Symmetry induced hole-spin mixing in quantum dot molecules

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We investigate theoretically the spin purity of single holes confined in vertically coupled GaAs/AlGaAs quantum dots (QDs) under longitudinal magnetic fields. A unique behavior is observed for triangular QDs, by which the spin is largely pure when the hole is in one of the dots, but it becomes strongly mixed when an electric field is used to drive it into molecular resonance. The spin admixture is due to the valence band spin-orbit interaction, which is greatly enhanced in C3h symmetry environments. The strong yet reversible electrical control of hole spin suggests that molecules with C3-symmetry QDs, like those obtained with [111] growth, can outperform the usual C2-symmetry QDs obtained with [001] growth for the development of scalable qubit architectures.

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Single spins confined in III-V semiconductor QDs are currently considered as potential qubits for solid-state quantum information processing, which combine fast optical and electrical manipulation with prospects of scalability. In the last years, heavy hole spin qubits have emerged as a robust and long-lived alternative to nuclear spins. The spin admixture is due to the valence band spin-orbit interaction, which is greatly enhanced in C3h symmetry environments. The strong yet reversible electrical control of hole spin suggests that molecules with C3-symmetry QDs, like those obtained with [111] growth, can outperform the usual C2-symmetry QDs obtained with [001] growth for the development of scalable qubit architectures.

Although most works so far have focused on QDs grown along [001], it has been noted that the C2 point symmetry of such systems gives rise to a splitting of bright exciton states which limits the fidelity of optical hole spin preparation. No such splitting is however expected in [111] grown QDs owing to their higher (C3) symmetry, which hence become an alternative worth exploring. Early studies on single (In)GaAs/AlGaAs QDs grown along [111] have revealed that hole states have weak heavy hole-light hole (HH-LH) coupling due to the large aspect ratio, which is a prerequisite to obtain pure hole spins and minimize the impact of hyperfine interaction with the lattice nuclei. In turn, magneto-photoluminescence spectra have reported characteristic differences from [001] grown QDs which were ascribed to the influence of the C3 symmetric symmetry on the hole states.

In this paper, we move forward and study hole states confined in quantum dot molecules (QDM) formed by a pair of vertically stacked QDs grown along [111]. QDMs present several advantages over single QDs for qubit development, including readout independency from initialization and measurement protocols, higher fidelity of spin preparation and enhanced wavelength tunability with external electric fields, which greatly improves prospects of scalability. We consider [111] grown GaAs/AlGaAs QDMs with triangular shape, similar to those reported in Refs. 26,27, adding longitudinal magnetic and electric fields to control the Zeeman splitting and charge localization. Our calculations show that the HH spin purity is high when the hole is confined in individual QDs, but severe spin admixture takes place when the electric field is used to form molecular orbitals. The spin admixture follows from the formation of orbitals with approximate C3h point group symmetry, which enables otherwise forbidden spin-orbit interactions (SOI). The symmetry-induced SOI does not mix nearby Zeeman sublevels, but it couples bonding and antibonding molecular states split by the tunneling energy. This is in sharp contrast with usual [001] grown QDMs, with C2h symmetry, where tunneling is normally a spin-preserving process.

Since the activation of SOI mechanisms is generally associated with a descent of the system symmetry (e.g., system and bulk inversion asymmetry, QDM misalignment), the enhancement of SOI for C3 QDs is apparently counterintuitive. Yet, we observe strong spin admixture between hole states over 1 meV apart, which is 2.5 times greater than the largest spin-orbit anticrossing measured in [001] grown QDMs. We provide an explanation through group theory analysis of the multi-band k·p Hamiltonian for holes, showing this is an exclusive property of C3 systems, and discuss the implications of these findings for the development of hole spin qubit architectures.

The Hamiltonian we use to describe hole states reads:

\[ \mathbb{H} = \mathbb{H}_{BF} + \mathbb{H}_{B} + \mathbb{H}_{strain} + (V(r) + e(\phi_pz(r) - Fz)) \mathbb{I}. \]  

(1)

Here \( \mathbb{H}_{BF} \) is the four-band Burt-Foreman Hamiltonian for [111] grown zinc-blende crystals, which considers HH-LH subband coupling as in the Luttinger model but including position-dependent effective masses. \( \mathbb{H}_{B} \) represents the terms coming from a magnetic field applied along the growth (z) direction. \( \mathbb{H}_{strain} \) is the strain Hamiltonian, \( V(r) \) the band-offset potential, \( e \) the hole charge, \( \phi_pz \) the piezoelectric potential, \( F \) an axial electric field and \( \mathbb{I} \) a rank-4 identity matrix. Further details on the Hamiltonian can be found in the Supplemental Material. Hamiltonian (1) is solved numerically after obtaining the strain tensors and piezoelectric fields using the Comsol package. The eigenstates are Luttinger
spinors of the form:

\[ |n\rangle = \sum_{J_z=-3/2}^{3/2} f_{J_z}^n(r) |J = 3/2, J_z\rangle, \quad (2) \]

where \( f_{J_z}^n(r) \) is the envelope function associated to \( |J = 3/2, J_z\rangle \), the periodic function with Bloch angular momentum \( J_z \). \( J_z = \pm 3/2 \) correspond to spin up and spin down HH components, while \( J_z = \pm 1/2 \) correspond to LH components. The expectation value \( \langle J_z \rangle = \sum J_z f_{J_z}^n(r) f_{J_z}^* \) can be taken as a measure of the hole spin purity, with \( \langle J_z \rangle \approx \pm 3/2 \) indicating nearly pure HH spin up (\( \uparrow \)) or spin down (\( \downarrow \)) states.

For our calculations we consider pyramidal GaAs/Al_{0.3}Ga_{0.7}As QDMs with triangular confinement, similar to those obtained by metallorganic vapor deposition. The vertically stacked QDs are separated by a barrier of thickness \( d = 2 \) nm, although they are interconnected by a thin Al_{0.05}Ga_{0.95}As vertical quantum wire which enhances tunnel coupling -see structure in Fig. 1(a)-(c). A weak magnetic field of \( B = 0.2 \) T is applied along the coupling direction. Figure 1(a) shows the energy of the first four hole states under the influence of a vertical electric field \( F \). At \( F = 11 \) kV/cm we see two Zeeman-split doublets. The upper one (states \( |1\rangle \) and \( |2\rangle \)) corresponds to the main component of the hole spinor in the top QD, which is slightly bigger than the bottom QD. In turn, the lower doublet (states \( |3\rangle \) and \( |4\rangle \)) has the main component in the bottom dot. Since the increasing electric field favors the occupation of the bottom dot, a charge transfer anticrossing takes place at \( F = 14.7 \) kV/cm (resonant electric field, \( F_r \)), where bonding and antibonding molecular orbitals are formed. The behavior closely resembles that of [001] grown QDMs except for one anomaly: the Zeeman splitting of both doublets is quenched near the resonant electric field, see Fig. 1(a) inset.

