Confinement sensitivity in quantum dot singlet–triplet relaxation

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Abstract

Spin–orbit mediated phonon relaxation in a two-dimensional quantum dot is investigated using different confining potentials. Elliptical harmonic oscillator and cylindrical well results are compared to each other in the case of a two-electron GaAs quantum dot subjected to a tilted magnetic field. The lowest energy set of two-body singlet and triplet states are calculated including spin–orbit and magnetic effects. These are used to calculate the phonon induced transition rate from the excited triplet to the ground state singlet for magnetic fields up to where the states cross. The roll of the cubic Dresselhaus effect, which is found to be much more important than previously assumed, and the positioning of ‘spin hot-spots’ are discussed and relaxation rates for a few different systems are exhibited.

Keywords: spin–orbit, spin relaxation, Dresselhaus coupling, phonons

(Some figures may appear in colour only in the online journal)

1. Introduction

Quantum dots (QDs) created through semiconductor heterostructures are promising components in the fields of spintronics, quantum information and quantum computing [1, 2]. For time sensitive applications, as when operations are to be performed on excited electron states within their coherence time, a long relaxation time is a decisive property. Long lifetimes are expected if the decay requires a spin flip and experimental studies have here measured excited state relaxation on the microsecond to millisecond timescale [3–6], with the longest lifetimes for dots subjected to a magnetic field. It is important to understand, and to quantitatively model, the dominating electronic and spin-dependent relaxation processes.

The role of the magnetic field is to suppress direct spin exchange with bulk material nuclei [4]. The still occurring relaxation is in this situation believed to be dominated by phonon exchange, but this mechanism opens the route to spin flips only in combination with spin–orbit coupling. This has been an important area of study, both for single electron [7–9] and multi electron states [6, 10–12], where the role of the Rashba [13] as well as the Dresselhaus [14] mechanisms for spin–orbit interaction in semiconductors have been investigated.

For two electron GaAs dots in particular, relaxation times on the millisecond timescale have been demonstrated experimentally [6, 11], and the singlet–triplet energy-splitting, as well as relaxation rates, have been investigated in some detail for magnetic fields of up to a few Tesla. Previous theoretical efforts to model these studies have shown that the system seems to exhibit a smaller than expected Dresselhaus coefficient [15, 16] and that a cylindrical hard-wall potential reproduces the singlet–triplet energy-splitting more accurately than a harmonic oscillator potential [17]. In the present work, an elliptical cylinder potential is used to further study the realistic parameter range determining the energy splitting and relaxation rates of said system, motivated by the better energy-splitting agreement previously shown. Harmonic oscillator results are also produced using the same methods and the differences in results due to potential shape are investigated.

This paper starts with a brief model- and implementation section describing what physical effects are included and how they are implemented. The electron wave-functions are represented in a B-spline basis [18], allowing for an arbitrary potential shape. The use of a B-spline basis, for which exact derivation is possible, is also important for the evaluation of the cubic Dresselhaus contribution which requires high-order derivatives of the wave function. Magnetic effects, in the form...
of a tilted magnetic, as well as spin–orbit (SO) effects are included in the one-body Hamiltonian. All the one-particle contributions to the Hamiltonian are used to create a basis which in turn is used to include the Coulomb interaction through the full configuration interaction (FCI) method. The two-electron states corresponding to the singlet ground state and first excited triplet states are extracted and used to calculate the phonon induced relaxation rate from Fermi’s golden rule. Dot width, ellipticity and relative directions of the tilted magnetic field are varied to match the energy splitting profile from the experimental results.

Following the model and implementation section, the results show the calculated singlet–triplet energy splittings for different dot potentials and compared them to the experimental results by Meunier et al [11]. This is followed by relaxation rates calculated for the same systems.

2. Model and implementation

2.1. Model

The two electron Hamiltonian describing the quantum dot can be expressed as

\[ H = \sum_{i=1,2} h(\mathbf{r}_i) + \frac{e^2}{4\pi\varepsilon_0} \frac{\varepsilon}{|\mathbf{r}_1 - \mathbf{r}_2|}, \]

where \( \varepsilon \) is the relative permittivity of the semiconductor material and \( h(\mathbf{r}_i) \) is the one-particle Hamiltonian:

\[ h(\mathbf{r}_i) = \frac{1}{2m^*} \mathbf{\hat{H}}^2 + g^* \mu_B \mathbf{B} \cdot \mathbf{S} + \mathbf{V} + h_{SO}. \]

Here \( \mathbf{\hat{H}} \) is the momentum operator, \( \mathbf{S} \) is the spin operator vector, \( \mathbf{V} \) is the effective confinement potential, \( \mathbf{B} \) is the external magnetic field and \( h_{SO} \) is the spin–orbit interaction. Bulk material properties are used for the electron effective mass \( m^* \) and gyromagnetic ratio \( g^* \).

