Two Carriers in Vertically Coupled Quantum Dots: Magnetic Field Effect

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We study a two-charge-carrier (two holes or two electrons) quantum dot molecule in a magnetic field. In comparison with the electron states in the double quantum dot, the switching between the hole states is achieved by changing both the inter-dot distance and magnetic field. We use harmonic potentials to model the confining of two charge carriers and calculate the energy difference ΔE between the two lowest energy states with the Hund-Mulliken technique, including the Coulomb interaction. Introducing the Zeeman effect, we note a ground-state crossing, which can be observed as a pronounced jump in the magnetization at a perpendicular magnetic field of a few Tesla. The ground states of the molecule provide a possible realization for a quantum gate.

Keywords: Quantum Dots, Valence Band Mixing, Harmonic Potentials, Hund-Mulliken Technique, Switching, Phase Transition, Qubit, Magnetization.

RESEARCH ARTICLE

Quantum dots (QDs) are small quasi-zero-dimensional semiconductor structures with a discrete density of states and electronic shells similar to those of atoms.[1,2] These "artificial atoms" can be charged either with conduction electrons or with conduction electrons and valence band holes. The experiments and theory on electronic properties of QDs charged with electrons are progressing very rapidly.[1,3] Some experiments[4] aim at the creation of a dense electron-hole plasma in artificial atoms. Clearly, an understanding of interacting electrons and holes confined in QDs is needed. However, the confinement of the charge carriers can be achieved by electrical gating and/or a magnetic field. The dimensions of QDs are on the order of the Fermi wavelength. Typical laboratory magnetic fields ($B \approx 1$ T) correspond to magnetic lengths on the order of $l_B \approx 10$ nm, which is much larger than the Bohr radius of real atoms but the same size as artificial atoms. As a consequence, the dot spectrum depends strongly on the applied magnetic field.[5–7] In coupled QDs, which can be considered to some extent as artificial molecules, Coulomb blockade effects[8] and magnetization[9] have been observed, as has the formation of a delocalized "molecular state."[10] The spin $\frac{1}{2}$ of an electron is a "natural" representation of a qubit since it comprises exactly two levels; there are no additional degrees of freedom into which the system could "leak" and thereby cause errors in a quantum computation. These advantages have motivated the idea of spin-based solid-state quantum computation with the use of electron spins S_e in coupled QDs,[11–17] where the required two-spin coupling is provided by the Heisenberg exchange interaction between the two spins in adjacent QDs. The microscopic origin of the exchange coupling lies in the virtual tunneling of carriers from one QD to the other and back, and there are several external physical parameters (gate voltage, magnetic field, etc.) that can be used for controlled quantum gate operation.[17] QDs are the most attractive structure in terms of miniaturization, scaling up, electronic and optical control of the qubits, and their logic operations. Therefore, an approach to quantum computing based on QD spin is especially interesting. We concentrate in this paper on the magnetic properties of pairs of QDs in which two particles (holes or electrons) are vertically coupled and are subject to the Coulomb interaction.

Magneto-optical experiments have revealed unusually long spin coherence times exceeding 100 ns in the case of n-doped semiconductors and hole spin coherence times in p-doped GaAs quantum wells of 700 ps (Ref. 18) and hence support the conjecture that coherence also exists in this latter case; thus total hole spins (or, rather, total angular momentum J_h) in semiconductors may be a possible candidate for a scalable quantum computer architecture. Motivated by this, and under some approximations, we show that the total angular momentum J_h of two holes confined in vertically coupled QDs may be considered as a quantum gate (in spite of the many problems induced by the spin-orbit coupling, the strong nonparabolicity subband caused by the mass's anisotropy, coherence, and, in general, the complexity of the valence band structure). We use the Hund-Mulliken technique, thus exploiting the analogy between QDs ("artificial atoms"[19]) and natural

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atoms to calculate the energy difference of two vertically coupled QDs containing two particles as a function of the inter-dot distance (d) and magnetic field (B). From the energy spectrum, we derive the equilibrium magnetization as a function of the magnetic field to measure the charge carrier's total angular momentum J_h and S_e.

