Effective theory of monolayer TMDC double quantum dots

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Abstract

Monolayer transition metal dichalcogenides (TMDCs) are promising candidates for quantum technologies, such as spin qubits in quantum dots, because they are truly two-dimensional semiconductors with a direct band gap. In this work, we analyse theoretically the behaviour of a double quantum dot (DQD) system created in the conduction band of these materials, with two electrons in the (1,1) charge configuration. Motivated by recent experimental progress, we consider several scenarios, including different spin–orbit splittings in the two dots and including the case when the valley degeneracy is lifted due to an insulating ferromagnetic substrate. Finally, we discuss in which cases it is possible to reduce the low energy subspace to the lowest Kramers pairs. We find that in this case the low energy model is formally identical to the Heisenberg exchange Hamiltonian, indicating that such Kramers pairs may serve as qubit implementations.

1. Introduction

Monolayers of transition metal dichalcogenides (TMDCs) are a class of 2D materials with very interesting electronic and optical properties [1, 2]. They are atomically thin semiconductors with a direct band gap and two-fold degenerate valleys in the Brillouin zone [3, 4]. Early studies discovered that their intrinsic spin–orbit interaction splits the spin states in the valence band and that it is possible to optically manipulate the spin and valley degrees of freedom (DOF) [5, 6]. Further theoretical works suggested that the conduction band should be split as well due to the spin–orbit coupling [7, 8]. The conduction band spin–orbit splitting was also supported by recent experiments [9].

The possibility to construct purely two-dimensional, electrostatically defined quantum dots (QDs) is one of the reasons that makes monolayer TMDCs so attractive, at least from a fundamental point of view. Compared to III–V semiconductors, such as GaAs [10–12], TMDCs have several isotopes with vanishing nuclear spin, thus lacking hyperfine interactions with the electronic spin. Moreover, TMDCs comprise an additional valley pseudospin. Although these two features are common to several other systems used for QDs, such as Si/SiGe quantum wells [13–16], graphene [17–22] and carbon nanotubes (CNTs) [23–26], TMDCs are special because they exhibit very strong spin–orbit coupling (SOC). Theoretical investigation of QDs in TMDCs started with QDs in gated nanoribbons [27] and the magnetic field dependence of the single-electron spectrum [7] and it now includes studies of valley hybridisation [28], flake QDs of triangular and hexagonal shape and nanoribbons [29, 30], the valley Zeeman effect [31], optical control of a spin-valley qubit [32], spin-degenerate regimes for small QDs in a magnetic field [33], spin relaxation [34], electric control of a spin-valley qubit [35] and a model of valley qubit [36]. On the experimental side, gating monolayer TMDCs is not straightforward [38, 40] and low material quality has hindered the experimental study of the intrinsic properties of these materials for a long time. However, recently there has been a significant progress in the fabrication process of nanostructures in TMDCs. This has enabled the creation of single QDs on monolayer [37, 41] or trilayer TMDCs [40], double QD experiments with tunable coupling strength between the dots [39, 41] and the observation of gate-controlled Coulomb blockade effect [37–41].

In this paper we study double quantum dots (DQDs) taking into account the spin–orbit coupling and both spin and valley DOF. We note that, although single [25] and double [42–44] quantum dots in CNTs have been studied, where the low energy theory shares some similarities with TMDCs, little is known about TMDC DQDs and the role of the exchange interaction. For simplicity we restrict each dot to the lowest orbital and we consider the system filled with two electrons. Starting from the model introduced in [45], we add the spin–orbit interaction and we find a low energy effec-
tive Hamiltonian for the case where each dot is occupied by one electron. We also investigate the situations where the spin–orbit splitting is different for each dot and where we include the effects of a magnetic field. Furthermore, for large spin–orbit splittings it is convenient to focus on a smaller subset of states formed by the two lowest Kramers pairs of the system. In this smaller subspace we find that in most cases the interaction is formally identical to the Heisenberg exchange interaction, which was suggested to perform a CNOT gate between spin qubits [46]. This supports the idea that the lowest Kramers pair can serve as a qubit for TMDC, as was proposed in [7].

This paper is organised as follows. In section 2 we introduce the system Hamiltonian, we present a naming conventions for certain useful projection operators and we briefly evoke the results for the zero spin–orbit splitting case. Then, in section 3 we present the effective Hamiltonians for the case where each QD is occupied by one electron, for three different situations: when the spin–orbit splitting is equal in both dots, when the spin–orbit splitting is different and when the TMDC is deposited on an insulating ferromagnetic substrate or placed in an external magnetic field. Afterwards, we explore in section 4 under which conditions it is possible to focus on smaller effective Hamiltonians for these three situations. Finally, in section 5 we present our conclusions.

2. Model

2.1. Basic definitions

We give here the definitions for the different terms of the Hamiltonians that describe the various scenarios studied in this work. The operators \( c_{j\tau\sigma}^{\dagger} \) annihilate (create) an electron in QD \( j \) with valley \( \tau \) and spin \( \sigma \). Here \( j = L \) (R) refers to the left (right) QD; \( \tau = K \) (\( \bar{K} \)) indicates the positive (negative) valley and \( \sigma = \uparrow (\downarrow) \) specifies spin up (spin down).

The on-site Coulomb repulsion between electrons in the same QD is captured by the Hubbard Hamiltonian,

\[
H_U = \frac{U}{2} \sum_{j=L,R} n_j (n_j - 1),
\]

where \( U > 0 \) is the positive charging energy of the dot and the number operator is defined as

\[
n_j = \sum_{\tau=K,\bar{K}} \sum_{\sigma=\uparrow,\downarrow} c_{j\tau\sigma}^{\dagger} c_{j\tau\sigma}.
\]

A detuning term specifies the energy difference \( \varepsilon \) between the dots,

\[
H_\varepsilon = \frac{\varepsilon}{2} (n_L - n_R).
\]

Electron-hopping from one dot to the other is accounted for by a tunneling term that preserves spin and valley,

\[
H_t = \sum_{\tau,\sigma} \left( t_{\tau\sigma} \tilde{c}_{L\tau\sigma}^{\dagger} \tilde{c}_{R\tau\sigma} + \text{h.c.} \right),
\]

where the tunneling coefficient \( t \) is generally a complex number.

