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Model expressions for the spin-orbit interaction and phonon-mediated spin dynamics in quantum dots

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Abstract. Model expressions for the spin-orbit interaction (SOI) in a quantum dot are obtained. The resulting form does not neglect cubic terms and allows for a generalized structural inversion asymmetry. We also obtain analytical expressions for the coupling between states for the electron-phonon interaction and use these to derive spin-relaxation rates, which are found to be qualitatively similar to those derived elsewhere in the literature. We find that, due to the inclusion of cubic terms, the Dresselhaus contribution to the ground state spin relaxation disappears for spherical dots. A comparison with previous theory and existing experimental results shows good agreement thereby presenting a clear analytical formalism for future developments. Comparative calculations for potential materials are presented.

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1. Introduction

The control of spin in electronic and photonic devices is key to the future development of spintronic devices for dense optical communications and quantum informatics applications. Quantum dots (QDs) offer a promising means of achieving this, since the mechanisms leading to spin relaxation are greatly reduced in these localized states. To tailor spin control in QDs, we need to understand these spin relaxing mechanisms and develop models that can be easily applied to novel materials and heterostructures.

Two temperature regimes relevant to different applications may be distinguished. At low temperatures (typically less than 10 K), where many of the experimental investigations have been conducted, electronic spin in a QD is a promising candidate for a quantum bit (qbit) for quantum computing and quantum information applications. At higher temperatures (room temperature), there is particular interest in the control of spin via optical orientation in photonic devices, such as vertical cavity surface emitting lasers (spin VCSELS [1–6]) for optical communication systems and classical information processing. Whilst most of the experimental studies of spin dynamics in QDs are at temperatures less than 10 K and often in the presence of a magnetic field, spin-VCSELS operate at room temperature and typically with no field applied, so a good understanding of the temperature dependence of the relaxation rates is required.

Spin dynamics in semiconductor QDs are intimately linked to the spin-orbit interaction (SOI), which modifies the conduction band, providing a path for spin relaxation via general scattering processes due to the quantum dot states existing in an admixture of opposite spin-states. The two principal contributions to the SOI are due to bulk inversion asymmetry (BIA) in the crystal and structural inversion asymmetry (SIA) in nanostructures, known respectively as Dresselhaus [7] and Bychkov-Rashba [8] SOIs. In a quantum well (QW), these perturbations to the Hamiltonian are averaged over the growth direction and, very often, the remaining cubic terms in the Dresselhaus SOI are neglected, leaving expressions linear in wavevector.

Whilst this method is uncontentious for 2D electrons, it is also widely used in the context of spin relaxation in quantum dots (QDs) [9–24]. However, it may be argued that using a model for 2D electrons is not valid in the 0D case of a quantum dot and that a more appropriate form for the SOI should be used. Some authors, for instance Woods *et al* [25], do start with the full 3D expression for the SOI, although in this case the authors' results involve numerical terms for particular cases and would therefore be difficult to use for general modeling of experimental results.

In this paper, we set up a model of quantum dots based on harmonic oscillators and present a formulation of the SOI in the pseudo-zero dimensional case. A semi-analytical treatment, also based on harmonic oscillator states, has also been pursued by Planelles *et al* [26], who generate phonon scattering matrices via recursion. In the present work, we find explicit analytical expressions for the coupling of states via the SOI and the electron-phonon interaction and use these to derive expressions for the spin relaxation rates due to acoustic phonons. These are found to be qualitatively similar to

expressions derived elsewhere in the literature, showing that this approach may be used consistently without negating previous work.

The rôle of the cubic Dresselhaus term has been treated numerically by Hansen *et al* [27], who address the case of doubly occupied quantum dots. It has also been found to be important in the coherent coupling of quantum dots by Stano *et al* [28], who also pursue a numerical approach. Krich *et al* [29] find that the cubic Dresselhaus is significant in inducing anti-crossings between the energies of different orbital levels with opposite spins in the presence of an applied magnetic field and find that it plays an important role in electronic transport properties through quantum dots. Here, we find analytical expressions for spin relaxation between spin states in the same orbital level in singly occupied dots.

2. Spin-orbit interaction

The Dresselhaus SOI may be given by [30]

$$H_{\mathrm{D}} = \gamma_{\mathrm{D}} \boldsymbol{\sigma} \cdot \boldsymbol{\kappa}, \tag{1}$$

where $\boldsymbol{\sigma}$ is the Pauli pseudo-vector for the spin and the components of the orbital vector $\boldsymbol{\kappa}$ are given by cyclic permutation of

$$\kappa_x = k_x \left(k_y^2 - k_z^2 \right), \tag{2}$$

where k_i are the components of wavevector. Here x, y and z label the crystallographic axes. γ_{D} is the the Dresselhaus coefficient (with dimensions of $[EL^3]$) and, in the case that the spin-orbit energy Δ is much less than the energy gap $\epsilon_g$may be given by $\gamma_{\mathrm{D}} = 2\hbar^3 \Delta / \left(3 m_{cv} \epsilon_g \sqrt{2 m \epsilon_g} \right)$, where m_{cv} is a parameter of the Kane model.

Inversion symmetry may also be broken by an electric field $\mathbf{E}$, giving rise to the Bychkov-Rashba SOI. This electric field may arise due to the spatial variation of the band edge in asymmetric nanostructures and hence the effect is referred to as structural inversion asymmetry (SIA) SOI. The contribution to the Hamiltonian is given by [32]

$$H_{\mathrm{BR}} = \alpha_0 \boldsymbol{\sigma} \cdot (\mathbf{k} \times \mathbf{E}), \tag{3}$$

where $\alpha_0 = e\eta(2-\eta)P^2/(3m_0^2\epsilon_g^2)$, e is the elementary charge, m_0 is the free electron mass, $\eta = \Delta/(\epsilon_g + \Delta)$ and P is the interband momentum matrix element.

