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LETTER TO THE EDITOR

Donors confined by spherical quantum dots and located anywhere

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Abstract. Using proper basis functions, a variational calculation is performed for ground and excited states of a shallow donor confined by a GaAs–$Ga_{1-x}Al_xAs$ spherical quantum dot with radius R_0 and displaced from the centre of the dot by a distance D. Calculated results show that the ordering of quantum levels is dependent on R_0 and D (the so-called quantum-size effect) and that the variation of the binding energies of p and d states with D is different from that of s states. There are maxima of binding energies for p and d states at the proper positions of the donor ion inside the quantum dot.

Advances in two-dimensional quantum wells (2D QWs) and one-dimensional quantum-well wires (1d QWWs) have stimulated interest in the problem of calculating the energy levels of 'confined quantum systems', such as hydrogenic impurity states in a confined region. The impurity states in 2D QWs and superlattices have been calculated by a number of authors [1–3]. In the last few years there has been increasing interest in the study of the electronic properties of QWWs. Several calculations [4–6] have been performed for electron and impurity levels of QWWs. Recently, much attention has been given to the electronic structure of quantum-dot structures owing to their potential application in electronic devices. For instance, confined donors, acceptors and excitons in quantum dots have been studied [7–11].

The exact solutions of hydrogenic donors located at the centres of spherical quantum dots (SQDs) have been obtained by us earlier. However, the impurities could be located anywhere in SQDs, and the ground-state level and the level ordering will change as the location shifts to the edge or out of the SQDs. It should be interesting therefore to extend our early work [7] to a systematic investigation of the positional dependence of ground and excited donor states in SQDs with a finite barrier height. In this letter, we focus attention on the level ordering and binding energies of a donor off centre in an SQD. A variational method with the use of proper basis functions, which are the exact eigenfunctions of a central donor with an effective ion charge in the SQD, is used to determine the quantum levels.

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

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

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

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following form:

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

where V_0 is the barrier height and can be obtained from a fixed ratio of the band-gap discontinuity. In this letter, effective atomic units are used so that all energies are measured in units of the effective Rydberg Ryd* and all distances are measured in units of effective Bohr radius a^*. We should point out that the polarization and image-charge effects have been ignored as is reasonable for the GaAs–$Ga_{1-x}Al_xAs$ QW systems. The Schrodinger-like equation is expressed as

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

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

The Hamiltonian can be rewritten as

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

with

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

$$H' = 2w/r - 2/|r - D|. \tag{6}$$

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 [7]. 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 (7) includes only the terms with a fixed magnetic quantum number m, i.e., $m_1 = m_2 = \ldots = m_f = m$. According to the variational principle, it is straightforward to obtain the equation

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

with

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

The condition that (8) has non-zero solution is

$$||H'_{ij} - (E - E_j)\delta_{ij}|| = 0. \tag{10}$$

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

$$\psi_i(r, \theta, \phi) = R_{n_i l_i}(r) Y_{l_i m_i}(\theta, \phi) \tag{11}$$

where $Y_{l_i m_i}(\theta, \phi)$ and $R_{n_i l_i}(r)$ are the spherical harmonic and radial wave functions, respectively. Using the relation

$$\frac{1}{|r - r'|} = \sum_{l=0}^{\infty} \frac{r_<^l}{r_>^{l+1}} P_l(\cos \Theta) \tag{12}$$

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

$$H'_{ij} = 2w \int_0^{\infty} R_{n_i l_i} R_{n_j l_j} r \, dr \, \delta_{l_i l_j} - 2 \sum_{l=|l_i - l_j|}^{l_i + l_j} (-1)^m \left[(2l_i + 1)(2l_j + 1)\right]^{1/2} \begin{pmatrix} l_i & l_j & l \\ 0 & 0 & 0 \end{pmatrix}$$

$$\times \begin{pmatrix} l_i & l_j & l \\ -m & m & 0 \end{pmatrix} \left[\frac{1}{D^{l+1}} \int_0^D R_{n_i l_i} R_{n_j l_j} r^{l+2} \, dr + D^l \int_D^{\infty} R_{n_i l_i} R_{n_j l_j} \frac{1}{r^{l-1}} \, dr \right] \tag{13}$$

where $\begin{pmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{pmatrix}$ is the Wigner $3j$ symbol. Then the energy levels are obtained by solving (10) numerically. For central donors in SQDs, i.e., $D = 0$, the Hamiltonian H of (1) becomes H_0 of (5) with $w = 1$. As mentioned above, the exact solutions can be obtained (7). The quantum levels are dependent on the principal and radial quantum numbers n and l and degenerate with respect to the magnetic quantum number m, so the levels are denoted by $E_{nl}(w = 1)$. We have 1s, 2p, 3d, 2s and 4f level states, and so on, if the usual notation of l, i.e., s, p, d, f and the principal quantum number n are used for the level notation. For $D \neq 0$, the degeneracy of $E_{nl}(w = 1)$ is partially removed. The quantum levels obtained by solving (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 non-degenerate.