The intrinsic spin–orbit coupling is modeled by a simple time-reversal symmetric (\( \mathcal{T} \)-symmetric) spin-splitting: \( \Delta_{\tau\sigma} \) [7].

\[
H_\Delta = \sum_{j,L,R} \tilde{c}_{j\tau\sigma}^{\dagger} \tilde{c}_{j\tau\sigma} \Delta_{j,\tau\sigma},
\]

where \( \Delta_{j,\tau\sigma} \) is the energy splitting of the spin states in the set \( \mathcal{P} = \{ \ket{K \uparrow}, \ket{\bar{K} \downarrow} \} \) are shifted by the energy \( +\Delta \), while the Kramers pairs states in the set \( \mathcal{N} = \{ \ket{K \downarrow}, \ket{\bar{K} \uparrow} \} \) are shifted by the energy \( -\Delta \). We call \( \mathcal{P} \) the positive Kramers pair and \( \mathcal{N} \) the negative Kramers pair. For the double dot system,

\[
H_\Delta = \sum_{j,L,R} \sum_{\tau,\sigma} \tilde{c}_{j\tau\sigma}^{\dagger} \tilde{c}_{j\tau\sigma} \Delta_{j,\tau\sigma}.
\]

Equation (5) assumes that the spin–orbit splitting is the same for every dot, which is usually the case for dots created on the same material. In case the spin–orbit splitting is different we use the following generalisation,

\[
H_{\Delta_L,\Delta_R} = \sum_{j,L,R} \sum_{\tau,\sigma} \tilde{c}_{j\tau\sigma}^{\dagger} \tilde{c}_{j\tau\sigma} \Delta_{j,\tau\sigma},
\]

where \( \Delta_L \) and \( \Delta_R \) are the spin–orbit splittings in the left and right QD respectively.

We also consider the coupling of spin and valley to an external magnetic field. The corresponding spin Zeeman term is given by

\[
H_S = \sum_{j,L,R} \sum_{\tau,\sigma} \tilde{c}_{j\tau\sigma}^{\dagger} \tilde{c}_{j\tau\sigma} \hbar h_{jL} \sigma_\tau \sigma, \sigma_{\tau\sigma},
\]

where \( h_{jL} \) and \( h_{jR} \) are two vectors of coupling constants for left and right QD respectively and \( \sigma \) is the vector of Pauli matrices acting on spin.

The valley Zeeman term is the valley counterpart of the spin Zeeman but considering only the \( z \)-Pauli matrix acting on the valley. This is motivated by [7] and recent experiments [47, 48],

\[
H_V = \sum_{j,L,R} \sum_{\tau,\sigma} \tilde{c}_{j\tau\sigma}^{\dagger} \tilde{c}_{j\tau\sigma} \hbar h_{jL} \sigma_{\tau\sigma}, \sigma_{\tau\sigma},
\]

where \( h_{jL} \) and \( h_{jR} \) describe the valley splittings in the left and right dot. A concise outline of the interactions and the DOF of our DQD model is shown in figure 1.
\[ |t| \ll |U \pm \varepsilon|, \tag{9} \]

2.2. Naming basis states and projection operators

It is natural to refer to certain operators that will appear as projectors on specific states. We present a convention to name all 28 states of the total Hilbert space and we give the projection operators that will be useful later.

Because of the Pauli exclusion principle, not all the states in the \((1, 1)\)-subspace are allowed to tunnel to \((0, 2)\) or \((2, 0)\)-states, but only those which are antisymmetric in spin and valley \([44, 49]\). Following the antisymmetric nature of the tunneling states, we begin by introducing a basis consisting of states that are symmetric or antisymmetric in both spin and valley: \(|sV sS(nL, nR)\rangle\), where \((nL, nR)\) is the charge configuration and \(sV (sS)\) indicates the exchange symmetry of valley (spin) DOF. The exchange symmetry can be either that of a singlet \((S)\) or that of a triplet \((T)\); see table 1 for the definitions of these states. When \((nL, nR) = (1, 1)\) we omit the indication of the charge configuration in the subscript. The equal spin–orbit coupling term \(\Delta_0\) (equation \(5\)) is not diagonal in this basis. In order to work with a basis that makes \(\Delta_0\) diagonal, in the \((1, 1)\)-subspace we substitute \(|ST0\rangle, |T0S\rangle, |SS\rangle\) and \(|T0T0\rangle\) with the following states (see appendix A),

\[ |n_{\pm}\rangle = (|ST0\rangle \pm |T0S\rangle)/\sqrt{2}, \tag{10a} \]

\[ |\pi_{\pm}\rangle = (|T0T0\rangle \pm |SS\rangle)/\sqrt{2}. \tag{10b} \]

Here \(|n_{\pm}\rangle, |\pi_{\pm}\rangle\) are antisymmetric (symmetric) spin–valley states which are superpositions of only positive (subscript +) or negative (subscript −) Kramers pairs, see also appendix B. Note that \(|n_{\pm}\rangle, |\pi_{\pm}\rangle\) are odd (even) under the time-reversal operator \(\mathcal{T}\). For these states there is no defined exchange symmetry for spin or valley alone. States analogous to \(|n_{\pm}\rangle\) are defined in the \((2, 0)\) and \((0, 2)\)-subspaces. In table 2, we list all these states grouped by charge configuration and by symmetry under exchange.

Projection operators on each one of the antisymmetric \((1, 1)\)-states are easy to obtain (see appendix B),

\[ P_{|TS\rangle} = \frac{1}{16} (1 \pm \tau_L) (1 \pm \tau_R) (1 - \sigma_L \cdot \sigma_R), \tag{11a} \]

\[ P_{|StS\rangle} = \frac{1}{16} (1 \pm \sigma_L) (1 \pm \sigma_R) (1 - \tau_L \cdot \tau_R), \tag{11b} \]

\[ P_{|n_{\pm}\rangle} = \frac{1}{16} (1 - \tau_L \tau_R) (1 - \sigma_L \sigma_R) \times (1 \pm \tau_L \sigma_L \tau_R \sigma_R), \tag{11c} \]

where \(\tau_i, (\sigma_i)\) is the ith Pauli matrix acting on valley (spin) in QD \(j = L, R\) and \(\sigma, \tau\) are vectors of Pauli matrices acting on spin or valley respectively. We note that the projection operator over the whole antisymmetric subspace of the \((1, 1)\)-sector can be written as \([45]\) (see appendix B),

\[ P_{\text{as}} = (3 - \sigma_L \cdot \sigma_R - \tau_L \cdot \tau_R - (\sigma_L \cdot \sigma_R)(\tau_L \cdot \tau_R))/8, \tag{12} \]

2.3. Zero spin–orbit splitting \((\Delta = 0)\)

We finish this section by briefly considering the case where there is no spin–orbit splitting \((\Delta_L = \Delta_R = 0)\). The total Hamiltonian is then,

\[ H_{\text{tot}} = H_U + H_c + H_t, \tag{13} \]

where \(H_U, H_c\) and \(H_t\) are defined in equations \((1), (3)\) and \((4)\) respectively. This model describes DQD
The states are grouped as (1,1)-states (antisymmetric and symmetric), (2,0)-states and (0,2)-states (only antisymmetric).