We first start our calculation by considered the problem of a single-particle Hamiltonian of the charge carriers (electron or hole) confined in the double-dot system. For simplicity, we first study the problem of an electron. In a magnetic field $B = (0, 0, B)$ that corresponds to the vector potential $A = B(-y, x, 0)/2$, the one-particle Hamiltonian by which we describe a single electron in the upper ($+d/2$) or lower ($-d/2$) dot of the double-dot system is

$$H_{\pm d/2}(\rho, z) = \frac{1}{2m}\left(p - \frac{eA(\rho, z)}{c}\right)^2 + V_L(\rho, z) + V_V(\rho, z) \tag{1}$$

where m is the electron effective mass, and the potential V_L describes the lateral confinement, whereas V_V models the vertical double-well structure with quantization energy $\hbar w_z$. For the lateral confinement we choose the parabolic potential,

$$V_L(\rho) = \frac{mw_z^2}{2}\begin{cases}\alpha_{0+}^2\rho^2, & z > 0\\ \alpha_{0-}^2\rho^2, & z < 0\end{cases} \tag{2}$$

where we have introduced the anisotropy parameters $\alpha_{0\pm}$ determining the strength of the vertical relative to the lateral confinement. Note that for dots of different size ($\alpha_{0+} \neq \alpha_{0-}$) the potential (Eq. (2)) is not continuous at $z = 0$. The lateral effective Bohr radii $R_{B\pm} = \sqrt{\hbar/mw_z\alpha_{0\pm}}$ are a measure of the lateral extension of the electron wave function in the dots. In describing the confinement V_V along the inter-dot axis, we have used a (locally harmonic) double-well potential of the form

$$V_V(z) = \frac{mw_z^2}{2d^2}(z^2 - (d/2)^2)^2 \tag{3}$$

which, in the limit of a large inter-dot distance $d \gg R_{B\pm}$, separates (for $z \approx \pm d/2$) into two harmonic wells (one for each dot) of frequency w_z. The ground-state Fock-Darwin solution,[20]

$$\varphi_{\pm d/2}(\rho, z) = (mw_z/\pi\hbar)^{3/4}\sqrt{\alpha_\pm}\exp -(mw_z/2\hbar) \times \{\alpha_\pm\rho^2 + (z \mp d/2)^2\} \tag{4}$$

with $w_L(B) = (eB/2mc)$ is the Larmor frequency, and the parameter $\alpha_\pm(B) = \sqrt{\alpha_{0\pm}^2 + w_L^2(B)/w_z^2}$ describes the compression of the one-particle wave function perpendicular to the magnetic field. $\varphi_{+d/2}(\rho, z)$ is the ground-state wave function of the particle confined in the upper part of the double-dot system, whereas $\varphi_{-d/2}(\rho, z)$ corresponds to the lower dot. In this case (d is infinite) and when the QDs are equivalent, the bound states of the discrete spectrum are twofold degenerate: the particle can be found either in one dot or in the other, and when the dots are different in size, the larger dot is energetically favorable and the particle can be found in it. At finite d, the previous eigenstates are not longer eigenstates of the coupled-dot Hamiltonian, $H_{\pm d/2}(\rho, z)$. Let $\varphi_{+d/2}(\rho, z)$ be the wave function of the particle centered at ($z = d/2$), despite the exponential decay displayed by $\varphi_{+d/2}(\rho, z)$, when $|z - d/2| > R_{B_\pm}/2$, $\varphi_{+d/2}(\rho, z)$ centered on ($z = -d/2$) is different from zero. Thus $\varphi_{+d/2}(\rho, z)$ is not proportional to $H_{\pm d/2}\varphi_{+d/2}(\rho, z)$. On the other hand, the overlap s defined by $s = \langle\varphi_{+d/2}(i)|\varphi_{-d/2}(i)\rangle$ is different from zero, where (i) is a short notation for (ρ_i, z_i). A nonvanishing overlap s implies that the electron can tunnel between the dots. If d is large enough, we may expect coupling between the dots due to the tunnel effect. From the exact solution of $H_{\pm d/2}(i)$,

$$\Phi(i) = \beta\varphi_{+d/2}(i) + \theta\varphi_{-d/2}(i) \tag{5}$$

we only retain for the lowest states of the double QD a linear combination of the ground states of the isolated dots; the best coefficients (β, θ) will be those that give the minimum value to the orbital energy. The energy ε_+ corresponds to $\Phi_+(i) = \beta_+\varphi_{+d/2}(i) + \theta_+\varphi_{-d/2}(i)$, whereas ε_- corresponds to $\Phi_-(i) = \beta_-\varphi_{+d/2}(i) + \theta_-\varphi_{-d/2}(i)$.

