Binding and transition energies of off-center D^- impurity states in quantum wells and magnetic fields

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The binding and transition energies of D^0 and D^- off center in GaAs-$Ga_{1-x}Al_xAs$ quantum wells (QW's) in a magnetic field are reported. It is found that a magnetic field can make the binding energy of D^- off center in QW's not only increase but also decrease and a very strong magnetic field can dissociate a D^- into an electron and a D^0. On the basis of analysis of the variation of D^0 and D^- transition energies with magnetic field, a method is proposed for experimentalists to confirm the D^- states in QW's.

Stimulated by interest in the physics and technological applications, researchers have fabricated and investigated many interesting low-dimensional semiconductor microstructures. Many investigations of neutral shallow donors D^0 in GaAs-$Ga_{1-x}Al_xAs$ quantum wells (QW's) with and without doping in strong magnetic fields have been undertaken. Recently, negative donors D^0, i.e., neutral donors that bind an additional electron, have already been observed and identified in multiple QW's.[1,2] All of the theoretical and experimental studies[1-14] have shown that the binding energy is much larger for the lowest D^- singlet state than for the triplet state and that a magnetic field can introduce more bound states. The binding energies of both D^0 and D^- centers increase with increasing the field and the ratio of D^- to D^0 binding energy in a strong magnetic field approaches a constant value which is strongly dependent on the dimensionality. No information seems to be available concerning the properties of D^0 and D^- off center in QW's in a strong magnetic field. It is of both theoretical and experimental interest to know the electronic structures and the off-center effect. In the present work, the field effect on the binding and transition energies of D^0 and D^- off center in QW's which is in sharp contrast to that mentioned above, is studied. On the basis of the results obtained theoretically here, a method is proposed for experimentalists to confirm the D^- states in QW's.

Within the framework of an effective-mass approximation, the Hamiltonian of a D^0 off center in GaAs-$Ga_{1-x}Al_xAs$ QW's in a magnetic field B perpendicular to the interfaces can be written as

$$H(\rho,\phi,z)=-\frac{1}{\rho}\frac{\partial}{\partial\rho}\left[\rho\frac{\partial}{\partial\rho}\right]-\frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}-\frac{\partial^2}{\partial z^2}-\frac{2}{[\rho^2+(z-z_0)^2]^{1/2}}+V(z)+\frac{\gamma^2}{4}\rho^2+\gamma L_z \quad (1)$$

with

$$V(z)=\begin{cases}V_0 & \text{if } |z|\geq L/2\\ 0 & \text{if } |z|<L/2\end{cases}, \quad (2)$$

where γ is the reduced magnetic field and the relationship between B and γ is shown below. γL_z is the Zeeman term and V_0 is the barrier height which can be obtained from a fixed ratio of the band-gap discontinuity. L and z_0 are, respectively, a well width and an off-center distance of donor ion in the QW's, i.e., a distance along the z axis from the center of the QW's. We have ignored the Zeeman spin energy, which does not affect the binding energies. In the theoretical calculation, the effective atomic units are used so that all energies are measured in units of the effective Rydberg Ry* ($m^*e^4/2\hbar^2\epsilon^2$) and all distances are measured in units of effective Bohr radius a^* ($\epsilon\hbar^2/e^2m^*$). The m^* and ϵ are the electronic effective mass and the dielectric constant of the QW's. It should be pointed out that the effective-mass difference between GaAs and $Ga_{1-x}Al_xAs$ materials and the polarization and image charge effects have been ignored. The reduced magnetic field γ introduced above is equal to $\hbar\omega_c/2$ Ry* where $\omega_c=eB/m^*c$ is the cyclotron frequency. For comparing theoretical results with experimental data, Ry*, and a^* can be taken to be, respectively, 5.8 meV and 100 Å for the GaAs-$Ga_{1-x}Al_xAs$ QW's and then, according to the relationship between B and γ, $\bar{\gamma}=1$ corresponds to $B=6.75$ T.