We first consider the case where the spin–orbit coupling act – spin–orbit splitting reported in section 2.3. Exchange interaction and symmetric spin–orbit coupling act independently of each other. We obtain the effective Hamiltonian:

\[ H_{\text{eff}} = -JP_{\text{as}} + \Delta \Sigma, \]

where \( f \) is the same constant defined in (15), \( P_{\text{as}} \) is given in (12), \( \Delta \Sigma = H_{\Delta}|_{(1,1)} = \Delta (\tau_3\sigma_3 + t_{\text{Ryd}}\sigma_0) \) and \( H_{\Delta}|_{(1,1)} \) is the restriction of \( H_{\Delta} \) to the (1,1)-subspace.

It is simple to evaluate the consequences of equation (18) since it is diagonal in the states of table 2. Assuming now that \( \Delta > 0 \), the ground state is \( |n_{-}\rangle \) because it is the only state that is shifted down by both \(-J\) and \(-2\Delta\), thus the ground state space is one dimensional, see figures 2(b) and (c). The dimensionality of the ground state space changes from one to six only when \( \Delta \) is exactly equal to zero (see figure 2(a)) and this degeneracy is lifted linearly in \( \Delta \). As shown in figures 2(b) and (c), there are five groups of excited states with various degrees of degeneracy and whose relative distances in energy depend on the sizes of \( \Delta \) and \( J \). For later comparison, we observe that in this case call this symmetric spin–orbit splitting. The total Hamiltonian is

\[ H_{\text{tot}} = H_U + H_e + H_i + H_{\Delta}, \]

where \( H_{\Delta} \) is defined in equation (5). This Hamiltonian describes DQDs created in TMDCs \([3–7,9]\), but it may also be used to study DQDs in CNTs \([25,42–44,50,51]\).

Symmetric spin–orbit coupling \( H_{\Delta} \) shifts those states formed by two elements of the negative (positive) Kramers pair by \(-2\Delta + 2\Delta \), while it leaves unchanged those states formed by one element of the negative Kramers pair and one element of the positive Kramers pair (see table 2). In order to identify the (1,1)-sector as our low energy subspace (LES) we have to guarantee first of all that no (2,0) or (0,2)-state is lower in energy than any (1,1)-state. Looking at table 2 we see that this condition is met when

\[ 4|\Delta| < U - |\epsilon|. \]

When both (9) and (17) are satisfied, it follows that (2,0) and (0,2)-states are energetically unfavorable and tunneling out from the (1,1)-sector is strongly suppressed. However, virtual tunneling processes must be taken into account. It is important to notice that antisymmetric (1,1)-states can tunnel only to their (0,2) and (2,0) counterpart states that have the same spin and valley configuration. This is due to the spin- and valley-preserving nature of the tunneling term of equation (4): there is no transition from a negative Kramers pair to a positive Kramers pair and viceversa. Therefore, all the energy differences between initial and final states in a virtual tunneling process do not depend on \( \Delta \) and the exchange interaction that emerges does not change from the case of zero spin–orbit splitting reported in section 2.3. Exchange interaction and symmetric spin–orbit coupling act independently of each other. We obtain the effective Hamiltonian:

\[ H_{\text{eff}} = -JP_{\text{as}} + \Delta \Sigma, \]

where \( f \) is the same constant defined in (15), \( P_{\text{as}} \) is given in (12), \( \Delta \Sigma = H_{\Delta}|_{(1,1)} = \Delta (\tau_3\sigma_3 + t_{\text{Ryd}}\sigma_0) \) and \( H_{\Delta}|_{(1,1)} \) is the restriction of \( H_{\Delta} \) to the (1,1)-subspace.

It is simple to evaluate the consequences of equation (18) since it is diagonal in the states of table 2. Assuming now that \( \Delta > 0 \), the ground state is \( |n_{-}\rangle \) because it is the only state that is shifted down by both \(-J\) and \(-2\Delta\), thus the ground state space is one dimensional, see figures 2(b) and (c). The dimensionality of the ground state space changes from one to six only when \( \Delta \) is exactly equal to zero (see figure 2(a)) and this degeneracy is lifted linearly in \( \Delta \). As shown in figures 2(b) and (c), there are five groups of excited states with various degrees of degeneracy and whose relative distances in energy depend on the sizes of \( \Delta \) and \( J \). For later comparison, we observe that in this case call this symmetric spin–orbit splitting. The total Hamiltonian is

\[ H_{\text{tot}} = H_U + H_e + H_i + H_{\Delta}, \]

where \( H_{\Delta} \) is defined in equation (5). This Hamiltonian describes DQDs created in TMDCs \([3–7,9]\), but it may also be used to study DQDs in CNTs \([25,42–44,50,51]\).
each antisymmetric state is separated from its symmetric counterpart by the same energy $J$. In the case $\Delta < 0$, the results are analogous, but the ground state is $|n_+\rangle$.

Regarding the experimental situation, the few available experiments indicate $U \approx 2$ meV for QDs created in WSe$_2$ [37] and MoS$_2$ [39-41] with QD radii around 100 nm. On the other hand, the theory predicts $2\Delta$ to be about 3 meV for MoS$_2$ and larger for other compounds [7, 52]. Therefore, at the moment, condition (17) seems not to be satisfied for TMDs, but further experiments with smaller QD sizes may lead to higher charging energies. However, when equation (17) is not satisfied, a consistent effective theory can be derived in a subspace of the $(1,1)$-sector, as explained in section 4.

3.2. Asymmetric spin–orbit splitting ($\Delta_L \neq \Delta_R$)

Suppose now that the spin–orbit strength is different in the two dots, $\Delta_L \neq \Delta_R$, this is the case of asymmetric spin–orbit splitting. The total Hamiltonian is

$$ H_{\text{tot}} = H_U + H_\varepsilon + H_I + H_{\Delta_L, \Delta_R}, $$

where $H_\varepsilon$ has been substituted by $H_{\Delta_L, \Delta_R}$ defined in equation (6).