Compared with the binding energy of a donor centre in a 3D, 2D or 1D system, the binding energy of the corresponding donor states in the SQD can be defined by

$$E_B(n, l, m) = E_{nl}(w = 0) - E_{nlm} \tag{14}$$

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

$$k_0 + K_0 \tan(k_0 R_0) = 0 \qquad \text{if } l = 0 \tag{15a}$$

$$ik_l h_l(iK_l R_0) j_{l-1}(k_l R_0) + K_l h_{l-1}(iK_l R_0) j_l(k_l R_0) = 0 \qquad \text{if } l \geqslant 1 \tag{15b}$$

$$k_l = E^{1/2} \qquad K_l = (V_0 - E)^{1/2} \tag{16}$$

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

The ground and excited states have been calculated for GaAs–$Ga_{1-x}Al_xAs$ SQDs of $V_0 = 80$ Ryd* with $R_0 = 1$ and $3a^*$, respectively. It is found that the energy levels $E_{nl}(w = 1)$ split as the donor ion moves away from the centre of the sphere and that E_{nl0} is the lowest level among E_{nlm} because of the donor ion located on the z-axis. The $E_{nl}(w = 0)$ and $E_B(n, l, 0)$ have also been calculated as shown in tables 1 and 2. It is readily seen that for SQDs with $R_0 = 1a^*$, the ordering of $E_{nl}(w = 1)$ is the same as that of $E_{nl}(w = 0)$ and it is 1s, 2p, 3d, 2s, ... However, the situation is different for SQDs with $R_0 = 3a^*$. The ordering of $E_{nl}(w = 0)$ is 1s, 2p, 3d, 2s, ... while that of $E_{nl}(w = 1)$ is 1s, 2p, 2s, 3d, ... What we have mentioned above means that the ordering of $E_{nl}(w = 0)$ cannot be changed by adding a donor in the centre of an SQD as the R_0 is smaller or the confinement is stronger, and the situation is different as the R_0 is larger.

Table 1. Binding energies $E_B(n, l, 0)$ of a donor off centre in an SQD of $V_0 = 80$ Ryd* with $R_0 = a^*$. The corresponding quantum levels $E_{nl}(w = 0)$ and $E_{nl}(w = 1)$ are also shown and $E_{nl0} = E_{nl}(w = 0) - E_B(n, l, 0)$. D is the distance of the donor ion along the z-axis from the centre of the sphere. Effective atomic units, i.e., effective Rydberg Ryd* and Bohr radius a^*, are used.

			$E_B(n, l, 0)$							
nl	$E_{nl}(w = 0)$	$E_{nl}(w = 1)$	D=0	D=0.1	D=0.3	D = 0.5	D=0.7	D=0.9	D=1.5	D=2.0
1s	7.957	3.337	4.620	4.568	4.256	3.715	3.000	2.322	1.342	0.989
2p	16.225	12.875	3.350	3.399	3.677	3.842	3.559	2.815	1.486	1.045
3d	26.593	23.670	2.923	2.945	3.150	3.492	3.591	3.001	1.536	1.046
2s	31.425	25.828	5.597	5.381	4.233	3.156	2.546	2.123	1.308	0.923

Table 2. The same as table 1 but for an SQD of V_0= 80 Ryd* with $R_0 = 3a^*$.

			$E_B(n, l, 0)$							
nl	$E_{nl}(w = 0)$	$E_{nl}(w = 1)$	D=0	D=0.5	D=1.0	D = 1.5	D=2.0	D=2.5	D=4.0	D=7.0
1s	1.019	−0.872	1.891	1.824	1.763	1.628	1.356	0.982	0.511	0.282
2p	2.085	0.848	1.237	1.273	1.333	1.367	1.319	1.102	0.551	0.294
3d	3.429	2.363	1.066	1.084	1.168	1.254	1.290	1.148	0.554	0.294
2s	4.074	1.989	2.085	1.861	1.489	1.147	0.931	0.785	0.478	0.262

With increasing distance D, the binding energies $E_B(n, 0, 0)$ of s states decrease while those of p and d states increase continuously until their maxima, and then decrease, as shown in tables 1 and 2. This is easy to understand if we note that the donor ion is on the z-axis and that the square maxima of wavefunctions of s states are at the origin and those of p and d states with $m = 0$ are at positions of the z-axis. It is interesting to note that for the case of $R_0 = 3a^*$, the ordering of E_{nl0} becomes the same as that of $E_{nl}(w = 0)$ as the donor ion moves away from the centre of the sphere to reach a fixed point. However, the ordering of E_{nl0} does not change for the case of $R_0 = a^*$ as the donor ion moves away from the centre.

In summary, we have for the first time reported calculation results of ground states and excited states with $m = 0$ of donors confined by GaAs–$Ga_{1-x}Al_xAs$ SQDs of V_0 = 80 Ryd* with $R_0 = a^*$ and $3a^*$ and located inside and outside SQDs. The results have clearly demonstrated the so-called quantum-size effect. The ordering of quantum levels is dependent on the radius R_0 and the distance D and the ordering of E_{nl0} can vary between $R_0 = a^*$ and $3a^*$ cases. The binding energies of s states decrease continuously with increasing D while there are maxima for those of p and d states with $m = 0$. Finally, it is worthwhile to point out that the calculation method with the proper H_0, i.e., a proper set of basis functions, is useful for studying electronic structures of impurities off centre or others in QW structures. Using the proper H_0 of (5) is important for obtaining better results. It is best to take $w = 1$ and 0 for the calculation of quantum levels of a donor inside and outside the SQDs, respectively. In fact, the results should be much better with the use of a proper w in H_0 of (5).

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