{r}) = |g\rangle$ is an electron wave function. The spin-phonon interaction due to small oscillations at $\mathbf{r}_j$ can be expressed in terms of the dilatation operator $\Delta = \langle g | \Delta | g \rangle$ if the mean internuclear distance ($\approx n_I^{-1/3}$, $n_I$ is the nuclear spin concentration) is shorter than the wavelength of relevant phonons; i.e., $H_{hf}^{SL} = \Omega s$, where $\Omega = \hat{n} a_{hf} \Delta \sqrt{I(I+1)n_I}/V_{QD}^{hf}$. Here, the unit vector $\hat{n}$ is directed along the effective nuclear field defined by Eq. (33) and $V_{QD}^{hf} = \left(\int |\psi(\mathbf{r})|^4 d^3\mathbf{r}\right)^{-1}$. Calculation of the phase relaxation rate for the case of a shallow donor results in the expression, which is similar to Eq. (27),

$$T_{2,0}^{-1} = \frac{\xi_{hf} \tau_{ph}}{3} \sqrt{1 + \frac{T^2}{T_L^2}}, \quad (34)$$

$$\xi_{hf} = \frac{I(I+1)n_I a_{hf}^2}{6\pi^2 \hbar \rho V_{QD}^{hf} c_0^2 d_0^2}, \quad (35)$$

Similarly, one can find the approximate rate for an electron localized in a QD through an analogy with Eq. (28),

$$T_{2,0}^{-1} = \xi_{hf} \tau_{ph}^{-1} \sqrt{c_{hf}^2 + d_{hf}^2 \frac{T^2}{T_L^2}}, \quad (36)$$

where $c_{hf} = 3.7 - 68\epsilon^2$, $d_{hf} = 2.7 - 9.8\epsilon^2$, and $\epsilon \lesssim 0.1$. Numerical estimations provided for a donor in Si and GaAs in terms of Eq. (34) indicate inefficiency of this mechanism with a very long relaxation time (about $10^{14}$ s and $10^8$ s, respectively). Hence, this mechanism can be neglected in most cases.

Along with the Hamiltonian given in Eq. (24), the spin-two-phonon interaction $H_{SL}^{(2)} = \sum D_{ijklmn} \mu_B B_i s_j u_{kl} u_{mn}$ can also contribute to spin decoherence ($D_{ijklmn}$ is the spin-two-phonon coupling constants). Now the fluctuating effective field takes the form $\Omega_\alpha = \sum_{p,p'} W_{\alpha p,pp'}^{p,p'} Q_p Q_{p'}$, where $W_{\alpha p,pp'}^{p,p'}$ are the matrix elements of $H_{SL}^{(2)}$. Hence the Fourier image
of the correlation function $\gamma_{\mu \nu}(\omega)$ [Eq. (13)] is expressed in terms of phonon correlation functions $\langle (Q_{p_1} Q_{p_2}) (\tau) Q_{p_3} Q_{p_4} \rangle_\omega$. Its calculation performed in a harmonic approximation leads to a simple approximation for the spin phase relaxation rate

$$T_{2,0}^{-1} = \frac{\mu_B^2 B^2 D^2}{21 \rho^2 c_T^2} \left( \frac{k_B T}{\hbar c_T} \right)^7 .$$

(37)

The parameter $D$ can be estimated as $D = 3(g - 2)C^2/E_g^2$ ($g$, $C$, and $E_g$ are the electron $g$ factor, deformation potential and energy gap). Numerical evaluation of Eq. (37) at low temperatures ($T = 4$ K) predicts a long relaxation time. In the case of GaAs and $B = 1$ T, one can find $T_{2,0} \approx 3 \times 10^5$ s that is too long to be of any experimental or practical interest.

V. PRECESSION FLUCTUATIONS

This section analyzes the elastic process listed as "Local field steps" in Table I. The specific spin-phonon interactions under consideration are the indirect mechanisms resulting from "g-tensor fluctuation" and "hyperfine field fluctuation" (see Table II) via the stochastic transitions between different electronic states.

A. Qualitative consideration

Consider an electron spin $s$ under the influence of a magnetic field directed along the $z$ axis whose strength fluctuates in a step-wise fashion in time (see Fig. 4). In this case, the projection of electron spin on the $z$ axis $s_z$ is conserved and no longitudinal relaxation occurs. Nevertheless, the phase of electron spin will change randomly with the Zeeman frequency fluctuation $\delta \Omega$ resulting in a decoherence rate of $T_{2,0}^{-1} \sim \delta \Omega^2 \tau_c$. Here, $\tau_c$ is the correlation time of the fluctuation. As discussed earlier, the origin of $\delta \Omega$ can be the phonon-mediated stochastic transitions between different orbital states. Since the spin splitting characteristics vary with the orbital states, electrons experience uncontrollable changes in the precession frequency (thus, the phase) while undergoing spin-independent phonon scattering between them. In the case of a QD, this process may play an important role owing to the shallow energy levels and the dependence of $g$-tensor or hyperfine constant on the electronic orbital states.
B. Theoretical model

We begin the quantitative analysis by defining the Hamiltonian \( H \) over the basis functions consisting of a few lowest electronic states \( |n\rangle \), which are involved due to the phonon-assisted transitions. We also consider that the single-electron problem in the QD gives a doubly degenerate energy spectrum \( E_n \) with the eigenstates \( |n\rangle \) in the absence of the Zeeman energy and the hyperfine interaction. When a magnetic field is applied, it is conveniently assumed that the differences in the spin splitting of the electronic states \( |n\rangle \) and \( |n'\rangle \) are small with respect to the energy intervals \( |E_n - E_{n'}| \). The specific nature and type of the QD is unimportant for the analysis. With these conditions, the total Hamiltonian can be expressed as

\[
H = H_s + H_e + H_{ph} + H_{e-ph}. \tag{38}
\]

The first term \( H_s \) is the spin (or pseudospin) energy Hamiltonian which can be reduced to the form \( H_s = \hat{\Omega}s \) in the most general case with \( \hat{\Omega}_z = \langle \uparrow |H_s| \uparrow \rangle - \langle \downarrow |H_s| \downarrow \rangle \) as discussed in Eq. (2). Its projection on the lowest electronic states \( |n\rangle \) reads (see, for comparison, Refs. 86 and 85)

\[
\hat{\Omega} = \sum_{n,n'} |n\rangle \Omega^{n,n'} \langle n' |, \tag{39}
\]

where \( \Omega^{n,n'} \) are the matrix elements of the effective field (in units of energy) taken between the \( |n\rangle \) and \( |n'\rangle \) states. The spin-independent (i.e., orbital) electron energies describe the Hamiltonian \( H_e \)

\[
H_e = \sum_n E_n |n\rangle \langle n|. \tag{40}
\]

The Hamiltonians of the lattice and electron-phonon interactions have the usual form

\[
H_{ph} = \sum_p \omega_p (a_p^+ a_p + \frac{1}{2}); \tag{41}
\]

\[
H_{e-ph} = \sum_{p,n,n'} B_{p,n,n'}^n |n\rangle \langle n'| (a_p^+ + a_{-p}). \tag{42}
\]

Here, \( B_{k,k'}^p \) is the matrix element of the electron-phonon interaction that depends on the material parameters and the geometry of the QD. The last three terms of Eq. (38) constitute the Hamiltonian of the dissipative sub-system \( H_L \) (= \( H_e + H_{ph} + H_{e-ph} \)) following the notation of Eq. (2).
Thus, the problem of spin relaxation is reduced to calculating the correlation functions of the effective field operator with the Hamiltonian $H_L$ of the dissipative subsystem. Clearly, the derivation depends strongly on the specific form of $H_L$, the energy spectrum, and the quantity of electron states considered. Keeping this context in mind, we consider the simpler problem of electron fluctuations between only two discrete states $|n\rangle = |g\rangle$ or $|e\rangle$ that are the ground and the first excited electronic energy states (with an energy separation of $\delta_0$). The corresponding Zeeman frequencies are denoted $\Omega_z^{(g)}$ [ = $\Omega_z^{g,g}$ in Eq. (39)] and $\Omega_z^{(e)}$ ( = $\Omega_z^{e,e}$), respectively. Such a simplification allows us to easily perform all the necessary calculations in an analytical form. In addition, most of the important physics of the mechanism under consideration can be obtained in the framework of this two-level model.