When we build up a "molecule" starting from two "atomic" systems of this type, the Hamiltonian for the two interacting electrons in the potential of double QDs, including the Coulomb interaction, is

$$H = H_{\pm d/2}(1) + H_{\pm d/2}(2) + \frac{e^2}{\varepsilon|r_1 - r_2|} \tag{6}$$

where ε is the dielectric constant and $H_{\pm d/2}(i)$ is the single-particle Hamiltonian given by (Eq. (1)), for which we know the exact solution. Using the Hund-Mulliken approach, we generate four basis functions with respect to which we diagonalize the two-particle Hamiltonian H. Finally, we can calculate the two lowest energy states and therefore the energy difference ΔE.

The Zeeman splitting $H_Z = g\mu_B BS_e$ is not included in the two-particle Hamiltonian (Eq. (6)), since in the absence of spin-orbit coupling one can treat the orbital problem separately and include the Zeeman interaction later (which we will do when we study the low-energy spectra and magnetization). Here we have denoted the effective Lande factor by g and the Bohr magneton by μ_B.

As far as the hole states are concerned, we use the Luttinger Hamiltonian H_L, which describes the Γ_g^v valence band with intrinsic angular momentum of the hole $j_h = 3/2$ (with the z projection $m_h = \pm 3/2$ for heavy hole, hh, and $m_h = \pm 1/2$ for light hole, lh).[21] We neglect the split of the band, which in GaAs is 340 meV below the

valence band edge.[22] The confinement of the carriers is of the form $V^{hh,lh}(\rho,z) = V_{\mathrm{L}}^{hh,lh}(\rho,z) + V_{\mathrm{V}}^{hh,lh}(\rho,z)$, where

$$V_{\mathrm{L}}^{hh,lh}(\rho,z) = \frac{m_0}{(\gamma_1 \pm \gamma_2)} \frac{(w_z^{hh,lh})^2}{2} \begin{cases} (\alpha_{0+}^{hh,lh})^2\rho^2, & z>0 \\ (\alpha_{0-}^{hh,lh})^2\rho^2, & z<0 \end{cases} \tag{7}$$

describes the lateral heavy (light) hole confinement, whereas

$$V_{\mathrm{V}}^{hh,lh}(\rho,z) = \frac{m_0}{(\gamma_1 \mp 2\gamma_2)} \frac{(w_z^{hh,lh})^2}{2(d)^2} \left(z^2 - \left(\frac{d}{2}\right)^2\right)^2 \tag{8}$$

describes the vertically heavy (light) hole confinement and m_0 is the free electron mass. The Hamiltonian of 3D confined hole states reads

$$H_{\pm d/2}^{\mathrm{L}} = \frac{\hbar^2}{m_0} \begin{pmatrix} H_{hh} & R & S & 0 \\ R^* & H_{lh} & 0 & S \\ S^* & 0 & H_{lh} & -R \\ 0 & S^* & -R^* & H_{hh} \end{pmatrix} \tag{9}$$

where

$$H_{hh,lh} = -\frac{1}{2}\left[\frac{\partial}{\partial z}(\gamma_1 \mp 2\gamma_2)\frac{\partial}{\partial z} + \frac{1}{\rho}\frac{\partial}{\partial \rho}(\gamma_1 \pm \gamma_2)\rho\frac{\partial}{\partial \rho} - \frac{(\gamma_1 \pm \gamma_2)}{\rho^2}\hat{l}_z^2\right] + V^{hh,lh}(\rho,z) + (\gamma_1 \pm \gamma_2)\left[\frac{1}{2}\left(\frac{\omega_c}{2}\right)^2\rho^2 + \left(\frac{\omega_c}{2}\right)\hat{l}_z\right] \tag{10}$$

