Electrically tunable g-factors in quantum dot molecular spin states

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We present a magneto-photoluminescence study of individual vertically stacked InAs/GaAs quantum dot pairs separated by thin tunnel barriers. As an applied electric field tunes the relative energies of the two dots, we observe a strong resonant increase or decrease in the g-factors of different spin states that have molecular wavefunctions distributed over both quantum dots. We propose a phenomenological model for the change in g-factor based on resonant changes in the amplitude of the wavefunction in the barrier due to the formation of bonding and antibonding orbitals.

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Quantum Dots and Quantum Dot Molecules (QDMs) have proven to be a versatile medium for isolating and manipulating spins, which are of great interest for quantum information processing. In particular, photoluminescence (PL) spectra have been used in self-assembled QDMs to observe coherent tunneling of electrons and identify spin interactions through fine structure. Electrical control of isolated spins through their g-factors is highly desirable for implementation of quantum gate operations. To date, electrical control of g-factors has only been observed in ensembles of electrons in quantum wells and isolated quantum dots. We present a striking electric field resonance in the g-factor for molecular spin states confined to a single quantum dot molecule.

To our knowledge this is the first observation of electrical control over the g-factor for a single confined spin. Moreover, the isolation of a single QDM allows us to spectrally resolve and identify individual molecular spin states that have different g-factor behaviors. In Fig. 1a, we indicate molecular spin states of both the neutral exciton (X0), one electron recombining with one hole) and positive trion (X+, electron-hole recombination in the presence of an extra hole) at zero magnetic field. The different electric field dependences of the g-factors for these states is apparent in Fig. 1b, where the splitting of PL lines increases for some molecular spin states and decreases for others. This electric field dependence is nearly an order of magnitude larger than previously reported in quantum wells. The effect arises from the formation of bonding and antibonding orbitals, which results in a change in the amplitude of the wavefunction in the barrier at resonance.

Our QDMs consist of two vertically stacked self-assembled InAs dots truncated at a thickness of 2.5 nm and separated by a 2 nm GaAs tunneling barrier. As an applied electric field tunes the relative energies of the two dots, strong tunnel coupling between the hole states creates the molecular spin states. Unlike samples with a thicker tunnel barrier, the states retain molecular character throughout the observed range of electric fields. We present data from a single molecule, but the universality of the behavior has been verified by detailed studies of 7 other molecules from the same sample. We first explain the spectra and molecular spin states at B = 0 T. We then describe the magnetic field dependence and propose a phenomenological model for the electric field dependent Zeeman splitting.

In Fig. 2 we show all PL lines from X0 and X+ at B = 0 T. These lines are identified by their relative energies, the power dependence of their intensities and the electric field dependence of the anticrossings. The X0 lines (Fig. 2a) show a clear anticrossing at Fx0. The X+ lines (Fig. 2b) have a more complicated pattern because anticrossings occur in both the initial (one electron and two holes) and final (one hole) states.

To explain the origin of the X+ molecular spin states...
If there is one hole in each dot, singlet (both in the bottom dot) or metric hole spin wavefunction ($\downarrow \uparrow$) between dot centers and (0 wavefunction ($\downarrow \uparrow$) the total spin projection. Singlets, which have total spin 0, will be denoted $X_S$ to distinguish them from the ±1/2 triplets.

The final state has only a single hole with relative energies given by the Hamiltonian:

$$\hat{H}^h = \begin{pmatrix} 0 & t_h \\ t_h & edF \end{pmatrix}$$

in the spin degenerate $\frac{1}{2}, \frac{1}{2} \frac{1}{2}, \frac{1}{2} \frac{1}{2}$ basis, where $t_h$ is the tunneling energy for a bare hole, $d$ is the distance between dot centers and $F$ is the applied electric field. The energies of the final states as a function of electric field are shown in Fig. 2b. The formation of bonding and antibonding states at the anticrossing point is illustrated.

The initial state ($X^+$) contains one electron and two holes. For simplicity we present only the electron-spin-up case, which is degenerate with the spin-down case at zero magnetic field. If both holes are in the same dot, the Pauli principle requires singlet configurations: $\uparrow \uparrow 0 \uparrow - \downarrow$ (both in the bottom dot) or $\uparrow \downarrow 0 \downarrow$ (both in the top dot).

If there is one hole in each dot, singlet ($\uparrow \downarrow 0 \downarrow$) and triplet ($\uparrow \downarrow \downarrow 0$, $\downarrow \uparrow \downarrow 0$) configurations are possible. By $\uparrow \downarrow 0 \downarrow$ we mean the antisymmetric hole spin wavefunction ($\downarrow \uparrow - \uparrow \downarrow$). $\uparrow \downarrow 0 \downarrow$ means the symmetric hole spin wavefunction ($\downarrow \uparrow + \uparrow \downarrow$).