There is theoretical evidence that for (rather small) QDs on TMDs the spin–orbit strength $\Delta$ depends on the radius of the QD [35]. Therefore, the Hamiltonian in equation (19) can reflect a situation where the two dots have different sizes. Changing the size of the QD would allow for a smooth and tunable adjustment of $\Delta$. Another situation where $\Delta_L \neq \Delta_R$ can be relevant is when the dots would be created in lateral heterojunctions such that each dot is created in different types of TMDs, which intrinsically possess different spin–orbit splittings [52]. Lateral heterojunctions of different TMDs have already been demonstrated by several groups [53-55]. To our knowledge no experiment has been reported yet involving such a DQD. We notice that for heterojunctions the situation might be further complicated by mismatch in the energy band gap, acting as a detuning away from the interface, and by likely non-negligible differences in the Coulomb charging energy $U$ of the two compounds.

We point out that $H_{\Delta_L, \Delta_R}$ is not diagonal in the basis of the states listed in table 2. Each of the antisymmetric states $|T_+ S\rangle, |T_- S\rangle, |S T_+\rangle, |S T_-\rangle$ is mixed with its symmetric counterpart and the off-diagonal elements are $\pm (\Delta_L - \Delta_R)$. This is relevant to determine the ground state, but not for the form of the effective Hamiltonian, because the mixing happens inside the $(1,1)$-subspace. Another difference is that, in contrast to table 2, the $\pm 2\Delta$ on the diagonal of the Hamiltonian are replaced by $\pm (\Delta_L + \Delta_R)$ in the $(1,1)$-subspace, by $\pm 2\Delta_L$ in $(2,0)$ and by $\pm 2\Delta_R$ in $(0,2)$. When condition (9) is also valid, we can ensure that $(1,1)$-states are lower in energy than $(2,0)$ and $(0,2)$ states by requiring that

$$ \max\{|\Delta_L + \Delta_R|, |\Delta_L - \Delta_R|\} + 2 \max\{|\Delta_L|, |\Delta_R|\} < U - |\varepsilon|. $$

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**Figure 2.** Level structure of $(1,1)$-states for the symmetric spin–orbit splitting case at fixed (small) detuning. We show three relevant cases with increasing spin–orbit splitting ($\Delta$) from left to right. Thick horizontal lines illustrate the energy levels. Degenerate states are shown as a group of thick lines close together with a number indicating the degree of degeneracy. The colored energy levels indicate the antisymmetric state $|n\rangle$, while the blue energy levels are the symmetric states $|\pi\rangle$. In (a) the spin–orbit splitting is zero and there are only two degenerate energy levels, separated by the exchange energy $J$. The higher (lower) energy corresponding to the symmetric (asymmetric) states. In (b) both symmetric and antisymmetric energy levels are separated in three groups, $|n\rangle$ becomes the ground state and the first excited states are antisymmetric. In (c), when $2\Delta$ is greater than the exchange energy $J$, the $N \times N$-sector becomes the LES and the first excited states are symmetric.

- $\Delta = 0$
- $2\Delta < J$
- $2\Delta > J$
In this case we can use the Schrieffer–Wolff transformation [56–58] to obtain the effective Hamiltonian. The term $H_{\Delta_L,\Delta_R}$ has three significant consequences. Firstly, the following matrix element differences now depend on $\pm(\Delta_L - \Delta_R)$,

$$E_{n,\pm} - E_{n,\pm}(\pm e) = \pm(\Delta_L - \Delta_R) - U + \varepsilon, \quad (21a)$$

$$E_{n,\pm} - E_{n,\pm}(\pm e) = \mp(\Delta_L - \Delta_R) - U - \varepsilon, \quad (21b)$$

where $E_{n,\pm} = \langle \psi | H_{\text{eff}} | \psi \rangle$. Secondly, in order to apply perturbation theory to the $(1,1)$-subspace, the coupling constants must satisfy the conditions $|t| \ll |U \pm |\varepsilon + \Delta_L - \Delta_R||$ and $|t| \ll |U \pm (\varepsilon - \Delta_L + \Delta_R)|$ simultaneously. In turn, these imply that $(\Delta_L - \Delta_R) \ll U$. Lastly, the exchange energies relative to $|n,\pm\rangle$ acquire a dependence on the asymmetry of the spin–orbit splittings,

$$J_{n,\pm} = \frac{4|t|^2 U}{(U + \varepsilon \pm \Delta_L \mp \Delta_R)(U - \varepsilon \mp \Delta_L \pm \Delta_R)}, \quad (22)$$

see equation (15). The exchange energies for states $|ST\rangle \pm |ST\rangle$ and $|ST\rangle \mp |ST\rangle$ are not affected.

An effective Hamiltonian can be written in a compact form in the following way,

$$H_{\text{eff}} = -JP_{n,\pm} + \Delta_L \Sigma_L \mp \Delta_R \Sigma_R - (J_{n,\pm} - J)P_{n,\pm}, \quad (23)$$

where $P_{n,\pm}$ and $P_{n,\pm}$ are the projectors on $|n,\pm\rangle$ and $|n,\pm\rangle$ defined in equation (11c) and in this case we write the restriction $H_{\Delta_L,\Delta_R}(1,1) = \Delta_1 \Sigma_L \pm \Delta_R \Sigma_R$, with $\Sigma_j = \tau_j \sigma_j$. By diagonalizing the Hamiltonian (23), we obtain the energy levels shown in figure 3.

Interestingly, the two levels that appear at energy 0 and energy $-J$ for the symmetric spin–orbit splitting case move to the energies,$$
\frac{-J}{2} \pm \sqrt{J^2 + 4(\Delta_L - \Delta_R)^2} = \frac{-J}{2} \pm \frac{\Phi}{2}. \quad (24)
$$

This is a consequence of the level repulsion due to the off-diagonal $\pm(\Delta_L - \Delta_R)$ matrix elements coming from $H_{\Delta_L,\Delta_R}$. However, the degrees of degeneracy remain the same. Looking at equation (24) and at figure 3, we realise that, for positive $\Delta_L + \Delta_R$, the ground state is $|n,\pm\rangle$, unless $|\Delta_L - \Delta_R|$ is larger than $\Delta_L + \Delta_R$, in which case the states $|T+,S\rangle |T-,S\rangle |ST\rangle \pm |ST\rangle$ (or their symmetric counterparts) span a 4-fold degenerate ground state space.