In this work we focus on two effective confinement potentials; the two-dimensional harmonic oscillator

\[ V_{HO}(\mathbf{r}_i) = \frac{m^*\omega^2}{2} (\hat{x}_i^2 + \hat{y}_i^2), \]

where \( \omega \) is the harmonic oscillator frequency and \( \hat{x} \) is the dot ellipticity; and the hard wall cylindrical potential

\[ V_{HW}(\mathbf{r}_i) = \frac{m^*\omega^2}{2} (\hat{x}_i^2 + \frac{\hat{y}_i^2}{N^2}) \frac{r_0^{2(N-1)}}{r_0^{2(N-1)}}, \]

where \( N \) is a large integer, \( r_0 \) is the dot radius and \( \omega \) is used to tune the dot to match the harmonic oscillator at \( r_0 \). The hard wall potential is not a true step function at \( |\mathbf{r}_i| = r_0 \) but has a softness that is decreased with a high \( N \) value.

The magnetic field \( \mathbf{B} \) is tilted from the \( z \)-axis by an angle \( \theta \) and azimuthally from the \( x \)-axis by \( \phi \), so that:

\[ \mathbf{B} = B_0 (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta), \]

and the vector potential \( \mathbf{A} = \nabla \times \mathbf{A} \) can thus be chosen as

\[ \mathbf{A} = B_0 \left( \frac{\pi \cos \phi \sin \theta - y \cos \theta, x \cos \theta}{2}, \right. \]

\[ \left. - z \sin \phi \sin \theta, y \cos \phi \sin \theta - x \sin \phi \sin \theta \right). \]

Equipped with the vector potential we can expand the kinetic energy operator as:

\[ \frac{1}{2m^*} \mathbf{\hat{H}}^2 = -\frac{\hbar^2}{2m^*} \nabla^2 + \frac{e^2}{2m^*} \mathbf{A}^2 + \frac{e^2}{2m^*} \mathbf{B} \cdot \mathbf{\nabla}. \]

In the following we choose to study a QD restricted in the \( z \)-dimension, often orders of magnitude thinner than wide. Aiming for the 2D-limit we write the wave function as:

\[ \Psi(\mathbf{r}) = \psi(\mathbf{r}, y) \delta(z), \]

where the \( z \)-component is assumed to be in the shape of a delta function. The effective diamagnetic term in the \( xy \)-plane is obtained from \( A^2 \) in the limit \( z \rightarrow 0 \) which takes the form of an anisotropic harmonic oscillator potential in the \( xy \)-plane:

\[ A^2 = \frac{B_0}{4} \left( (x^2 + y^2) \cos^2 \theta + (y \sin \phi - x \cos \phi)^2 \sin^2 \theta \right). \]

This \( A^2 \)-potential has an elliptical cross section and the minor axis lies along projection of the magnetic field in the \( xy \)-plane (i.e. in the \( \phi \) direction).

Due to the frozen wave function in the \( z \)-dimension, electron motion is restrained within the \( xy \)-plane. This will reduce the angular momentum operator to \( \mathbf{L} = L_z \), leaving the angular Zeeman term:

\[ \frac{e}{2m^*} \mathbf{B} \cdot \mathbf{L} = \frac{e}{2m^*} \cos \theta B_0 \mathbf{L}_z. \]

With \( \mathbf{B} \) placed along the \( z \)-axis, the electron spin states are eigenstates to the \( \mathbf{S} \) operator, leading to the Zeeman spin term shifting the states \( | \pm 1/2 \rangle \) by the energy:

\[ \mp g^* \mu_B B_0 \hbar/2. \]

With an inclined magnetic field the \( \mathbf{S} \)-eigenstates will no longer be eigenstates to the Zeeman operator, instead the Zeeman spin term will couple the \( | \pm 1/2 \rangle \) states by:

\[ g^* \mu_B \mathbf{B} \cdot \mathbf{S} = g^* \mu_B B_0 \left( \cos \phi \sin \theta \mathbf{S}_z \right. \]

\[ \left. + \sin \phi \sin \theta \mathbf{S}_x + \cos \phi \mathbf{S}_y \right). \]

Note that this alone will not mix spins, but rather lead to spin up and down states (as defined by the direction of the magnetic field) that are linear combinations of the \( \mathbf{S}_z \)-eigenstates according to:

\[ | \uparrow \rangle = \cos(\theta/2)|+1/2\rangle - \sin(\theta/2)|-1/2\rangle, \]

and

\[ | \downarrow \rangle = \sin(\theta/2)|+1/2\rangle + \cos(\theta/2)|-1/2\rangle, \]

with the same energy splitting as without any field inclination.