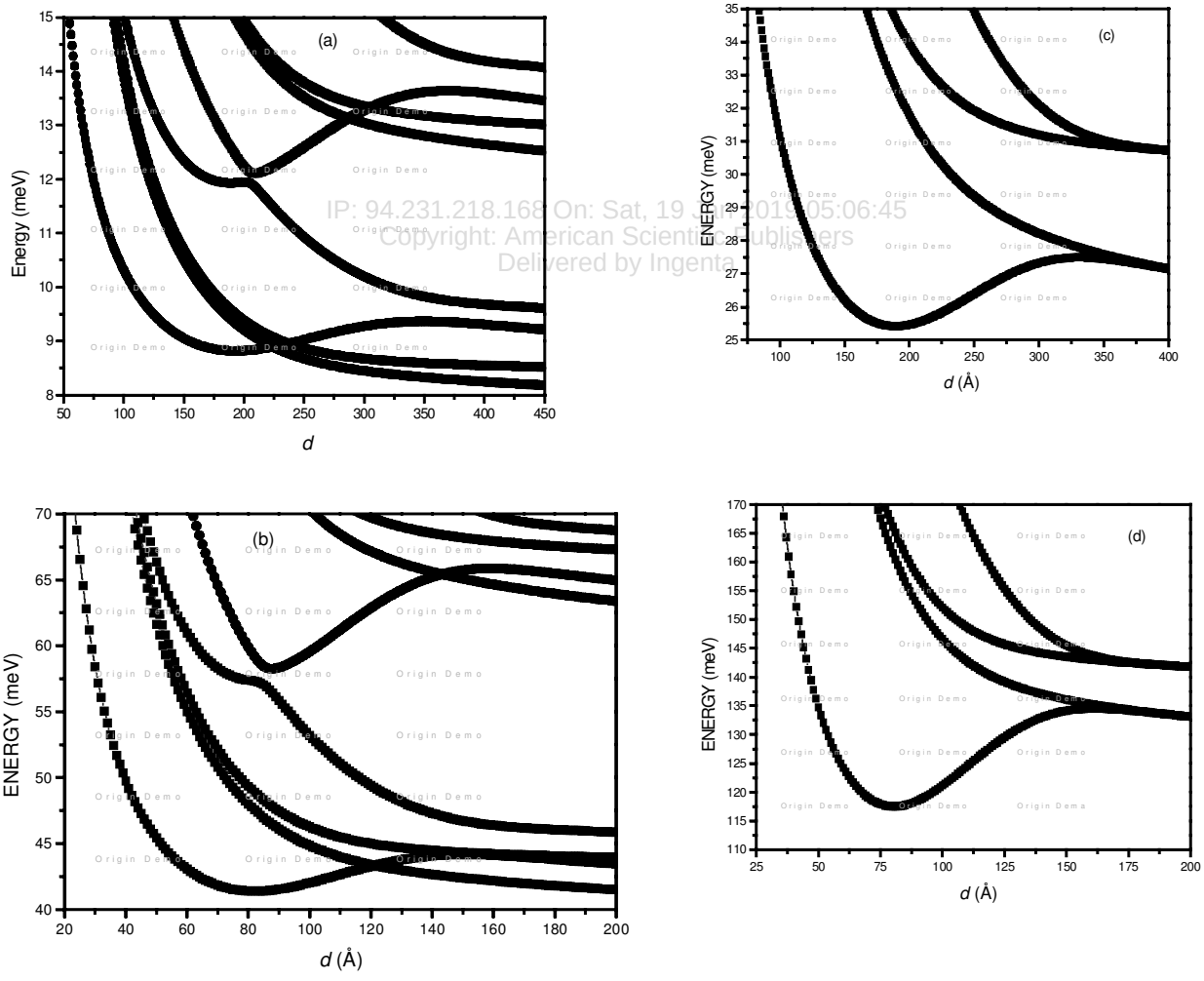

Fig. 1. Energy as a function of the inter-dot distance. Left graphs (a and b): Spectrum for the lowest eight hole levels. The first one corresponds to the *hh* state. In the weak confinement $\hbar w_z^{hh(lh)} = 3$ meV (12.5 meV), the lowest level crossing occurs at $d = 224$ Å, whereas in the strong confinement $\hbar w_z^{hh(lh)} = 16$ meV (66.71 meV) the ground state changes from $J_h = 0$ to $J_h = 1$ at about 120 Å. The anisotropy parameter is $\alpha_{\pm}^{hh(lh)} = 0.67(0.085)$. Right graphs (c and d): Two-electron spectra, again plotted as a function of the inter-dot distance in weak and strong confinement, that is, $\hbar w_z^e = 16$ meV and 90 meV, respectively. Note that two-electron and two-hole spectra are clearly distinguishable; in particular, there is no ground-state crossing for two electrons.

$$R = \frac{\sqrt{3}}{2}\hat{D}\bar{\gamma}\hat{D}, \quad S = \frac{\sqrt{3}}{2}\left(\hat{D}\gamma_3\frac{\partial}{\partial z} + \frac{\partial}{\partial z}\gamma_3\hat{D}\right) \tag{11}$$

$$\hat{D} = e^{-i\theta}\left[\frac{\partial}{\partial\rho} + \frac{1}{\rho}\hat{l}_z + \frac{\omega_c}{2}\rho\right] \tag{12}$$

$\hat{l}_z = -i\frac{\partial}{\partial\theta}$ is the component of the orbital angular momentum and $\omega_c = eB/c$ is the (free electron) cyclotron frequency. The atomic Zeeman splitting is incorporated by adding the terms $\kappa\omega_c m_h$ to the diagonal of $H^L_{\pm d/2}$. $(\gamma_1, \gamma_2, \gamma_3, \kappa)$ are the Luttinger parameters. In writing (Eq. (11)), we have adopted the axial approximation,[21] by inducing the parameter $\bar{\gamma} = (\gamma_2 + \gamma_3)/2$.