We want to briefly mention here that a generalisation of the asymmetric spin–orbit coupling which preserves the $T$-symmetry, including couplings to the in-plane components of the spins, does not affect the exchange interaction. Consider

$$H_{\Delta_{L\pm},\Delta_{R\pm}} = \sum_j \Delta_j \cdot \sum_{\tau,\sigma_1,\sigma_2} c_{j \tau \sigma_1}^\dagger \tau (\tau)_{\sigma_1 \sigma_2} c_{j \tau \sigma_2} \quad (25)$$

where $\Delta_j$ is the vector of the three coupling constants $\Delta_{j\pm}, \Delta_{j\pm}, \Delta_{j\pm}$ for QD $j$, multiplied by the vector of spin Pauli matrices $\sigma$. Equation (25) resembles the spin Zeeman term (see equation (7)), but it also preserves $T$ -symmetry because of the $\tau_j$ that multiplies the spin Pauli matrices. Qualitatively, a term like this should appear in the Hamiltonian of TMDC DQD systems for which it is possible to define an average local tilt and an average local curvature of the TMDC sheet for each dot [59]. However, tilt and curvature add couplings between conduction and valence band as well and one would need to check whether these couplings are small enough to be neglected. This is not the focus of this work and we only want to stress that substituting $H_{\Delta_L,\Delta_R}$ with $H_{\Delta_L,\Delta_R,\Delta_R}$ in equation (19) does not modify the exchange interaction given in equation (25).

3.3. Spin and valley Zeeman and symmetric spin–orbit coupling

In this section we investigate the influence of spin and valley Zeeman terms on the symmetric spin–orbit splitting case. The total Hamiltonian for this scenario reads

$$H_{\text{sym}} = H_U + H_L + H_T + H_{\Delta} + H_S + H_V, \quad (26)$$

where $H_S$ and $H_{\Delta}$ are defined by equations (7) and (8) respectively. This is the case depicted in figure 1.

This model approximates at least two similar but distinct situations. First, when a monolayer TMDC is placed in a non-uniform and weak magnetic field, the spin of the electron in each dot couples to the local field in all three spacial directions through the spin Zeeman interaction $H_S$. On the other hand, here we assume that the valley DOF couples only to perpendicular magnetic field. Indeed, in [7] it was shown that such coupling exists due to orbital effects. Since in-plane magnetic field does not lead to orbital effects in the strict two-dimensional limit, we assume that it does not couple to the valley. In contrast to [7] here we only consider the lowest orbital state in each of the dots but we also take into account the Coulomb charging energy $U$. The other possible situation that came to our attention is that of monolayer TMDC deposited on a ferromagnetic insulator, e.g. europium oxide (EuO). Such a situation was considered in [60], where equation (7) was used to model the giant and tunable valley splitting. A Rashba term is also included, mixing conduction and valence band, that we do not consider here. Although [60] does not consider the valley Zeeman term, we note that it is allowed in the effective Hamiltonian of the TMDC because the ferromagnetic substrate breaks time-reversal symmetry. The presence of such magnetic exchange field in TMDC and ferromagnetic semiconductor heterostructures has been recently demonstrated[47, 48].

Now both $H_S$ and $H_V$ are non–diagonal in the basis of states presented in table 2 and they mix a large number of states. We do not list all the newly introduced matrix elements but we observe that they are confined inside the charge sectors $(0,0)$, $(1,1)$ and $(2,0)$, as $H_S$ and $H_V$ do not contain terms of the form $c_{j \tau \sigma}^\dagger c_{k \tau ' \sigma'}$. 

### Table 2: Basis States of the Hamiltonian

| Charge Sector | States |
|---------------|--------|
| $(0,0)$       | $|0,0\rangle$ |
| $(1,1)$       | $|1,1\rangle$ |
| $(2,0)$       | $|2,0\rangle$ |
Figure 3. Comparison of the energy levels for symmetric (left) and asymmetric (right) spin–orbit splitting cases. The energy levels from left to right are aligned in a way to preserve the sum \( \Delta_L + \Delta_R \) between the two cases (equal to \( 2\Delta \) for the symmetric spin–orbit splitting case). The left hand side is equivalent to the case of \( 2\Delta > J \) depicted in figure 2(c). The exchange energies \( J \) and \( J_{\text{tot}} \) are defined in equations (15) and (22), where \( \Phi = \sqrt{P + 4(\Delta_L - \Delta_R)^2} \), see equation (24).

(4.1) Symmetric spin–orbit splitting

(\( \Delta_L = \Delta_R = \Delta \))

In the case of symmetric spin–orbit splitting (\( \Delta_L = \Delta_R = \Delta \), see \( H_{\text{tot}} \) in equation (16)) there is no matrix element that allows a transition between \( \mathcal{N} \) and \( \mathcal{P} \). Assuming equation (9) and \( 2\Delta > J \) (which is usually the case for TMDCs at low detuning) we see from figure 2(c) that the subspace spanned by the states of \( \mathcal{N} \times \mathcal{N} \) (also called \( \mathcal{N} \times \mathcal{N} \)-sector) is the LES, where

\[
\mathcal{N} \times \mathcal{N} = \{ |K; \downarrow; \mathcal{K}; \downarrow\rangle, |K; \downarrow; \mathcal{K}; \uparrow\rangle, |\mathcal{K}; \uparrow; \mathcal{K}; \uparrow\rangle, |\mathcal{K}; \uparrow; \mathcal{K}; \downarrow\rangle \},
\]

with \( |\tau_1; \sigma_1; \tau_2; \sigma_2\rangle = c_{\tau_1; \sigma_1} c_{\tau_2; \sigma_2} |0\rangle \). Now we present the effective Hamiltonian when the system is restricted to this LES.

We can easily identify the negative Kramers pair \( \mathcal{N} \) as a spin-1/2 DOF with

\[
|\tilde{\uparrow}\rangle \equiv |\mathcal{K}; \uparrow\rangle, \quad |\tilde{\downarrow}\rangle \equiv |\mathcal{K}; \downarrow\rangle,
\]

so that the effective basis \( \mathcal{N} \times \mathcal{N} \) can be seen as the basis of all the states of two spins,

\[
\mathcal{N} \times \mathcal{N} = \{ |\tilde{\uparrow}; \tilde{\downarrow}, |\tilde{\downarrow}; \tilde{\uparrow}, |\tilde{\uparrow}; \tilde{\uparrow}, |\tilde{\downarrow}; \tilde{\downarrow}\rangle \}. \tag{32}
\]