Hereinafter, it is convenient to introduce Pauli matrices $\sigma_1$, $\sigma_2$, $\sigma_3$ on the basis $|e\rangle$, $|g\rangle$, where according to the definition, $\sigma_1$, $\sigma_2$, $\sigma_3$ are invariant with respect to the coordinate system rotation in contrast to actual spin matrices $\mathbf{s}$. Then, the Hamiltonian of the dissipative subsystem takes the form

$$H_L = H_{ph} + \frac{1}{2}\delta_0\sigma_3 + \Sigma_p B_p \sigma_1(a^+_p + a^-_p).$$

(43)

The electron spin Hamiltonian $H_s$ can now be split into the steady part $H_S$ and fluctuating component $H_{SL}$ with

$$H_S = \frac{1}{2}(\Omega^e + \Omega^g)\mathbf{s} \hat{1}$$

(44)

and

$$H_{SL} = \frac{1}{2}(\Omega^{(e)} - \Omega^{(g)})\mathbf{s} \sigma_3,$$

(45)

where $\hat{1}$ represents the unity matrix in the basis $|e\rangle$, $|g\rangle$. According to Eq. (43), we must find

$$V = H_{SL} - \langle H_{SL} \rangle = \mathbf{\Omega} \mathbf{s}$$

from Eq. (45) that determines $\mathbf{\Omega} = \frac{1}{2}(\Omega^{(e)} - \Omega^{(g)})(\sigma_3 - \langle \sigma_3 \rangle)$ with $\langle \sigma_3 \rangle = -\tanh(\delta_0/2k_B T)$. The correlation functions in Eq. (13) now takes the form

$$\gamma_{\mu \nu}(\omega) = \frac{1}{4} (\Omega^{(e)}_{\mu} - \Omega^{(g)}_{\mu})(\Omega^{(e)}_{\nu} - \Omega^{(g)}_{\nu})J_\omega(T);$$

(46)

$$J_\omega(T) = \langle [\sigma_3(\tau) - \langle \sigma_3 \rangle][\sigma_3(\tau) - \langle \sigma_3 \rangle]\rangle_\omega.$$  

(47)

The function $J_\omega(T)$ can be obtained by using the double-time Green’s function $G(t, t') = \langle [\sigma_3(t) ; \sigma_3(t')]\rangle$ with $H_L$. The final expression takes the following form:

$$J_\omega(T) = \frac{1 - \langle \sigma_3 \rangle^2}{\pi n(\omega)} \frac{\tau_c}{\omega^2 \tau_c^2 + 1};$$

(48)

$$\tau_c^{-1} = 2\pi \sum_p |B_p|^2 \langle Q_p(\tau)Q_{-p}\rangle_{\delta},$$

(49)
where the difference between \( \delta' = \delta_0 - \frac{1}{2}(\Omega^{(e)} - \Omega^{(g)}) \) and \( \delta_0 \) will be ignored in the subsequent consideration. In the harmonic approximation, one can find

\[
\langle Q_p(\tau)Q_{-p}\rangle' = (2N_p + 1)\delta (\omega_p - \delta_0),
\]

with the phonon population factor \( N_p = \langle a^+_p a_p \rangle \) for mode \( p \). The parameter \( \tau_c \) has the simple physical meaning of the correlation time caused by phonon-assisted transitions between the \( |g\rangle \) and \( |e\rangle \) states.

**C. Temperature dependence of phase relaxation**

Actually, Eqs. (46)-(50) describe the problem under consideration in a very general form. As a result, an analysis on the temperature dependence can be provided even before the details of the phonon-mediated mechanism that induces the effective field fluctuation are determined. Specifically, the correlation time \( \tau_c \) given in Eq. (49) can be written as

\[
\tau_c \tanh \left( \frac{\delta_0}{2k_B T} \right)
\]

with the aid of Eq. (50), where \( \tau_\delta = \left[ 2\pi \sum_p |B_p|^2 \delta (\omega_p - \delta_0) \right]^{-1} \) is the lifetime of the excited electron state with respect to the transition to the ground state through phonon emission in the limit \( T \to 0 \). Thus, for the \( \omega = 0 \) component (e.g., \( \gamma_{zz}^0 \)), the temperature dependence of spin relaxation is reduced to

\[
J_0(T) = \frac{\tau_\delta}{\pi} F \left( \frac{\delta_0}{2k_B T} \right); \quad F(x) = \left( 1 - \tanh^2 x \right) \tanh x.
\]

Figure 5 shows the numerical evaluation of \( J_0(T) \) as a function of \( T \) assuming \( \delta_0 = 1 \) meV. A maximum is observed at a temperature near \( k_B T = \delta_0 \) with distinctive slopes on each side. The left of the peak corresponds to the reduced hopping from the \( |g\rangle \) to \( |e\rangle \) state that decreases the difference \( \Omega^{(g)} - \langle \Omega^{(g)} \rangle \) (or the amplitude of fluctuations). Hence, in the limit \( T \ll \delta_0 \) the effective field fluctuations are frozen and our mechanism becomes ineffective as \( F(\delta_0/2k_B T) \to \exp(-\delta_0/k_B T) \). The slow negative slope on the high temperature side [i.e., \( F(\delta_0/2k_B T) \to \delta_0/2k_B T \)] arises due to the well-known effect of dynamical fluctuation averaging, which becomes more pronounced with an increase in temperature.

**D. Effect of g-factor fluctuation**

The general theory discussed above is applied to a specific mechanism of phase relaxation, which stems from the hopping between the excited and ground states with different \( g \)-factors.
In the most general case, the reason for such a difference is the g-factor dependence on the energy separation between the discrete electronic levels and the nearest spin-orbital split electronic band. For technologically significant III-V compounds, where the interaction with the valence band edge determines the deviation of electron g-factor from the free electron Landé factor \( g_0 \approx 2.0023 \), one can find the amplitude of the fluctuation \( \Delta g = (dg/dE_g)\delta_0 \) with \( dg/dE_g = (g_0 - g)(\Delta_{so} + 2E_g)/E_g(\Delta_{so} + E_g) \), where \( E_g \) is the band gap and \( \Delta_{so} \) the spin-orbital splitting of the valence band; we also assume the inequality \( \delta_0 \ll E_g \). In the case of a Si QD, \( E_g \) is a splitting of the \( \Delta \) point in the Brillouin zone. Then, the final equation for the phonon-assisted rate of phase relaxation caused by the fluctuations of Zeeman splitting is given by

\[
T_{2,Z}^{-1} = \frac{(g_0 - g)^2}{4g^2} \left( \frac{\delta_0(\Delta_{so} + 2E_g)}{E_g(\Delta_{so} + E_g)} \right)^2 \omega_0^2 \tau_\delta \left( \frac{\delta_0}{2k_BT} \right),
\]

where \( \hbar \omega_0 = g\mu_B B \). One can see that our mechanism reveals a quadratic dependence of \( T_{2,Z}^{-1} \) on the applied magnetic field \( B \) in contrast to the \( \sim B^5 \) dependence found in the previous calculations of longitudinal spin-lattice relaxation through the direct (one-phonon) processes.