The Zeeman splitting suppression can be seen as a vanishing effective g-factor. Unlike in previous reports, however, the origin of this effect cannot be ascribed to the different g-factor of the QD and barrier materials as the QDs and the vertical wire connecting them have similar composition. Because electrically tunable g-factors are of interest for spin manipulation, we further investigate into the origin of this phenomenon. Fig. 1(b) shows the hole Bloch angular momentum expectation value of the four states under consideration. As can be seen, away from the resonant field, \( \langle J_z \rangle \) gradually approaches \( \pm 3/2 \), indicating that the hole states confined in individual QDs are nearly HH states with fairly pure spin. In the vicinity of \( F_r \), however, \( \langle J_z \rangle \approx 0 \), which means that for molecular states the spin becomes completely mixed.

For comparison, since [111] grown QDs normally have either triangular or hexagonal shape, in Fig. 1(c)-(d) we study a QDM formed by hexagonal QDs. One can see that no spin mixing takes place near \( F_r \) in this case. Actually, the behavior is now the same as observed in vertically aligned [001] grown QDMs with \( C_{2h} \) symmetry QDs, where tunneling is a spin preserving process. It follows that the strong spin mixing in Fig. 1(b) is not a consequence of the [111] crystal orientation, but rather of the triangular envelope confinement. In fact, we also observe it for triangles grown along [001], see Supplementary Material. It does not result from the presence or absence of strain either, as similar results are obtained using lattice mismatched materials such as InAs/GaAs. Likewise, it is not induced by the magnetic field, as \( H_B \) is a diagonal term which does not couple different spinor component. It does not take place in single QDs either. It is an exclusive property of QDMs with triangular confinement.

Further insight into the hole spin mixing mechanism is obtained in Fig. 2 which plots the weight of the four spinor components corresponding to each of the states \( |1\rangle \) to \( |4\rangle \) of the triangular QDM. Two conclusions can be extracted: (i) LHs play a minor role in all cases, the mixing is essentially between HH components with orthogonal spin projections \( J_z = 1/2 \); (ii) states \( |1\rangle \) and \( |4\rangle \) (panels (a) and (d)) seem to exhibit complementary behavior, and so do \( |2\rangle \) and \( |3\rangle \) (panels (b) and (d)). This suggests that the spin mixing is due to independent interactions between \( |1\rangle \) and \( |4\rangle \) on the one hand, and \( |2\rangle \) and \( |3\rangle \) on the other.

To understand the origin of the HH spin mixing we resort to a point group theory analysis. Eq. 1 Hamiltonian can be simplified as \( H \approx H_{L,K} + H_B + (V(r) + eFz) \), where we have disregarded strain terms -which are weak for GaAs/AlAs heterostructures- and approximated \( H_{BF} \) by the (con-
stant mass) Luttinger-Kohn Hamiltonian:

$$\mathcal{H}^{[111]}_{LK} = -\frac{1}{2} \begin{pmatrix} \hat{P} + \hat{Q} - \hat{S} & \hat{R} & 0 \\ -\hat{S}^\dagger & \hat{P} - \hat{Q} & \hat{R} \\ 0 & \hat{R}^\dagger & \hat{S}^\dagger \end{pmatrix}$$

(3)

with

$$\hat{P} + \hat{Q} = (\gamma_1 \pm \gamma_3)(k_x^2 + k_y^2) + (\gamma_1 \mp 2\gamma_3)k_z^2$$

$$\hat{R} = -\frac{1}{\sqrt{3}}(\gamma_2 + 2\gamma_3)k_z^2 + \frac{2\sqrt{3}}{\gamma_3}(\gamma_2 - \gamma_3)k_+k_z$$

(4)

$$\hat{S} = -\frac{2\sqrt{3}}{\gamma_3}(\gamma_2 - \gamma_3)k_+^2 + \frac{\sqrt{3}}{\gamma_3}(2\gamma_2 + \gamma_3)k_-k_z$$

where $\gamma_1, \gamma_2, \gamma_3$ are the Luttinger parameters and $k_{\pm} = k_x \pm ik_y$. One can then see that $\mathcal{H}$ has $C_3$ point symmetry set by the confining potential $V(r)$. Near the resonant electric field, however, an additional approximate symmetry must be considered. Even if the two QDs forming the QDM are not identical, the electric field restores an effective parity symmetry, the bonding and antibonding HH molecular orbitals forming even and odd functions with respect to a mirror plane in between the QDs. The corresponding point group is then $C_{3h}$. Note that for $\mathcal{H}^{[111]}_{LK}$ to hold exact $C_{3h}$ symmetry, we need to impose the so-called axial approximation ($\gamma_2 = \gamma_3$), which is actually valid for many III-V materials, such as GaAs. Actually, we do not impose exact symmetry in the numerical calculations and nevertheless, the obtained results reveal, as expected, a high degree of symmetry.

The anticrossing of Fig. 1(a) can be rationalized considering the symmetry of the hole spinors in the double group $C_{3h}$. Within this group [1] and [4] have $E_{-3/2}$ symmetry, while [2] and [3] have $E_{3/2}$ symmetry. The different symmetry of [1] and [2] ([3] and [4]) explains the lack of interaction within the Zeeman doublets, inspite of the quasi-degeneracy. It also becomes clear why [1] and [4] ([2] and [3]) interact separately, as observed in Fig. 2.

The above picture differs from the widely studied [001] grown QDMs with circular confinement, where the symmetry of all four spinorial states involved in the molecular anticrossing is different, which results in the absence of interaction and hence spin-preserving tunneling. Sizeable hole spin mixing has been observed only in QDMs with significant misalignment, because the symmetry is then completely reduced ($C_1$ point group). Nevertheless, the largest spin anticrossing measured for such system is 0.4 meV, corresponding to InAs QDMs with anomalously large lateral offset. This is 2.5 times smaller than the 1 meV gap we estimate in Fig. 1 and 6 times smaller than the 2.5 meV we predict for InAs/GaAs QDMs.

To explain the unusual strength of the spin-orbit coupling in triangular QDMs, we can examine the envelope symmetry of $\mathcal{H}^{[111]}_{LK}$ (within axial approximation) and the ensuing eigenfunctions. In the $C_{3h}$ point group, the terms
of Eq. (3) form basis of the following irreducible representations:

\[ \Gamma_{H_{kk}} = \begin{pmatrix} A' & E''_0 & E'_+ & 0 \\ E''_+ & A' & 0 & E''_+ \\ E'_- & 0 & A' & E''_0 \\ 0 & E'_- & E'_+ & A' \end{pmatrix}. \]  

(5)

A remarkable consequence is that for any hole state \( |n⟩ \), the envelope functions of the spin up and down HH components, \( f^u_{3/2} \) and \( f^d_{3/2} \) in Eq. (2), must have the same symmetry except for the even/odd parity (e.g. \( A' \) and \( A'' \)). This is a key factor in determining the strength of the spin admixture, as we show in the perturbative analysis below.