3. Quantum dot model

The exact form of the confining potential in a QD is difficult to specify. In self-assembled dots, strain fields are expected to be important along with any induced electric field due to band edge variation between different materials. However, to second order, any confining potential may be approximated as parabolic at the potential minimum, whether this is due to strain or an applied electric field. To a reasonable approximation then, we may model the electronic functions as harmonic oscillators, as observed by

Teichmann *et al* [31] via scanning tunneling microscopy in the case of InAs dots embedded in an AlAs matrix.

To this end, we treat a dot as a 3D parabolic potential, characterized by angular frequencies ω_x, ω_y and ω_z in each of the Cartesian directions. For simplicity, we shall take these to be coincident with the crystallographic axes. For spherical dots, this would be irrelevant. However, dots are usually grown in a 2D wetting layer and are squashed in the growth direction. As this is often the [100] direction, our simplification is appropriate. The actual geometry of the dot, which may, for instance, be in the shape of a truncated pyramid or disc is then approximated by a spheroid. For other growth directions other than [100] the model will have to be modified appropriately.

The QD orbital states are given by a product of 1D harmonic oscillator states. The wavefunction in the x direction, for instance, is given by the Hermite function

$$\phi_n(x) = \frac{1}{\left(2^n n!\right)^{1/2}} \left(\frac{\alpha_x^2}{\pi}\right)^{1/4} \mathrm{e}^{-\alpha_x^2 x^2/2} H_n\left(\alpha_x x\right), \tag{4}$$

where $\alpha_x = (m^*\omega_x/\hbar)^{1/2}$, m^* is the effective mass in the dot and the H_n are Hermite polynomials.

Applying the properties of Hermite polynomials and using $\mathbf{k} = -i\nabla$, we may derive the following useful results (for $\phi_n(x)$)

$$\begin{aligned} \langle \phi_m \left| k_x \right| \phi_n \rangle \;&=\; -\,\mathrm{i} \left(\frac{\alpha_x^2}{2}\right)^{1/2} \\ &\times\; \left[n^{1/2} \delta_{m,n-1} - (n+1)^{1/2}\, \delta_{m,n+1} \right] \end{aligned} \tag{5}$$

and

$$\begin{aligned} \left\langle \phi_m \left| k_x^2 \right| \phi_n \right\rangle \;&=\; -\,\frac{\alpha_x^2}{2} \Big[\left\{ n\,(n-1) \right\}^{1/2} \delta_{m,n-2} \\ &-\; (2n+1)\, \delta_{m,n} \\ &+\; \left\{ (n+1)\,(n+2) \right\}^{1/2} \delta_{m,n+2} \Big]. \end{aligned} \tag{6}$$

and similarly for the wavefunctions in the y and z directions. Writing m in terms of n and vice versa, it is straightforward to show that

$$\langle \phi_m \left| k_x \right| \phi_n \rangle = -\left\langle \phi_n \left| k_x \right| \phi_m \right\rangle \tag{7}$$

and

$$\left\langle \phi_m \left| k_x^2 \right| \phi_n \right\rangle = \left\langle \phi_n \left| k_x^2 \right| \phi_m \right\rangle. \tag{8}$$

We may denote the total QD spatial state vectors explicitly by $|\phi_{ijk}\rangle$, where the i, j and k are integers labeling the 1D solutions in the x, y and z directions respectively. For brevity, we may shorten this using a vector notation $\mathbf{n} = (i, j, k)$ when the explicit

indices are not needed. On the occasions when they are, we will use the shorthand notation $\langle A\rangle_{mn} \equiv \langle \phi_m | A | \phi_n \rangle$ for the inner product of an operator A with 1D states.

For the unperturbed states, the spin state of the QD will not depend on the orbital part. We may then take the total state to be a tensor product of the orbital states and spinors $|\phi_{\mathbf{n},\chi}\rangle = |\phi_{\mathbf{n}}\rangle |\chi\rangle$, where χ labels the spin. Taking the inner product of (1) and (2) for the Dresselhaus SOI with states $\mathbf{m}$ and $\mathbf{n}$, we generate the matrix elements

$$\langle \phi_{\mathbf{m},\chi'} | H_{\mathrm{D}} | \phi_{\mathbf{n},\chi} \rangle = \gamma_{\mathrm{D}} \langle \chi' | \boldsymbol{\sigma} | \chi \rangle \cdot \langle \phi_{\mathbf{m}} | \boldsymbol{\kappa} | \phi_{\mathbf{n}} \rangle , \tag{9}$$

where, employing the orthonormality of the harmonic oscillator states (for $\mathbf{m} = (i', j', k')$),

$$\begin{aligned} \langle \phi_{i'j'k'} | \kappa_x | \phi_{ijk} \rangle &= \langle k_x \rangle_{i'i} \\ &\times \left[\left\langle k_y^2 \right\rangle_{j'j} \delta_{k'k} - \left\langle k_z^2 \right\rangle_{k'k} \delta_{j'j} \right] . \end{aligned} \tag{10}$$

Using (7) and (8), we obtain the important results

$$\langle \phi_{\mathbf{n}} | H_{\mathrm{D}} | \phi_{\mathbf{n}} \rangle = 0 \tag{11}$$

and

$$\langle \phi_{\mathbf{m}} | H_{\mathrm{D}} | \phi_{\mathbf{n}} \rangle = - \langle \phi_{\mathbf{n}} | H_{\mathrm{D}} | \phi_{\mathbf{m}} \rangle . \tag{12}$$

Of particular interest are the matrix elements between the ground and first excited states. Using (5) and (6) we find, for $\mathbf{n} = (1, 0, 0)$,

$$\langle \phi_{000} | \kappa_x | \phi_{100} \rangle = -\frac{\mathrm{i}\alpha_x}{\sqrt{2}} \left(\frac{\alpha_y^2}{2} - \frac{\alpha_z^2}{2} \right) \tag{13}$$

and

$$\langle \phi_{000} | \kappa_y | \phi_{100} \rangle = \langle \phi_{000} | \kappa_z | \phi_{100} \rangle = 0, \tag{14}$$

with similar expressions for the other first excited states. Note that for a spherical dot we would have $\alpha_y = \alpha_z$ and the coupling between states would be zero. This result was also noted by Planelles *et al* [26], who also retained the cubic Dresselhaus terms. This has important consequences for drastically reducing the rôle of the Dresselhaus SOI in mediating spin-flip transitions in an isotropic dot, as will be seen later. Generally, however, as previously mentioned, the dot is often squashed in the growth direction of the wetting layer, so some residual coupling will remain.

