PHYSICAL REVIEW B VOLUME 50, NUMBER 7 15 AUGUST 1994-I

Spectrum and binding of an off-center donor in a spherical quantum dot

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(Received 26 January 1994)

The energy levels and binding energies of an off-center donor in a GaAs-$Ga_{1-x}Al_xAs$ spherical quantum dot are calculated by a linear variational method. The results clearly show the quantum size effect. The location effects of a donor ion on splitting, level ordering, and binding of the donor states are studied. It is found that the alteration of the position of a single Coulomb center can largely change the single-electron spectrum in a quantum dot with a larger radius.

I. INTRODUCTION

Stimulated by interest in physics and technological applications of low-dimensional quantum structures, calculations have been performed for electron states and related problems in two-dimensional quantum wells (2D QW's), one-dimensional quantum-well wires (QWW's), and quantum dots (QD's).[1-15] Understanding the impurity states in the confined systems is an important problem in semiconductor physics. The impurity states in 2D QW's and superlattices have been calculated by a number of authors,[2,5,6] and several calculations[3,7,8] have been performed for electron and impurity levels of QWW's. Recently, the electronic structure of the quantum dot, especially the donor states, acceptor states, and excitons, has received much attention.[9-15]

The exact solutions of hydrogenic donors located at the centers of spherical quantum dots (SQD's) have been obtained earlier by us.[9,10] However, the impurities could be located anywhere in the SQD's, and the ground-state level and the level ordering will change as the location shifts to the edge or out of the SQD's as mentioned in Ref. 9. Very recently, several authors have calculated the ground energies of the donors at a distance D from the centers in SQD's with an infinite barrier. At the same time, a general variational method with the use of basis functions not satisfying the boundary condition has been developed.[14] An improved approach has also been discussed in a recent paper.[15]

To our knowledge, there have been no calculated results about the excited states of off-center donors in QD's. It should be interesting, therefore, to extend our early work[9] to a systematic investigation of the positional dependence of ground and excited donor states in SQD's with a finite barrier height and, then, to show the coupling and competition effect of Coulomb potential with confinement one and the location effect on level ordering and binding energies. In addition, the ground states obtained can be compared with others[14,15] and the barrier-height effect on the quantum levels can be studied.

In Sec. II of this paper, the Hamiltonian and the calculation method are presented. The energy-level structure and binding energies are shown and discussed in Sec. III and Sec. IV, followed by a summary in Sec. V.

II. HAMILTONIAN AND CALCULATION METHOD

Within the framework of an effective-mass approximation, the Hamiltonian of a hydrogenic donor in a GaAs-$Ga_{1-x}Al_xAs$ spherical quantum dot can be written as

$$H=-\nabla^2-\frac{2}{|\mathbf{r}-\mathbf{D}|}+V(r) , \tag{1}$$

where the donor ion is located a distance D along the z axis from the center of the sphere. The potential $V(r)$ is taken to be spherically symmetric in the present work and has the form as follows:

$$V(r)=\begin{cases} V_0 & \text{if } r\geq R_0 , \\ 0 & \text{if } r<R_0 , \end{cases} \tag{2}$$

where V_0 is the barrier height and can be obtained from a fixed ratio $Q=0.7$ of the band-gap discontinuity $\Delta E_g=1.247x$ eV between GaAs and $Ga_{1-x}Al_xAs$ (Ref. 16) and, then, $V_0=0.8729x$ eV. In this paper, the effective atomic units are used so that all energies are measured in units of the effective Rydberg $\mathcal{R}$ and all distances are measured in units of effective Bohr radius a^*. The $\mathcal{R}$ and a^* can be determined by $m^*e^4/2\hbar^2\varepsilon^2$ and $\varepsilon\hbar^2/m^*e^2$, where m^* and ε are, respectively, the electronic effective mass and the dielectric constant of GaAs material and equal to $0.067m_e$ and 13.18.[16] Then, the $\mathcal{R}$ and a^* are equal to 5.2 meV and 104 Å, respectively. We should point 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 Schrödinger-like equation is expressed as

$$H\Psi(r,\theta,\varphi)=E\Psi(r,\theta,\varphi) . \tag{3}$$

For the symmetry, the eigenstates of H can be labeled by magnetic (m) quantum numbers. Because the radius and angle variables do not separate, Eq. (3) cannot be solved exactly. Therefore, approximation methods should be used.