2.1.1. The spin–orbit interaction. To achieve spin-mixing we need an interaction that couples the spin and spatial components of the wave function. Electrons moving in an electric
field generate a magnetic field which can couple to the spin magnetic-moment, and for a 2D-dot a net effect can be shown to arise when there is an asymmetry of the potential in the direction perpendicular to the plane of the semiconductor heterostructure. Depending on if the asymmetry originates from the heterostructure itself or from the bulk crystal structure the effects are represented by distinct operators and are called Rashba [13] or Dresselhaus coupling [14] respectively. For a 2D system the latter consist of two types of terms: one which depends linearly on the momenta in the $xy$-plane and one that depends on the square of the momentum in one of the two directions times the linear momentum in the other direction. We can thus write the spin–orbit Hamiltonian as the sum of three terms, $h_{SO} = h_R + h_{D1} + h_{D3}$, including the Rashba:

$$h_R = \frac{\alpha}{\hbar}\langle \hat{\pi}_x \hat{\sigma}_y - \hat{\pi}_y \hat{\sigma}_x \rangle,$$

(15)

the linear Dresselhaus:

$$h_{D1} = \frac{\gamma}{\hbar^2} \langle \hat{\pi}_x^2 \hat{\sigma}_y - \hat{\pi}_y^2 \hat{\sigma}_x \rangle,$$

(16)

and the cubic Dresselhaus:

$$h_{D3} = \frac{\gamma}{\hbar^3} \left[ (\hat{\pi}_x \hat{\sigma}_y \hat{\sigma}_z - \hat{\pi}_z \hat{\sigma}_x \hat{\sigma}_y) + (\hat{\pi}_y \hat{\sigma}_z \hat{\sigma}_x - \hat{\pi}_x \hat{\sigma}_y \hat{\sigma}_z) \right].$$

(17)

where $\alpha$ and $\gamma$ are the Rashba and Dresselhaus coefficients, $\hat{\pi}_{x,y,z}$ are the momentum operators and $\hat{\sigma}_{x,y,z}$ are the Pauli spin matrices. The Rashba interaction conserves $m_l + m_s$ and linear Dresselhaus interaction conserves $m_l - m_s$. The cubic Dresselhaus operator couple states $|m_l, m_s\rangle$ according to the selection rules [19]:

$$|m_l, +1/2\rangle \rightarrow |m_l - 1, -1/2\rangle, |m_l + 1, +1/2\rangle, |m_l - 1, +1/2\rangle, |m_l - 3, +1/2\rangle.$$  

(18)

The important point here is that when the one-particle states are no longer either spin-up or spin-down, but mixtures of the two, it follows that the two-particle states will be mixtures of spin-polarized and spin-unpolarized configurations and the total spin will cease to be a good quantum number. Although the effect of the spin–orbit interaction on the energy is very small it can thus have a profound effect on the life time of excited states since it opens up new decay paths.

Previous studies on similar systems have shown the Dresselhaus coupling to be the dominating spin–orbit interaction in this regime [20]. The ratio between the Dresselhaus and Rashba coefficients may however still be of interest when studying an anisotropic QD under a tilted and rotated magnetic field [12], small Rashba coefficients have been tested and found not to change the results significantly and will not be investigated further in this paper. Often the cubic Dresselhaus term is assumed to be a small correction that can be disregarded, but Krich and Halperin [21] have shown it to be of some significance. Our results indicate also that the cubic Dresselhaus term can have a mayor impact. In this context we note that there is an error in the implementation of this term by Hansen et al [15] and we will thus not compare with their findings below.

2.1.2. Electron–phonon interaction. Finally we need to consider the precise routes to spin-relaxation that have opened through the inclusion of spin–orbit interaction. The dominating scattering mechanisms, in systems similar to the ones studied here (with discrete energy states with a spacing of a few meV), have been shown to be through acoustic-phonons [4]. Meunier et al [11] also conclude that acoustic phonons are the only available phonons for the energy range explored in their experiment. The two most important phonon-electron couplings have previously been determined to be through deformed potential and piezoelectric coupling [22–24]. The phonon induced singlet–triplet relaxation rate can be estimated from Fermi’s golden rule:

$$\Gamma_i = \frac{V}{4\pi^2\hbar} \int d^3q |M_i(q)|^2 \left| \langle S|\hat{H}_{ph}|T_i \rangle \right|^2 \delta(\Delta E_{ST} - \hbar\omega),$$

(19)

for triplet states $T_{+1}$, $T_0$ and $T_{-1}$. $S$ and $T_i$ denote here the main character of the state. Since the phonon coupling is spin-conserving the relaxation is proportional to the usually tiny admixture of the opposite spin-character introduced through spin–orbit coupling. For example: a predominantly $T_{+1}$ state with an admixture of singlet character ($|\Psi_{T_{+1}} + \delta \Psi_S\rangle$) can relax to the singlet ground state, $\Psi_{S,S}$, with a rate proportional to $|\delta \Psi_S|\langle \hat{H}_{ph}|\Psi_{S,S}\rangle|^2$.