In this basis, the spin–orbit coupling always assumes the value of \(-2\Delta\), which we ignore in the following section.
since it is just an energy shift. It is well known that the effective Hamiltonian for two electrons with only the spin DOF in two QDs with only tunneling, detuning and Hubbard potential is given by \[ H_{\text{eff}} = iS_L \cdot \tilde{S}_R, \] with the usual exchange energy \( J \) multiplied by the projector on the singlet state \(|S\rangle\), the only antisymmetric state for two spins (\( S_L \) is the vector of spin operators acting on QD \( L \)). Analogously, using the following operators,
\[
\tilde{S}_\alpha = \sigma_x, \quad \tilde{S}_\beta = \tau_x\sigma_x, \quad \tilde{S}_\gamma = \tau_y\sigma_y, \tag{33}
\]
the Hamiltonian of the system restricted to basis \( N \times N \) is
\[
H_{\text{eff}, N \times N} = J \tilde{S}_L \cdot \tilde{S}_R, \tag{34}
\]
where \( \tilde{S}_\mu = \frac{i}{2} \tilde{S}_\mu \) is the spin operator proportional to the new Pauli operator \( \tilde{S}_\mu, i = x, y, z \), acting on QD \( j = L, R \). The ground state space is one-dimensional as for the full \((1, 1)\)-subspace. Indeed, the ground state is the same,
\[
|\tilde{S}\rangle = \left( |\uparrow; \uparrow\rangle - |\downarrow; \downarrow\rangle \right) / \sqrt{2} = \left( |K \downarrow; K \uparrow\rangle - |K \uparrow; K \downarrow\rangle \right) / \sqrt{2} = |n_-\rangle. \tag{35}
\]

\[ \begin{pmatrix}
-J - A & \delta h^x_L - \delta h^y_L & 0 \\
\delta h^y_L - \delta h^z_L & -A & 0 \\
0 & 0 & 0
\end{pmatrix} \begin{pmatrix}
0 \\
0 \\
-H^+_z + H^-_z - A_+
\end{pmatrix}, \tag{40}
\]

Since \((\Delta_L - \Delta_R)/(U \pm \varepsilon) \ll 1\), we can write
\[
J_{|n_-\rangle} \approx J \left(1 - 2\varepsilon \frac{\Delta_L - \Delta_R}{U^2 - \varepsilon^2} \right). \tag{39}
\]

The effect of an almost symmetric spin–orbit coupling on the exchange energy is that of fine tuning around the value of \( J \) by a small positive or negative quantity, depending on the sign of \( \varepsilon \) and \( \Delta_L - \Delta_R \). Moreover, as \( \varepsilon \to 0 \), \( J_{|n_-\rangle} \to J \). The above results remain valid for the generalisation to \( H_{\Delta_L, \Delta_R, \varepsilon} \) (equation (25)).

### 4.3. Spin and valley Zeeman and symmetric spin–orbit coupling

We now consider the total Hamiltonian of equation (26) restricted to the \( N \times N \)-sector. The results in this section are valid when the spin and valley Zeeman coupling constants are weak compared to the spin–orbit strength: \(|h_{S\ell}|, |h_{V\ell}| \ll 2\Delta, j = L, R, i = x, y, z\). We show directly and discuss the effective Hamiltonian for the subspace spanned by \( N \times N \) as a \( 4 \times 4 \) matrix. The columns are associated, from left to right, to the rotated basis states \(|n_-\rangle, |n_\downarrow\rangle, |T_+\rangle, |T_-\rangle\).

\[
H_{\text{eff}, N \times N} = J H_{S\ell} + H_{V\ell} + H_{\text{orb}}, \tag{41a}
\]
\[
\delta h^x_L = h_{S\ell} \pm ih_{S\ell}, \tag{41b}
\]
\[
h_{S\ell} = h_{S\ell} \pm ih_{S\ell}, \tag{41c}
\]

with \( \ell = S, V, j = L, R \) and \( A_+ \) and \( A_- \) are given by
\[
A_\pm = \frac{h^x_L h^y_R + h^y_L h^z_R}{2\Delta \pm H^2_x - H^2_z}. \tag{42a}
\]
\[
A = \frac{A_- + A_+}{2} = \frac{2\Delta h^x_L h^y_R + h^y_L h^z_R}{4\Delta^2 - (H^2_x - H^2_z)}. \tag{42b}
\]

Since \(|h_{S\ell}| \ll 2\Delta\), we can use the approximation \( A_+ \approx A_\approx A \), thus the contribution of \( A \) to equation (40) is an energy shift that can be ignored. Therefore, in this case we can also write

\[
H_{\text{eff}, N \times N} = J H_{S\ell} + H_{S\ell} |N \times N \rangle \langle N \times N| + H_{V\ell} |N \times N \rangle \langle N \times N|, \tag{43}
\]

where \( H_{S\ell} |N \times N \rangle = H_{V\ell} |N \times N \rangle \) are the restrictions of \( H_S \) and \( H_V \) to the \( N \times N \)-sector.

Given the generality of \( H_{\Delta_L, \Delta_R, \varepsilon} \) (equation (25)) as a \( T \)-symmetric term and the fact that \( H_{\Delta_L, \Delta_R, \varepsilon} \) and \( H_S \) only differ in the presence of \( \tau_z \) that multiplies the
spin Pauli matrices, we can assert that the new matrix elements ($H^0_T$, $H^1_T$, $\delta h^0_T$, $\delta h^1_T$, $A_\pm$ and $A_\mp$) that appear in equation (40) as compared to equation (34), are allowed only by breaking the time-reversal symmetry.

5. Conclusions

Motivated by recent experimental works [37–41], we have studied and presented the influence of spin–orbit coupling on the low energy properties of a DQD system with spin and valley DOF in the $(1, 1)$ charge configuration. In our analysis we have also explored the possibility of a different spin–orbit splitting in each dot and we have included a $T$-symmetry breaking magnetic field. In addition, we have discussed under which conditions the LES orbit splitting in each dot and we have included a –also explored the possibility of a different spin configuration. In our analysis we have found that an equal spin–orbit splitting in each dot has no effects on the induced exchange interaction with respect to the case without spin–orbit coupling. On the other hand, asymmetric spin–orbit splitting, spin Zeeman and valley Zeeman modify the exchange coupling constants of three different pairs of antisymmetric states respectively. The modification of the exchange energies is similar for all these three pairs of states.