Hamiltonian \( \mathcal{H} \) can be split as a sum of diagonal and off-diagonal terms, \( \mathcal{H} = \mathcal{H}_0 + \mathcal{H}' \), the latter being responsible for band coupling. If we disregard \( \mathcal{H}' \), the levels anticrossing at \( F_t \) are those represented at the top of Fig. 3(a), namely two Zeeman doublets formed by HHs with opposite spin \( J_z = \pm 3/2 \) but the same rotational symmetry (\( A \)) and parity (\( ' \) or \( '' \)). Each doublet is split by a Zeeman term \( \Delta B \) and separated from each other by an amount \( \Delta t \), where \( t \) is the HH tunneling integral. Considering \( \mathcal{H}' \) as a perturbative term, the mixing between the spin up ground state \( |k^{(0)}⟩ = (A', +3/2) \) and any spin down HH state \( |j^{(0)}⟩ \) is given by:

\[ |k^{(2)}⟩ = \sum_{i \neq k} \left( \sum_{j \neq k} \langle i|\mathcal{H}'|j^{(0)}⟩ \langle j^{(0)}|\mathcal{H}'|k^{(0)}⟩ \right) |i⟩ \]

(6)

where \( |j^{(0)}⟩ \) is the \( j \)-th intermediate state and \( E_j^{(0)} \) its corresponding energy. Notice that the strength of coupling is inversely proportional to \( \Delta E_{hh} = E_j^{(0)} - E_i^{(0)} \), i.e. the energy difference between the spin up and down HH states.

As explained with detail in the Supplemental Material, the symmetry of \( \mathcal{H}' \) operators, off-diagonal terms of Eq. (4), translates into selection rules which make the numerator of Eq. (6) vanish for all except the two paths plotted with thick arrows in Fig. 3(a). Both paths involve excited LHs as intermediate states, and the spin down HH is \( |i⟩ = (A'', -3/2) \), i.e. a state participating in the molecular anticrossing. This implies \( \Delta E_{hh} \) is small (few meV at most), and is in contrast with other point symmetries, where selection rules lead to coupling with higher excited HH states, so that \( \Delta E_{hh} \) is much larger. For example, if we consider QDMs with circular QDs (point group \( C_{\infty h} \)), the HH components coupled by \( \mathcal{H}' \) no longer have the same rotational symmetry, but they differ by three quanta of azimuthal angular momentum \( M_z \). Consequently, there is no coupling between the \( M_z = 0 \) HHs forming the molecular anticrossing. Having a QDM structure is also essential, as then \( A' \) and \( A'' \) are roughly split by the tunneling energy \( \Delta t \), which can be made small enough for the SOI to be efficient. By contrast, in single QDs the strong vertical confinement would lead to several meV splitting.

The suppression of the Zeeman splitting observed in Fig. 3(a) can be also understood from the perturbative analysis. As indicated in Fig. 3(b), the band coupling occurs between HH states belonging to different doublets. Because \( \Delta E_{hh} \) is smaller for the innermost states \( (2t - \Delta B) \) than for the outermost ones \( (2t + \Delta B) \), the interaction is stronger, leading to an effectively reduced Zeeman splitting, \( \Delta B \).

It is clear from the discussions above that tunneling must be an important parameter to control the strength of the spin mixing. One might then expect that spin mixing is enhanced for long interdot distances \( d \), when tunneling energy \( t \) is small. Fig. 3(d) shows the spin purity of the ground state HH for QDMs with different \( d \) at resonant electric field. Interestingly, the maximum spin mixing is found at intermediate distances, \( d \approx 2 \text{nm} \). This follows from the characteristic, non-monotonous decay of hole tunneling in QDMs. As shown in the schematic of Fig. 3(c), there is a critical distance \( d_c \) where bonding and antibonding hole states are reversed. At this point, \( t \) has a relative minimum combined with large wave function delocalization, which enables the strong spin mixing. For \( d < d_c \), \( t \) increases rapidly, reducing the interaction. For \( d > d_c \), \( t \) eventually decreases but so does the wave function delocalization. As a result we gradually retrieve the single QD limit, where spin mixing is weak.

Electrical control of hole spins in QDMs has been proposed as a key ingredient for scalable qubit architectures. So far, however, only [001] grown QDMs have been considered, where the main source of spin mixing was misalignment between the vertically stacked dots which is a difficult parameter to regulate experimentally. The \( C_{3h} \)-symmetry-induced spin mixing described here arises as a more robust and manageable mechanism. It can also help increase the fidelity of spin control gate operations, as this requires the spin mixed states to (i) be energetically well resolved, and (ii) be able to form indirect excitons with large optical dipole strength. As for (i), we predict strong mixing between states 1 meV away from each other, larger than any previous measurement. As for (ii), unlike in misaligned QDMs, the spin mixing we describe is strong at the resonant electric field, where direct and indirect excitons have comparable optical strength. Another advantage is the possibility to use weak magnetic fields, which limits the influence of the g-factor inhomogeneity of different QDs in the qubit scaling.

In summary, we have shown that triangular QDs can be used to build QDMs with electrically controllable hole spin. The hole spin is well defined inside the individual QDs, but the formation of delocalized molecular orbitals with \( C_{3h} \) symmetry enables SOI induced mixing with unprecedented strength. The reversible control, the strength of the interaction and the robust nature of the spin mixing mechanism imply that holes in triangular QDMs, like those obtained with [111] growth, form a promising system for quantum information processing with some advantages as compared to circular [001] grown QDMs.
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Supplemental Material for: Symmetry induced hole-spin mixing in quantum dot molecules

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I. THEORETICAL MODEL AND PARAMETERS

The Hamiltonian we use to describe hole states in QDs grown along the [001] direction is (atomic units):

\[
H^{[001]} = H^{[001]}_{BF} + H^{[001]}_B + H^{[001]}_{strain} + (V(r) + e(\phi_{pz}^{[001]}(r) - F z)) \mathbb{I}.
\]  

(1)

Here \(H^{[001]}_{BF}\) is the four-band Burt-Foreman Hamiltonian for zinc-blende crystals, which considers HH-LH subband coupling and position-dependent effective masses:

\[
H^{[001]}_{BF} = \begin{pmatrix}
\frac{3}{2} \sum_i k_i^2 - M & \frac{L - M}{3} & \frac{i}{3} (k_x(N - N')k_y - k_y(N - N')k_x) & 0 \\
\frac{L - M}{3} & \frac{3}{2} \sum_i k_i^2 - M & 0 & \frac{i}{3} (k_y(N - N')k_x - k_x(N - N')k_y) \\
\frac{i}{3} (k_x(N - N')k_y - k_y(N - N')k_x) & 0 & \frac{3}{2} \sum_{i < j} (k_i(N + N')k_j + k_j(N + N')k_i) & 1 \\
0 & \frac{i}{3} (k_y(N - N')k_x - k_x(N - N')k_y) & 1 & \frac{3}{2} \sum_{i < j} (k_i(N + N')k_j + k_j(N + N')k_i)
\end{pmatrix}
\]  