The Hamiltonian can be rewritten as

$$H=H_0+H' \ , \tag{4}$$

with

$$H_0=-\nabla^2-\frac{2w}{r}+V(r) \tag{5}$$

and

$$H'=\frac{2w}{r}-\frac{2}{|\mathbf{r}-\mathbf{D}|} \ , \tag{6}$$

where w can be taken as a variational parameter for a better calculation. In the present work, w is equal to 1 and 0 for the calculation of quantum levels of a donor inside and outside the spherical quantum dot, respectively. Let us consider a linear variational function of the form

$$\Psi_m=\sum_{i=1}^{f} c_i\psi_i \ , \tag{7}$$

where ψ_i is the ith exact normorthogonal eigenstate of H_0 with eigenenergy E_i, which can be obtained exactly.[9] The principal, orbital, and magnetic quantum numbers of ψ_i are n_i, l_i, and m_i, respectively. In the problem considered, the summation in Eq. (7) includes only the terms with a fixed magnetic quantum number m, i.e., $m_1=m_2=\cdots=m_f=m$. In addition, it is interesting to point out that ψ_i and $\nabla\psi_i/m^*$ (Ref. 9) and, then, Ψ_m and $\nabla\Psi_m/m^*$ are, respectively, continuous at $r=R_0$.

According to the variational principle, it is straightforward to obtain the equation

$$\sum_{j=1}^{f}[H'_{ij}-(E-E_j)\delta_{ij}]c_j=0 \ , \quad i=1,2,\ldots,f \ , \tag{8}$$

with

$$H'_{ij}=\langle\psi_i|H'|\psi_j\rangle \ . \tag{9}$$

The condition that this set of equations has nonzero solution leads to the secular equation of finite degree f,

$$\|H'-B\|=0 \ , \tag{10}$$

where matrix elements of H' are H'_{ij} of Eq. (9) and B is a diagonal matrix, i.e.,

$$B_{ij}=(E-E_j)\delta_{ij} \ , \quad i,j=1,2,\ldots,f \ . \tag{11}$$

In order to calculate H'_{ij} of Eq. (9), ψ_i is written in the form

$$\psi_i(r,\theta,\varphi)=R_{n_il_i}(r)Y_{l_im_i}(\theta,\varphi) \ , \tag{12}$$

where $Y_{l_im_i}(\theta,\varphi)$ and $R_{n_il_i}(r)$ are the spherical harmonic and radial wave function, respectively. Using the relation

$$\frac{1}{|\mathbf{r}-\mathbf{r}'|}=\sum_{l=0}^{\infty}\frac{r_<^l}{r_>^{l+1}}P_l(\cos\Theta) \ , \tag{13}$$

where $r_<=\min(r,r')$, $r_>=\max(r,r')$, and Θ is the angle between $\mathbf{r}$ and $\mathbf{r}'$, a form of H'_{ij}, which is useful for the numerical calculation, is given by

$$H'_{ij}=2w\int_0^{\infty}R_{n_il_i}R_{n_jl_j}r\,dr\,\delta_{l_il_j}-2\sum_{l=|l_i-l_j|}^{l_i+l_j}(-1)^m[(2l_i+1)(2l_j+1)]^{1/2}\begin{bmatrix} l_i & l_j & l \\ 0 & 0 & 0\end{bmatrix}\begin{bmatrix} l_i & l_j & l \\ -m & m & l\end{bmatrix}$$
$$\times\left[\frac{1}{D^{l+1}}\int_0^{D}R_{n_il_i}R_{n_jl_j}r^{l+2}dr+D^l\int_D^{\infty}R_{n_il_i}R_{n_jl_j}\frac{1}{r^{l-1}}dr\right] \ . \tag{14}$$