An estimation of the excited state lifetime \( \tau_\delta \) can be performed in terms of a deformation potential interaction and a model of lateral carrier confinement. The matrix element of the corresponding electron-phonon interaction between the \( |g\rangle \) and \( |e\rangle \) states is provided in Ref. \([17]\) as \( B_q = iC\sqrt{\hbar q/2\rho v_\parallel}V_0J_{osc} \), where \( J_{osc} = J_{osc}(q) \) is a form factor, \( C \) is the deformation potential, and \( \rho, v_\parallel \) and \( V_0 \) are the density, longitudinal sound velocity, and volume of crystal, respectively. A straightforward calculation of inverse lifetime results in the expression

\[
\tau_\delta^{-1} = \frac{C^2q_3^2\alpha}{32\pi^2\hbar \rho v_\parallel^2} \int_0^1 (1 - z^2)e^{-\alpha(1-z^2)}dz,
\]

where \( q_\delta = \delta_0/\hbar v_\parallel \), \( \alpha = \delta_0/2m_e v_\parallel^2 \), and \( m_e \) is the lateral effective mass.

To show the efficiency of the mechanism under consideration, we calculate the relaxation rates in terms of Eqs. (52) and (53) for a GaAs QD with \( \delta_0 = 1 \) meV and the magnetic field \( B = 1 \) T as a function of temperature (Fig. 6, curve 1). A similar calculation is provided for a Si QD with \( g_0 - g_\parallel = 0.0131 \) and \( g_0 - g_\perp = 0.0141 \) (Fig. 6, curve 2). A comparison between the result of Fig. 6 and the \( T_1 \) calculation shows that the phonon-induced g-factor fluctuation via excited states can control the phase relaxation in a Si QD at \( T \gtrsim 2 \) K (i.e., \( T_{2,Z} < T_1 \)) in spite of a small \( g_0 - g \) (see Sec. VII for a detailed quantitative analysis).
in the case of a GaAs QD, this elastic process prevails over the spin-flip transitions at \( T \gtrsim 1 \) K and \( B \leq 1 \) T.

### E. Effect of nuclear field fluctuation

A similar fluctuation arises in the nuclear hyperfine field when the electron undergoes uncontrollable transitions between the orbital \(|e\rangle\) and \(|g\rangle\) states. As shown in Eq. (33), the hyperfine interaction is influenced by the electronic envelope functions evaluated at the location of the nuclear spins. Hence, the difference in \( \Psi_e (r) \) and \( \Psi_g (r) \) leads to a dispersion of the hyperfine field whose mean value is given as \( \delta \Omega_n = a_{hf} \sqrt{\frac{2}{3}} I(I+1) \kappa n_I / V_{QD} \). Here, \( I \) and \( n_I \) are the nuclear spin and its concentration in a QD of volume \( V_{QD} \) as discussed before; the dimensionless parameter \( \kappa = V_{QD} \int (|\Psi_e (r)|^2 - |\Psi_g (r)|^2)^2 d^3r \) is equal to \( 9/16 \pi \) within the approximations of Ref. 17. If we set \( \Omega_z^{(e)} - \Omega_z^{(g)} = \delta \Omega_n \) in Eq. (46), we readily find the following estimation

\[
T_{2,hf}^{-1} = \frac{\kappa}{6} I(I+1) a_{hf}^2 n_I^{-1} V_{QD} \tau_0 F \left( \frac{\delta_0}{2k_B T} \right).
\]

This equation shows the independence of spin relaxation \( (T_{2,hf}) \) on the magnetic field when the process is induced by the hyperfine interaction. An estimation of Eq. (54) (with an appropriate averaging over \( ^{69}\text{Ga}, ^{71}\text{Ga} \) and \( ^{75}\text{As} \)) for a GaAs QD with a typical size of \( L_{xy} = 50 \) nm, \( L_z = 5 \) nm and \( \delta_0 = 1 \) meV gives \( T_{2,hf} = 8 \times 10^{-5} \) s, \( 2 \times 10^{-7} \) s, and \( 1.7 \times 10^{-8} \) s for \( T = 1 \) K, 2 K, and 4 K, respectively. A similar estimation performed for a Si QD with the same dimensions and temperatures results in \( T_{2,hf} = 29 \) s, \( 9 \times 10^{-2} \) s and \( 6.5 \times 10^{-3} \) s, respectively. Clearly, Eq. (54) can provide a dominant contribution for spin relaxation at low fields as it does not depend on the applied magnetic field. In the case of the Si QD, a detailed quantitative analysis is provided in Sec. VII.

### VI. INTERVALLEY TRANSITIONS

In this section, we consider one particular but important case that is applicable to the semiconductors with multiple equivalent energy minima. The basic principle is the same as that discussed in the previous section, i.e., via the phonon-mediated stochastic transitions. The difference is that the present case takes into account the intervalley transitions between
the so-called "valley-split" states. Although normally prohibited, these processes become possible in the presence of point defects. Of the two potential mechanisms, the hyperfine field fluctuation is suppressed due to the equivalence of the involved valleys. However, the "g-tensor fluctuation" mechanism maintains its significance as the degeneracy of these states is lifted (i.e., "valley split" with different energies) in asymmetrical structures.

A. Qualitative consideration

Following the brief discussion given above, this mechanism can occur in semiconductors with multiple equivalent energy minima. As a specific example, a QD grown along the [001] (i.e., z) direction with six equivalent minima near the X point is considered (i.e., Si-like); the cases with different crystallographic symmetries can be analyzed with a similar treatment. Subsequently, the valley-orbital structure of the QD assumes the ground state formed by two equivalent valleys [001] and [001̅] with the wave functions ψ_{001} and ψ_{001̅}. Any center-asymmetrical potential \( V(r) \) (with respect to the z direction) removes the degeneracy over the equivalent valleys with a valley-orbital splitting \( \Delta E_{v-v} \), thus forming the even and odd states \( \psi_\pm = (\psi_{001} \pm \psi_{001̅})/\sqrt{2} \). In the case of a triangular potential \( V(r) \), an estimation predicts the splitting of \( \Delta E_{v-v} = l_{v-v} dV(r)/dz \) with \( l_{v-v} \approx 0.5 \) Å in Si. Clearly, one can see that a moderately asymmetrical confinement potential can result in an energy splitting sufficient large to induce the "g-tensor fluctuation" under an appropriate lifetime \( \tau_δ \). However, the phonon induced transitions between the valley-orbital states with a different parity are normally forbidden or sufficiently suppressed.

This restriction is lifted when one considers a structure composed of solid solutions. For example, the Ge atoms in a Si_{1-x}Ge_x QD with \( x \ll 1 \) (that offers a promising candidate for solid-state QC) can be treated as the point defects in the Si lattice. The phonon scattering with point defects due to the lattice anharmonicity results in the appearance of high-frequency harmonics that can transfer electrons between the equivalent valleys [001] and [001̅] or between the valley-orbital states with a different parity. Figure 7 presents a diagram of the process responsible for these transitions. As schematically illustrated, the third-order anharmonicity splits a phonon resonant with \( \Delta E_{v-v} \) into two high-frequency virtual phonons. One of them is "frozen" due to the static deformation surrounding the defect, while the other phonon induces the electron to undergo an intervalley scattering.
The transfer between the states with an energy separation leads to the fluctuation in the electron precession frequency as described in the previous section.