(2)

with \(\mathbb{I}\) a rank-4 identity matrix, \(\{A, B\} = \frac{1}{2} (AB + BA)\), \(X_\pm = X_x \pm X_y\) and \(\mathbb{J}_i\) the angular momentum \(i\)-component matrix:

\[
\mathbb{J}_x = \begin{pmatrix}
0 & \frac{\sqrt{3}}{2} & 0 & 0 \\
\frac{\sqrt{3}}{2} & 0 & 1 & 0 \\
0 & 1 & 0 & \frac{\sqrt{3}}{2} \\
0 & 0 & \frac{\sqrt{3}}{2} & 0
\end{pmatrix}, \quad \mathbb{J}_y = \begin{pmatrix}
0 & -i \frac{\sqrt{3}}{2} & 0 & 0 \\
i \frac{\sqrt{3}}{2} & 0 & -i & 0 \\
0 & i & 0 & -i \frac{\sqrt{3}}{2} \\
0 & 0 & -i \frac{\sqrt{3}}{2} & 0
\end{pmatrix}, \quad \mathbb{J}_z = \begin{pmatrix}
\frac{3}{2} & 0 & 0 & 0 \\
0 & \frac{1}{2} & 0 & 0 \\
0 & 0 & -\frac{1}{2} & 0 \\
0 & 0 & 0 & -\frac{3}{2}
\end{pmatrix}
\]  

(3)

By inserting eq. (3) in eq. (2) we end up with the following matrix representation:

\[
H^{[001]}_{BF} = - \begin{pmatrix}
\hat{P}' & \hat{S}^- & -\hat{R} & 0 \\
\hat{S}^+ & \hat{P}' & -\hat{C} & -\hat{R} \\
-\hat{R}^\dagger & -\hat{C}^\dagger & \hat{P}' & -\hat{S}^+ \\
0 & -\hat{R}^\dagger & -\hat{S}^+ & \hat{P}'
\end{pmatrix}
\]  

(4)
where,

\[ P' = \frac{1}{2}(k_x(L + M)k_x + k_y(L + M)k_y + k_z(2M)k_z) + \frac{i}{2}(k_x(N - N')k_y - k_y(N - N')k_x) \]

\[ P'' = \frac{1}{6}(k_x(L + 5M)k_x + k_y(L + 5M)k_y + 2k_z(2L + M)k_z) + \frac{i}{6}(k_x(N - N')k_y - k_y(N - N')k_x) \]

\[ P''^* = \frac{1}{6}(k_x(L + 5M)k_x + k_y(L + 5M)k_y + 2k_z(2L + M)k_z) - \frac{i}{6}(k_x(N - N')k_y - k_y(N - N')k_x) \]

\[ P'^* = \frac{1}{2}(k_x(L + M)k_x + k_y(L + M)k_y + k_z(2M)k_z) - \frac{i}{2}(k_x(N - N')k_y - k_y(N - N')k_x) \]

\[ R = \frac{1}{2\sqrt{3}}[k_x(L - M)k_x - k_y(L - M)k_y - i(k_x(N + N')k_y + k_y(N + N')k_x)] \]

\[ R^\dagger = \frac{1}{2\sqrt{3}}[k_x(L - M)k_x - k_y(L - M)k_y + i(k_y(N + N')k_x + k_x(N + N')k_y)] \]

\[ S_- = -\frac{1}{\sqrt{3}}[k_xNk_z + k_zN'k_-] \]

\[ S^\dagger_- = -\frac{1}{\sqrt{3}}[k_zNk_+ + k_+N'k_z] \]

\[ S_+ = -\frac{1}{\sqrt{3}}[k_xNk_+ + k_+N'k_z] \]

\[ S^\dagger_+ = -\frac{1}{\sqrt{3}}[k_zNk_- + k_-N'k_z] \]

\[ C = -\frac{1}{3}(k_z(N - N')k_- - k_-(N - N')k_z) \]

\[ C^\dagger = -\frac{1}{3}(k_+(N - N')k_z - k_z(N - N')k_+) \]

(5)

with \( L, M, N, N' \) being the Stravinou-van Dalen mass parameters.

By setting the parameters constant, Eq. (I) Hamiltonian turns into the Luttinger Kohn Hamiltonian: The Stravinou-van Dalen parameters are then related to the Luttinger parameters \( \gamma_1, \gamma_2, \gamma_3 \) as \( L - M = -3\gamma_2, 3L + M = -2\gamma_1 - 5\gamma_2, N - N' = 1 + \gamma_1 - 2\gamma_2 - 3\gamma_3 \) and \( N + N' = -3\gamma_3 \).

\[ \mathbb{H}^{[001]}_B \] represents the terms coming from a magnetic field applied along the growth (\( z \)) direction, \( B^z \)

\[ \mathbb{H}_B = -\left( \frac{B^2}{8}(x^2 + y^2) + \frac{B}{2}(xk_y - yk_x) \right) \left( (\gamma_1 - \frac{5}{2}\gamma_2)\mathbb{I} + \gamma_2\mathbb{J}_z^2 \right) + \kappa\mu_BB\mathbb{J}_z. \]

(6)

with \( \mathbb{I} \) a rank-4 identity matrix, \( \mathbb{J}_z \) the angular momentum \( z \)-component matrix (\( J = 3/2 \)), \( \kappa = 4/3 \) the effective g-factor and \( \mu_B \) the Bohr magneton.

\[ \mathbb{H}^{[001]}_{\text{strain}} \] is the strain Hamiltonian, formally identical to Eq. (I) with the products \( k_i k_j \) replaced by the strain tensor component \( \epsilon_{ij} \). \( e \) and \( \phi_{pz} \) are the hole charge and strain-induced piezoelectric potential -which is also diagonal-, \( F \) an axial electric field and \( V(\mathbf{r}) \) the band-offset potential. For the triangular QDMs we use a structure like that obtained by metallorganic vapor deposition. The structure is illustrated in Fig. (I) using similar geometry and composition to the pyramidal QDMs of Refs. Thus, the QDs are made of GaAs, the barrier of Al\(_{0.3}\)Ga\(_{0.7}\)As and the vertical wire connecting the dots of
Al\textsubscript{0.05}Ga\textsubscript{0.95}As. We note that the vertical wire plays no critical role in the phenomena we describe. The robustness of the results are checked by carrying out additional calculations with triangular round edges and also breaking the symmetry with two round and a sharp edge (not shown). For comparison, we also study QDMs made of hexagonal QDs. The sides of the hexagons are taken to have the same dimensions as those in Fig. 1, and a hexagonal wire is used which preserves the \( C_6 \) symmetry.