(See the Appendix.) Then the energy levels are obtained by solving Eq. (10) numerically. For central donors in SQD's, i.e., $D=0$, the Hamiltonian H of Eq. (1) becomes H_0 of Eq. (5) with $w=1$. As mentioned above, the exact solutions can be obtained. The quantum levels are dependent on the principal and orbital quantum numbers n and l, and degenerate with respect to the magnetic quantum number m, so that the levels are denoted by $E_{nl}(w=1)$. We have $1s,2p,3d,2s,4f$ levels states, and so on, if the usual notation of l, i.e., $s,p,d,f,\ldots$ and the principal quantum number n are used for the level notation. For $D\neq0$, the degeneracy of $E_{nl}(w=1)$ is partially removed. The quantum levels obtained by solving Eq. (10) can be denoted by E_{nlm} even though l is not a good quantum number. E_{nlm} is degenerate with respect to m and $-m$, and E_{nl0} is nondegenerate.

Compared with the binding energy of a donor center in a three-, two-, or one-dimensional system, the binding energy of the corresponding donor states in the SQD can be defined by

$$E_B(n,l,m)=E_{nl}(w=0)-E_{nlm} \ , \tag{15}$$

where $E_{nl}(w=0)$ is the energy level as there is no Coulomb potential in the Hamiltonian of Eq. (1), i.e., $w=0$ in H_0 of Eq. (5). The equation of eigenenergies $E_{nl}(w=0)$ is as follows:

$$k_0+K_0\tan(k_0R_0)=0 \quad \text{if } l=0 \ , \tag{16a}$$

$$ik_lh_l(iK_lR_0)j_{l-1}(k_lR_0)+K_lh_{l-1}(iK_lR_0)j_l(k_lR_0)=0 \quad \text{if } l\geq1 \ , \tag{16b}$$

and

$$k_l=E^{1/2} \ , \quad K_l=(V_0-E)^{1/2} \ , \tag{17}$$

where j_l and h_l are the lth-order spherical Bessel functions and Hankel functions of the first kind, respectively.

III. SPLITTING AND ORDERING

We have performed a numerical calculation for GaAs-$Ga_{1-x}Al_xAs$ SQD's of R_0 being equal to $1a^*$ and $3a^*$ with $V_0=80\mathcal{R}$ ($x \approx 0.45$). In Figs. 1(a) and 1(b), we have plotted the ground and excited energy levels of a donor as a function of D for $R_0=1a^*$ with $V_0=80\mathcal{R}$, and only the states with $m=0,\pm1,\pm2$ are shown. The corresponding results of $R_0=3a^*$ with $V_0=80\mathcal{R}$ are in Figs. 2(a) and 2(b).

The splitting of energy levels can be easily seen from the figures as the impurity ion is removed from the center. For example, p states are split into two levels with $m=0$ and ±1 and d states are split into three levels with $m=0,\pm1,\pm2$. It is interesting to note that E_{nl0} is the lowest level among E_{nlm}. However, there is no splitting for s states. All of the energy levels approach the exact ones $E_{nl}(w=1)$ as D approaches zero. On the other hand, the $l+1$ splitting levels come near and approach $E_{nl}(w=0)$ as D approaches infinity.