The corresponding Hamiltonian of a D^- off center in the QW's is as follows:

$$H=H(\rho_1,\phi_1,z_1)+H(\rho_2,\phi_2,z_2)+\frac{2}{[\rho_{12}^2+(z_1-z_2)^2]^{1/2}}, \quad (3)$$

where $\rho_{12}=|\rho_1-\rho_2|$.

Equation (1) can be rewritten as

$$H(\rho,\phi,z)=H_0(\lambda,\alpha,\rho,\phi,z)+H'(\lambda,\alpha) \quad (4)$$

with

$$H_0(\lambda,\alpha,\rho,\phi,z)=-\frac{1}{\rho}\frac{\partial}{\partial\rho}\left[\rho\frac{\partial}{\partial\rho}\right]-\frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}-\frac{2\lambda}{\rho+\alpha}-\frac{\partial^2}{\partial z^2}+V(z)+\frac{\gamma^2}{4}\rho^2+\gamma L_z \quad (5)$$

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and

$$H'(\lambda,\alpha)=[2\lambda/(\rho+\alpha)]-\{2/[\rho^2+(z-z_0)^2]^{1/2}\} . \quad (6)$$

Exact solutions of H_0 with a well-defined magnetic quantum number m can be obtained.[15] Taking the eigenfunctions $\psi(z)\psi_0(\alpha,\lambda,\rho)$ and $\psi(z)\psi_1(\alpha,\lambda,\rho)e^{-i\phi}$ of the ground s-like ($m=0$) and first excited p^--like ($m=-1$) states of $H_0(\lambda,\alpha,\rho,\phi,z)$ as trial functions with variational parameters α and λ, the energies $E(D^0,0)$ and $E(D^0,1)$ of the ground and first excited states of $H(\rho,\phi,z)$ can be obtained by the variational calculation. Then, the binding energies $E_B(D^0,0)$ and $E_B(D^0,1)$ and the $1s\rightarrow 2p^-$ transition energy $\Delta E(D^0)$ of off-center D^0 in QW's in a magnetic field γ are, respectively, given by

$$E_B(D^0,0)=E(e,0)-E(D^0,0) , \quad (7)$$

$$E_B(D^0,1)=E(e,0)-E(D^0,1) , \quad (8)$$

and

$$\Delta E(D^0)=E(D^0,1)-E(D^0,0) , \quad (9)$$

where $E(e,0)$ is the ground-state energy of an electron in the QW's in the magnetic field γ. It is interesting to point out that the $1s\rightarrow 2p^+$ transition energy is exactly equal to $\Delta E(D^0)$ plus 2γ if the band nonparabolicity is neglected.[17]

With the use of the forms of Eqs. (5) and (6), Eq. (3) can be rewritten as

$$H=\tilde{H}(\lambda_1,\lambda_2,\alpha_1,\alpha_2)+H_{in}(\lambda_1,\lambda_2,\alpha_1,\alpha_2) \quad (10)$$

with

$$\tilde{H}(\lambda_1,\lambda_2,\alpha_1,\alpha_2)=H_0(\lambda_1,\alpha_1,\rho_1,\phi_1,z_1)+H_0(\lambda_1,\alpha_2,\rho_2,\phi_2,z_2) \quad (11)$$

and

$$H_{in}=H'(\lambda_1,\alpha_1)+H'(\lambda_2,\alpha_2)+2/[\rho_{12}^2+(z_1-z_2)^2]^{1/2} . \quad (12)$$