Applying similar considerations to the Bychkov-Rashba SOI, we have

$$\langle \phi_{\mathbf{m},\chi'} | H_{\mathrm{BR}} | \phi_{\mathbf{n},\chi} \rangle = \alpha_0 \langle \chi' | \boldsymbol{\sigma} | \chi \rangle \cdot \langle \phi_{\mathbf{m}} | (\mathbf{k} \times \mathbf{E}) | \phi_{\mathbf{n}} \rangle , \tag{15}$$

where the exact form will depend on the spatial dependence of the electric field. In the particular case where **E** varies only over its direction of application (as usually assumed for QWs), which we shall take to be the z direction, we have

$$\begin{aligned}\langle \phi_{\mathbf{m}} | H_{\mathrm{BR}} | \phi_{\mathbf{n}} \rangle &= \alpha_0 \langle E_z \rangle_{k'k} \\ &\times \left(\langle k_y \rangle_{j'j} \delta_{i'i} \mathbf{e}_x - \langle k_x \rangle_{i'i} \delta_{j'j} \mathbf{e}_y \right). \end{aligned} \tag{16}$$

Since the Hermite functions are real, $\langle E_z \rangle_{k'k} = \langle E_z \rangle_{kk'}$, so in this case we have $\langle \phi_{\mathbf{m}} | H_{\mathrm{BR}} | \phi_{\mathbf{n}} \rangle = -\langle \phi_{\mathbf{n}} | H_{\mathrm{BR}} | \phi_{\mathbf{m}} \rangle$, as we did for the Dresselhaus SOI. Note, however that if E_z has any x or y dependence, this condition may no longer hold, which has consequences for spin relaxation between same orbital states.

4. Phonon mediated spin relaxation

In bulk materials and QWs, where the electronic wavefunctions have an extended nature, the dominant pathways to spin relaxation are usually the D'yakonov-Perel [33] and Elliot-Yafet mechanisms [34, 35]. The former relies on the precession of the spin in an effective magnetic field dependent on wavevector and so is suppressed in the reduced **k**-space of the dot. The Elliot-Yafet mechanism relies on the states of the system residing in an admixture of spin states, so that coupling between opposite spin states may occur by scattering via non-magnetic potentials (i.e. that would not ordinarily flip spin). Such a situation still pertains in QDs. However, due to the localized nature of the dots, scattering by localized defects and charge centers is not so significant. Phonon scattering, on the other hand, is still pertinent, particularly at high temperature, which is of relevance for the dynamics of spin-VCSELS. We therefore focus here on phonon-mediated spin relaxation.

The unperturbed eigenstates of the QD are pure spin states. The effect of the spin-orbit interaction is then to mix these pure states. The perturbed QD states may then be given via first order perturbation theory as

$$\begin{aligned}|\psi_{\mathbf{n},+}\rangle &= |\phi_{\mathbf{n},+}\rangle + \sum_{\epsilon_{\mathbf{m}} \neq \epsilon_{\mathbf{n}}} |\phi_{\mathbf{m},+}\rangle \frac{\langle \phi_{\mathbf{m},+} | H_{\mathrm{SO}} | \phi_{\mathbf{n},+} \rangle}{\epsilon_{\mathbf{n},+} - \epsilon_{\mathbf{m},+}} \\ &+ \sum_{\epsilon_{\mathbf{m}} \neq \epsilon_{\mathbf{n}}} |\phi_{\mathbf{m},-}\rangle \frac{\langle \phi_{\mathbf{m},-} | H_{\mathrm{SO}} | \phi_{\mathbf{n},+} \rangle}{\epsilon_{\mathbf{n},+} - \epsilon_{\mathbf{m},-}}, \end{aligned} \tag{17}$$

where $H_{\mathrm{SO}} = H_{\mathrm{D}} + H_{\mathrm{BR}}$. The relevant energy scales here are the energy spacings between QD levels $\hbar\omega = \hbar^2/(m^* L^2)$, where $L = 1/\alpha$ is a characteristic length scale for the dots. The perturbing energy due to the spin orbit interaction will then be $\Delta\epsilon \sim \gamma_D / L^3$, so the justification of the first order perturbation treatment requires $\Delta\epsilon/(\hbar\omega) = \gamma_D m^*/(\hbar^2 L) \ll 1$ (note that L must be the largest length characterizing the dots, i.e. the diameter rather than height, corresponding to the smallest energy

separations). Using GaAs as an example with a characteristic length scale of ~ 10 nm, $\Delta\epsilon/(\hbar\omega) \sim 10^{-3}$.

The admixture of spin states seen in (17) provides a mechanism for spin relaxation analogous to the Elliot process [34], whereby components of opposite spin may be coupled by ordinary scattering interactions. In a QD, an important mechanism for this will be phonon scattering.