Guided by the investigation of phonon-induced electron relaxation in quantum dots made by Climente et al [9] we include relaxation through three phonon effects; deformation potential coupling

$$|M_1(q)|^2 = \frac{\hbar^2}{2\rho c V} |q|^2,$$

(20)

longitudinal piezoelectric coupling

$$|M_2(q)|^2 = \frac{32\pi^2\hbar^2}{c^2\rho c V} \left( 3q_y q_z \right)^2 |q|^2,$$

(21)

and transversal piezoelectric coupling

$$|M_3(q)|^2 = \frac{32\pi^2\hbar^2}{c^2\rho c V} \left( q_y^2 q_z^2 + q_z^2 q_x^2 + q_x^2 q_y^2 \right),$$

(22)

The last term is counted twice to account for two identical phonon modes [25]. Here $\Delta E_{ST}$ is the singlet–triplet energy splitting, $q = (q_x, q_y, q_z)$ is the phonon momentum and the states couple through the $\hat{H}_{ph} = \sum_{i=1,2} e^{-i\mathbf{q}\cdot \mathbf{r}}$ operator. Linear dispersion is used so that the phonon momentum is matched to the energy splitting, $|\mathbf{q}| = \Delta E / (\hbar c)$, where $c_i$ is the longitudinal speed of sound for $i = 1, 2$, and the transversal speed of sound for $j = 3$. $V$ is the normalization volume that will be cancel out. $\Xi_d$ is the deformation potential constant, $\rho$ is the material mass density and $h_{14}$ is the piezoelectric constant.

Only one-phonon relaxation processes has been investigated here. We note that Trif et al [26] have found that two-phonon processes can be of quite some importance for very
low B-fields, but since the subject here is strong magnetic fields we have not investigated this further.

2.2. Implementation

The one-particle Schrödinger equation, equation (2), is solved through diagonalization of the Hamiltonian matrix within a numerical B-spline [18] basis, where the polynomial basis allows for integration to machine precision through Gaussian quadrature. We note that the spin–orbit interaction is included already at this level and that spin-mixing thus is allowed within the full basis. It has earlier been shown in the literature [24, 27] that a large number of basis functions are needed in order to achieve convergence for the spin–orbit coupling even when only the lowest few many-body states are of interest.

Using a restricted set of the one-electron states obtained, all possible two-electron Slater determinants are constructed, and the full configuration interaction generalized eigenvalue equation is set up and solved through diagonalization of the two-particle Hamiltonian, matrix, see equation (1). The selection of one-electron orbitals to include is made by choosing a maximum number of their \( n_x + n_y \) quantity and monitoring the convergence in two-electron energy to within one percent. Here \( n_{x/y} \) denote the quantum numbers for the one dimensional well or harmonic oscillator potential. The \( n_x + n_y \) quantity is quite constant for a specific state even in the presence of an elliptic potential and tilted magnetic field. In close proximity to any avoided crossings created e.g. by the spin–orbit interactions, \( n_x + n_y \) will change between the avoiding states.

We model a GaAs dot using bulk values for the effective mass, \( m^* = 0.067 m_e \), relative permittivity, \( \varepsilon = 14.4 \) and effective gyromagnetic ratio \( g^* = -0.44 \). For the Dresselhaus parameter, we choose values of; \( \gamma = 27.5 \) eV \( \mathbf{\AA}^{-1} \), in accordance to previous experimental and theoretical results in GaAs, [28–30]; and \( \gamma = 9 \) eV \( \mathbf{\AA}^{-1} \), from previous system specific results, [11, 15, 16].

For the phonon transition calculations, a crystal density of \( d = 5310 \) kg \( \mathbf{m}^{-3} \), deformation potential constant \( \Xi_d = 6.7 \) eV, piezoelectric constant \( h_{14} = 1.4 \) V \( \mathbf{m}^{-1} \) and sound velocities \( c_L = 4720 \) m \( \mathbf{s}^{-1} \) longitudinal and \( c_T = 3340 \) m \( \mathbf{s}^{-1} \) transversal are employed.

3. One-electron spectrum

A study of the one-electron spectrum of the potentials will yield some important information on how the various parts of the one-electron Hamiltonian will effect the states. The key effect being the spin–orbit interaction and the avoided crossings appearing when it is included.

3.1. Harmonic oscillator confinement

The symmetric harmonic oscillator spectrum in figure 1 is familiar for a magnetic field perpendicular to the plane. The angular momentum Zeeman-splitting, linearly dependent on the B-field, dominates at low energies, but is overshadowed by the \( A^2 \) term at stronger fields, and at very high field strengths, Landau levels start forming. An important property of the harmonic oscillator is the equal-distance level spacing at zero magnetic field. A consequence of which is that the linear Zeeman splitting will create points where states bunch together and cross. One such point in figure 1 is around 1.26 T where states differing by multiples of \( \Delta m_L = 3 \) will cross.

By zooming in on the vicinity of the \( m_L = +2, -1, \) and \(-4\) crossing, we see that the cubic Dresselhaus effect is creating an avoided crossing between the \( |n, m_L, m_s\rangle = |0, +2, \uparrow\rangle \) and \( |1, -1, \uparrow\rangle \) states as well as between the \( |1, -1, \downarrow\rangle \) and \( |0, -4, \uparrow\rangle \) states, as expected from the selection rules. In the vicinity of these crossings, the electron spins will be heavily mixed. Since all such \( \Delta m_L = 3 \) crossings are located at the same magnetic field strength, this should result in a ‘spin hot-spot’ and many-electron states formed from these one-electron orbitals cannot be eigenstates to \( \hat{S}^2 \) any more.