Using the exact eigenfunctions of the ground ($m=0$) and first excited ($m=-1$) states of $H_0(\lambda_1,\alpha_1,\rho_1,\phi_1,z_1)$ and $H_0(\lambda_2,\alpha_2,\rho_2,\phi_2,z_2)$, the Chandrasekhar-type trial functions[16] with well-defined total magnetic quantum number $M=m_1+m_2$ can be obtained. The trial function Ψ_0 of the lowest singlet s-like ($M=0$) state is given by

$$\Psi_0=A_0(1+c\rho_{12})\psi(z_1)\psi(z_2)[\psi_0(\alpha_1,\lambda_1,\rho_1)\psi_0(\alpha_2,\lambda_2,\rho_2)+\psi_0(\alpha_1,\lambda_1,\rho_2)\psi_0(\alpha_2,\lambda_2,\rho_1)] \quad (13)$$

while the trial functions Ψ_1^+ and Ψ_1^- of singlet and triplet p^--like ($M=-1$) states are given by

$$\Psi_1^{\pm}=A_1(1+c\rho_{12})\psi(z_1)\psi(z_2)\times[\psi_0(\alpha_1,\lambda_1,\rho_1)\psi_1(\alpha_2,\lambda_2,\rho_2)e^{-i\phi_2}\pm\psi_0(\alpha_1,\lambda_1,\rho_2)\psi_1(\alpha_2,\lambda_2,\rho_1)e^{-i\phi_1}] . \quad (14)$$

Here $\psi(z)\psi_0(\alpha,\lambda,\rho)$ and $\psi(z)\psi_1(\alpha,\lambda,\rho)e^{-i\phi}$ are the eigenfunctions of the ground and first excited states of $H_0(\alpha,\lambda,\rho,\phi,z)$, respectively, and λ_1, λ_2, α_1, α_2, and c are variational parameters. A_0 and A_1 are the normalization constants. Then the variational energy $E(D^-,0)$ of the singlet s-like ($M=0$) state and the variational energies $E^+(D^-,1)$ and $E^-(D^-,1)$ of the singlet and triplet p^--like ($M=-1$) states can be obtained.

Once $E(D^-,0)$ and $E^-(D^-,1)$ are obtained, the binding energies $E_B(D^-,0)$ and $E_B(D^-,1)$ of singlet s-like and triplet p^--like states are given by

$$E_B(D^-,0)=E(D^0,0)+E(e,0)-E(D^-,0) \quad (15)$$

and

$$E_B(D^-,1)=E(D^0,0)+E(e,0)-E^-(D^-,1) , \quad (16)$$

respectively. In addition to spin conservation, the dipole-created D^- transitions in Faraday geometry obey the $\Delta M=\pm 1$ selection rule. The D^- transitions of $\Delta M=+1$ and -1 are analogous to the $1s\rightarrow 2p^+$ and $1s\rightarrow 2p^-$ D^0 transitions.[5,17] The p^-- and p^+-like states are exactly separated by the cyclotron energy 2γ for parabolic conduction bands. The $1s\rightarrow 2p^-$ D^- transition energy $\Delta E(D^-)$ is defined by

$$\Delta E(D^-)=E^+(D^-,1)-E(D^-,0) , \quad (17)$$

where $E^+(D^-,1)$ is the energy of the singlet p^--like state. Then, the $1s\rightarrow 2p^+$ D^- transition energy is equal to $\Delta E(D^-)$ plus 2γ.

The ground ($m=0$) and first excited ($m=-1$) states of D^0 off center in QW's of $V_0=80$ Ry* with $L=2a^*$ have been calculated for $z_0/a^*=0.0$, 0.25, 0.5, 0.75, and 0.95, respectively. In Fig. 1, the binding energies $E_B(D^0,0)$ and $E_B(D^0,1)$ have been plotted as a function of γ. It is readily seen that all of them increase with increasing γ and the values are larger for smaller z_0 than for larger z_0. However, the variation with z_0 is much larger for the ground states than for the first excited states. It is easy to understand if we note that the $\psi_1(\alpha,\lambda,\rho)$ of excited states is more extended than the $\psi_0(\alpha,\lambda,\rho)$ of ground states and thus, z_0 has a strong effect on the Coulomb increation energies of D^0 ground states.