For the case of $R_0=1a^*$, the level ordering from 1s up to 4d states is all the same for $D=0$ and ∞, i.e., 1s,2p,3d,2s,4f,3p,5g,4d, as shown in Figs. 1(a) and 1(b). For, however, the case of $R_0=3a^*$, the ordering of $D=0$

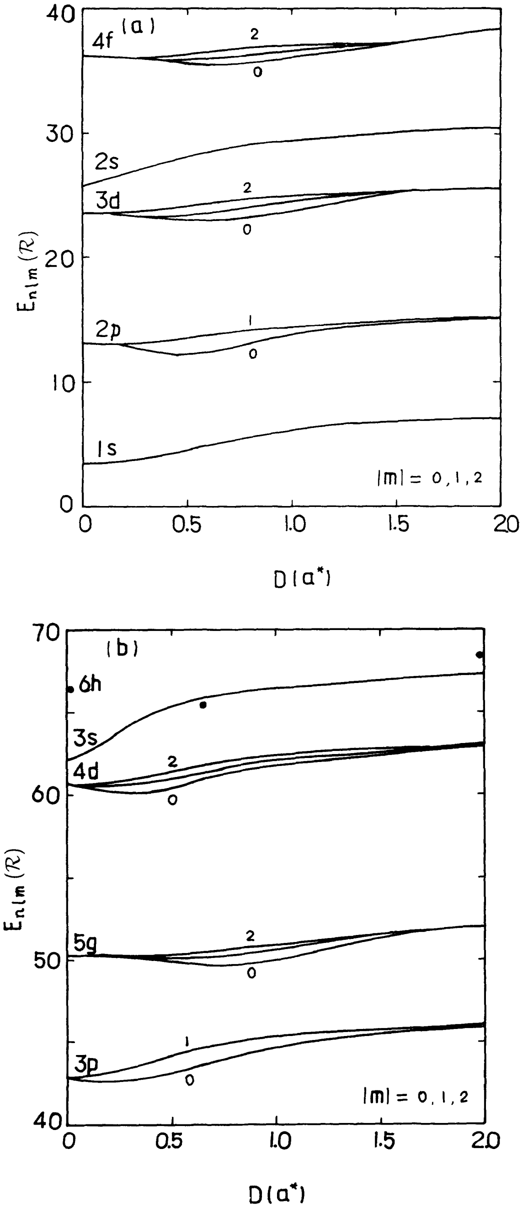

FIG. 1. Energy levels E_{nlm} of 1s, 2p states and so on (a) and 3p, 5g states and so on (b) with $|m|=0,1,2$ as a function of D for an off-center donor in SQD's of $R_0=1a^*$ with $V_0=80\mathcal{R}$. Solid circles show the levels of 6h states with $m=0$ (see the text).