Let us denote a general electron-phonon interaction by $U_{\pm\mathbf{q},b}$, where $\mathbf{q}$ is the phonon wavevector, b is the mode branch and the $\pm$ signs denote emission (upper sign) or absorption (lower sign) of a phonon respectively (and should not be confused with the labeling of spin). We then denote the matrix elements for coupling between perturbed states by $V^{\chi'\chi\pm}_{\mathbf{mn},\mathbf{q},b} \equiv \langle \psi_{\mathbf{m},\chi'} | U_{\pm\mathbf{q},b} | \psi_{\mathbf{n},\chi} \rangle$ and unperturbed states by $U^{\chi'\chi\pm}_{\mathbf{mn},\mathbf{q},b} \equiv \langle \phi_{\mathbf{m},\chi'} | U_{\pm\mathbf{q},b} | \phi_{\mathbf{n},\chi} \rangle$. Note that the latter are diagonal in spin, so would produce no spin flipping on their own. These elements then have the form

$$\langle \phi_{\mathbf{m}} | U_{\pm\mathbf{q},b} | \phi_{\mathbf{n}} \rangle = \pm A_{\pm\mathbf{q},b} \left\langle \phi_{\mathbf{m}} \left| e^{\pm \mathrm{i}\mathbf{q}\cdot\mathbf{r}} \right| \phi_{\mathbf{n}} \right\rangle, \tag{18}$$

where $A_{\pm\mathbf{q},b}$ is peculiar to the particular phonon process. The coupling elements may be given explicitly as a product of the elements for 1D states

$$\begin{aligned} \left\langle \phi_m \left| e^{iqx} \right| \phi_n \right\rangle &= \frac{e^{-q^2/(4\alpha_x^2)}}{\sqrt{2^{m+n}}} \\ &\times \sum_{k=0}^{\min(m,n)} \frac{2^k \sqrt{m!n!}}{(m-k)!(n-k)!k!} \\ &\times \left(\mathrm{i}\frac{q}{\alpha} \right)^{m+n-2k}. \end{aligned} \tag{19}$$

Note that the factor $e^{-q^2/(4\alpha_x^2)}$ limits the coupling to $q \ll 2\alpha_x$. For acoustic phonon interactions (for which the phonon energy is linear in q), this means phonon energies much less than the QD energy spacings $\hbar\omega_x$ and hence these processes are inefficient for energy relaxation between orbital levels.

From (17), the spin-preserving transitions are given by $V^{\chi\chi\pm}_{\mathbf{mn},\mathbf{q},b} \approx U^{\chi\chi\pm}_{\mathbf{mn},\mathbf{q},b}$, whilst, defining $H^{\chi'\chi}_{\mathbf{mn}} \equiv \langle \phi_{\mathbf{m},\chi'} | H_{\mathrm{SO}} | \phi_{\mathbf{n},\chi} \rangle$, the matrix element for phonon-mediated spin-flipping is

$$V^{\chi'\chi}_{\mathbf{mn}} = \sum_{\epsilon_{\mathbf{k}} \neq \epsilon_{\mathbf{m}}} \frac{H^{\chi'\chi}_{\mathbf{mk}} U^{\chi\chi}_{\mathbf{kn}}}{\epsilon_{\mathbf{m}} - \epsilon_{\mathbf{k}} + \mathrm{i}\Gamma} + \sum_{\epsilon_{\mathbf{k}'} \neq \epsilon_{\mathbf{n}}} \frac{U^{\chi'\chi'}_{\mathbf{mk}'} H^{\chi'\chi}_{\mathbf{k}'\mathbf{n}}}{\epsilon_{\mathbf{n}} - \epsilon_{\mathbf{k}'} - \mathrm{i}\Gamma}, \tag{20}$$

where the explicit phonon labeling has been dropped for brevity. Here, the energy term Γ splits the degeneracy of the spin states. In previous treatments [11] the degeneracy of the states has been lifted by the Zeeman energy $g\mu_B B$ (where g is the g-factor, μ_B is the Bohr magneton) in the presence of an applied magnetic field B. Since the spin states are

Kramers conjugate pairs, taking the complex conjugate reverses the sign on the Zeeman energy in the same manner as iΓ. Hence we may take $\Gamma = -\mathrm{i}g\mu_B B$. Alternatively, we may leave interpret Γ as an energy broadening, which is typically modelled in this way via the introduction of an imaginary energy. More generally, we may put $\Gamma \to \Gamma - \mathrm{i}g\mu_B B$ to model both simultaneously.

We note, however, that since $H^{\chi'\chi}_{\mathbf{mn}} = -H^{\chi'\chi}_{\mathbf{nm}}$, if $\Gamma = 0$, then the element for same orbital levels will be zero. This is known as Van Vlech cancellation [36, 37] and is a key factor for the suppression of spin relaxation in dots. Note that if the Bychkov-Rashba SOI does not meet this cancellation condition due to the spatial dependence of the electric field, there will then be significant coupling between exactly degenerate states.

Using the model described above, we may then proceed to either derive explicit expressions for each scattering mechanism, or solve (20) computationally. In either case, the rates for energy relaxing processes will be small, except for polar optical phonon scattering where the phonon energy matches the QD energy separations - indicative of the so-called phonon bottleneck [38].

For the time being, we may simply consider spin-flipping in the orbital ground state $\mathbf{m}$ = (0,0,0). We may approximate this by considering only the first excited states in each direction $\mathbf{k}$ = (1,0,0), (0,1,0), (0,0,1). In this case, we have

$$V^{\chi'\chi}_{\mathbf{00}} = -2\mathrm{i}\Gamma \sum_{\mathbf{k}} \frac{H^{\chi'\chi}_{\mathbf{0k}} U^{\chi\chi}_{\mathbf{k0}}}{\hbar^2\omega^2_{\mathbf{k}} + \Gamma^2},$$

where $\omega_{(1,0,0)} = \omega_x$ etc. Rather than derive general expressions for an arbitrary spin basis, we shall assume that the spin is quantized along the z direction and use (16) for H_{BR}. Using the shorthand $\langle \phi_{\mathbf{m},} |O| \phi_{\mathbf{n}} \rangle = \langle O \rangle_{\mathbf{mn}}$ for a general operator O, the terms for the SOI are

$$\langle \phi_{\mathbf{m},\pm} | H_{\mathrm{D}} | \phi_{\mathbf{n},\mp} \rangle = \gamma_{\mathrm{D}} \left(\langle \kappa_x \rangle_{\mathbf{mn}} \mp \mathrm{i} \langle \kappa_y \rangle_{\mathbf{mn}} \right) \tag{21}$$

and

$$\langle \phi_{\mathbf{m},\pm} | H_{\mathrm{BR}} | \phi_{\mathbf{n},\mp} \rangle = \alpha_0 \left(\langle k_y E_z \rangle_{\mathbf{mn}} \pm \mathrm{i} \langle k_x E_z \rangle_{\mathbf{mn}} \right). \tag{22}$$