Using the trial functions of Eqs. (13) and (14), the singlet and triplet states of D^- off center in the QW's of $L=2a^*$ have been calculated for the same z_0, i.e., 0.0, $0.25a^*$, $0.5a^*$, $0.75a^*$, and $0.95a^*$, respectively. In the absence of a magnetic field, i.e., $\gamma=0$, $E_B(D^-,0)$ is, re-

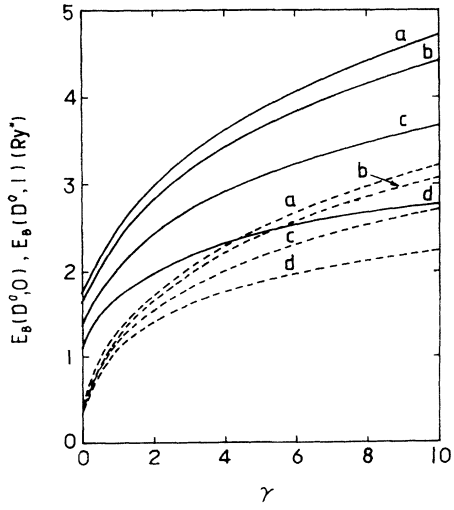

FIG. 1. Binding energies $E_B(D^0,0)$ (solid lines) and $E_B(D^0,1)$ (dashed lines) of D^0 off center in the QW's vs γ for $z_0/a^*=0.0$, 0.25, 0.5, and 0.75 indicated by a, b, c, and d on the lines, respectively.

spectively, equal to 0.219, 0.180, 0.107 , 0.055, and 0.033 Ry* for $z_0/a^*=0.0$, 0.25, 0.50, 0.75, and 0.95. It is interesting to point out that the $E_B(D^-,1)$ in the QW's with $\gamma=0$ is almost independent on z_0 and about 0.03 Ry*. This might be understood if we note the following facts. First, for the triplet p^--like state, both one-electron orbitals in Eq. (14) are orthogonal to each other and thus, the one-electron exchange integral of the ion potential $-2/[\rho^2+(z-z_0)^2]^{1/2}$ is equal to zero. Second, for $\gamma=0$, the outer one-electron ($m=-1$) and two-electron Coulomb integrals are slightly dependent on z_0 and their sum can be almost independent of z_0. Third, the two-electron exchange integral is very small and almost independent of z_0 at $\gamma=0$. The value of 0.03 Ry* is quite different from zero in pure two-dimensional (2D) and 3D cases, i.e., in QW's of $V_0=\infty$ with $L=0$ and ∞, respectively. It is an interesting quantum-size effect. Considering that a smaller energy scale is involved in $E_B(D^-,1)$ with $\gamma=0$ and that the differences between exact and variational values can be different for $E(D^0,0)$ and $E^-(D^-,1)$ in Eq. (24) even though the cancellation law of the differences in the equation does exist due to using the same kind of one-electron functions, it should be interesting to give the more exact values of $E(D^0,0)$ and $E^-(D^-,1)$ of $\gamma=0$ with a more elaborate method.

As shown in Fig. 2, the variations of the binding energies $E_B(D^-,0)$ and $E_B(D^-,1)$ with γ are quite different for the singlet s-like ($M=0$) and triplet p^--like ($M=-1$) states and for different z_0. With increasing γ, the $E_B(D^-,0)$ and $E_B(D^-,1)$ of $z_0=0.0$ and $0.25a^*$ increase and the $E_B(D^-,0)$ is always larger than the $E_B(D^-,1)$ while the binding energy $E_B(D^-,1)$ of $z_0=0.5a^*$ increases continually and the $E_B(D^-,0)$ increases rapidly until a maximum value and, then, decreases slowly. For $z_0=0.5a^*$, there is an intersection of both of the curves at a point of $\gamma\approx5.5$. Furthermore, there are maxima for both $E_B(D^-,0)$ and $E_B(D^-,1)$ of $z_0=0.75a^*$ and there is no particular bound D^- state for $z_0=0.75a^*$ with $\gamma>9.2$ and $z_0=0.95a^*$ with $\gamma>3.8$, respectively. It means that the triplet states of D^- off center in QW's can be bound more strongly than the singlet ones due to an applied strong magnetic field and that for a fixed z_0, an applied magnetic field can make the binding energies of D^- off center in QW's not only increase but also decrease and a very strong magnetic field can dissociate a D^- off center in a QW into an electron and a D^0 off center in the QW. It is in sharp contrast to that of D^- centers ($z_0=0$) in QW's in strong magnetic fields.[1-14]