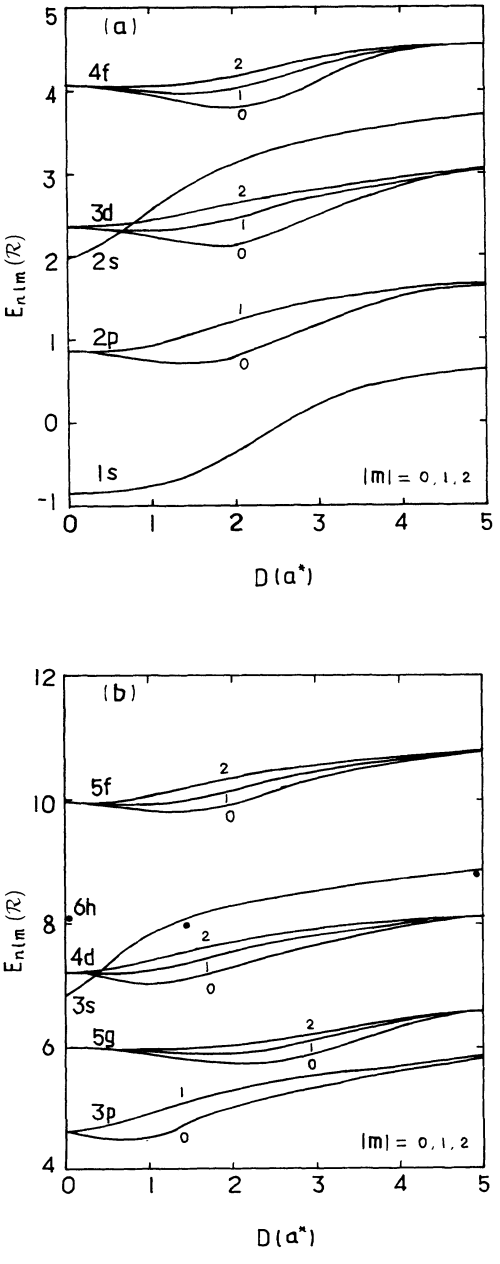

FIG. 2. Same as Fig. 1 but for that of $R_0=3a^*$ with $V_0=80\mathcal{R}$.

is $1s,2p,2s,3d,4f,3p,5g,3s,4d,\ldots$ and that of $D=\infty$ is $1s,2p,3d,2s,4f,3p,5g,4d,3s$ and so on. The ordering difference can be explained as follows. For $R_0=1a^*$, the confining potential V_0 is much stronger than the Coulomb potential, and the structure of levels is less affected by the position of the donor ion, whereas for $R_0=3a^*$, the coupling between confining and Coulomb potentials is stronger and leads to the change of level ordering as the donor is moved. An obvious feature of Figs. 2(a) and 2(b) is the intersection of s states and d states, which alters the sequence. The crossover of $2s$ and $3d$ occurs at about $0.7a^*$, and that of $3s,4d$ at about $0.4a^*$. This is a result of the coupling and competition of Coulomb potential with the confinement one. Moreover, the effect of the two potentials on energy levels is related to the corresponding wave functions. Only s state wave functions have nonzero values at the origin, so the Coulomb potential around the center has greater influence on s states than others. It is the reason why the energies of s states change more remarkably along with changing D than the others. The quantum size effect, which exists commonly in microstructures, mainly stems from the wave functions depending on some factors sensitively when the size of the system reduces to the particular range. For the above problem, the radius R_0 of QD's is the sensitive factor. In addition to the effect of R_0, the finite barrier height has an effect on the values of levels and the ordering of higher levels too. The effect can be illustrated by the result of $w=0$. For example, the value of $4d$, $6h$, and $3s$ levels of $R_0=1a^*$ with $V_0=\infty$ are equal to $82.719\mathcal{R}$, $87.531\mathcal{R}$, and $88.826\mathcal{R}$, and those with $V_0=80\mathcal{R}$, $64.207\mathcal{R}$, $68.929\mathcal{R}$, and $68.070\mathcal{R}$, respectively. The ordering of $6h$ and $3s$ levels is reversed as V_0 becomes finite. The same phenomenon exists also for $R_0=3a^*$, if V_0 reduces to a lower value, for example, $10\mathcal{R}$. The change of energy ordering might lead to some influence on the property of quantum dots as R_0 and V_0 are in an appropriate range.

Up to now, only the ground state of an off-center donor ion in an infinite well has been studied. It is helpful to compare the ground levels obtained in the present work with those in Ref. 15. The ground energies of an off-center donor for $D=0.5$, and $1.0a^*$ in a QD of $R_0=3a^*$ with $V_0=80\mathcal{R}$ is, respectively, equal to $-0.805\mathcal{R}$ and $-0.743\mathcal{R}$. It is agreeable with those ($-0.827\,84\mathcal{R}$ and $-0.756\,80\mathcal{R}$) in Ref. 15 since the finite-barrier effect is small in this case.