In the absence of coupling between heavy-hole and light-hole valence bands, the method for obtaining the hole eigenstates proceeds along the same lines as for electrons,[23] whereas when the coupling between *hh* and *lh* states is introduced through operators R and S, the eigenstates of the Luttinger Hamiltonian can be written in the form of four-component spinors.

When we turn to the system of two holes in double QD, the problem of finding the hole eigenstates seems more difficult than that of two electrons. To simplify the calculation and to relate this to the case of electrons (with which we compare our results), the problem can be solved as follows: first we neglect the coupling between *hh* and *lh* bands, since the eigenfunctions are similar to (Eq. (4)), except for the introduction of a few modifications induced by the mass's anisotropy. For this, we assume a low-temperature description where $kT \ll \hbar w_z^{hh(lh)}$, so that we can restrict ourselves to the two lowest orbital eigenstates of $H = H^L_{\pm d/2}(1) + H^L_{\pm d/2}(2) + e^2/\varepsilon|r_1 - r_2|$, one of which is $J_h = 0$ and the other is $J_h = 1$. We neglect the multiplet states that correspond to $J_h = 2$ and $J_h = 3$.

Throughout this paper, we have evaluated our results for two vertically equal small (large) GaAs QDs ($\gamma_1 = 6.85$, $\gamma_2 = 2.1$, $\gamma_3 = 2.9$, $\varepsilon = 13.1$), 79 Å (183 Å) in

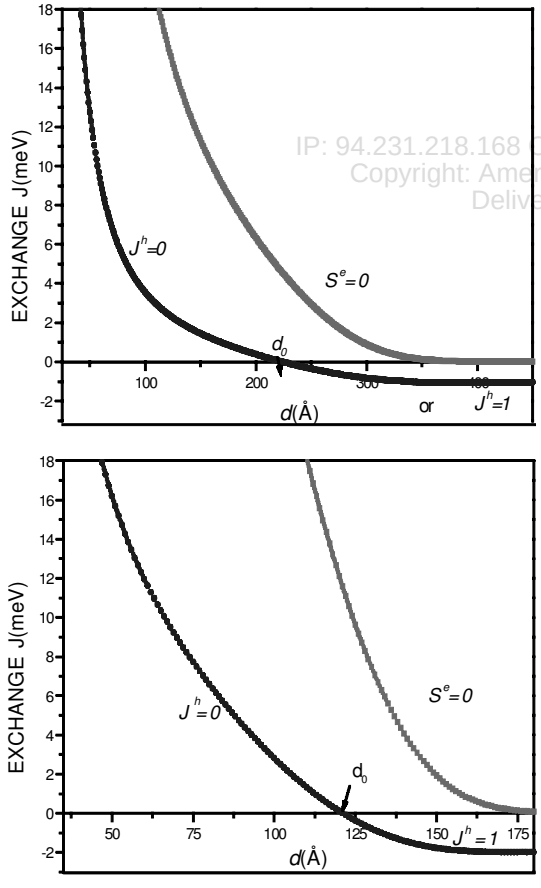

Fig. 2. The energy difference as a function of the inter-dot distance. Left (right) graph: Spectrum of two carriers confined in two vertically coupled large (small) GaAs QDs. The circle symbols correspond to hole carriers, the square to electrons. For p-type material, the energy difference is switched at $d \approx d_0$, whereas for electrons the ground state is a singlet state with $S_e = 0$. We see clearly that ΔE decreases as d increases; this effect is more significant for small QDs where the Coulomb interaction between the two carriers is completely neglected.