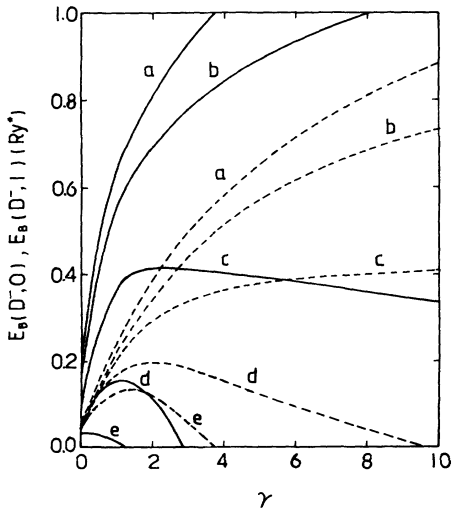

FIG. 2. Binding energies $E_B(D^-,0)$ (solid lines) and $E_B(D^-,1)$ (dashed lines) of D^- off center in the QW's vs γ for $z_0/d^*=0.0$, 0.25, 0.5, 0.75, and 0.95 indicated by *a*, *b*, *c*, *d*, and *e* on the lines, respectively.

What has been mentioned above can be understood on the basis of the following. In QW's, z_0 has much less effect on the one-electron Coulomb and exchange integrals of $-2/[\rho^2+(z-z_0)^2]^{1/2}$ for more extended orbitals than for more localized ones and z_0 has only a weak effect on the two-electron Coulomb and exchange integrals under strong fields. Therefore, both $E_B(D^-,0)$ and $E_B(D^-,1)$ are much more sensitive to z_0 in strong magnetic fields than in weak ones and the effect of z_0 on $E_B(D^-,0)$ is larger than that on $E_B(D^-,1)$.

For $z_0=0$, the experimental magneto-optical D^- transition energies (the position of the D^- peak, i.e., so-called peak B)[1,2,18-20] have been obtained in good agreement with variational quantum Monte Carlo results.[3,17] Therefore it is interesting to compare our theoretical results of $z_0=0$ with experimental results and to determine if the calculation method and the trial functions used here are suitable or excellent for D^- states in GaAs-$Ga_{1-x}Al_xAs$ QW's in a magnetic field. For a D^- center in the QW's with $L=2a^*$ (200 Å) in magnetic fields $\gamma=1$ and 2 (6.75 and 13.5 T), the $1s\rightarrow2p^-$ transition energy obtained by us is, respectively, equal to 0.756 and 0.950 Ry* (35.4 and 44.4 cm^{-1}), about the same as the experimental data (about 36 and 43 cm^{-1} of $L=194$ Å) (Ref. 17) and slightly less than the variational quantum Monte Carlo results.[17] Then, it would be expected that using the trial

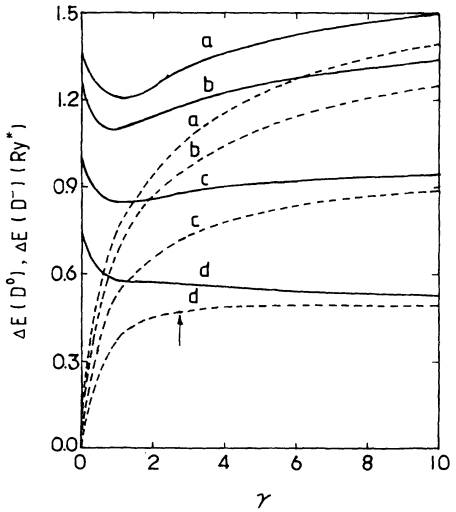

FIG. 3. $1s\rightarrow2p^-$ transition energies $\Delta E(D^0)$ (solid lines) and $\Delta E(D^-)$ (dashed lines) of D^0 and D^0 off center in the QW's vs γ for $z_0/a^*=0.0$, 0.25, 0.5, and 0.75 indicated by *a*, *b*, *c*, and *d* on the lines, respectively. The arrow shows a point where the singlet bound state of $z_0/a^*=0.75$ disappears.

functions and the calculation method, reasonable results can be obtained for the singlet and triplet states of an off-center D^- in the QW's in a magnetic field.