For [111] grown QDs, the hole Hamiltonian becomes:

\[
H^{[111]} = H^{[111]}_{BF} + H^{[111]}_B + H^{[111]}_{\text{strain}} + (V(r) + e (\phi^{[111]}_{pz}(r) - F z)) \mathbb{I}.
\]  

(7)

where we have obtained \( H^{[111]}_{BF} \) and \( H^{[111]}_{\text{strain}} \) from Eq. (2) by writing \( k_i \) and \( J_i \) appearing in the Hamiltonian as a function of the new coordinates \( k'_i \) and \( J'_i \) according to,

\[
k = M k' \quad \quad J = M J'
\]

(8)

where \( M \) is the rotation matrix\(^7\)

\[
M = \begin{pmatrix}
\frac{1}{\sqrt{6}} & -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{3}} \\
\frac{1}{\sqrt{6}} & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{3}} \\
-\frac{\sqrt{2}}{3} & 0 & \frac{1}{\sqrt{3}}
\end{pmatrix}.
\]

(9)

Once we reach the new matrices, the prime is removed from \( k_i \) for the sake of a better presentation. The Burt-Foreman Hamiltonian in the [111] direction then reads:

\[
H^{[111]}_{BF} = -\frac{1}{2} \begin{pmatrix}
\hat{P}' & -\hat{S}_- & \hat{R} & 0 \\
-\hat{S}_- & \hat{P}' & \hat{C} & \hat{R} \\
\hat{R} & \hat{C} & \hat{P}' & \hat{S}_+ \\
0 & \hat{R} & \hat{S}_+ & \hat{P}'
\end{pmatrix}
\]

(10)
where,

\[
\hat{P}^r = [k_x(\gamma_1 + \gamma_3)k_x + k_y(\gamma_1 + \gamma_3)k_y + k_z(\gamma_1 - 2\gamma_3)k_z] - i [k_x(\gamma_1 - 2\gamma_2 - 3\gamma_3)k_y - k_y(\gamma_1 - 2\gamma_2 - 3\gamma_3)k_x]
\]

\[
\hat{P}^\Lambda = [k_x(\gamma_1 - \gamma_3)k_x + k_y(\gamma_1 - \gamma_3)k_y + k_z(\gamma_1 + 2\gamma_3)k_z] - \frac{1}{3} [k_x(\gamma_1 - 2\gamma_2 - 3\gamma_3)k_y - k_y(\gamma_1 - 2\gamma_2 - 3\gamma_3)k_x]
\]

\[
\hat{S}^- = -\frac{1}{\sqrt{3}} \{ (k_-k_z - k_zk_-) + \sqrt{2} [k_x(\gamma_2 - \gamma_3)k_x - k_y(\gamma_2 - \gamma_3)k_y + i (k_x(\gamma_2 - \gamma_3)k_y + k_y(\gamma_2 - \gamma_3)k_x)]
\]

\[
\hat{S}^+ = -\frac{1}{\sqrt{3}} \{ (k_+k_z - k_zk_+) + \sqrt{2} [k_x(\gamma_2 - \gamma_3)k_x - k_y(\gamma_2 - \gamma_3)k_y - i (k_x(\gamma_2 - \gamma_3)k_y + k_y(\gamma_2 - \gamma_3)k_x)]
\]

\[
\hat{R} = -\frac{1}{\sqrt{3}} \{ k_-(\gamma_2 + 2\gamma_3)k_- - \sqrt{2} [k_x(\gamma_2 - \gamma_3)k_+ + k_y(\gamma_2 - \gamma_3)k_z] \}
\]

\[
\hat{\mathcal{O}} = -\frac{2}{3} [k_z(\gamma_1 - 2\gamma_2 - 3\gamma_3)k_- - k_-(\gamma_1 - 2\gamma_2 - 3\gamma_3)k_z]
\]

(11)

The $H_B$ Hamiltonian rotation requires a special care. It was obtained in\(^2\) by initially disregarding the effect of the remote bands and later enclosing it, replacing the mass by the effective mass. Then, since we rotate the crystalline structure keeping the axes fixed and then the Bloch functions, the form of $H_B$ does not change. However, the effect of the remote bands does change, so that one should replace $\gamma_2$ by $\gamma_3$ in the expression of the effectives masses. This modification must be introduced in $H_B^{[001]}$ to reach $H_B^{[111]}$.

To calculate the strain $\epsilon_{ij}$ in $H_{strain}^{[111]}$, we take the elastic constants of $[001]$ grown heterostructures, $C^{[001]}$, and rotate the axes. The resulting elastic constants $C^{[111]}$ are related to $C^{[001]}$ by:

\[
C^{[111]}_{ijkl} = \sum_{a,b,c,d} M_{ia} M_{jb} M_{kc} M_{ld} C^{[001]}_{abcd}
\]

(12)

Likewise, for the piezoelectric potential we rotate the axes and obtain:

\[
p_i = \sum_k e_{ij}^{[111]} \epsilon_{jk}
\]

(13)

with $e_{ij}^{[111]} = \sum_{a,b,c} M_{ia} M_{jb} M_{kc} e_{abc}^{[001]}$.

Hamiltonians (11) and (17) are solved numerically after obtaining the strain tensors and piezoelectric fields using the Comsol 4.2 package. The material parameters of GaAs, AlAs and InAs are taken from Ref\(^8\), except for the crystal density, dielectric constant and piezoelectric coefficient, which are obtained from Ref\(^9\). Linear interpolations are used for all alloys parameters. Luttinger parameters are inferred from the linearly interpolated masses.
II. CHARACTER TABLE AND PRODUCT TABLE FOR THE DOUBLE GROUP $\tilde{C}_{3h}$

The character table we use is:

\[
\begin{array}{cccccccccccccc}
\tilde{C}_{3h} & E & C_3^+ & C_3^- & S_3^+ & S_3^- & \tilde{E} & \tilde{C}_3^+ & \tilde{C}_3^- & \tilde{S}_3^+ & \tilde{S}_3^- & \text{basis} \\
A' & 1 & 1 & 1 & 1 & 1 & 1 & 1 & 1 & 1 & 1 & J_z \\
E_+ & 1 & \omega & \omega^* & 1 & \omega & \omega^* & 1 & \omega & \omega^* & 1 & x + iy \\
E_- & 1 & \omega^* & \omega & 1 & \omega^* & \omega & 1 & \omega^* & \omega & 1 & x - iy \\
A'' & 1 & 1 & -1 & -1 & -1 & 1 & 1 & 1 & -1 & -1 & z \\
E'_+ & 1 & \omega & \omega^* & -1 & -\omega & -\omega^* & 1 & \omega & \omega^* & -1 & -\omega & -\omega^* & J_x + iJ_y \\
E'_- & 1 & \omega^* & \omega & -1 & -\omega^* & -\omega & 1 & \omega^* & \omega & -1 & -\omega^* & -\omega & J_x - iJ_y \\
E_{-1/2} & 1 & -\omega & \omega^* & i & -i\omega & i\omega^* & -1 & \omega & -\omega^* & -i & i\omega & -i\omega^* & J_{-1/2} \\
E_{1/2} & 1 & -\omega^* & \omega & -i & i\omega & -i\omega^* & -1 & \omega^* & -\omega & i & -i\omega^* & i\omega & J_{1/2} \\
E_{5/2} & 1 & -\omega & \omega^* & -i & i\omega & -i\omega^* & -1 & \omega & -\omega^* & i & -i\omega & i\omega^* & \\
E_{-5/2} & 1 & -\omega^* & \omega & i & -i\omega & i\omega & -1 & \omega & -\omega^* & -i & i\omega^* & -i\omega & \\
E_{-3/2} & 1 & 1 & -1 & 1 & -1 & 1 & -1 & 1 & -1 & i & -i & i & \\
E_{3/2} & 1 & -1 & 1 & -i & i & -i & -1 & 1 & -1 & i & -i & i & \\
\end{array}
\]

where $\omega = e^{i\frac{2\pi}{3}}$ and $J_i$ is the $i$-th component of the angular momentum.