### B. Analysis of intervalley transition

The estimation of spin decoherence rate due to the transitions between the "valley-split" states can be performed in terms of Eq. (52) when the lifetime $\tau_\delta$ is calculated taking into account the valley-orbital structure of a QD. However, direct application of Eq. (50) to Eq. (49) would resulting the vanishingly small matrix elements $B_q$ with $\hbar \omega_q = \Delta E_{v-v'}$ for the case of intervalley transitions. In order to take into consideration the process depicted in Fig. 7, one must supplement the Hamiltonian [Eq. (41)] with terms accounting for the phonon scattering at the defects:

$$H_3 = \sum_{q,q'} F_{q,q'-q} Q_q Q_{q'},$$

(55)

$$F_{q,q',q'-q} = \frac{\hbar}{2\rho V_0} \frac{C_{q,q'}}{\sqrt{\omega_q \omega_{q'}}},$$

(56)

where the matrix element of phonon scattering is

$$C_{q,q'} = i \frac{g_C}{2a^3} \sum_a (V_{q'-q} a)(e_q a)(e_{q'} a)(q a)(q' a)[(q - q') a].$$

(57)

Here $g_C$ is a constant, $a$ the unit vectors connecting neighboring lattice atoms, and $V_q$ a Fourier transformation of the static displacement $y(r)$ caused by point defects. Additionally, a more general expression is applied for the correlation function (see, for example, Ref. 79 and 80) in place of Eq. (50) that is obtained in a harmonic approximation:

$$\langle Q_p(\tau) Q_{-p}\rangle_\omega = \frac{1}{\pi} \frac{(2N_p + 1) \Gamma_p(\omega)}{(\omega^2 - \omega_p^2)^2 / \omega_p^2 + \Gamma_p^2(\omega)}.$$

(58)

The phonon damping rate $\Gamma_p(\omega)$ must be calculated in terms of the interaction specified by Eqs. (55)-(57). Following the works of Refs. 80 and 97, it is given as

$$\Gamma_p(\omega) = 8\pi \sum_{q'} |F_{p,p',q'-q}|^2 [\delta(\omega - \omega_{p'}) - \delta(\omega + \omega_{p'})].$$

(59)

in terms of the Hamiltonian $H_{ph} + H_3$ [Eqs. (41) and (55)].

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31
For simplicity, we ignore the lattice anisotropy and consider the displacement \( y(\mathbf{r}) \) in the model of isotropic medium:

\[
y(\mathbf{r}) = \sum_{j=1}^{N_D} A_D \frac{\mathbf{r} - \mathbf{R}_j}{|\mathbf{r} - \mathbf{R}_j|^3},
\]

(60)

where \( A_D \) is a mismatch in the lattice volume between the host and substitutional impurity atoms. As it plays the role of deformation charge, this parameter (i.e., \( A_D \)) can be found from the analysis of electron paramagnetic resonance linewidth. The Coloumbic deformation field resulting from Eq. (60) gives a well-known expression

\[
V_q = 4\pi i A_D V_0 \sum_{j=1}^{N_D} e^{i\mathbf{q}\mathbf{R}_j} \frac{\mathbf{q}}{q^2}.
\]

(61)

Then, substitution of Eqs. (56), (57) and (61) into Eq. (59) and follow-up integration over the phonon spectrum in the Debye model (i.e., \( \omega_p = \omega_{q, \kappa} = c_\kappa q \)) results in

\[
\Gamma_{\kappa, q}(\omega) = \frac{27\pi}{16} n_D \Sigma_{\kappa} \frac{A_D^2 g_C^2}{\rho^2 c_L^2} \omega^3 \omega_q,
\]

(62)

where \( n_D \) is the concentration of point defects. The coefficient \( \Sigma_{\kappa} \) in this equation takes into account the contributions of the phonon modes with different polarizations. In the approximation of an isotropic medium, one can find \( \Sigma_L = S_{LL} + (c_L/c_T)^2 S_{LT} \), where the sums \( S_{\kappa\kappa'} = \left\langle \left\{ \sum_a (e_{\mathbf{q}\kappa}^* \mathbf{a}) (e_{\mathbf{q}\kappa'}^* \mathbf{a}) (\mathbf{q}^\kappa \mathbf{a}) (\mathbf{q}^\kappa' \mathbf{a}) (|\mathbf{q} - \mathbf{q}'| \mathbf{a})^2 \right\}^2 \right\rangle_{(4\pi)^2} \) account for the polarization mixing; \( \mathbf{a} = \mathbf{a}/a; \mathbf{q} = \mathbf{q}/q; \quad ... \Rightarrow (4\pi)^2 \) means averaging over the directions of \( \mathbf{q} \) and \( \mathbf{q}' \). A numerical estimation for the diamond lattice gives \( S_{LL} = 0.789 \) and \( S_{LT} = 0.239 \); hence, \( \Sigma_L = 1.29 \) for Si. Finally, the Fourier transformation of the correlation function [Eq. (58)] is obtained as

\[
\langle Q_p(\tau) Q_{-p}(\omega) \rangle = \frac{27}{16\hbar} (2N_p + 1) n_D \Sigma_{\kappa} \frac{A_D^2 g_C^2}{\rho^2 c_L^2} \omega^3 \omega_q.
\]

(63)

for the phonons connecting the equivalent valleys.

The next step is to evaluate the matrix element of the intervalley electron-phonon interaction [see Eq. (49)]. It assumes the form

\[
B_p = \sqrt{\frac{\hbar}{2\rho V_0 \omega_p}} (\Xi_d + \frac{1}{3} \Xi_u) q \delta_{\kappa, L} \Phi(\mathbf{q}),
\]

(64)

where \( \Xi_d \) and \( \Xi_u \) are the deformation potential constants and the form factor \( \Phi(\mathbf{q}) = \langle \psi_\mp | e^{i\mathbf{q}\mathbf{r}} | \psi_\pm \rangle \) selects the wave vectors close to the \( k \)-space separation \( \Delta k_v \) of the two minima. Assuming a QD with the lateral and transversal dimensions of \( L_{xy} \) and \( L_z \), one can obtain

\[
\Phi(\mathbf{q}) \approx \exp\left[\frac{L_{xy}^2}{4}(q_x^2 + q_y^2) - \frac{L_z^2}{4}(q_z + \Delta k_v)^2\right].
\]

(65)
The appropriate $\Delta k_v$ takes the value of $\approx 0.3\pi/a_L$ ($a_L$ the lattice constant) considering the Umklapp process. Substitution of Eqs. (62) and (64) into Eq. (58) results in the contributions of two different types. The first is a direct process and, as mentioned earlier, has only a minor significance to $\tau_c$ with a characteristic factor $\sim \exp[-L^2/2\Delta k_v^2] \ll 1$. On the other hand, the second type accounts for the electron interaction with virtual phonons of energy $\omega_q \gg \Delta E_{v-v}$. This contribution is calculated with the help of Eqs. (63)-(65):

$$\tau_c = \tau_\delta \tanh \frac{\Delta E_{v-v}}{2k_BT},$$  \hspace{1cm} (66)

$$\tau_\delta^{-1} = C_{iv}nD\gamma^2 A_D^2 \Xi^2 \Delta E_{v-v}^3 \frac{\hbar^4 V_Q D c_L^5}{\rho V_Q D c_L^5}$$  \hspace{1cm} (67)

When deriving Eq. (66), an estimation $g_c = 24\gamma\rho c^2$ from Ref. 82 is used that defines the constant $C_{iv} \approx 210$.