Using (19), the phonon coupling elements between the ground and first excited states are

$$\left\langle \phi_{100} \left| e^{\mathrm{i}\mathbf{q}\cdot\mathbf{r}} \right| \phi_{000} \right\rangle = \frac{\mathrm{i}q_x f_{\mathbf{q}}}{\sqrt{2}\alpha_x} \tag{23}$$

etc., where

$$f_{\mathbf{q}} = \exp\left(-\frac{q_x^2}{4\alpha_x^2} - \frac{q_y^2}{4\alpha_y^2} - \frac{q_z^2}{4\alpha_z^2} \right). \tag{24}$$

As a further simplification, we assume that the dot is an oblate spheroid with $\omega_x = \omega_y = \omega_0$ and $\omega_z > \omega_0$. We then obtain

$$V^{\pm\mp}_{\mathbf{00}} = -\frac{\mathrm{i}\Gamma}{\hbar^2\omega_0^2 + \Gamma^2} A_{\mathbf{q},b} \left(\gamma_{\pm} q_x \pm \mathrm{i}\gamma^*_{\pm} q_y \right) f_{\mathbf{q}}, \tag{25}$$

where

$$\gamma_{\pm} = \gamma_{\mathrm{D}} \left(\frac{\alpha_{xy}^2 - \alpha_z^2}{2} \right) \pm \mathrm{i}\alpha_0 \left\langle E_z \right\rangle_{00} . \tag{26}$$

At this point, it is worth pointing out that, had we followed the usual 2D treatment for the SOI, the term $\gamma_{\mathrm{D}}\alpha_z^2/2$ would be proportional to a factor β_{D} in the linear part of the 2D Dresselhaus SOI, whilst $\gamma_{\mathrm{D}}\alpha_{xy}^2/2$ would correspond to the cubic term, which is typically neglected. Furthermore, the quantity $\alpha_0 \left\langle E_z \right\rangle_{00}$ equates to the usual definition of the Bychkov-Rashba coefficient α_{BR} [32].

The spin relaxation rate is then found from Fermi's Golden Rule

$$\begin{aligned} W_{00}^{\pm\mp} = & \frac{2\pi}{\hbar} \left(\frac{\Gamma}{\hbar^2\omega_0^2 + \Gamma^2} \right)^2 |\gamma_{\pm}|^2 \\ & \times \sum_{\mathbf{q},b} |A_{\mathbf{q},b}|^2 \left(q_x^2 + q_y^2 \right) f_{\mathbf{q}}^2 \delta \left(\Gamma - \hbar\omega_{\mathbf{q},b} \right) \end{aligned} \tag{27}$$

where $\hbar\omega_{\mathbf{q},b}$ is the phonon energy. (Note that here we tacitly assume $\Gamma = |\Gamma|$ in the case that Γ is complex). For acoustic phonons, we may approximate the scattering as elastic, so $\hbar\omega_{\mathbf{q},b} = \hbar v_{\mathbf{q},b} q \to 0$ and $f_{\mathbf{q}} \to 1$. Since the spin-flip rates are equal, we may drop the $\pm$ for denoting spin.

Reintroducing the notation to denote absorption and emission, the rate for piezoelectric scattering in zinc blende materials may then be found to be

$$\begin{aligned} {W_{00}}^{\pm}(\mathrm{PE}) = & \left(\frac{\Gamma}{\hbar^2\omega_0^2 + \Gamma^2} \right)^2 |\gamma|^2 \sum_b \frac{\kappa_b}{c_b} \frac{e^2 e_{14}^2}{3\pi\hbar\varepsilon^2} \\ & \times \left(n(\Gamma/\hbar) \pm \frac{1}{2} + \frac{1}{2} \right) \left(\frac{\Gamma}{\hbar v_b} \right)^3 , \end{aligned} \tag{28}$$

where e is the electronic charge, e_{14} is the piezoelectric coefficient and ε is the permittivity. Since we are interested in the case of single occupied dots, there will be no screening via the redistribution of free electrons. The dimensionless constants κ_b and averaged elastic constants c_b arise out of the spherical averaging of the electronic coupling over propagation direction [39]. Here $\kappa_{\mathrm{L}} = 12/35$ and $\kappa_{\mathrm{T}} = 16/35$ for longitudinal and transverse modes respectively, whilst

$$c_{\mathrm{L}} = c_{11} + \frac{2}{5} \left(c_{12} + 2c_{44} - c_{11} \right) \tag{29}$$

and

$$c_{\mathrm{T}} = c_{44} - \frac{1}{5} \left(c_{12} + 2c_{44} - c_{11} \right) , \tag{30}$$

where the c_{ij} are the usual elastic coefficients. The v_b are averaged mode velocities and are given in terms of the material density ρ by $v_b = \sqrt{c_b/\rho}$.

The phonon occupation number

$$n(\omega) = \frac{1}{\exp(\hbar\omega/(k_B T)) - 1} \tag{31}$$

is the Bose-Einstein factor in terms of the phonon energy $\hbar\omega$ and the thermal energy $k_B T$, where k_B is Boltzmann's constant and T is the temperature. Note that if Γ is taken to be proportional to the magnetic field B, (28) reproduces the B^5 dependency found in Ref. [11] and elsewhere. However, this dependency only holds at very low temperature. At higher temperatures, $n(\omega) + 1 \approx n \approx k_B T/(\hbar v q) \to k_B T/\Gamma$, via the action of the Dirac delta function in the integral, which reduces the dependency to Γ^4 as well as introducing a linear dependence on temperature. Such magnetic field and temperature dependencies of the spin relaxation rate have been observed by Cheng *et al* [40] in GaAs QDs.