The table of products of $\tilde{C}_{3h}$ irreducible representations is:
III. SYMMETRY OF THE HAMILTONIAN AND THE WAVE FUNCTIONS

The employed $H_{BF}$ Hamiltonian is mass-position-dependent. Actually, in our system the mass parameters are constant within every domain, and have a sudden jump at the edge between neighboring domains. For the sake of easiness, to discuss on symmetry, we consider the Luttinger-Kohn constant-mass parameters $H_{LK}$ limit of $H_{BF}$.

The symmetry of the $H_{LK}$ Hamiltonian (eqs. 4, 10 employing constant mass parameters) including a triangular confining potential and an axial magnetic field is $C_3$. However, within the axial approximation $10$ ($\gamma_2 = \gamma_3$) it reaches $C_{3h}$. The symmetries of their matrix element operators can then be calculated from the above character table and the expressions on eqs. 5, 11 (assuming constant mass parameters):

$$
\Gamma_{H_{LK}} = \begin{pmatrix}
A' & E'' & E_+ & 0 \\
E'' & A' & 0 & E_+ \\
E_+ & 0 & A' & E'' \\
0 & E_+ & E'' & A'
\end{pmatrix}.
$$

Accordingly, the symmetry of the envelope bonding/anti-bonding ground state functions
we have:

\[
\begin{pmatrix}
A' (b) \\
E''_+ (a) \\
E'_- (b) \\
A'' (a)
\end{pmatrix}
\quad \text{and} \quad
\begin{pmatrix}
A'' (a) \\
E'_+ (b) \\
E''_- (a) \\
A' (b)
\end{pmatrix}
\]

where the labels \((a), (b)\) indicates the bonding/anti-bonding character of the components. These envelope components, combine with the Bloch functions yielding the wave function.

The Bloch functions are built as the symmetry-adapted product of \(J = 1\) angular momentum functions and the \(J = 1/2\) spin functions. However, the presence of the mirror symmetry \(\sigma_h\) allows to employ bonding (\(\chi(\sigma_h) = 1\)) and anti-bonding (\(\chi(\sigma_h) = -1\)) \(J = 1\) angular momentum functions. For example, employing anti-bonding angular momentum functions we have:

\[
|3/2, 3/2\rangle = -\frac{1}{\sqrt{2}}(X + iY) \uparrow \quad \quad \quad |3/2, -3/2\rangle = \frac{1}{\sqrt{2}}(X - iY) \downarrow \quad \quad \quad |3/2, 1/2\rangle = \sqrt{\frac{3}{8}}|Z \uparrow\rangle - \frac{1}{\sqrt{8}}(X + iY) \downarrow \quad \quad \quad |3/2, -1/2\rangle = \sqrt{\frac{3}{8}}|Z \downarrow\rangle + \frac{1}{\sqrt{8}}(X - iY) \uparrow
\]

The bonding \(J = 1\) angular momentum functions are like the antibonding where \(X, Y,\) and \(Z\) are replaced by \(J_x, J_y,\) and \(J_z\). As a result, the table of products allows us to determine the symmetries of the Bloch functions:

\[
\begin{align*}
|3/2, 3/2^a\rangle & \to E_{3/2} & |3/2, 3/2^b\rangle & \to E_{-3/2} \\
|3/2, 1/2^a\rangle & \to E_{-5/2} & |3/2, 1/2^b\rangle & \to E_{1/2} \\
|3/2, -1/2^a\rangle & \to E_{5/2} & |3/2, -1/2^b\rangle & \to E_{-1/2} \\
|3/2, -3/2^a\rangle & \to E_{-3/2} & |3/2, 3/2^b\rangle & \to E_{3/2}
\end{align*}
\]

The Bloch functions symmetries required to combine with the envelope components eq. \(17\) (left) yielding \(E_{3/2}\) and \(E_{-3/2}\), and those required to combine with eq. \(17\) (right) yielding \(E_{3/2}\) and \(E_{-3/2}\), are:

\[
\begin{pmatrix}
E_{3/2} \\
E_{-5/2} \\
E_{5/2} \\
E_{-3/2}
\end{pmatrix}
\quad \text{and} \quad
\begin{pmatrix}
E_{-3/2} \\
E_{1/2} \\
E_{-1/2} \\
E_{3/2}
\end{pmatrix}
\quad \text{and} \quad
\begin{pmatrix}
E_{3/2} \\
E_{-5/2} \\
E_{5/2} \\
E_{-3/2}
\end{pmatrix}
\]

\[\text{(20)}\]
IV. SPIN MIXING IN QDMS GROWN ALONG [001]

In the paper we have considered triangular QDs grown along the [111] direction because they are formed naturally in that direction. The physics leading to hole spin mixing is however connected with the envelope symmetry, and does not depend on the crystal orientation. To illustrate this point, in Fig. 2 we plot the expectation value of the Bloch angular momentum \( \langle J_z \rangle \) as a function of the interdot distance \( d \) for the upper Zeeman doublet of GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As QDMs (states |1⟩ and |2⟩). All the parameters are taken as in Fig. 3(d) of the paper, except that now the QDM is grown along [001] -i.e. Hamiltonian \( \mathbb{H}_{[001]} \) instead of \( \mathbb{H}_{[111]} \). As can be seen, the picture is qualitatively the same as that obtained for [111]

![FIG. 2: Calculated spin purity of states |1⟩ and |2⟩ for different interdot distances at the resonant electric field for a triangular QDM grown along [001].](image)

grown QDMs (Fig.3(d) in the paper). The main difference is that the critical distance \( d_c \), where the bonding-antibonding reversal takes place and hole spin mixing is maximized, is now shifted towards longer interdot distances. This is because the effective masses of HH along [001] are lighter than those along [111]. Therefore, the tunneling is stronger.