The initial state ($X^+$) configurations are possible. The relative energies of the initial states are given by the Hamiltonian:

$$\hat{H}^{X^+} = \begin{pmatrix} \Gamma_1 & t_{X^+} & 0 & 0 & 0 \\ t_{X^+} & edF & J^h & 0 & 0 \\ 0 & J^h & E_{+1/2} & 0 & 0 \\ 0 & 0 & 0 & 0 & E_{-5/2} \\ 0 & t_{X^+} & 0 & 0 & 0 \end{pmatrix}$$

in the $\uparrow \uparrow \uparrow \uparrow \uparrow$, $\uparrow \downarrow \downarrow \uparrow \downarrow$, $\downarrow \uparrow \uparrow \uparrow \downarrow$, $\downarrow \downarrow \uparrow \uparrow \downarrow$, $\uparrow \downarrow \downarrow \downarrow \downarrow$, $\downarrow \uparrow \downarrow \downarrow \downarrow$ basis. $\Gamma_1$ and $\Gamma_2$ are due to Coulomb interactions, $t_{X^+}$ is the Coulomb-corrected tunneling energy for a hole in the presence of an electron and additional hole, $E_k = edF + mJ^h$ with $m = 0, +1, -1$ for $k = 1/2, -5/2, +7/2$. $J^h$ is the exchange energy between an electron and hole in the same dot.

FIG. 2: Energies extracted from Fig. 1. (a) $X^0$ lines anticross at $F_{X^0}$, where the direct (lower inset) and indirect (upper inset) transitions are degenerate. (b) $X^+$ initial states anticross at $F_{X^+}$ and near 90 kV/cm. Final (hole) states anticross at $F_h$. Inset: charge distribution for the circled singlet and triplet transitions.

FIG. 3: Calculated zero magnetic field energies of spin-degenerate (a) initial states ($X^+$) and (b) final states ($h^+$) inset. Labels indicate the dominant basis state for easy comparison to Eq. 2. Insets in (b) show the bonding (lower) and antibonding (upper) wavefunctions (green) and probability distributions (red). (c) Fine structure of singlet and triplet states at $F = 79.2$ kV/cm. Arrows indicate optically allowed transitions to the bonding final state.
singlet states, the triplet states do not mix and are not affected by these anticrossings. This creates a "kinetic" splitting between triplet and singlet states. An example is indicated by the dashed oval in Fig. 2b, where the lower energy singlet line remains separated from the two optically allowed triplet lines, which are split by electron-hole exchange. The fine structure of the corresponding molecular spin states. Fig. 4a shows the states of Fig. [19], we calculate the magnetic field dependence of the states is shown in Fig. 3c.

FIG. 4: (a) Magnetic field dependence of initial ($X^+$) and final ($h$) states for the singlet and triplet transitions at an electric field of 79.2 kV/cm (schematic Fig. 3c). Vertical lines indicate spin allowed recombinations. (b) Calculated (colored orientations of a single hole, split by 3c, which split into doublets with an applied longitudinal molecular spin states. Fig. 4a shows the states of Fig. 3c, which split into doublets with an applied longitudinal molecular spin states. Fig. 4a shows the states of Fig. [19], we calculate the magnetic field dependence of the states is shown in Fig. 3c.

FIG. 5: Zeeman splitting and corresponding g-factor, $g_T$, as a function of electric field at B = 6 T.

mentally observed PL energies are given by the symbols, with the diamagnetic shift (10.9 $\mu$eV/T$^2$) subtracted.

The g-factor resonances are clearly evident in Fig. 5, where the symbols plot the measured energy splitting of the X$^0$ and two $X^+$ Zeeman doublets at B = 6 T as a function of electric field. Strong enhancement or suppression of the splitting is observed at $F_h$ and $F_{X^0}$. All of the data can be qualitatively explained by a phenomenological model of the formation of bonding and antibonding orbitals, which results in resonant changes in the amplitude of the wavefunction in the barrier. We focus first on transitions involving a bonding orbital (open symbols), which have an increased amplitude of the wavefunction in the barrier (Fig. 5, lower inset).

The wavefunction for the bonding orbital of a single hole can be written as $\Psi_B = a|1\rangle + b|2\rangle$, where $|1\rangle$ and $|2\rangle$ are the basis states of Eq. 1, the wavefunctions for holes localized in the two different dots. The coefficients $a$ and $b$ are functions of electric field determined by Eq. 1. The electric field-dependent g-factor for a hole in a bonding orbital is given by $g^B_h(F) = \langle \Psi_B | g_h(z) | \Psi_B \rangle$, where $g_h(z)$ is the hole g-factor as a function of position in the sample. $g_h(z)$ is taken as a phenomenological parameter, in part because the degree of alloying between the nominally pure InAs dots and GaAs barrier is unknown.