The case of the elliptic harmonic oscillator with a tilted magnetic field, figure 2, has at a first glance a similar
spectrum as the symmetric case. Noticeable differences being an overall scaling since the magnetic z-component is dampened by a factor \( \cos \theta \); and the splitting of states at low field strength due to the ellipticity. Since the angular momentum operator, \( \hat{L}_z \), is independent of \( r \), it commutes with the circular symmetric harmonic oscillator potential, and it is possible to choose eigenfunctions that are simultaneously eigenfunctions to the Hamiltonian and to \( \hat{L}_z \). An elliptic potential, however, does not commute with \( \hat{L}_z \). No common eigenfunctions can then be found and the elliptical states at low magnetic field strengths are highly mixed in \( m_l \), and as a consequence they respond weakly to the Zeeman effect. Once the magnetic field strength is strong enough to dominate over the potential, states can once again be approximately described by their \( m_l \) quantum number, and will start to split linearly with \( \cos \theta B_0 \).

The crossing points seen in the symmetrical harmonic oscillator will however persist, but at scaled field strengths, since these are a result of the linear Zeeman splitting that still dominates in this region. The crossing at 1.26 T in figure 1, has for instance been shifted to 2.11 T in figure 2.

A more detailed investigation of the state crossings reveals that many more avoided crossings are created by the spin–orbit interaction in the elliptic case. Since the spin states are a linear combination of the \( \hat{S}_z \) eigenstates, they both will couple to states of \( \Delta m_l = 3 \). Also second order effects appear, coupling states of \( \Delta m_l = 6 \). This will result in strong spin mixing occurring over a broader magnetic field range, resulting in a larger spin hot-spot than in the case with a circular symmetric confinement potential.

3.2. Cylinder well confinement

Figure 3 shows the one-electron energy spectra for a circular well with a magnetic field perpendicular to the plane. The one-electron state energies at zero magnetic field will differ between the circular well and harmonic oscillator. Since the electrons in the well are strictly confined within the radius of the dot, states with higher radial quantum numbers will have
higher energies than their harmonic oscillator counterparts. The resulting energy level structure, and degeneracies will therefore differ.

The angular components of the circular quantum well eigenstates are equivalent to the harmonic oscillator polar angular components, and are hence eigenfunctions to the angular momentum operator. This results in a Zeeman splitting for the circular well states, much in the same manner as for the harmonic oscillator, until the \( A^2 \)-term once again starts to dominate, and Landau levels begin forming.

An important consequence of not having equidistant energy levels at zero magnetic field is the lack of high degeneracy at certain field strengths, as those found in the harmonic oscillator. This should reduce the effect of spin hot-spots, and instead spread out the spin mixed states over the entire spectra. In the zoomed in part of figure 3, an avoided crossing due to the cubic Dresselhaus interaction can be seen between the \( |1, -1, \downarrow \rangle \) and \( |0, -4, \uparrow \rangle \) states.

The ellipticity and tilted field added to figure 4 produces similar results to those found in the elliptic harmonic oscillator. The shift due to the scaled \( z \)-component of the magnetic field will not behave quite as linearly though, and some state crossings will disappear or be shifted far from the magnetic area of interest.

As in the harmonic oscillator case, the spin states, being linear combinations of the \( S_z \) eigenstates, will form avoided crossings with more states for a tilted magnetic field, as compared to the case with a field perpendicular to the \( xy \)-plane.

4. Two-electron results

The four lowest two-particle states obtained by diagonalization of the two-particle Hamiltonian matrix, see equation (1), are: a singlet state, which is the ground state for modest B-fields, and the Zeeman-split triplet states, with \( M_S = -1, 0, 1 \). The position of the three latter with respect to the ground states is shown as a function of magnetic field in figure 5.

In the following we want to compare our calculations to the experiment by by Meunier et al [11], and the first question is which of the states that were really addressed there. They claim to populate all three triplet states, and that the measurements are done on an average over these. We doubt that this is the case and that the most likely triplet state to populate should be the lowest energy spin polarized state.

We draw this conclusion from the way the experiment is performed. As described in Hanson et al [6] it starts with a single electron trapped in the dot, a second electron is then allowed to tunnel into the dot, creating either a singlet or triplet state. After a variable waiting time (allowing for relaxation if it was the excited states that was formed) an electron is allowed to tunnel out and a change in current is observed with a quantum point contact. Based on the tunneling rate the two-electron state is said to be determined. The important point now is that the first electron can be assumed to be in a definite state, presumably the one-particle ground state since it has occupied the dot for some time. In the presence of a magnetic field it will thus be in the state \( |g, \uparrow \rangle \). The second electron can populate the