Indeed, if we take constant mass parameters in \( \mathbb{H}_{BF}^{[001]} \), eq. 4 we obtain the corresponding Luttinger-Kohn Hamiltonian:
\[
\mathbb{H}^{[001]}_{LK} = - \begin{pmatrix}
\hat{P} + \hat{Q} & -\hat{S} & \hat{R} & 0 \\
-\hat{S}^\dagger & \hat{P} - \hat{Q} & 0 & \hat{R} \\
\hat{R}^\dagger & 0 & \hat{P} - \hat{Q} & \hat{S} \\
0 & \hat{R}^\dagger & \hat{S}^\dagger & \hat{P} + \hat{Q}
\end{pmatrix}
\]  
(21)

with
\[
\hat{P} \pm \hat{Q} = \left[ (\gamma_1 \pm \gamma_2)(k_x^2 + k_y^2) + (\gamma_1 \mp 2\gamma_2)k_z^2 \right] / 2
\]
\[
\hat{R} = \left[ -\sqrt{3}\gamma_2(k_x^2 - k_y^2) + i2\sqrt{3}\gamma_3k_zk_y \right] / 2
\]
\[
\hat{S} = \gamma_3\sqrt{3}(k_x - i k_y)k_z
\]  
(22)

Also, if we take constant mass parameters in \(\mathbb{H}^{[111]}_{BF}\), eq. [10], we obtain:
\[
\mathbb{H}^{[111]}_{LK} = -\frac{1}{2} \begin{pmatrix}
\hat{P} + \hat{Q} & -\hat{S} & \hat{R} & 0 \\
-\hat{S}^\dagger & \hat{P} - \hat{Q} & 0 & \hat{R} \\
\hat{R}^\dagger & 0 & \hat{P} - \hat{Q} & \hat{S} \\
0 & \hat{R}^\dagger & \hat{S}^\dagger & \hat{P} + \hat{Q}
\end{pmatrix}
\]  
(23)

with
\[
\hat{P} \pm \hat{Q} = (\gamma_1 \pm \gamma_3)(k_x^2 + k_y^2) + (\gamma_1 \mp 2\gamma_3)k_z^2
\]
\[
\hat{R} = -\frac{1}{\sqrt{3}}(\gamma_2 + 2\gamma_3)k_x^2 + \frac{2\sqrt{2}}{\sqrt{3}}(\gamma_2 - \gamma_3)k_z
\]
\[
\hat{S} = -\frac{2\sqrt{2}}{\sqrt{3}}(\gamma_2 - \gamma_3)k_y^2 + \frac{2}{\sqrt{3}}(2\gamma_2 + \gamma_3)k_z
\]  
(24)

By comparing \(\hat{P} + \hat{Q}\) in Eq. (22) with Eq. (24), one can note the different HH effective masses in the z direction: \(1/(\gamma_1 - 2\gamma_2)\) vs \(1/(\gamma_1 - 2\gamma_3)\).

It is worth noting that the \(\hat{R}\) operator in \(\mathbb{H}^{[001]}_{LK}\) does not have \(C_3\) rotational symmetry (the \(C_3\) character table can be obtained from that of \(C_{3h}\) by just considering rotations and keeping rows 1-3,7-8 and 12). However, for many III-V materials including GaAs, \(\gamma_2 \approx \gamma_3\) and one can approximate them both by \(\bar{\gamma} = (\gamma_2 + \gamma_3)/2\) in the \(\hat{R}\) and \(\hat{R}^\dagger\) matrix elements, thus yielding \(\mathbb{H}^{[001]}_{LK}\) with axial symmetry,\(^{10}\) that is reduced to \(C_3\) (or \(C_{3h}\)) symmetry by the triangular confining potential and the magnetic field. On the other hand, Hamiltonian \(\mathbb{H}^{[111]}_{LK}\) does have \(C_3\) symmetry, but –as discussed in the paper– the axial approximation in both \(R\) and \(S\) matrix elements is needed to display exact \(C_{3h}\) symmetry.

In short, in both crystallographic directions the Hamiltonian has approximate \(C_{3h}\) symmetry, which becomes exact if the axial approximation is assumed. Similar considerations on the axial approximation hold for the strain terms.
V. SPIN MIXING IN INAS/GAAS QDMS

Next, we consider InAs/GaAs QDMS grown along [001], similar to those obtained by self-assembled growth\textsuperscript{11} but with triangular (pyramidal) QD shape. Unlike for GaAs/AlGaAs, no interdot wire is present in this case. On the other hand, strain and piezoelectricity now play a significant role.

Figure 3 shows the spin purity of the four first hole states as a function of an external electric field. One can see that also in this case there is a strong spin mixing ($|\langle J_z \rangle| \ll 3/2$). An inspection of the individual spinor components (not shown) reveals that also for InAs/GaAs LH components play a minor role. Most of the mixing in Fig. 3 comes from admixture between HH $J_z = +3/2$ and $J_z = -3/2$ components, following the symmetry-induced mechanism described in our paper.

![Figure 3](image.png)

FIG. 3: (a) and (b): hole energy levels and Bloch angular momentum expectation value of a triangular InAs/GaAs QDM grown along [001], as a function of a vertical electric field. The insets in (a) show the hole localization for each doublet.
It is remarkable that the spin mixing here takes place for bonding and antibonding states split by more than 2 meV. Besides, the spin mixing takes place over a wide window of electric fields (wider than for GaAs). This is because of the stronger SOI of InAs as compared to GaAs. These results suggest that the eventual design of triangular InAs/GaAs QDMs would also form a promising system for hole spin manipulation.

VI. PERTURBATIONAL ESTIMATE OF THE SPIN MIXING STRENGTH

In this section we expand the discussion of Fig. 3 based on perturbation theory. The goal is to show that the spin in triangular QDMs is much stronger than in circular QDMs.

The Hamiltonian describing the hole states in a QDM, be it $H_{[001]}$ or $H_{[111]}$, can be split as:

$$ H = H_0 + H'. $$

Here $H_0$ are the diagonal terms, whose eigenfunctions are single-band HH or LH states:

$$ |\Psi_{HH\uparrow}^{0}(X_p)\rangle = f_{3/2}(X_p) \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \quad |\Psi_{LH\uparrow}^{0}(X_p)\rangle = f_{1/2}(X_p) \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}, $$

$$ |\Psi_{LH\downarrow}^{0}(X_p)\rangle = f_{-1/2}(X_p) \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix}, \quad |\Psi_{HH\downarrow}^{0}(X_p)\rangle = f_{-3/2}(X_p) \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}, $$

where $f_{Jz}(X_p)$ is the envelope function of $X$ rotational symmetry and $p$ parity. In turn, $H'$ represents the off-diagonal terms of $H$, coming from $H_{BF}$ and $H_{strain}$. This term is responsible for the band coupling. Without loss of generality, because the symmetry of $H_{BF}$ and $H_{strain}$ is the same, in what follows we consider GaAs/AlGaAs QDMs, where $H_{strain}$ is negligible. The analysis is further simplified replacing $H_{BF}$ by its constant mass analogue, $H_{LK}$, Eqs. (21) or (23) within axial approximation. This leads to the following expression for $H'$:
\[
\mathbb{H}' = -\begin{pmatrix}
0 & -S & R & 0 \\
-S^\dagger & 0 & 0 & R \\
R^\dagger & 0 & 0 & S \\
0 & R^\dagger & S^\dagger & 0
\end{pmatrix}
\] (27)

Within this approximation the matrix element operators \( R \) and \( S \) are just proportional to \( k_- \) and \( k_- k_z \), respectively.