IV. BINDING

Studying the binding energy defined as Eq. (15), we find that the binding energy increases with increasing principal quantum number n for fixed l and $D=0$, i.e., $E_B(1,0,0)<E_B(2,0,0)<E_B(3,0,0)$, $E_B(2,1,0)<E_B(3,1,0)<E_B(4,1,0)$ and so on, as shown in Figs. 3(a) and 3(b), because of the confining potential V_0. It is quite different from the situation of an isolated hydrogen atom and its binding energy decreases with increasing n. As R_0 is in a proper range, the motion of electron is mainly restricted in the well. Studying the radial wave function $R_{nl}(r)$, it can be found that the position of the maximum of $|R_{nl}(r)|^2$ approaches the origin gradually with increasing n for fixed l. Therefore, the donor ions at the origin influence the states with larger n more strongly. It explains the changing tendency of binding energy. The phenomenon mentioned above is a conspicuous character of electronic structures in quantum dots. However, the binding energies of all states close to each other as D is larger. For instance, they are between $0.9\mathcal{R}$ and $1.05\mathcal{R}$ for $R_0=1a^*$ with $D=2a^*$ and between $0.27\mathcal{R}$ and $0.29\mathcal{R}$ for $R_0=3a^*$ with $D=7a^*$. It is evident for large D that the binding energy is approximately equal to $2/D$ (in units of $\mathcal{R}$), which is the Coulomb energy of two point

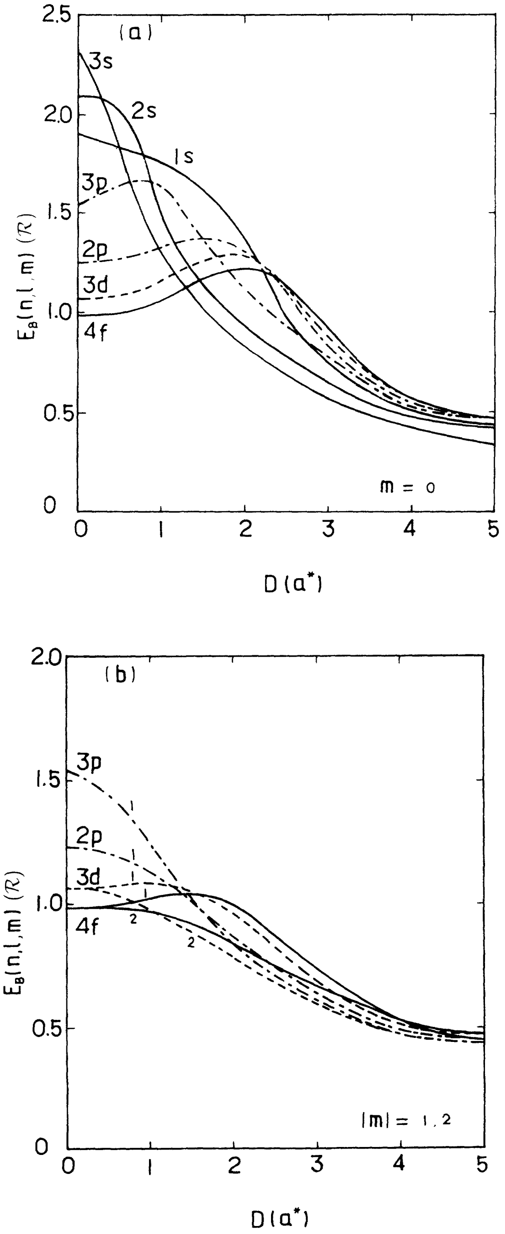

FIG. 3. Binding energies $E_B(n,l,m)$ of $1s$, $2s$, $3s$, $2p$, $3p$, $3d$, and $4f$ states with $m=0$ (a) and $2p$, $3p$, $3d$, and $4f$ states with $|m|=1$ and 2 (b) as a function of D for an off-center donor in SQD's of $R_0=3a^*$ with $V_0=80\mathcal{R}$.