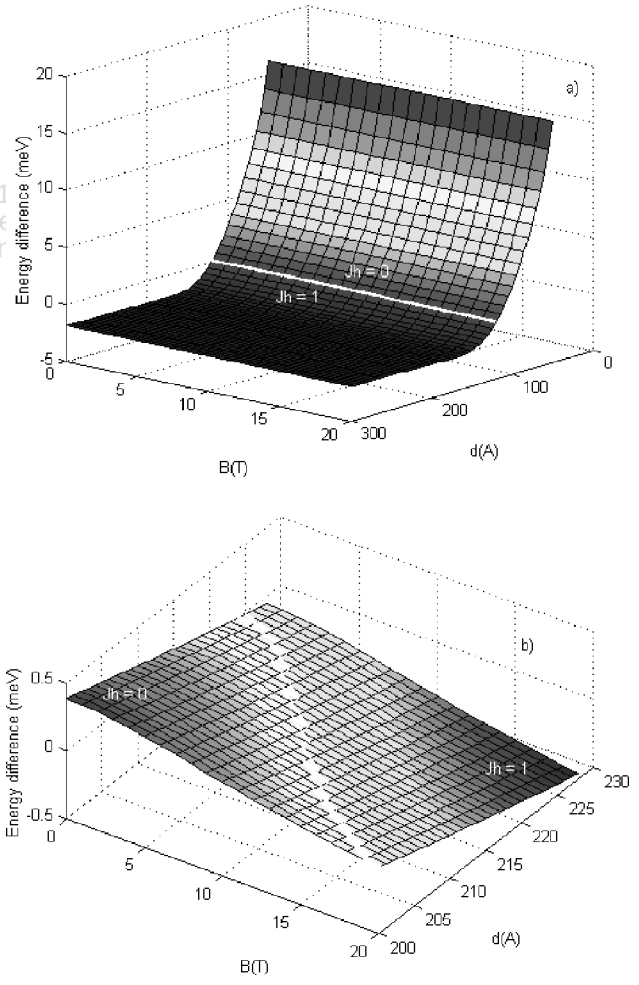

Fig. 3. The energy difference as a function of the magnetic field B and inter-dot separation d. The white lines in the graphs (a and b) separate the $J_h = 0\,(\Delta E > 0)$ and $J_h = 1\,(\Delta E < 0)$ ground state. The parameter for this plot corresponds to a system of two equal large GaAs dots (as in Fig. 1). Switching by magnetic field depends strongly on d; it changes as one moves from $d = 200$ Å to 225 Å (graph b).

diameter and 35 Å (81.93 Å) high. This choice respects the preceding approximations.

At zero magnetic field, we plot in Figure 1 the energy spectrum as a function of the inter-dot distance. Figure 1a and b displays the energy levels corresponding to the hole, whereas Figure 1c and d corresponds to the electron states. For both large (Fig. 1a and c) and small (Fig. 1b and d) QD, that is, in the weak and strong confinement regimes, respectively, the mixing between heavy-hole and light-hole states dominates the Coulomb interaction on neighboring QDs. One can see that for the large $d > 0$, the energies are shifted down and level crossing occurs. At some of the level crossings a gap opens. This is due to the lower symmetry of the problem for $d > 0$. For large d the mixing vanishes and the ground states stabilize to a *hh* state. On the other hand, the energy decreases as d increases and a spontaneous "phase transition" appears at $d \approx d_0$ in each regime. A direct comparison with electrons is depicted in Figure 1 (right graph), which shows that independently of the dot size the lowest singlet state is always located below the energy level of the triplet state. At a large inter-dot distance the singlet and triplet states are nearly degenerate, that is, their energy levels lie very close to, but do not cross, each other.

We see the energy difference as the cause of the energy separation between the two lowest energy states. The most remarkable feature of $\Delta E(d)$ is the change of the sign from positive to negative at $d \approx d_0$; we note that this phenomenon is depicted only for p-type material. However, for $d < d_0$, ΔE is positive, and the level with $J_h = 0$ is the ground state. For $d > d_0$, ΔE is negative, a phase transition appears, and the ground state is $J_h = 1$. The typical behavior of the energy difference as a function of the inter-dot distance is shown in Figure 2. However, there is a strong decrease in ΔE as a function of d. This can be understood from the fact that very distant QDs interact only weakly and the energy difference of the states is smaller. For the electronic spectrum the ground state is always found to be a singlet state, and evidently the energy difference remains greater than or equal to zero.[16]

We study now the effect of the magnetic field B and band mixing on the energies of holes. We concentrate on the $J_h = 0$ regime, and we show that for specific controllable hole parameters such as inter-dot