Making this higher temperature approximation, the spin flip rate due to both emission and absorption of piezoelectric phonons reduces to

$$W_{00}(\mathrm{PE}) = \frac{\Lambda_{\mathrm{PE}}}{\left(1 + \Gamma^2/(\hbar^2\omega_0^2)\right)^2} \left(\frac{\Gamma}{\hbar\omega_0}\right)^4 \frac{k_B T}{\hbar}, \tag{32}$$

where

$$\Lambda_{\mathrm{PE}} = \frac{2\left|\gamma\right|^2 e^2 e_{14}^2}{3\pi\hbar^3\varepsilon^2\rho} \sum_b \frac{\kappa_b}{v_b^5}. \tag{33}$$

Note that this dimensionless value is still dependent on the dot geometry since γ depends the dot dimensions via (26). Moreover, we might also realistically expect the elastic constants, density and mode velocities to be modified by strain.

For deformation potential acoustic phonon scattering, only longitudinal modes contribute (so $b = \mathrm{L}$) and the spin-flip rate is found to be

$$\begin{aligned} {W_{00}}^{\pm}(\mathrm{DP}) = & \left(\frac{\Gamma}{\hbar^2\omega_0^2 + \Gamma^2}\right)^2 \left|\gamma\right|^2 \frac{\Xi^2}{3\pi\hbar\rho v_{\mathrm{L}}^2} \\ & \times \left(n(\Gamma/\hbar) \pm \frac{1}{2} + \frac{1}{2}\right) \left(\frac{\Gamma}{\hbar v_{\mathrm{L}}}\right)^5. \end{aligned} \tag{34}$$

where Ξ is the deformation potential. Here we have a Γ^7 dependency, reducing again to Γ^6 and a linear T dependence as the temperature increases above a few kelvin. Making the same approximations as above, we have

$$W_{00}(\mathrm{DP}) = \frac{\Lambda_{\mathrm{DP}}}{\left(1 + \Gamma^2/(\hbar^2\omega_0^2)\right)^2} \left(\frac{\Gamma}{\hbar\omega_0}\right)^6 \frac{k_B T}{\hbar}, \tag{35}$$

where

$$\Lambda_{\mathrm{DP}} = \frac{2\left(\hbar\omega_0\right)^2 \left|\gamma\right|^2 \Xi^2}{3\pi\hbar^5\rho v_{\mathrm{L}}^7}. \tag{36}$$

5. Results and discussion

5.1. Zero phonon line broadening

Similar results to (28) and (34) have been given elsewhere, where $\Gamma = g\mu_B B$ in terms of the g-factor, Bohr magneton μ_B and magnetic field B. However, experimental results [40] typically show some spin relaxation even at $B = 0$ and interpreting Γ in terms of the broadening of QD level suggests the possibility of a temperature dependent relaxation process. It would then be important to distinguish between the homogeneous broadening of a single dot from the inhomogeneous broadening of an array of dots.

In previous works [11, 25], the temperature dependence of the spin relaxation has been modeled in terms of two-phonon processes. However, since the broadening of the energy level can itself be explained in terms of two-phonon processes [41], it may be that the two approaches have the same underlying physics.

The proper treatment of this phenomenon is beyond the present scope of this paper and would need to address not just the Lorentzian broadening of the zero-phonon line (ZPL) but the wider phonon sidebands, both of which depend on temperature. Moody *et al* [42] fit the homogeneous broadening in GaAs QDs to a model of the form

$$\Gamma(T) = \Gamma_0 + \sum_i \Gamma_i n\left(\omega_i\right), \tag{37}$$

where $n\left(\omega_i\right)$ is again the Bose-Einstein factor and $\hbar\omega_i$ are the energy separations between the ground state and higher lying states. This gives a temperature dependence becoming roughly linear above about 20 K and a value of $\Gamma = 0.4$ meV at 50 K. A similar, phenomenological formula had been used earlier by Sanguinetti *et al* [43] for the ZPL broadening of individual GaAs QDs

$$\Gamma(T) = \Gamma_0 + aT + bn\left(\epsilon_{\mathrm{A}}/\hbar\right), \tag{38}$$

where ϵ_{A} is a characteristic activation energy. The authors' noted that this activation energy was, to within experimental error, equal to the LO phonon energy, consistent with the idea that the broadening is due to two-phonon interactions with higher lying levels (and virtual levels) via polar optical phonons. Similar values to Moody *et al* were found for the broadening, going up to ~ 1 meV at 100 K. In the low temperature regime, where most experimental determinations of spin relaxation have been conducted, the constant term is typically $\Gamma_0 \approx 0.1$ meV. The Zeeman energy will start becoming comparable to this around 2 T and above.

A difficulty with determining the Zeeman energy exactly is that the g-factor of the electron in the QD is likely to be very different to the bulk value due to confinement effects. Indeed, measurements of g-factor in quantum wells [44] show the value of the g-factor changing sign (passing through zero) as the well width is varied. Further studies may be needed to fully determine the g-factors in different QDs to properly quantify the magnetic field dependence of the spin dynamics.

Table 1. Parameters for some III-V materials. Note that signs are omitted for e_{14} and Ξ.

Parameter	GaAs	InAs	AlAs	InSb
γ_D (eV·Å^3)	18.00[a]	27.18[b]	18.53[b]	490.00[c]
e_{14} (Cm^{-2})[d]	0.16	0.045	0.225	0.07
Ξ (eV)[e]	7.17	5.08	5.64	6.94
$\varepsilon/\varepsilon_0$[d]	12.90	15.15	10.06	16.80
c_{11} (GPa)[e]	122.10	83.29	125.00	68.47
c_{12} (GPa)[e]	56.60	45.26	53.40	37.35
c_{44} (GPa)[e]	60.00	39.59	54.20	31.11
ρ (g·cm^{-3})[d]	5.32	5.68	3.76	5.77
A_{R} (meV^{-2})	0.32	3.51	0.042	3.95

[a] Wang *et al* [45] (measured)
[b] Winkler [46] (calculated)
[c] Kallaher *et al* [47] (measured)
[d] Ioffe NSM archive `www.ioffe.ru/SVA/NSM/Semicond/`
[e] Vurgaftman *et al* [48]

In order to use the ZPL broadening for Γ, we note that a fairly good fit to Sanguinetti *et al*'s data may still be obtained on dropping the term linear in temperature from (38). We therefore propose the following form

$$\Gamma(T) = \Gamma_0 + \Gamma_{\mathrm{LO}} n\left(\omega_{\mathrm{LO}}\right), \tag{39}$$

where $\hbar\omega_{\mathrm{LO}}$ is the LO phonon energy. We may then choose values commensurate with the fitted data of $\Gamma_0 \approx 0.1$ meV and $\Gamma_{\mathrm{LO}} \approx 35$ meV (it is tempting to set this as the LO phonon energy as well given the similarity, though there is no obvious reason why this might be so).