We also found that TMDCs satisfy the conditions to restrict the LES to the $\mathcal{N} \times \mathcal{N}$-sector, where the effective Hamiltonian for the symmetric spin–orbit coupling case is formally identical to the Heisenberg exchange interaction between two spin–only qubits in valley non-degenerate materials. This renders the Kramers pair an ideal implementation of a qubit in TMDC, as was suggested in [7]. A recipe for a CNOT gate with these states is readily available from the original Loss and DiVincenzo proposal for spin–only qubits [46]. If, in addition, the $\tau_x$ operation could be effectively implemented (theoretical proposals to achieve this include the use of impurities [35, 61] or the use of oscillating confinement potentials [36]), then we would also have a full set for single qubit operations. Moving to the asymmetric spin–orbit coupling we found that only the exchange energy is affected, while the form of the exchange Hamiltonian remains unchanged. The spin–orbit coupling asymmetry offers a way to tune the exchange energy other than the detuning of the dots. Moreover, for the spin and valley Zeeman case, new couplings appeared in the reduced effective Hamiltonian, which originate from the breaking of the time-reversal symmetry.

It is interesting to note that the calculations of [30] suggest the existence of QD states in finite flakes that are localised to the edges of the flakes and whose energies are in the bulk band gap. To our knowledge it is not known if states similar to those of [30] can exist for the gate defined QDs examined here. The consideration of this question is beyond the scope of the present work.

Finally, we expect the results of this work to be useful to understand transport properties of TMDC DQDs. In particular, the existence of a spin-valley blockade might be investigated similarly to earlier studies for CNT DQDs [44, 62–64].

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Appendix A. Eigenstates of spin–orbit coupling

In section 2.2 we introduced the basis states $|s, \delta S \rangle_{n_0, n_1}$, where spin and valley DOF have either singlet or triplet configurations. Not all these states are eigenstates of the spin–orbit coupling defined in equation (5). We show this for the $(1, 1)$-states $|ss \rangle$, $|\ell S_0 \rangle$, $|T_0 S \rangle$ and $|T_0 T_0 \rangle$. As explained for equation (18), the restriction of equation (5) to the $(1, 1)$-subspace is given by $H_{\Delta}^{(1,1)} = \Delta (\tau_L \sigma_L + \tau_R \sigma_R)$. Before applying this operator to the investigated states, we illustrate the action of $\tau_x$ and $\sigma_x$, $j = L, R$, on a singlet state using the definition given in table 1,

$$\tau_x |S \rangle = \tau_x (|K \rangle + |K \rangle) / \sqrt{2} = (|K \rangle + |K \rangle) / \sqrt{2} = |T_0 \rangle_V. \quad (A.1)$$

It is easy to prove the related identities, $$\tau_R |S \rangle = \tau_R (|S \rangle - |S \rangle) = |S \rangle - |S \rangle,$$

Then it follows that $|SS \rangle$ cannot be an eigenstate of $H_{\Delta}$, because

$$H_{\Delta} |SS \rangle = \Delta (\tau_L \sigma_L + \tau_R \sigma_R) |SS \rangle = \Delta (\ell T_0 + T_0 \ell T_0) = 2\Delta |T_0 T_0 \rangle. \quad (A.2)$$

Similarly, we also obtain $H_{\Delta} |T_0 T_0 \rangle = 2\Delta |SS \rangle$, $H_{\Delta} |ST_0 \rangle = 2\Delta |T_0 S \rangle$, $H_{\Delta} |T_0 S \rangle = 2\Delta |ST_0 \rangle$. To conclude, we see that the state $|n_+ \rangle$, given by the linear combination $|ST_0 \rangle + |T_0 S \rangle$ (up to a normalisation factor), is an eigenstate of $H_{\Delta}$ and likewise for the other states in equation (10). The eigenvalues are given by $H_{\Delta} |n_+ \rangle = \pm 2\Delta |n_\pm \rangle$ and $H_{\Delta} |n_\pm \rangle = \pm 2\Delta |n_\pm \rangle$, as illustrated in table 2.

Appendix B. Projectors

Here we explain how to find a compact form for projection operators on spin-valley states which have a particular structure, such as the $(1, 1)$ antisymmetric states described in this paper. For the sake of clarity we list them here explicitly:
\[ |T_+S\rangle = (|K \uparrow; K \downarrow\rangle - |K \downarrow; K \uparrow\rangle)/\sqrt{2}, \quad (B.1a) \]
\[ |T_-S\rangle = (|K \uparrow; K \downarrow\rangle - |K \downarrow; K \uparrow\rangle)/\sqrt{2}, \quad (B.1b) \]
\[ |ST_+\rangle = (|K \uparrow; K \downarrow\rangle - |K \downarrow; K \uparrow\rangle)/\sqrt{2}, \quad (B.1c) \]
\[ |ST_-\rangle = (|K \downarrow; K \downarrow\rangle - |K \downarrow; K \downarrow\rangle)/\sqrt{2}, \quad (B.1d) \]
\[ |n_+\rangle = (|K \uparrow; K \downarrow\rangle - |K \downarrow; K \uparrow\rangle)/\sqrt{2}, \quad (B.1e) \]
\[ |n_-\rangle = (|K \downarrow; K \downarrow\rangle - |K \downarrow; K \downarrow\rangle)/\sqrt{2}, \quad (B.1f) \]

with \(|\tau_{1\sigma_1}; \tau_{2\sigma_2}\rangle = \frac{1}{2} \gamma_{1\sigma_1, \gamma_{2\sigma_2}} [0, 0]|0\rangle\rangle\). The operators that project on these states are shown in equation (11). We can follow two approaches to obtain them, one is more intuitive and gives compact results, assembling projectors on larger parts of the Hilbert space, but in practice it works only when the states we are considering have a structure for which we already know the correct basic projectors. The other one is rather formal but general, however it does not give the projectors in a compact form.

In order to show that equations (11) are really the projectors we are looking for, we will present here the intuitive way to derive them, using symmetries in the structure of states (B.1). Take \(\tau_x\), this operator has eigenvalue \(+1\) when the state is in valley \(|K\rangle\) and eigenvalue \(-1\) when the state is in valley \(|K\rangle\). Thus, \((1 + \tau_x)/2\) is the projector on all those states which have an electron in the \(j\)th QD in valley \(|K\rangle\) and \((1 - \tau_x)/2\) projects on states where the electron in QD \(j\) has valley \(|K\rangle\). Analogous considerations hold for the spin operator \(\sigma_z\) and the spin states \(|\uparrow\rangle, |\downarrow\rangle\). In other words, we wrote down projectors on states which have a certain valley or possess a certain spin in a specific dot. Now focus on \(\tau_x \tau_y \tau_z\), this string of operators has eigenvalue \(+1\) when both electrons are in the same valley \((|KK\rangle\) or \(|\bar{K}\bar{K}\rangle)\) and eigenvalue \(-1\) when the valleys are different \((|K\bar{K}\rangle\) or \(|\bar{K}K\rangle)\). Then, operators \((1 + \tau_x \tau_y \tau_z)/2\) project on states whose valleys are the same \((+)\) or are opposite \((-)\). Similar considerations are valid for their spin counterparts.