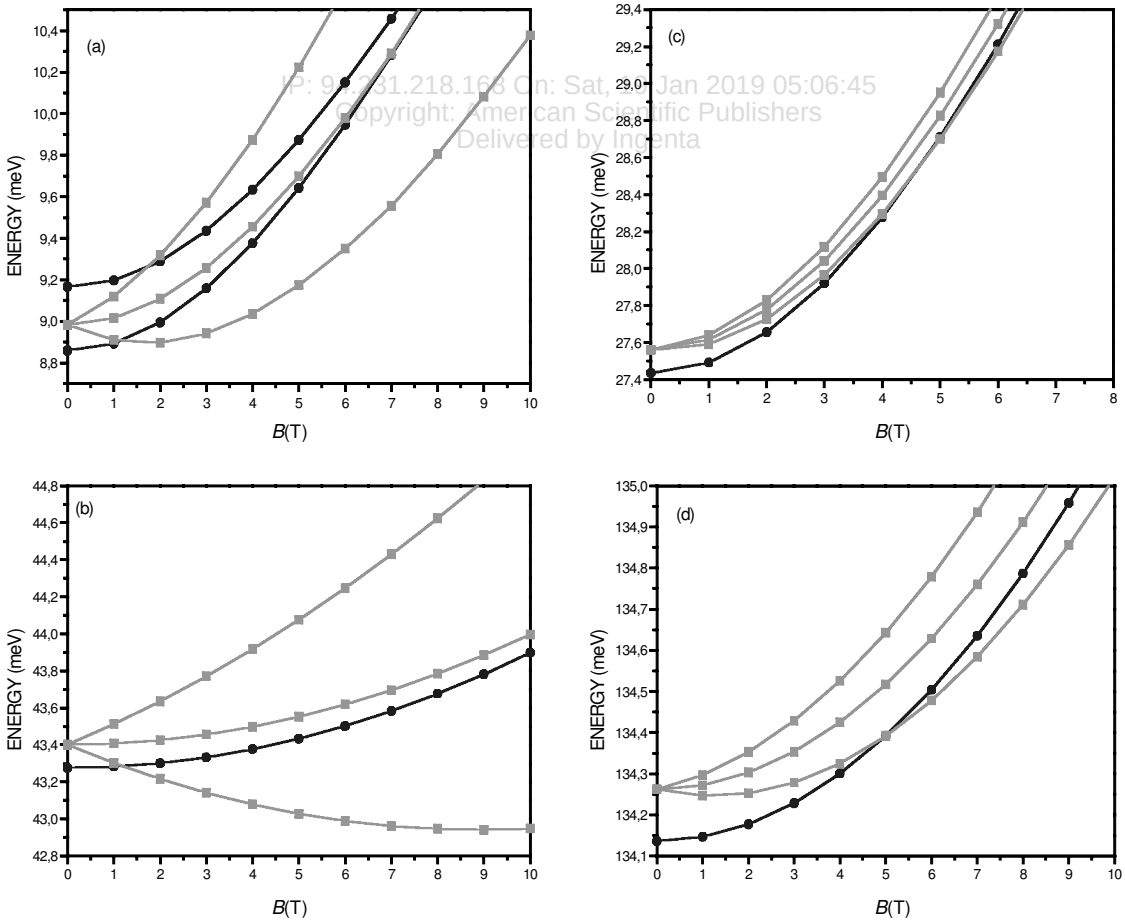

Fig. 4. Field dependence of the lowest carrier levels for two vertically coupled GaAs dots (same parameters as in Fig. 2), including the Zeeman coupling with $\kappa = -1.84$ (Ref. 26) and $g = -0.44$. The circle symbols correspond to $J_h = 0$ ($S_e = 0$) and the square symbols to $J_h = 1$ ($S_e = 1$). Under the influence of the magnetic field, the hole's ground state (a, b) changes from $J_h = 0$ to $J_h = 1$ at about 1 T, and it changes around 5 T for electrons (c, d).

coupling, confinement, and dot dimension, the magnetic field applied perpendicular to the double QDs can be a "switch" for the ΔE coupling. Because of this, switching the total angular momentum in an experimental setup is not easy, but switching J_h by B or d is the most natural choice. The energy difference ΔE between the lowest $J_h = 0$ and $J_h = 1$ states is plotted in Figure 3. For electrons we guarantee that the $S_e = 0$ state remains lower with arbitrary d and B.[24] However, Figure 4 illustrates a crossing between the lowest ground states, which is obtained with the contribution of the Zeeman splitting of the form $g\mu_B BS_e$ in the case of two electrons and $\kappa\mu_B BJ_h + (e/c\hbar)q(J_h)^3 B$ for holes, where q is the Luttinger parameter. We note that the q material parameter is very small for the semiconductor of interest here and it can be neglected. In the $J_h = 1$ regime, the switching between the two lowest states is detected at high magnetic fields (about 36 T), which is why we neglect it.