5.2. Comparison of phonon processes

We may compare the relative importance of deformation potential and piezoelectric phonon scattering for flipping spin by defining the ratio of acoustic phonon rates

$$R_{\mathrm{A}} \equiv \frac{W_{00}(\mathrm{DP})}{W_{00}(\mathrm{PE})} = A_{\mathrm{R}}\Gamma^2, \tag{40}$$

where

$$A_{\mathrm{R}} = \frac{\Xi^2\varepsilon^2}{\hbar^2 e^2 e_{14}^2 v_{\mathrm{L}}^2}\left(\sum_b \kappa_b \frac{v_{\mathrm{L}}^5}{v_b^5}\right)^{-1}. \tag{41}$$

The factor A_{R} is independent of geometry and for GaAs is found to be $A_{\mathrm{R}} = 0.32\,\mathrm{meV}^{-2}$, so that we may conclude that piezoelectric scattering is the dominant process in this case. Further ratios are given in table 1 for other selected III-V materials. As can be seen, deformation potential scattering is of much greater importance in InAs and InSb.

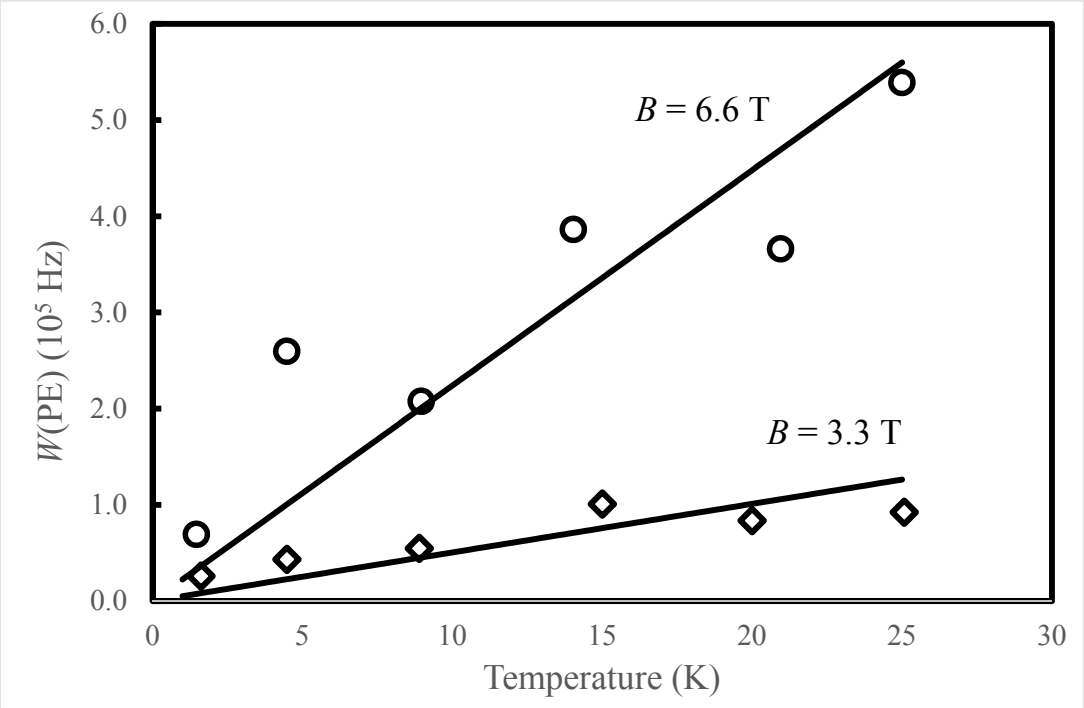

Figure 1. Fit of the scattering rate for piezoelectric phonon assisted spin relaxation via the Dresselhaus SOI in GaAs to experimental data by Cheng *et al* [40] for $B = 3.3$ T (diamonds) and $B = 6.6$ T (circles). Fitting parameters used were $g = 0.4$, $\alpha_z = 1/4$ nm^{-1} and $\alpha_{xy} = 1/18$ nm^{-1}.

5.3. Temperature dependence of spin relaxation in GaAs

To confirm the validity of the expressions derived here, we have fitted the temperature dependence of the spin relaxation rate in GaAs measured by Cheng *et al* [40] using (32) for piezoelectric scattering assuming only the Dresselhaus SOI. Characteristic length scales for the quantum dot are given by the α_i^{-1} and we have used $\alpha_z^{-1} = 4$ nm and $\alpha_{xy}^{-1} = 18$ nm. Note that these need not be the exact dimensions of the dot, so long as they are of comparable size. We have also used $g = 0.4$, which is a typical value for GaAs, although exact values in a QD are subject to variation. Finally, the literature abounds with different experimental and theoretical determinations of the Dresselhaus coefficient in GaAs. We have chosen that value of 18 eVÅ^3 used by Wang *et al* [45], since this is about midway between the range of values.

The results of the fit are shown in figure 1. Here we have incorporated both the Zeeman energy $g\mu_B B$ and the ZPL broadening via (39) in our calculations. We are also able to fit the magnetic field dependence, although the best fit requires modifying the values of α_z and α_{xy}. Given the possible error in g and γ_D, this remains reasonable. We note that the incorporation of broadening naturally accounts for the field independent constant added by Cheng *et al* in their fitting of the data.