TABLE I. Maxima of $E_B(n,l,0)$ and minima of E_{nl0} with the corresponding positions D_m.

nl		1s	2p	3d	2s	4f	3p	5g	4d	3s
$R_0=1a^*$	D_m	0	0.51	0.64	0	0.68	0.30	0.71	0.40	0
	E_{nl0}	3.337	12.383	22.981	25.828	35.439	42.474	49.758	60.207	62.299
	$E_B(n,l,0)$	4.620	3.842	3.612	5.597	3.480	4.542	3.320	4.000	5.771
$R_0=3a^*$	D_m	0	1.60	1.88	0	2.01	0.73	2.04	1.08	0
	E_{nl0}	−0.872	0.717	2.132	1.989	3.821	4.499	5.741	7.015	6.849
	$E_B(n,l,0)$	1.891	1.368	1.297	2.085	1.219	1.658	1.168	1.517	2.311

charges separated by a distance of D. The value of all of the binding energies estimated by $2/D$ is, respectively, equal to $1\mathcal{R}$ and $0.28\mathcal{R}$ for $D=2a^*$ and $7a^*$, and close to that mentioned above.

As shown in the figures, the binding energies reduce with increasing $|m|$ for fixed l and n and it is consistent with the ordering mentioned in Sec. III. Furthermore, the binding energies of states with $|m|=l$ reduce monotonously with increasing D, while all of the other states have maximum binding energies at some D not equal to zero. The maximum value is larger for smaller $|m|$. The above character can be explained as follows. According to the angular distribution of wave function, the states with $m=0$ mainly distribute along the z axis and the other states are more and more distant from the z axis with increasing $|m|$. So, the donor ion influences the state with smaller $|m|$ more effectively and, then, a larger binding energy is gained. The maximum binding energies of several states with $m=0$ and the corresponding levels are shown in Table I for both cases of $R_0=1a^*$ and $3a^*$. It is readily seen that for a fixed $(n-l-1)$ with $m=0$, i.e., fixed radial-node numbers of wave functions, the position of maximum binding energy increases and the maximum value decreases with increasing l. It is easy to understand if we note that for the fixed $(n-l-1)$ the position of the maximum of $|R_{nl}(r)|^2$ increases with l. It is readily seen that the maximum binding energies and their positions of higher levels with the same $(n-l-1)$ are close to each other.

It can be easily seen that four values, i.e., both energies of $D=0$ and $D=\infty$, the minimum (maximum binding) energy and its position, are important for determining the D dependence of a level and the level order. The energies of $D=0$ and ∞ can be calculated exactly. The maximum binding energy and its position of a level can be obtained by solving Eq. (10) with different D. Those of a higher level with larger l, however, can be estimated on the basis of the features of binding energies mentioned above. Then, the range of a higher level can be determined. For example, the range of $6h$ levels can be obtained by using the energies of $D=0$ and ∞, the estimated maximum binding energies and positions, and the approximate binding energies $2/D$ of a larger D, as shown in Figs. 1(b) and 2(b). It is interesting to note that the $3s$ levels can intersect with the $6h$ level in Fig. 1(b) and with $4d$ and $6h$ levels in Fig. 2(b), respectively. The feature means that a single Coulomb center has more important effects on the level ordering for a larger QD than for a smaller one.

V. SUMMARY

Using a linear variational method, we have reported the calculated results of ground states and excited states of donor confined by GaAs-Ga$_{1-x}$Al$_x$As SQD and located inside and outside of the SQD. The results have clearly demonstrated the so-called quantum size effect. The splitting and ordering of quantum levels is dependent on the radius R_0, the distance D, and the barrier height V_0.

For states with fixed l and n, the binding energies decrease with increasing $|m|$. The binding energies of states with $|m|=l$ decrease monotonously with increasing D, while all of the other states have maximum binding energies at some D not equal to zero. The maximum values are larger for smaller $|m|$. It is also seen that the maximum binding energies and their positions of higher levels with the same $(n-l-1)$ are close to each other. The character of binding energies can be used to estimate the minimum value of a higher level and its position.

The most interesting and inspiring feature of the D dependence of the energy-level structure is the intersection of some levels. It means that a single Coulomb center with a proper D can largely change the single-electron spectrum of a quantum dot with proper R_0 and V_0 and, then, the property. The concept might be useful for designing some devices in the future. In addition to the effect of a single Coulomb center, the effect of the potential shape of a spherically symmetric quantum dot and that of nonspherically symmetry of QD's on the spectrum can be important in some other kinds of quantum-dot systems. It is an interesting subject to study.