We now suggest one type of spin measurement of magnetization. We can determine the measurement of the carrier total angular momentum (J_h or S_e) with the magnetization's measurement. The magnetization of the system is given by $M = -\partial E_g/\partial B$, where E_g (approximated by $kT \ll \hbar w_z$) is the ground-state energy of the double QDs. Because of the crossing between the lowest states, the magnetization spectrum (Fig. 5) shows a jump that marks a phase transition. It is also found that the magnetization has a linear relation with the magnetic field in the low-field region. In the limit of a weak magnetic field, it is elementary to show that the magnetization of two electrons (or holes) is in proportion to the magnetic field,[25]

$$M \approx -\alpha B, \qquad \alpha = \left\langle \psi_g \left| \frac{e^2}{2m}\left(|r_1|^2 + |r_2|^2\right) \right| \psi_g \right\rangle \quad (13)$$

where ψ_g is the ground state of the two carriers. It is straightforward to see that α also measures the extension of the ground state. The magnetization reveals whether the ground state of the coupled-dot system corresponds to $J_h = 0$ ($S_e = 0$) or $J_h = 1$ ($S_e = 1$) states.

In summary, using the Hund-Mulliken approach, we have calculated the energy difference as a function of the inter-dot distance for carriers confined in a pair of vertically coupled QDs and have compared the two-hole spectra (ΔE becomes negative) with the two-electron spectra (ΔE remains positive for arbitrary inter-dot distance). For two-hole filling in the presence of a magnetic field, the ground-state switching from $J_h = 0$ to $J_h = 1$ states occurs at fields of a few Tesla, depending on the strength of the confinement, the distance coupling, and the dot size. However, $\Delta E(d, B)$ changes sign (reflecting a crossing between the two lowest states); in addition to being our fundamental interest, this dependence opens up the possibility of using holes in coupled QDs as quantum devices, which can be operated by magnetic field or inter-dot distance. The behavior of the energy difference ΔE between the two lowest energy hole states leads to a quite peculiar phase transition. In the magnetic field effect, we show that, for a specific choice of GaAs dots, the magnetic field can be a switching "on-off" of the energy ΔE. The switching is depicted only for p-type material. Moreover, with additive cooperation of the Zeeman splitting, the crossing between the lowest levels is obtained at a relatively small critical magnetic field (about 1 T for holes and 5 T for electrons). Detailed information on the nature of the ground state of double QDs may be obtained by measuring the magnetization.

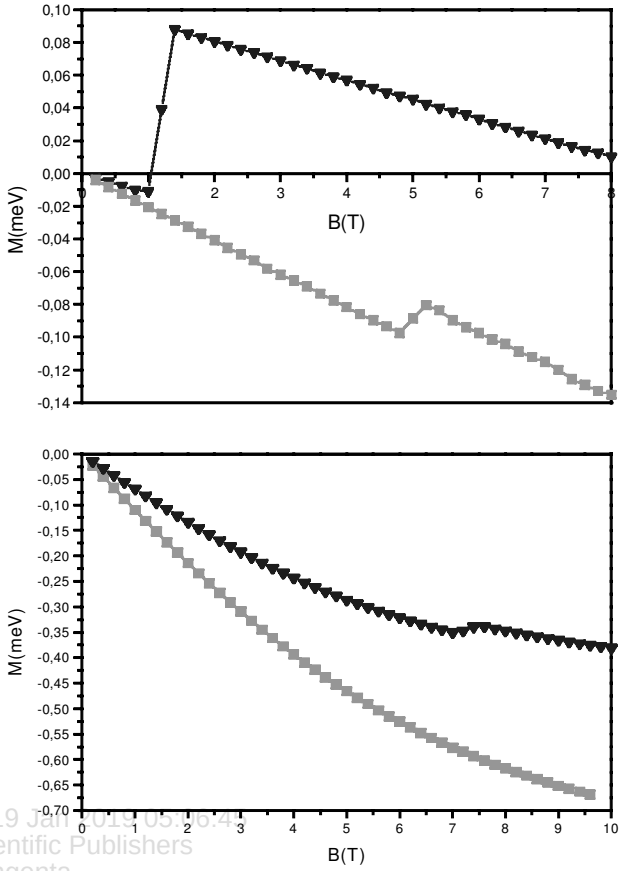

Fig. 5. Magnetization (M) as a function of the B field for vertically coupled large GaAs QDs containing two holes (triangle down symbols) and two electrons (square symbols) at $T \sim 0$ K. The two lowest ground states crossings are induced by the Zeeman splitting (first graph), causing a jump in the magnetization around 1 T for holes and 5 T for electrons, but no such signature occurs for electrons in the absence of the Zeeman effect (second graph).

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