**Figure 4.** Above: The one-electron spectrum of a elliptic well, with \( r_0 = 44 \text{ nm} \) and \( \delta = 1.05 \), as a function of the strength of a tilted magnetic field with angles \( \theta = 60^\circ \) and \( \phi = 90^\circ \), with Dresselhaus coefficient \( \gamma = 9 \text{ eV } \AA^3 \). Below: Enhancement of the dashed area in the figure above. Avoided crossings between several states due to the spin states not being pure \( S_z \) eigenstates. Linestyles as in figure 3.
and triplet states are the first excited harmonic oscillator, with a tilted magnetic field with angles \( \phi = 90^\circ \) and \( \delta = 1.1 \), as a function of the strength of a tilted magnetic field, with angles \( \theta = 55^\circ \) and \( \phi = 90^\circ \), with Dresselhaus coefficient \( \gamma = 9 \) eV Å\(^3\). The triplet splitting, where a large angle results in a weaker magnetic dependence. The inclination angle is thus rather strictly limited to the 55\(^\circ\) and 60\(^\circ\) found here for the two confinement potentials. The in-plane, azimuthal angle (not known from the experiment [11]) will determine the direction of the elliptic \( \Delta^2 \) potential, and will shift \( m_l \) states somewhat. It will however be a small effect compared to the effect from the Zeeman splitting when operating within the field range studied here. Still, variations of the azimuthal angle shows that a large angle (\( \sim 90^\circ \)) produces an energy splitting that matches the experiment for a rather large inclination angle, approaching the nominal one quoted in the experiment (at least for the cylindrical well potential), while a smaller azimuthal angle only produces a matching energy splitting if the inclination angle is tuned down, away from the experimental results. We have thus chosen to use an azimuthal angle of \( \sim 90^\circ \) in all the calculations. In conclusion we find thus, as was also concluded previously [17], that the splitting in the cylindrical well potential matches the experimentally measured inclination angle better than the harmonic, case although it is still somewhat on the low side.

The azimuthal angle has been chosen to to Our approach has then been to vary it and chosen the one that produces the best energy-splitting match with as high an inclination angle as possible (where the ellipticity introduced through the azimuthal angle dominates over a possible confinement ellipticity). The relaxation rates are strongly dependent on the energy splitting and the main effect from the azimuthal angle is indeed due to its effects on the splitting.

As a final remark regarding the magnetic field inclination angle, we note that both the investigated effective potentials

### 4.1 Singlet–triplet splitting

The experimental results used for comparison [11] come from a system with few parameter details. The inclination of the magnetic field, measured through Shubnikovde Haas oscillations, is 68 ± 5\(^\circ\), with an unknown azimuthal angle. From the singlet–triplet energy splitting measured and the constant splitting at low magnetic fields, we estimate the harmonic oscillator strength to be around \( \hbar \omega = 3 \) meV with an ellipticity \( \delta = 1.1 \), or a well radius of \( r_0 = 44 \) nm with an ellipticity \( \delta = 1.05 \). These are rough estimates, but lead to energy splittings as those seen in figure 5, which are in good agreement with the experiment.

If other parameters are used the energy splitting curve will change, leading to a less good agreement. For example: First, if the dot is widened, i.e. the radius is increased or the oscillator strength is decreased, the Coulomb repulsion between the electrons will decrease. Since the ground state singlet has a larger correlation energy than the excited triplet, it will be effected more by the potential change, resulting in a translation of the energy splitting curve to a lower energy, which will worsen the comparison with the experimental data.

Second, the ellipticity will both affect the energy splitting at zero field, and at what field strength the magnetic field starts to decrease the splitting. As seen in figures 2 and 4, the first excited one-electron state shifts down in energy when an ellipticity is introduced. This reduces the total energy of the triplet state since it is dominated by a configuration where one of the two electrons occupies this orbital. The splitting plateau lasts until the one-electron Zeeman term is large enough to start shifting the energies as discussed in the previous section. The ellipticity \( \delta = 1.1 \) in the harmonic case, and \( \delta = 1.05 \) for the well, were found to give the best agreement with the experimental data.

Third, the dominating magnetic effect in the investigated field range comes from the linear Zeeman splitting which is directly scaled by the inclination angle with a factor of \( \cos \theta \). Unsurprisingly the inclination angle will then also affect the singlet–triplet splitting, where a large angle results in a weaker magnetic dependence. The inclination angle is thus rather strictly limited to the 55\(^\circ\) and 60\(^\circ\) found here for the two confinement potentials. The in-plane, azimuthal angle (not known from the experiment [11]) will determine the direction of the elliptic \( \Delta^2 \) potential, and will shift \( m_l \) states somewhat. It will however be a small effect compared to the effect from the Zeeman splitting when operating within the field range studied here. Still, variations of the azimuthal angle shows that a large angle (\( \sim 90^\circ \)) produces an energy splitting that matches the experiment for a rather large inclination angle, approaching the nominal one quoted in the experiment (at least for the cylindrical well potential), while a smaller azimuthal angle only produces a matching energy splitting if the inclination angle is tuned down, away from the experimental results.