APPENDIX

Using Eqs. (12) and (13), Eq. (9) becomes

$$H'_{ij}=2w\int_0^\infty R_{n_il_i}R_{n_jl_j}r\,dr\,\delta_{l_il_j}-2\sum_{l=0}^{\infty}\left[\int Y^*_{l_im}(\theta,\varphi)Y_{l_jm}(\theta,\varphi)P_l(\cos\theta)d\Omega\right]$$

$$\times\left[\frac{1}{D^{l+1}}\int_0^D R_{n_il_i}R_{n_jl_j}r^{l+2}dr+D^l\int_D^\infty R_{n_il_i}R_{n_jl_j}\frac{1}{r^{l-1}}dr\right], \qquad \text{(A1)}$$

where $P_l(\cos\theta)$ is l-Legendre function. According to

$$\int_0^{2\pi}\int_0^{\pi} Y_{l_1m_1}Y_{l_2m_2}Y_{l_3m_3}\sin\theta\, d\theta\, d\varphi=\left[\frac{(2l_1+1)(2l_2+1)(2l_3+1)}{4\pi}\right]^{1/2}\begin{bmatrix} l_1 & l_2 & l_3\\ 0 & 0 & 0\end{bmatrix}\begin{bmatrix} l_1 & l_2 & l_3\\ m_1 & m_2 & m_3\end{bmatrix}, \tag{A2}$$

where

$$\begin{bmatrix} l_1 & l_2 & l_3\\ m_1 & m_2 & m_3\end{bmatrix}$$

is the Wigner 3-j symbol,

$$\int Y^*_{l_im}Y_{l_jm}P_l(\cos\theta)d\Omega=(-1)^m[(2l_i+1)(2l_j+1)]^{1/2}\begin{bmatrix} l_i & l_j & l\\ 0 & 0 & 0\end{bmatrix}\begin{bmatrix} l_i & l_j & l\\ -m & m & 0\end{bmatrix}, \tag{A3}$$

with

$$\begin{bmatrix} l_1 & l_2 & l_3\\ m_1 & m_2 & m_3\end{bmatrix}=(-1)^{l_1-l_2-m_3}\left[\frac{(l_1+l_2-l_3)!(l_2+l_3-l_1)!(l_3+l_1-l_2)!}{(l_1+l_2+l_3+1)!}\right]^{1/2}$$
$$\times\sum_{\kappa}(-1)^{\kappa}\left\{\frac{[(l_1+m_1)!(l_1-m_1)!(l_2+m_2)!(l_2-m_2)!(l_3+m_3)!(l_3-m_3)!]^{1/2}}{\kappa!(l_1+l_2-l_3-\kappa)!(l_1-m_1-\kappa)!(l_2+m_2-\kappa)!(l_3-l_2+m_1+\kappa)!(l_3-l_1-m_2+\kappa)!}\right\}, \tag{A4}$$

where only the finite number of κ is included in the summation since κ runs over all integers which do not lead to negative factorials. The l_1, l_2, l_3 and m_1, m_2, m_3 lead to nonzero 3-j symbol if they satisfy

$$l_1+l_2\geq l_3\ ,\quad l_2+l_3\geq l_1\ ,\quad l_3+l_1\geq l_2\ ,$$

and

$$l_1\geq|m_1|\ ,\quad l_2\geq|m_2|\ ,\quad l_3\geq|m_3|\ , \tag{A5}$$
$$m_1+m_2+m_3=0\ .$$

Therefore,

$$|l_i-l_j|\leq l\leq l_i+l_j \tag{A6}$$

is satisfied and the summation in Eq. (A1) has only finite terms.

Finally, Eq. (14) can be obtained by using Eqs. (A1), (A3), and (A6).

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