In Fig. 3, the $1s \rightarrow 2p^-$ transition energies $\Delta E(D^0)$ and $\Delta E(D^-)$ have been plotted as a function of γ for $z_0/a^* = 0.0$, 0.25, 0.5, and 0.75, respectively. It is obvious that the variations with γ are quite different for $\Delta E(D^0)$ and $\Delta E(D^-)$ before the $\Delta E(D^-)$ are close to the $\Delta E(D^0)$ with the same z_0. For example, the $\Delta E(D^0)$ of a D^0 center ($z_0 = 0$) in the QW's is, respectively, equal to 1.216 and 1.319 Ry* for $\gamma = 1$ and 3 and the difference is about 0.1 Ry* which is larger than that of D^0 off-center ($z_0 \neq 0$) in the QW's or D^0 in bulk GaAs. The $\Delta E(D^-)$ of D^- center ($z_0 = 0$) is, respectively, equal to 0.756 and 1.076 Ry* for $\gamma = 1$ and 3 and the difference 0.32 Ry*. Furthermore, the difference between $\Delta E(D^0)$ and $\Delta E(D^-)$ is 0.460 and 0.243 Ry* for $\gamma = 1$ and 3, respectively. It means that the variation of $\Delta E(D^0)$ with γ is much smaller than that of $\Delta E(D^-)$ and the increasing γ makes the D^- transition peak close to the corresponding D^0 peak. It is also found that for a fixed magnetic field, the difference between $\Delta E(D^-)$ and $\Delta E(D^0)$ decreases with increasing z_0 as shown in Fig. 3 and with increasing well width L, not shown in the figure. The positions of the $1s \rightarrow 2p^-$ transition peaks introduced by D^0 off center in the QW's, D^0 in the barriers ($z_0 > L/2$), and residual D^0 in the bulk buffer layer of the samples are almost independent of γ so that the D^- state with smaller z_0 in QW's can be experimentally distinguished from the D^0 ones and identified by the different variations of the $\Delta E(D^0)$ and $\Delta E(D^-)$ with γ.

The variations of both $1s \rightarrow 2p^-$ transition energies $\Delta E(D^0)$ and $\Delta E(D^-)$ with γ and the difference between $\Delta E(D^0)$ and $\Delta E(D^-)$ are smaller for a larger z_0 than for a smaller one. Therefore, it is not easy to distinguish the D^- transition from the D^0 transitions for a larger z_0 on the basis of the variations of $1s \rightarrow 2p^-$ transition energies with γ. Fortunately, it can be unambiguously identified by the disappearance of the transition peak (the singlet bound state) due to an applied strong magnetic field. This is in sharp contrast to previous results about D^- centers in QW's reported in the other papers to date.[1-14]

In conclusion, we have found that a strong magnetic field makes $E_B(D^0,0)$ and $E_B(D^0,1)$ of an off-center D^0 in QW's increase while it can make $E_B(D^-,0)$ and $E_B(D^-,1)$ of an off-center D^- in QW's not only increase but also decrease and that the variations of $E_B(D^-,0)$ and $\Delta E(D^0)$ with γ are different from those of $E_B(D^-,1)$ and $\Delta E(D^-)$, respectively. We have predicted that the singlet s-like and triplet p^--like bound states can disappear under very strong magnetic fields. On the basis of the analysis of the variation of $\Delta E(D^0)$ and $\Delta E(D^-)$ with γ, a method has been proposed for experimentalists to confirm the D^- states in QW's.

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