Considering \( \mathbb{H}' \) as a perturbative term, the mixing of states up to second order is given by:

\[
|\Psi_k^{(2)}\rangle = \sum_{i \neq k} \left( \sum_{j \neq k} \frac{\langle \Psi_i^{(0)} | \mathbb{H}' | \Psi_j^{(0)} \rangle \langle \Psi_j^{(0)} | \mathbb{H}' | \Psi_k^{(0)} \rangle}{E_k^0 - E_i^0} \right) |\Psi_i^{(0)}\rangle
\] (28)

Note that the coupling between \( |\Psi_{HH}^0 (X_i^{p_i})\rangle \) and \( |\Psi_{HH}^0 (X_k^{p_k})\rangle \) via \( \mathbb{H}' \) requires \( |\Psi_{LH}^0 (X_j^{p_j})\rangle \) as intermediate state, yielding the contribution:

\[
- \frac{\langle X_i^{p_i} | S | X_j^{p_j} \rangle \langle X_j^{p_j} | R | X_k^{p_k} \rangle}{\Delta E_{hh} \Delta E_{lh}}
\] (29)

where \( \Delta E_{hh} \) is the energy difference between the HH states \( |\Psi_{HH}^0 (X_i^{p_i})\rangle \) and \( |\Psi_{HH}^0 (X_k^{p_k})\rangle \), and \( \Delta E_{lh} \) that between \( |\Psi_{HH}^0 (X_k^{p_k})\rangle \) and \( |\Psi_{LH}^0 (X_j^{p_j})\rangle \).

Alternatively, this coupling can also be achieved with \( |\Psi_{LH}^0 (X_j^{p_j})\rangle \) as intermediate state, yielding the contribution:

\[
\frac{\langle X_i^{p_i} | R | X_j^{p_j} \rangle \langle X_j^{p_j} | S | X_k^{p_k} \rangle}{\Delta E_{hh} \Delta E_{lh}}
\] (30)

The matrix elements in the numerator of Eqs. (29), (30) determine the selection rules in the band coupling process. E.g. in the \( C_{3h} \) group, the matrix element operator \( S \) has \( E''_y \) symmetry (\( R \) has \( E'_z \)), see Eq. (16). Then, a totally symmetric \( A' \) \( \langle \Psi_{HH}^0 | \) ground state must couple, via \( S \), with a \( |\Psi_{LH}^0 \rangle \) state of symmetry \( E''_y \) \( (A' \otimes E''_y \otimes E''_y = A', \) otherwise the integral is zero). Next, \( \langle \Psi_{LH}^0 | \) with symmetry \( E''_y \) (the complex conjugate of that of \( |\Psi_{LH}^0 \rangle \)) must couple, via \( R \), with \( |\Psi_{HH}^0 \rangle \) of \( A'' \) symmetry \( (E''_y \otimes E'_z \otimes A'' = A', \) otherwise the integral is zero). Likewise, \( \langle \Psi_{HH}^0 \rangle \) of \( A' \) symmetry (note that \( A' \) and \( A'' \) are reals and then coincide with their complex conjugates) can couple via \( R \) with \( |\Psi_{LH}^0 \rangle \) of \( E'_z \) symmetry. Then, \( \langle \Psi_{LH}^0 \rangle \) of \( E'_z \) symmetry will couple, via \( S \), with \( |\Psi_{HH}^0 \rangle \) of \( A'' \) symmetry \( (E'_z \otimes E''_y \otimes A'' = A'). \)

The above reasonings lead us to define the \( C_{3h} \) allowed couplings, represented by thick vertical lines on the left side of Fig. 4. Blue and yellow arrows correspond to either contribution, eqs. (29) and (30). In the figure we have simplified the notation of the states in
eq. (26) to \((X^p, J_z)\), where \(X\) is the rotational symmetry of the envelope function \((A, E_+\) and \(E_-\) in \(C_3\) or \(M_z\) in \(C_\infty\)), \(p = \prime\) or \(\prime\prime\) represents the even/odd parity, and \(J_z\) indicates the non-zero component of the four-fold spinor.

One can now compare with the case of circular QDMs, where the group is \(C_\infty h\) and the envelope functions are labeled by \(M_z\) and parity. In this group, the matrix element operator \(R (S)\) is even (odd) and has \(M_z = -2\) (\(M_z = -1\)). Now the \(\langle \Psi^0_{HH\uparrow} \rangle\) ground state of 0' symmetry can couple via \(S (-1'')\) with \(|\Psi^0_{LH\downarrow}\rangle\) of symmetry 1''. Then, \(\langle \Psi^0_{LH\uparrow}\rangle\) of symmetry \(-1''\) couple, via \(R (-2')\), with \(|\Psi^0_{HH\downarrow}\rangle\) of 3'' symmetry (because \(-1 - 2 + 3 = 0\) and \(n \otimes \prime \otimes \prime\prime = \prime\)).

\(\langle \Psi^0_{HH\uparrow}\rangle\) 0' may also couple, via \(R (-2')\) with \(|\Psi^0_{LH\downarrow}\rangle\) 2'. In turn, \(\langle \Psi^0_{LH\downarrow}\rangle\) of \(-2'\) symmetry couples, via \(S (-1'')\), with \(|\Psi^0_{HH\downarrow}\rangle\) of 3'' symmetry.

Taking into account Fig. 4 and Eqs. (29) and (30) we can see that \(\Delta E_{hh}\) in the denominator involved in \(C_{3h}\) is quite smaller than that involved in \(C_{\infty h}\) and, therefore, the interaction should be much stronger.

As a matter of fact, the strong spin mixing at the resonant electric fields is a singular behavior of triangular QDMs. In \(C_{nh}\) symmetries with \(n > 3\) the spinor fourth component \((J_z = -3/2)\) has different rotational symmetry symmetry than the first one \((J_z = 3/2)\), as in the above discussed case of \(C_{\infty h}\). Then, for similar reasons, the coupling between the states belonging to the first two doublets are also forbidden. In the case of \(C_{2h}\) QDM the symmetries of \(S\) and \(R\) are \(B_g\) and \(A_g\) respectively. Bonding (anti-bonding) molecular orbitals are of \(A_g\) \((B_u)\) symmetry. Then, the doublet antibonding ground state (bonding first excited state) are \(B_u \uparrow\), \(B_u \downarrow\) \((A_g \uparrow\), \(A_g \downarrow)\). Therefore, any coupling among these four states is forbidden by symmetry, as can be easily checked with the help of Eqs. (29) and (30).

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FIG. 4: Diagram of single band hole energy levels under a longitudinal magnetic field for a QDM with symmetry $C_{3h}$ (left column) and $C_{\infty h}$ (right column). The labels $(X^p, J_z)$ indicate the symmetry of the levels. $X$ represents the rotational symmetry of the envelope function ($A, E_+,$ and $E_-$ in $C_3$ and $M_z$ in $C_{\infty h}$), $p = \prime$ or $\prime\prime$ represents the even/odd parity and $J_z$ indicates the non-zero component of the spinor (e.g. $J_z = 3/2$ means that the non-zero component is the first one, $J_z = 1/2$ the second, etc.) Thick arrows denote the symmetry allowed level couplings for the ground state, as inferred from Eqs. (29), (30).

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12 The spin admixture we report is proportional to the extradiagonal elements effective mass, $\gamma_2$ and $\gamma_3$. The value of these parameters is in turn modulated by the strength of the spin-orbit interaction. Then, the extent of the spin admixture is ultimately due to spin-orbit interaction.