The states with the simpler structure are \(|T_+S\rangle, |T_-S\rangle, |ST_+\rangle\) and \(|ST_-\rangle\). They all have one of the properties fixed for both dots (either positive valley, negative valley, spin up or spin down, respectively). Consider \(|T_+S\rangle\) (equation (B.1a)), for both dots the valley is \(|K\rangle\), but the spins are in a singlet state. The combination

\[ \frac{1}{4} (1 + \tau_x)(1 + \tau_y)(1 + \tau_z) \]

projects on all the states with positive valley in both dots. To complete the expression we multiply by the projector on the spin singlet, given by \((1 - \sigma_L \cdot \sigma_R)/4\).

Finally, the complete projector is

\[ P_{(T_+S)} = \frac{1}{16} (1 + \tau_x)(1 + \tau_y)(1 + \tau_z)(1 - \sigma_L \cdot \sigma_R), \]

as in (11a). The other very similar expressions in (11a) and (11b) follow the same derivation with the appropriate changes of signs and spin/valley operators.

States \(|n_\pm\rangle\) of equations (B.1e) and (B.1f) have a more complicated structure. First of all, they are both composed of states with opposite valley and opposite spin in the dots. The operator

\[ \frac{1}{4} (1 - \tau_x \tau_y \tau_z) \]

projects on the subspace spanned by \(|K \uparrow; \bar{K} \downarrow\rangle, |K \downarrow; \bar{K} \uparrow\rangle, |K \uparrow; \bar{K} \downarrow\rangle, |K \downarrow; \bar{K} \uparrow\rangle\). To distinguish between \(|n_+\rangle\) and \(|n_-\rangle\), we note that for \(|n_+\rangle\) the product of the eigenvalues of spin and valley inside each dot is positive, while for \(|n_-\rangle\) it is negative (this is why they get an energy shift of \(\pm 2\Delta\) by the spin–orbit coupling \(H_D\)). We use \(\tau_x \tau_y \tau_z\) to distinguish them and the projectors become

\[ \frac{1}{8} (1 - \tau_x \tau_y \tau_z)(1 - \sigma_L \sigma_R)(1 + \tau_x \tau_y \tau_z), \]

positive sign for the subspace spanned by \(|K \uparrow, \bar{K} \downarrow, |K \downarrow, \bar{K} \uparrow\rangle\) and negative sign for the subspace spanned by \(|K \downarrow, \bar{K} \uparrow, |K \uparrow, \bar{K} \downarrow\rangle\). Finally, the operator that identifies the correct superposition is \(\tau_x \tau_y \tau_z \sigma_L \sigma_R \sigma_R\), the same for both states. The total projectors \(|n\rangle\rangle\) are

\[ P_{|n\rangle\rangle} = \frac{1}{16} (1 - \tau_x \tau_y \tau_z)(1 - \sigma_L \sigma_R)(1 + \tau_x \tau_y \tau_z)(1 - \tau_x \tau_y \tau_z \sigma_L \sigma_R \sigma_R), \]

as in equation (11c).

Formally, the above results can also be obtained by noticing that the projectors we are looking for are spin–valley operators (i.e. acting on the \((1,1)\)-subspace) and every spin-and-valley operator \(P\) can be expanded in a linear combination of Pauli operator strings,

\[ P = \sum_{i,j,k,l=0,1,0,1} P_{ijkl} \tau_i \sigma_L \tau_j \sigma_R \tau_k \sigma_R, \]

where \(P_{ijkl}\) are complex coefficients. If \(|\psi\rangle\) is the state we want to project on, the projector is \(P = |\psi\rangle\langle\psi|\) and we can use the inner product provided by the trace operation to obtain the expansion coefficients,

\[ P_{ijkl} = \text{Tr} \left[ |\psi\rangle\langle\psi| \tau_i \sigma_L \tau_j \sigma_R \tau_k \sigma_R \right]. \]

Again, recollecting a compact expression from this set of coefficients requires some work (even when most of them are zeros) and does not give any physical insight.

For completeness, we briefly discuss \(P_{\sigma_0}\), the projector on the whole antisymmetric subspace of \((1,1)\)-states of equation (12). Although this operator could be obtained as the sum of the 6 projectors of equation (11) on the single, linearly independent antisymmetric states, we can write it down following another procedure, derived in [45]. It can be expressed as a combination of projectors on the singlet (S,
antisymmetric) and on the triplet ($T$, symmetric) subspaces for spin and valley,

$$P_{as} = P_{sp}^S p_{\text{valley}}^T + P_{sp}^T p_{\text{valley}}^S$$  \hfill (B.9)

Here we defined the spin projectors as

$$P_{sp}^S = \frac{1 - \sigma_L \cdot \sigma_R}{2}, \quad P_{sp}^T = \frac{3 + \sigma_L \cdot \sigma_R}{4}.$$  \hfill (B.10)

The valley projectors have the same form, only substituting $\tau$’s for $\sigma$’s. Equation (B.9) yields, then,

$$P_{as} = (3 - \sigma_L \cdot \sigma_R - \tau_L \cdot \tau_R - (\sigma_L \cdot \sigma_R)(\tau_L \cdot \tau_R))/(8).$$  \hfill (B.11)

**Appendix C. Conditions on spin and valley Zeeman coupling constants for (1,1)-subspace**

In section 3.3 we discussed the effects of the spin and valley Zeeman term on the exchange interaction in the case when the (1,1)-subspace is our LES. There we omitted to show the conditions to ensure that (1,1)-states are lower in energy than (2,0) and (0,2) states. We report them here. To simplify the discussion we assume that the spin Zeeman coupling constants associated with $x$- and $y$-Pauli matrices are small: $|\delta_{x,y}| \ll U$, $J = L, R$, $i = x, y$. Then, choosing an appropriate basis where $\sigma_x$ and $\tau_x$ are diagonal, it is easy to see that (1,1)-states are lower under the condition,

$$\max \{-2\Delta + |H_2 - H_V|, 2\Delta + |\delta H_2 - \delta H_V|, |\delta H_2 + \Delta H_V|, |\delta H_2 - \Delta H_V|\} + 2\max \{|\Delta|, |H_{2LZ}|, |H_{2VZ}|, |H_{2Sz}|, |H_{2Rz}|\} < U - |\varepsilon|,$$  \hfill (C.1)

where we used the same notation described in equation (41).

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