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As a final remark regarding the magnetic field inclination angle, we note that both the investigated effective potentials

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Figure 5. Above: The singlet–triplet splitting of an elliptic harmonic oscillator, with \( \hbar \omega = 2.96 \) meV and \( \delta = 1.1 \), as a function of the strength of a tilted magnetic field, with angles \( \theta = 55^\circ \) and \( \phi = 90^\circ \), with Dresselhaus coefficient \( \gamma = 9 \) eV Å\(^3\). The three triplet states are the first excited \( M_S = +1 \) (solid), \( M_S = 0 \) (dotted) and \( M_S = -1 \) (dashed) states. The circles show the experimental results [11]. Below: The singlet–triplet splitting for an elliptic well, with \( r_0 = 44 \) nm and \( \delta = 1.05 \), as a function of the strength of a tilted magnetic field with angles \( \theta = 60^\circ \) and \( \phi = 90^\circ \), an unpolared state is found to be created in addition to the lowest energy singlet state.

Due to the uncertainty regarding the individual population of the three triplet states, we have chosen to investigate all three states separately rather than averaging over them.

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\( \hbar \omega = 2.96 \) meV and \( \delta = 1.1 \), as a function of the strength of a tilted magnetic field, with angles \( \theta = 55^\circ \) and \( \phi = 90^\circ \), with Dresselhaus coefficient \( \gamma = 9 \) eV Å\(^3\). The triplet splitting, where a large angle results in a weaker magnetic dependence. The inclination angle is thus rather strictly limited to the 55\(^\circ\) and 60\(^\circ\) found here for the two confinement potentials. The in-plane, azimuthal angle (not known from the experiment [11]) will determine the direction of the elliptic \( \Delta^2 \) potential, and will shift \( m_l \) states somewhat. It will however be a small effect compared to the effect from the Zeeman splitting when operating within the field range studied here. Still, variations of the azimuthal angle shows that a large angle (\( \sim 90^\circ \)) produces an energy splitting that matches the experiment for a rather large inclination angle, approaching the nominal one quoted in the experiment (at least for the cylindrical well potential), while a smaller azimuthal angle only produces a matching energy splitting if the inclination angle is tuned down, away from the experimental results. We have thus chosen to use an azimuthal angle of \( \sim 90^\circ \) in all the calculations. In conclusion we find thus, as was also concluded previously [17], that the splitting in the cylindrical well potential matches the experimentally measured inclination angle better than the harmonic, case although it is still somewhat on the low side.

The azimuthal angle has been chosen to to Our approach has then been to vary it and chosen the one that produces the best energy-splitting match with as high an inclination angle as possible (where the ellipticity introduced through the azimuthal angle dominates over a possible confinement ellipticity). The relaxation rates are strongly dependent on the energy splitting and the main effect from the azimuthal angle is indeed due to its effects on the splitting.

As a final remark regarding the magnetic field inclination angle, we note that both the investigated effective potentials
are two dimensional, there might in addition be some effects from the confinement in the $z$-dimension that could better the agreement for both potentials. Inclusion of electron motion in the $z$-direction will allow the tilted field to couple not only to the in plane momentum, but also to the perpendicular one, possibly leading to a smaller singlet–triplet splitting, but has not been investigated further here.

4.2. Relaxation

We now study the relaxation rate as a function of the singlet–triplet splitting from the previous section.

If only the dominating configurations are considered for the triplet states, one would expect drastically different relaxation times [32] for $T_{-1}, T_0, T_{+1}$, however the combination of configuration interaction and spin–orbit mixing has been shown to give much less pronounced differences for $T_{-1}$ and $T_{+1}$ [27]. Our results are slightly different than what has been found in other studies. With only the linear Dresselhaus interaction present we find, in agreement with most of the literature [33, 34], that the $T_0$ state is much more long-lived than the other two, but when also the cubic Dresselhaus is included this is no longer the case. We note that Meunier et al [11] mention that they do not observe any slowly relaxing triplet component in the experiment. More recent experiments [31], also demonstrate shorter than expected $T_0$ lifetimes. One possibility is that this is due to a less efficient population of $T_0$ as discussed above, or that its relaxation is indeed faster than often believed. Below we discuss the relaxation for the the two different confinement potentials in more detail.

4.2.1. Harmonic oscillator confinement. In the case when only the linear Dresselhaus interaction is included, figure 6, upper panel, we find a relaxation maximum around 0.25 meV where the electronic state width matches the wavelength of the phonon. This is in agreement with the experimental results. When the singlet–triplet energy splitting approaches zero equation (19) predicts that the relaxation rate vanishes. The experiment shows indeed a declining rate, but it levels out and approaches a finite value at the smallest measured energy separations. As pointed out by Meunier et al [11] the measurement sensitivity is reduced for energy separations close to degeneracy, leading to larger error bars (not shown in figure 6) for the long life-times in this region. One can also argue that when the the singlet and triplet states are nearly degenerate they mix more efficiently and it might not be enough to consider the Dresselhaus mechanism. There is for example always the atomic spin–orbit coupling that might play a role. At the opposite end, at higher energy separations, both experiment and the calculation show a decreasing trend, as expected from equation (19). This trend is somewhat weaker in the experimental case which is explained by the fact that the maximum energy splitting corresponds to zero magnetic field and in this case it is well known that the dominating spin-relaxation mechanism is due to direct spin-exchange with the surrounding nuclear spins [4], which is not included the present study where we concentrate on the relaxation mechanisms when an external B-field is applied. The relaxation rates calculated for the spin polarized triplet state, $T_{+1}$ and $T_{−1}$, are fairly equal in respect to the energy splitting dependence, with the unpo­larized $T_0$ state mostly exhibiting a significantly smaller rate, as expected [33, 34]. Some structures in the curves seem dependent on the magnetic field rather than the phonon energy, namely the dip in rate for the $T_{+1}$ and $T_{−1}$ around 0.65 meV and the peaks in $T_{+1}$ and $T_0$ around 0.3 meV. These points correspond to magnetic field strengths of 1.1 and 2.1 T, where we also find the crossings of many of the one-electron basis states discussed in section 3.1.