### C. Spin decoherence due to intervalley transitions

With the intervalley transition mediated by the point defects as described above, we can now calculate the subsequent spin relaxation due to the associated fluctuation in the local magnetic field. Evidently, the hyperfine interaction does not take part in the process since the equivalent valley-orbital states possess identical electronic envelope functions. Hence, only the $g$-tensor fluctuations due to the valley split energy is considered. As in Sec. V, applying Eq. (66) to Eq. (52) solves the problem under the condition $\tau_c \ll T_2$. However, it should be noted that the formal calculation of $T_2$ using these equations violates the required inequality condition when $\Delta E_{v-v}$ is sufficiently small. Subsequently, the Markovian kinetic equations [Eqs. (18)-(20)] are unsuitable. Actually the spin decoherence time can be determinate by $\tau_c$ itself in the regime, where the result for $T_{2,Z}$ [see Eq. (52)] is much shorter than $\tau_c$, as schematically illustrated in Fig. 8. Here, the spin relaxation can be considered in two stages. The first is the ”pending” period of approximately $\tau_c$, during which the electron has not undergone the intervalley scattering. Obviously, no change occurs in the electron spin phase. Once the transition is made, the electron is subject to the effective magnetic field (due to the difference in the $g$ tensor) and starts to acquire the phase change. Within a typical time on the order of $\Delta T_2 \approx \hbar/|\Omega^{(e)} - \Omega^{(g)}| (\ll \tau_c)$, the electron fully loses the information regarding the initial state. The intermediate case $T_2 \approx \tau_c$ remains beyond the quantitative consideration. For simplicity, we apply an interpolation $T_{dc} \approx T_2 + \tau_c$. The calculated
decoherence time $T_{dc}$ for a Si$_{1-x}$Ge$_x$ QD is plotted as a function of $\Delta E_{v-v}$ (Fig. 9) and temperature (Fig. 10). Two cases of Ge composition ($x = 0.1$ and $x = 0.02$) are considered assuming the QD size of $L_{xy} = 50$ nm and $L_z = 5$ nm. Clearly, the proposed mechanism can significantly increase the spin relaxation rate in the mixed crystals. Even with the smaller "defect" concentration of 2 %, $T_{dc}$ can be reduced to the 10 msec range. Particularly, this rate is very strongly dependent on the magnitude of the splitting $\Delta E_{v-v}$ (in reference to the thermal energy). Hence, the control of the valley splitting will be essential for long coherence. For estimation, we assume $\Xi = 8.6$ eV, $\rho = 2.33$ g/cm$^3$, $c_L = 8.43 \times 10^5$ cm/s, $\gamma = 0.56$ and $C_{iv} \approx 210$. The deformation charge $A_D$ was found to be $1.2 \times 10^{-25}$ cm$^3$ for a Ge atom in Si from Ref. 98.

VII. COMPARISON BETWEEN DIFFERENT MECHANISMS

To gauge the significance of the elastic processes, the relevant transversal spin relaxation rates ($T_2^{-1}$) are obtained numerically in a Si QD (with $5 \times 50 \times 50$ nm$^3$) as a function of temperature, magnetic field, and the orbital energy separation. In addition to the three elastic processes described in this paper (i.e., anharmonic vibration, local field steps due to the $g$-tensor fluctuation and hyperfine field fluctuation), the contribution by the direct spin flip (that is the dominant inelastic process in Si) is taken into account for comparison. Since Si is not a mixed crystal and the defect density kept low for the QC application, the process via the intervalley transition is not considered. Figure 11 shows the calculated $T_2^{-1}$ vs. $B$ when the energy separation $\delta_0$ is fixed at 2 meV. For the range of temperatures considered in the study, it is apparent from the result that the direct spin-flip dominates at a sufficiently high magnetic field ($\gtrsim 1$ T) due to its strong dependence (e.g., $B^4$-$B^5$). For low fields, the anharmonic vibration (i.e., phonon decay) process tends to be significant particularly when the temperature is also low. As the temperature increases, the contributions due to the local field steps become prominent. While the $g$-tensor fluctuation mechanism plays a greater role at a stronger $B$, the field independent nature of the nuclear field makes it stand out in the other extreme. The corresponding map of the dominant process/mechanism in the $B$-$T$ parameter space ($\delta_0 = 2$ mev) is given in Fig. 12, clearly signifying the importance of elastic relaxation.

A similar comparison is provided in Fig. 13 as a function of the orbital energy separation $\delta_0$.
at three different temperatures. The magnetic field is set to 1 T. Note that the contribution due to the anharmonic vibration process is not shown as its relaxation rate falls below the range of presentation. From the figure, it is clear that the relaxation induced by the local field steps (via both the $g$-tensor and hyperfine field fluctuation mechanisms) are very sensitive to $\delta_0$; this can be readily understood as the process requires a round-trip transitions by overcoming $\delta_0$. Subsequently, its contribution is important only for a small $\delta_0$ (in reference to $k_B T$). Since the spin-flip rate is practically independent of $\delta_0$ and $T$ in the considered parameter range, the cross-over point between these rates monotonically moves to a higher $\delta_0$ with increasing $T$. By 4 K, the inelastic process drops out of the picture. Of the two elastic mechanisms, the $g$ tensor case seems to be relatively more efficient as $\delta_0$ increases. This may be due to the fact that the change in the $g$ tensor is directly proportional to the orbital energy separation. Figure 14 summarizes the dominant process/mechanism identified in the $\delta_0$-$T$ parameter space at $B = 1$ T. Considering the conditions typical for QC using an electron spin in a Si QD (e.g., $\delta_0 \lesssim 1$ meV, $T \lesssim 1$ K, $B \gtrsim 1$ T), it is expected that $T_2$ over 10 sec can be achieved. If a SiGe QD is used in place of Si, then the decoherence process can become more active depending on the valley-split energy. However, $T_2$ on the order of 10 msec or longer is still attainable. The spin relaxation in a GaAs QD is predicted to be much faster.

Table III summarizes the functional dependence of the dominant relaxation processes considered in this study. The zero-th power (e.g., $T^0$, $B^0$, $\delta_0^0$) denotes the independent nature to the corresponding parameter. The unique set of $T$-, $B$- and $\delta_0$-dependence will facilitate the experimental identification of each process. As the thermal energy provides the unwanted error/noise, the parameter space relevant to QC is where $T$ is much smaller than the other characteristic energies.

VIII. CONCLUSION

A group of elastic spin relaxation processes that do not involve the energy exchange between the Zeeman subsystem and the thermal reservoir, is studied in semiconductor QDs. Contrary to the common perception, these processes can play a dominant role in the electron spin decoherence under the certain conditions (magnetic field, temperature, orbital energy separation, etc.) as identified in the investigation. Particularly, the calculation results il-
illustrate the potential significance of an elastic decoherence mechanism originating from the intervalley transitions in semiconductor quantum dots with multiple equivalent energy minima (e.g., the X valleys in SiGe). The detailed understanding obtained in this work will help optimizing the QD based QC systems for long quantum coherence; for example, the Si based QDs can offer a spin relaxation time that is much longer than the GaAs counterparts. However, an additional, comprehensive effort is needed for a complete analysis of spin decoherence taking into account all the possible contributions and their manifestations.