If we assume that the g-factors for the holes localized in each dot are the same, we get $g^B_h(F) = g_h + 2ag_0z$, where $g_{12} = \langle 1 | g_h(z) | 2 \rangle$, the initial states are split by $g_e$, so the total transition g-factor is given by

$$g_T^B(F) = g_h + g^B_h(F) = g_T + \frac{2t_h g_{12}}{\sqrt{e^2d^2(\Delta F - F_h)^2 + 4t_h^2}}$$
where the explicit form for $2ab$ determines the lineshape centered around the anticrossing point $F_h$.

The lower black line in Fig. 3 is obtained by fitting Eq. 4 to $\Delta X^+_B$. Using the measured values of $t_h$ (0.86 meV) and $F_h$ (63.3 kV/cm) we find $g_{12} = 1.65$. The barrier contribution is positive, like the heavy hole g-factor in bulk GaAs ($\sim 1.05$) [21, 22]. Because $g_T$ and $g_{12}$ have opposite sign, the splitting reaches a minimum at $F_h$, where the amplitude in the barrier is at a maximum. The g-factor at the minimum is $-0.44$.

Because the $\pm 5/2$ triplet states recombine to the same bonding orbital of the final hole (Fig. 4), the model predicts the splitting of these lines as a function of electric field with no additional fitting. As shown in Fig. 4, the $\pm 5/2$ triplet states have an initial state splitting of $g_e + 2g_h$. The transition g-factor is therefore given by $g_T^B(F) = (g_e + 2g_h) - (g_h + 2abg_{12}) = g_T - 2abg_{12}$. This is shown by the upper black line, which matches $\Delta X^+_B(\pm 5/2)$, the observed splitting of the $\pm 5/2$ triplets. The maximum splitting corresponds to a g-factor of $-4.23$.

To apply the g-factor model to the bonding orbital of the neutral exciton (lower branch Fig. 4) we use the measured tunneling energy ($t_X = 0.58$ meV) and anticrossing field ($F_X = 82.1$ kV/cm). The lower red line in Fig. 5 shows the fit to $\Delta X^+_B$, the Zeeman splitting of PL lines from the bonding orbital. We find $g'_{12} = 1.32$. The electron-hole Coulomb interaction is responsible for the difference in tunneling energy and anticrossing field from the bare hole case and is also likely responsible for the difference between $g_{12}$ and $g'_{12}$. The g-factor at the minimum is $-0.59$.

Using this value of $g'_{12}$, the model immediately explains the increase in splitting for the antibonding orbital (upper branch Fig. 4), which has a reduced wavefunction amplitude within the barrier. The g-factor for the antibonding orbital is given by $g_T^B(F) = g_T - 2abg'_{12}$, which increases in magnitude at the resonant field because $g_T$ is negative and $g'_{12}$ is positive. This is shown by the upper red line in Fig. 5 which matches $\Delta X^+_A$, the Zeeman splitting for excitonic recombination from the antibonding orbital. The splitting increases to a maximum (g-factor $-3.35$) at the anticrossing point. The antibonding transitions for $X^+$ show similar behavior, but are too weak to obtain full resonance curves.

The overall agreement between the model and experimental data is quite good. There are some minor discrepancies, which highlight the need for a detailed theory, possibly requiring inclusion of excited states [23]. However, the agreement of the data with the resonance linewidths calculated using independently measured values of $t_X$ and $t_h$ is strong confirmation that the g-factor dependence does arise from the formation of bonding and antibonding orbitals. For $X^+$ the g-factor resonance arises from the wavefunction of the single hole in the final state, while for $X^0$ the orbital wavefunction of the hole is influenced by the additional electron.

We also studied samples in which electrons tunnel through the barrier, with an anticrossing energy ($\sim 2.3$ meV) comparable to that of the hole tunneling sample presented here ($\sim 1.7$ meV). This requires a thicker barrier (10 nm) because of the smaller electron effective mass. The electron wavefunction amplitudes in the barrier should be at least as large as the hole-tunneling case. However, the electron g-factor in bulk GaAs ($-0.44$) [24] is similar to the electron g-factor in InAs quantum dots ($-0.6$) and according to our model the contribution from the barrier should not significantly change the electron g-factor. We see no electric field dependence of the g-factor in these electron anticrossing samples. By adding aluminum to the barrier, the resonant contribution to the electron g-factor could be enhanced.

We have presented a resonant change in g-factor as a function of electric field for the molecular spin states of QDMs with a thin tunnel barrier. By studying single QDMs, we are able to identify the individual molecular spin states and the different resonant behavior of their g-factors. The results suggest that design of molecular spin states and tunnel resonances may provide new opportunities for combining optical and electrical control of confined spins.

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