For the calculations where the cubic Dresselhaus has been included, figure 6, lower panel, the parameter $\gamma$ has been tuned down so that the peak in relaxation roughly matches the experimental data. The avoided crossings become even more obvious when the cubic Dresselhaus term is included, where the crossing point of basis states of $\Delta n_l = 3$ becomes a spin hot-spot with a relaxation rate that does not match the

![Figure 6.](image-url)
A thickness corresponding to an oscillator strength enough it should be able to overshadow the cubic contribution. If large energy splitting from the experimental data, a smaller than expected field inclination angle was required. This shifted the $\Delta m_\text{I} = 3$ harmonic crossing point from 1.25 T in the case of a perpendicular magnetic field, to roughly 2.1 T with an inclination of $\theta = 55^\circ$. With the experimentally measured inclination angle of $\theta = 68^\circ$, this crossing should be found around 3.3 T, beyond the investigated magnetic field range. It is possible that some three dimensional, or other, effect not included can change the results to better match the measurements, and through that remove the issue with the cubic Dresselhaus effect spin hot-spot.

4.2.2. Cylinder Well confinement. The relaxation rate in the case of the cylinder well with only linear Dresselhaus interaction is similar to the harmonic case, as seen in figure 7 (upper panel). The peaks in the relaxation rates are due to the extraordinary strong singlet-triplet mixing that occurs close to the avoided crossings in the one-electron spectrum. They are however differently placed than in the harmonic oscillator case. Small structures in the experimental data may be indicative of the underlying one-electron avoided crossings, these are however not as obvious as in our calculations. The overall rate is higher than in the harmonic case when only including the linear interaction, indicating an even smaller Dresselhaus coefficient.

When the cubic effect is included, the avoided crossing points become more prominent since the total interaction is increased. More and larger peaks are present, especially for the $T_0$ and $T_{-1}$ states, to the extent that only the $T_{+1}$ state seems to match the experimental data. The main difference to the harmonic oscillator is the lack of a large surge in relaxation around the point of multiple crossings, which does not exist in the cylindrical well.

The difference in relaxation rate between low and high energy splittings is far larger than in the experimental data, however with a wider and flatter peak than in the harmonic oscillator case. Altering the modelled thickness of the dot and adjusting the Dresselhaus coefficient thereafter can possibly improve this.

5. Conclusion

Slightly different B-field inclination angles are needed to obtain a good fit to the experimental singlet–triplet splittings for the two investigated effective potentials. The hard wall cylindrical well shows better agreement with the nominal value of the experiment than the harmonic oscillator potential, likely due to the stricter electron confinement in the well potential. The still existing discrepancy between the calculated and measured inclination angle can possibly be explained by the lacking finite $z$-potential in the calculations.

Good agreement for the relaxation rate is achieved when only the linear Dresselhaus term is included in the computations, although a Dresselhaus coefficient of $\gamma = 27 \text{ eV } \AA^3$ produces somewhat larger relaxation rates than the experimental data, but it can be tuned down slightly for better agreement. Some magnetic field dependent features can be seen in the relaxation curves, where the avoided crossings...
in the one-particle spectrum occur. These peaks are thin and fairly weak, and may be hard to detect in experiments.

A main finding in the present study is that inclusion of the cubic Dresselhaus term significantly changes the results. For the harmonic confinement the results are clearly at odds with the experiment. For hard wall confinement the spin polarized $T_{2,1}$ still shows some resemblance to the experiment while the other states have several peaks that are not found in the experimental data. The qualitative differences between the two potential shapes indicate that the choice of potential may be very important when modelling few electron quantum dots. The peaks in the relaxation rates are due to the avoided-crossing in the one-electron spectra, where the electron spins become highly mixed due to the cubic Dresselhaus term. These spin hot-spots are gathered together in one large peak in the case of the harmonic oscillator, but spread out when an anharmonicity is introduced. The cubic Dresselhaus term in two dimensional systems has previously been investigated, also with results pointing towards its importance [21], however the most common approach has been to omit it when investigating this specific case [16] or related cases [34]. Altering the $z$-confinement will reduce the influence of the cubic term, but will need to be compensated with a smaller Dresselhaus coefficient.

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