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1. V. Cerletti, W. A. Coish, O. Guwat, and D. Loss, Nanotechnol. 16, R27 (2005).
2. K. A. Valiev, Physics-Uspekhi 48, 1 (2005).
3. I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. 76, 223 (2004).
4. F. X. Bronold, A. Saxena, and D. L. Smith, in Solid State Physics, edited by H. Ehrenreich and F. Spaepen (Academic, New York, 2004), vol. 58.
5. P. Facchi, S. Montangero, R. Fazio, and S. Pascazio, Phys. Rev. A 71, 060306 (2005).
6. X. Marie, T. Amand, P. Le Jeune, M. Paillard, P. Renucci, L. E. Golub, V. D. Dynnikov, and E. L. Ivchenko, Phys. Rev. B 60, 5811 (1999).
7. Y. G. Semenov, K. N. Borysenko, and K. W. Kim, Phys. Rev. B 66, 113302 (2002).
8. M. M. Glazov and E. Y. Sherman, Phys. Rev. B 71, 241312 (2005).
9. I. A. Merkulov, A. L. Efros, and M. Rosen, Phys. Rev. B 65, 205309 (2002).
10. Y. G. Semenov and K. W. Kim, Phys. Rev. B 67, 073301 (2003).
11. P.-F. Braun, X. Marie, L. Lombez, B. Urbaszek, T. Amand, P. Renucci, V. K. Kalevich, K. V. Kavokin, O. Krebs, P. Voisin, and Y. Masumoto, Phys. Rev. Lett. 94, 116601 (2005).
12. A. M. Tyryshkin, S. A. Lyon, A. V. Astashkin, and A. M. Raitsimring, Phys. Rev. B 68, 193207 (2003).
13. B. Georgeot, and D. L. Shepelyansky, Phys. Rev. Lett. 86, 2890 (2001).
14. M. S. Byrd, D. A. Lidar, L.-A. Wu, and P. Zanardi, Phys. Rev. A 71, 052301 (2005).
15. A. V. Khaetskii and Y. V. Nazarov, Phys. Rev. B 64, 125316 (2001).
16. C. Tahan, M. Friesen, and R. Joynt, Phys. Rev. B 66, 035314 (2002).
17. B. A. Glavin and K. W. Kim, Phys. Rev. B 68, 045308 (2003).
18. V. N. Golovach, A. Khaetskii, and D. Loss, Phys. Rev. Lett. 93, 016601 (2004).
19. V. I. Fal’ko, B. L. Altshuler, and O. Tsyplyat’ev, Phys. Rev. Lett. 95, 076603 (2005).
20. E. Y. Sherman and D. J. Lockwood, Phys. Rev. B 72, 125340 (2005).
21. R. Orbach, Proc. Roy. Soc. A 246, 458 (1961).
22. C. B. Finn, R. Orbach, and P. W. Wolf, Proc. Phys. Soc. 77, 261 (1961).
23. A. V. Khaetskii and Y. V. Nazarov, Phys. Rev. B 61, 12639 (2000).
24. M. C. Terrile, H. Panepucci, and R. A. Cavalho, Phys. Rev. B 15, 1110 (1977).
25. V. Y. Zevin, V. I. Konovalov, B. D. Shanina, Zh. Eksp. Teor. Fiz. 59, 561 (1970); Soviet physics,
26 R. Hernandez and M. B. Wallker, Phys. Rev. B 4, 3821 (1971).
27 W. Heitler, *The Quantum Theory of Radiation* (Dover, New York, 1984).
28 L. K. Aminov, Phys. Stat. Sol. B 50, 405 (1972).
29 A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).
30 S. A. Al'tshuler and B. M. Kozyrev, *Electron Paramagnetic Resonance in Compounds of Transition Elements* (Wiley, New York, 1974).
31 R. de Sousa and S. Das Sarma, Phys. Rev. B 67, 033301 (2003).
32 Y. C. Cheng and R. J. Silbey, Phys. Rev. A 69, 052325 (2004).
33 Y. G. Semenov and K. W. Kim, Phys. Rev. B 70, 085305 (2004).
34 D. Mozyrsky, S. Kogan, V. N. Gorshkov, and G. P. Berman, Phys. Rev. B 65, 245213 (2002).
35 Y. G. Semenov and K. W. Kim, Phys. Rev. Lett. 92, 026601 (2004).
36 J. R. Klauder and P. W. Anderson, Phys. Rev. 125, 912 (1962).
37 W. B. Mims, Phys. Rev. 168, 370 (1968).
38 M. Chiba and A. Hirai, J. Phys. Soc. Japan 33, 730 (1972).
39 R. de Sousa and S. Das Sarma, Phys. Rev. B 68, 115322 (2003).
40 A. V. Khaetskii, D. Loss, and L. Glazman, Phys. Rev. Lett. 88, 186802 (2002).
41 D. Gamliel and J. H. Freed, Phys. Rev. A 39, 3228 (1989).
42 Y. A. Serebrennikov and U. E. Steiner, Chem. Phys. Lett. 222, 309 (1994).
43 Y. A. Serebrennikov, Phys. Rev. Lett. 93, 266601 (2004).
44 D. Pines, J. Bardeen, and C. P. Slichter, Phys. Rev. 106, 489 (1957).
45 V. A. Abalmasov and F. Marquardt, Phys. Rev. B 70, 075313 (2004).
46 I. Waller, Zeits. f. Physik 79, 370 (1932).
47 I. Dzyaloshinskii, Phys. Chem. Solids 4, 241 (1958).
48 T. Moriya, Phys. Rev. 120, 91 (1960).
49 K. V. Kavokin, Phys. Rev. B 64, 075305 (2001).
50 D. V. Bulaev, D. Loss, Phys. Rev. Lett. 95, 076805 (2005).
51 A. F. Zinov’eva, A. V. Nenasheev, and A. V. Dvurechenskii, JETP Lett. 82, 302 (2005).
52 R. de L. Kronig, Physica 6, 33 (1939).
53 J. H. Van Vleck, Phys. Rev. 57, 426 (1940).
54 Y. A. Bychkov and E. I. Rashba, Pis’ma Zh. Eksp. Teor. Fiz. 39, 66 (1984) [JETP Lett. 39, 78.
(1984)].

55 M. I. D’yakonov and V. Y. Kocharovskii, Fiz. Tech. Poluprovodn. 20, 178 (1986) [Sov. Phys. Semicond. 20, 110 (1086)].

56 L. M. Wood, T. L. Reinecke, and Y. Lyanda-Geller, Phys. Rev. B 66, 161318 (2002).

57 S. I. Erlingsson and Y. V. Nazarov, Phys. Rev. B 66, 155327 (2002).

58 W. Yang and K. Chang, Phys. Rev. B 72, 075303 (2005).

59 A. A. Kiselev, E. L. Ivchenko, A. A. Sirenko, T. Ruf, M. Cardona, D. R. Yakovlev, W. Ossau, A. Waag, and G. Landwehr, J. Cryst. Growth 184/185, 831 (1998).

60 E. L. Ivchenko, A. A. Kiselev, and M. Willander, Solid State Commun. 102, 375 (1997).

61 A. D. Margulis and V. A. Margulis, Sov. Phys. Solid State 25, 934 (1983).

62 N. S. Averkiev and L. E. Golub, Phys. Rev. B 60, 15582 (1999).

63 N. G. Koloskova, Fiz. Tv. Tela. 5, 61 (1963) [Sov. Phys. Solid State 5, 40 (1963)].

64 Y. G. Semenov, Phys. Rev. B 67, 115319 (2003).

65 P. N. Argyries and P. L. Kelly, Phys. Rev. 134, A98 (1964).

66 P. Zwanzig, J. Chem. Phys. 33, 1338 (1960).

67 The existence of main axes for $g$-tensor can be proven at an arbitrarily low symmetry. See V. G. Grachev, Zh. Exp. Teor. Fiz. 92, 1834 (1987) [Sov. Phys. JETP 65, 1029 (1987)].