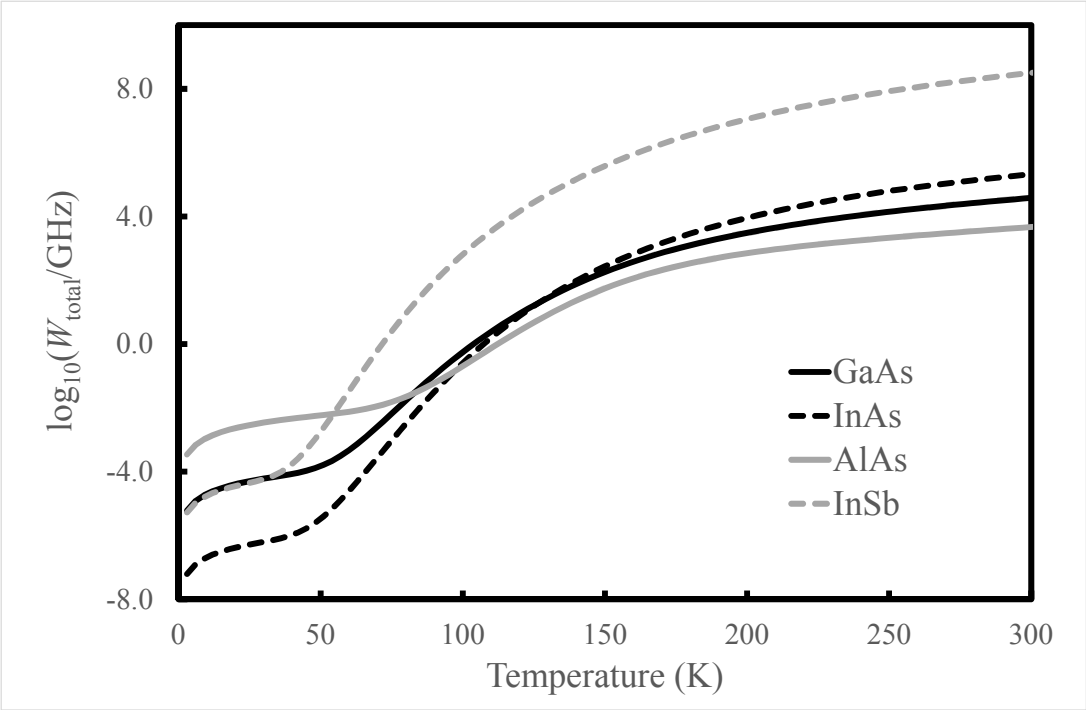

Figure 2. Extrapolated temperature dependence for the total spin relaxation rate (piezoelectric and deformation potential) due to the Dresselhaus SOI for selected semiconductors. Geometry-dependent parameters and g-factors are the same as used to fit the data in figure 1 for the sake of comparison.

5.4. Comparison of the Dresselhaus SOI in different materials

The lack of definitive values for γ_D becomes more acute when we broaden our gaze to other semiconductor materials, where there is currently a dearth of experimental studies. Moreover, there is a general disparity between theoretical and experimental determinations in GaAs that we might reasonable expect to extend to other materials. For instance, using the $\mathbf{k}\cdot\mathbf{p}$ method, Winkler [46] finds $\gamma_D = 27.6$ eVÅ^3, which we may compare with Walser *et al*'s experimental determination of 11 eVÅ^3 [44]. However, in the case of InAs and AlAs, we only have Winkler's determination, so we might expect this to be too large. Indeed, we may compare the theoretical value for InSb of 760 eVÅ^3 [46] to the experimental value of 490 eVÅ^3 determined by Kallaher *et al* [47]. In this latter case, we opt for the experimental value. The values chosen for each material are given in table 1.

A second point to make is that, in general, extrapolating a model beyond its tested range is generally bad practice. The temperature dependencies that we calculate are not then expected to be realistic at room temperature. Rather, the calculations are made as a basis for comparisons between different materials. In addition, we would expect the geometry of the dots and confining heterostructures to have a significant effect, which we cannot give a simple overview for. Hence, we only concentrate on the effect of the

material parameters and neglect the Bychkov-Rashba SOI from our calculations. The results are shown in figure 2 for both piezoelectric and deformation potential phonon scattering. We note the much greater rates associated with InSb, due mostly to the much greater Dresselhaus coefficient.

The calculated range of spin relaxation is over 12 orders of magnitude due to the high power dependence on Γ and the temperature dependence of Γ. This temperature dependence has only been observed up to around 100 K, so we must treat the extrapolation with caution. The general trend, however, suggests that spin relaxation may become acute at high temperatures, with relaxation times much less than 1 ps.

6. Conclusion

The results of (28) and (34) for the phonon-mediated spin relaxation rates in the QD ground state are qualitatively similar to those found elsewhere in the literature based on the 2D form for the spin orbit interaction and are consistent with experimental findings [40] when Γ is proportional to the magnetic field. In fact, some derivations [11] of these rates based on the 2D model implicitly invoke the same kind of 1D model for the coupling elements between states. The main differences here are that (*i*) we have constructed an explicit model for the QD states, (*ii*) we do not neglect the cubic terms of the Dresselhaus SOI, (*iii*) we allow for a general dependency of the electric field in the Bychkov-Rashba SOI and (*iv*) the quantization in the growth direction is achieved via the geometry of the dot rather than the surrounding well or wetting layer.

We have seen that a consequence of including the cubic Dresselhaus SOI is that the coupling due to bulk inversion symmetry will disappear for spherical dots when the factor $\alpha_{xy}^2 - \alpha_z^2 = 0$.

The formalism, whilst not represented comprehensively here, does allow for extension to a general dynamical model for dots of different ellipsoidal geometries. In addition, a general model may need to include two-phonon processes to account properly for the temperature dependence as well as electron-electron interactions to account for energy relaxation and interaction between dots.

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