68 A. Sher and H. Primakoff, Phys. Rev. 119, 178 (1960).

69 L. A. Khalfin, JETP Lett. 8, 65 (1968).

70 K. Urbanowski, Phys. Rev. A 50, 2847 (1994).

71 B. Misra and E. C. G. Sudarshan, J. Math. Phys. 18, 756 (1977).

72 The sign of exponent $-i$ in the similar expression of Ref. 64 corresponds to definition of Fourier transformation with sign $-i$ in exponent.

73 C. P. Slichter, Principles of Magnetic Resonance (Springer-Verlag, New York, 1992).

74 V. Zevin, Phys. Rev. B 11, 2447 (1975).

75 The anisotropy of Eq. (18) as well as the correlation between all the spin components of $\mathbf{s}$ are the main distinctions between these equations and the well-known Bloch-Redfield equations [see, for instance, K. Blum, Density Matrix Theory and Applications (Plenum, New York, 1996)].

76 G. E. Pikus and A. N. Titkov, in Optical Orientation, Ed. F. Meier and B. P. Zakharchenia (Elsevier, Amsterdam, 1984).

77 L. D. Landau and E. M. Lifshitz, Electrodynamics of Continuous Media (Pergamon, Oxford,
Note that the approximation of the deformation charge $A$ as a difference of volumes per Ge and Si atoms in the corresponding crystals leads to an overestimation $A \simeq 3 \times 10^{-24} \text{ cm}^3$. 

82 P. Carruthers, Rev. Mod. Phys. 33, 92 (1961).
83 C. J. Glassbrenner and G. A. Slack, Phys. Rev. 134, A1058 (1964).
84 J. Callaway, Phys. Rev. 113, 1046 (1959).
85 L. Roth, Phys. Rev. 118, 1534 (1960).
86 H. Hasegawa, Phys. Rev. 118, 1523 (1960).
87 D. K. Wilson and G. Feher, Phys. Rev. 124, 1068 (1961).
88 E. R. Glaser, T. A. Kennedy, B. Molnar, R. S. Sillmon, M. G. Spencer, M. Mizuta, and T. F. Kuech, Phys. Rev. B 43, 14540 (1991).
89 B. I. Kochelaev, Sov. Phys. JETP 37, 171 (1960).
90 C.-Y. Huang, Phys. Rev. 154, 215 (1967).
91 A. R. Bhatt, K. W. Kim, and M. A. Stroscio, J. Appl. Phys. 76, 3905 (1994).
92 A. A. Kiselev, E. L. Ivchenko, and U. Rössler, Phys. Rev. B 58, 16353 (1998).
93 D. Paget, G. Lampel, B. Sapoval, and V. I. Safarov, Phys. Rev. B 15, 5780 (1977).
94 G. Grosso, G. P. Parravicini, and C. Piermarocchi, Phys. Rev. B 54, 16393 (1996).
95 V. N. Smelyanskiy, A. G. Petukhov, and V. V. Osipov, Phys. Rev. B 72, 081304 (2005).
96 R. Vrijen, E. Yablonovitch, K. Wang, H. W. Jang, A. Balandin, V. Roychowdhury, T. Mor, and D. DiVincenzo, Phys. Rev. A 62, 012306 (2000).
97 M. A. Krivoglaz, *Theory of X-ray and Thermal-Neutron Scattering by Real Crystals* (Plenum, New York, 1969).
98 V. E. Kustov, M. G. Mil’vitskii, Y. G. Semenov, B. M. Turovskii, V. I. Shakhovtsov, and V. L. Shindich, Sov. Phys. Semicond. 20, 169 (1986).
99 Note that the approximation of the deformation charge $A$ as a difference of volumes per Ge and Si atoms in the corresponding crystals leads to an overestimation $A \simeq 3 \times 10^{-24} \text{ cm}^3$. 

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TABLE I: Main contributions to electron spin dephasing (with $T_2^*$) in a QD spin based QC system. The "random local fields" category denotes the processes that lead to phase diffusion of the multi-qubit system without causing decoherence in the individual qubits (i.e., spin). The decoherence or transversal relaxation ($T_2$) has contributions from both elastic and inelastic processes for a single spin. The latter involves spin flip transitions leading to the energy (or longitudinal) relaxation of the spin states (i.e., $T_1$). The # sign denotes the spin relaxation processes induced by the spin-phonon interactions. HFI symbolizes the hyperfine interaction.

| Dephasing ($T_2^*$) | Decoherence ($T_2$) | Random Local Fields |
|---------------------|---------------------|---------------------|
|                     | Inelastic process ($T_1$) | Elastic process |    |
| Direct spin-flip#   | Anharmonic vibration# | Local field steps# | $g$-factor dispersion |
| Orbach#             |                     | Spectral diffusion | Dispersion of HFI |
| Two-phonon#        |                     | Geometric phase#  | Impurity fields |
| Nuclear spins       |                     |                     | Inter-qubit interaction |
TABLE II: Various mechanisms of the spin-phonon interaction in a manner consistent to both elastic and inelastic processes listed in Table I (marked by #). SOI and HFI denote the spin-orbit and hyperfine interactions, respectively.

|                  | Spin-orbit interaction | Hyperfine interaction | Spin-spin interaction |
|------------------|------------------------|-----------------------|-----------------------|
| Direct           | SOI modulation         | HFI modulation        | Waller                |
| Admixture        | Kronig–Van Vleck       | Spin state mixing     | Exchange modulation   |
| Indirect         | g-tensor fluctuation   | Hyperfine-field fluctuation | Spin state mixing | Spin-spin fluctuation |
TABLE III: Functional dependence of the dominant spin-phonon processes on the relevant parameters considered in this study. The zero-th power (e.g., $T^0$, $B^0$, $\delta_0^0$) denotes the independent nature to the corresponding parameter. The temperature $T$, magnetic field $B$, and Zeeman frequency $\omega$ are expressed in units of energy. $T^*$ stands for a characteristic temperature, based on which the low and high temperature regimes are defined. Eqs refer to the corresponding equations given in the text.

| Process                  | Eqs | Temperature | Magnetic field | Energy separation |
|--------------------------|-----|-------------|----------------|------------------|
| Direct spin-flip         |     | $T^0$ $\omega$ $T^1$ $B^5$ $B^4$ | $T \ll T^* \ll T$ $T \ll \omega$ $T \gg \omega$ $T \ll \delta_0$ $T \gg \delta_0$ |
| Anharmonic vibration    | 28  | $T^0$ $T_{\text{eff}}$ $T^1$ $B^2$ $\delta_0^0$ | $T^0$ $T_{\text{eff}}$ $T^1$ $B^2$ $\delta_0^0$ |
| $g$-tensor steps        | 52  | $e^{-\delta_0/T}$ $\delta_0$ $T^{-1}$ $B^2$ $e^{-\delta_0/T}$ $\delta_0$ |
| HF-field steps           | 54  | $e^{-\delta_0/T}$ $\delta_0$ $T^{-1}$ $B^0$ $\delta_0^2 e^{-\delta_0/T}$ $\delta_0^{-1}$ |
FIG. 1: Schematic diagrams of one-phonon spin relaxation process. The vertical lines correspond to the localized electrons with orbital $n, n'$ and spin $s = \uparrow, \downarrow$ states; the dashed lines depict a phonon with the $q$ or $q'$ mode; and the circle represents a direct electron-phonon interaction. When the electronic transition due to the interaction with a phonon does not change the orbital states (i.e., $n = n'$), the diagrams describe the direct process of spin relaxation with the absorption (left panel) or the emission (right panel) of the phonon resonant with the Zeeman splitting. If $n$ and $n'$ corresponds to the ground and excited states respectively, the net effect of two depicted processes in tandem represents the Orbach process of spin relaxation. In this case (Orbach), the spin flip occurs only in one of the stages (either absorption or emission) while the other is spin independent.
FIG. 2: Schematic diagrams describing three types of spin flip $s \rightarrow s'$ Raman processes under the inelastic two-phonon scattering. (a) The spin-two-phonon interaction in the first order results in the direct Raman spin-relaxation process. (b) The process of second order on the one-phonon interactions assumes the electronic virtue transitions via intermediate states $n's''$; the interference of the processes, which corresponds to the braced diagrams, sometimes leads to the so-called Van Vleck cancellation and the distinctive dependence $\sim T^9$ for the relaxation rate. (c) The anharmonic spin relaxation Raman process implies an inelastic phonon scattering due to the third order anharmonicity with the creation of a virtual phonon $q''$, which in turn induces a spin flip via the spin-one-phonon interaction. Processes (a) and (c) are characterized by typical temperature dependence $\sim T^7$ for the Raman spin relaxation rate. As explained, the square represents the direct spin-two-phonon interaction, while the triangle stands for the inelastic phonon scattering due to the third order anharmonicity; other notification are the same as in Fig. 1.
FIG. 3: Schematic illustration of a single phonon contribution to the random spin phase shift. \( \Omega_{\text{inst}} \) is the instantaneous effective field of spin precession associated with a phonon. \( t_{11}, t_{12}, t_{13}, \ldots \), are the random instants of phonon generation, while \( t_{21}, t_{22}, t_{23}, \ldots \), of phonon annihilation. The vertical dotted lines depict the instants of the last full oscillations by the phonon before its disappearance. The residual oscillations (on the right of the dotted line) are not compensated and produce the net effect (\( \Omega_p \)), which influences the spin as a short pulses with a random amplitude at random times. The phase shift due to the influence of \( \Omega_p \) is a Markovian process at the time interval \( t \gg \tau_c \), where the phonon correlation time \( \tau_c \) is defined as a mean value of the differences \( t_{1i+1} - t_{1i} \) (thus, \( \tau_c = \tau_p \), the phonon relaxation time). The phase relaxation time is proportional to the mean value of the squared phase shift caused by a single pulse \( \langle \Delta \phi^2 \rangle \) and the rate of phonon relaxation \( \tau_p^{-1} \).
FIG. 4: Schematic diagram illustrating the process of precession fluctuations and their influence on spin phase relaxation. The model assumes sudden changes of the effective field $\Omega$ at random instances that lead to the uncontrollable phase disturbances. At the scale of time $t \gg \tau_c$ ($\tau_c$ is the mean interval for consecutive quantum leaps in $\Omega$), the Markovian process takes place that defines the relaxation rate proportional to $T_2^{-1} \sim \langle \Delta \phi^2 \rangle / \tau_c$, where $\langle \Delta \phi^2 \rangle$ is the mean value of the squared phase shift acquired between two successive sudden changes of $\Omega$. Taking into account that $\Delta \phi \approx \Omega \tau_c$, one can find the dependence $T_2^{-1} \sim \langle \Delta \phi^2 \rangle \tau_c$ typical for the process of precession fluctuation.
FIG. 5: Decoherence factor $F(\delta_0/2k_BT)$ [i.e., Eq. (51)] for the process of precession fluctuation as a function of temperature assuming $\delta_0 = 1$ meV.
FIG. 6: Spin decoherence rate via the $g$-tensor fluctuation mechanism [i.e., Eq. (52)] as a function of temperature for $\delta_0 = 1$ meV and $B = 1$ T. Curve 1 is for a GaAs QD, while curve 2 considers a Si QD with $g_0 - g_\parallel = 0.0131$ and $g_0 - g_\perp = 0.0141$. 
FIG. 7: Schematic illustration of the phonon-mediated intervalley relaxation process in a crystal with inhomogeneous deformations. The [001] and [00\bar{1}] denote the electron states in the two equivalent valleys; the circle represents the electron-phonon matrix element, the triangle depicts third-order anharmonicity, and the crossed circle is for the point defect. The real phonon \( q \) splits into two virtual phonons \( q' \) and \( q - q' \) due to the lattice anharmonicity. The latter is accommodated by the local deformation of the point defect that can be of short wavelength with \( |q'| \gg |q| \). Consequently, the other virtual phonon can have a momentum \( q' \) large enough to induce the intervalley transition.
FIG. 8: Diagram of phase relaxation in the case of inequality $T_2 \ll \tau_c$, where $T_2$ is the transversal relaxation time in the Markovian kinetic equations. In time of approximately $\tau_c$, the effective field is subject to a sudden large change. This causes relatively fast spin phase diffusion with the time scale of $\Delta T_2$. Hence, the duration of the two-step process, $\approx \tau_c$, can be associated with the phase relaxation time.
FIG. 9: Spin decoherence rate via the intervalley transitions as a function of valley-orbital splitting. Curve 1 is for Si$_{0.9}$Ge$_{0.1}$ (namely, the defect density of 10%), while Si$_{0.98}$Ge$_{0.02}$ is considered for curve 2.
FIG. 10: Spin decoherence rate via the intervalley transitions as a function of temperature. Curve 1 is for Si$_{0.9}$Ge$_{0.1}$, while Si$_{0.98}$Ge$_{0.02}$ is considered for curve 2.
FIG. 11: Spin decoherence rate vs. magnetic field for the dominant spin-phonon relaxation processes in a typical Si QD (of $5 \times 50 \times 50$ nm$^3$); line 1 – inelastic direct spin-flip process, line 2 – elastic process of acoustic-phonon anharmonic vibration, line 3 – elastic process of local field steps due to the hyperfine-field fluctuation, line 4 – elastic process of local field steps due to the $g$-tensor fluctuation. The temperatures are as shown in the figure; the energy splitting $\delta_0$ between the ground and the excited state is fixed at 2 meV. Since a Si QD is assumed without any point defects, the process via the intervalley transition is not considered.
FIG. 12: Phase diagram of dominant spin relaxation processes in the Si QD in the $B$-$T$ parameter space. The same conditions as in Fig. 11 are assumed (for example, $\delta_0 = 2$ meV). The unshaded region is where the elastic spin-phonon processes dominate.
FIG. 13: Spin decoherence rate vs. $\delta_0$ for the dominant spin-phonon relaxation processes in a typical Si QD (of $5 \times 50 \times 50$ nm$^3$); line 1 – inelastic direct spin-flip process, line 3 – elastic process of local field steps due to the hyperfine-field fluctuation, line 4 – elastic process of local field steps due to the $g$-tensor fluctuation. The temperatures are as shown in the figure; the magnetic field is fixed at 1 T. Since a Si QD is assumed without any point defects, the process via the intervalley transition is not considered.
FIG. 14: Phase diagram of dominant spin relaxation processes in the Si QD in the $\delta_0$-$T$ parameter space. The same conditions as in Fig. 13 are assumed (for example, $B = 1$ T). The unshaded region is where the elastic spin-phonon processes dominate.