

Size and shape effects of quantum dots on two-electron spectra

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The exact spectra of two electrons confined by two-dimensional and three-dimensional quantum dots (2D and 3D QD's) with parabolic potentials are obtained. Using the present results, the size and shape effects of QD's on the spectra are revealed. It is found that the spectra are dramatically changed with the variation of the dot size, and then the crossover of two levels can appear. The variation of spectra with size and the positions of crossover points are quite different between 2D and 3D QD's. It is well explained based on the study of electron-electron interaction energies in 2D and 3D QD's. The size and shape effects predict a possibility to observe phenomena related to electron-electron interactions in QD's. [S0163-1829(97)06120-1]

I. INTRODUCTION

Various designs, manufacturings, and studies of low-dimensional structures and materials have proliferated at an explosive rate because of interests in physics and in technological applications. Recently, advances in nanofabrication technology have made it possible to manufacture quantum dots (QD's) containing one, two, and more electrons, which are intensively investigated experimentally and theoretically. The experimental study of semiconductor QD's is expanding rapidly,¹⁻⁶ and electron-electron interaction and correlation effects are shown to be of great importance⁷⁻⁹ in such systems. In the meantime, a large number of theoretical investigations of electronic structures and related magnetic and optical properties in QD's have been performed to explain the experimental observations.

Semiconductor QD's are quite idealistic quasi-zero-dimensional structures to be studied since the effective-mass theory can be applied in a proper regime of quantum size. As is well known, the study of electronic structures in quantum-well structures with and without strong magnetic fields is an important problem in semiconductor physics. Quantum wells, in fact, under strong magnetic fields can form some kinds of QD's. Therefore, the studies of electronic structures in QD's containing more than one electron are of interest both in their own right and to understand the role of strong magnetic fields in quantum-well structures.

It is very important to have reliable methods to solve the many-electron problem and show characteristics of electronic structures in QD's. Previous approaches to the problem mainly include the "exact" numerical diagonalization,^{7,8} numerical simulations based on quantum Monte Carlo techniques,¹⁰ and Hartree-Fock calculations.^{7,11-13} For two electrons in spherical and circular QD's with parabolic potentials, the exact solutions can be obtained. To our knowledge, however, there has been no such systematic investigation related to the exact solutions of two electrons in such two-dimensional (2D) and three-dimensional (3D) QD's.

For the single-electron spectra of QD's, the size and shape effects have been studied and shown in detail.¹⁴ In order to exactly show the size and shape effects of QD's on the two-electron spectra and to better understand the characteristics of electron-electron interaction and correlation in confined systems, the energy levels of two electrons in 2D and 3D QD's are calculated by using the series expansion method in this paper. Based on the exact results, the interesting phenomenon is clearly revealed.

In Sec. II of this paper, the Hamiltonian and energy-level structures of two electrons in 2D and 3D QD's are presented. In Sec. III, the exact solutions are shown for two electrons in QD's with parabolic potentials. Main results are given and discussed in Sec. IV, followed by a summary in Sec. V.

II. HAMILTONIAN AND ENERGY LEVELS

Typically, the confinement potential seen by electrons in a QD is created by spatially extended charge distributions. For most QD's, a parabolic potential is a very good approximation to describe the confinement of the electrons.^{1,11} Hence, the forms of the Hamiltonian of two electrons in such parabolic 3D and 2D QD's are, respectively, as follows:

$$H_{3D} = -\nabla_1^2 - \nabla_2^2 + \frac{1}{4}\gamma^2 r_1^2 + \frac{1}{4}\gamma^2 r_2^2 + \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|}, \quad (1)$$

and

$$H_{2D} = -\nabla_1^2 - \nabla_2^2 + \frac{1}{4}\gamma^2 \rho_1^2 + \frac{1}{4}\gamma^2 \rho_2^2 + \frac{2}{|\rho_1 - \rho_2|}, \quad (2)$$

where the effective atomic units are used. The effective Rydberg Ry^* and the effective Bohr radius a^* are taken to be the energy and length units, respectively. It is easy to see that $\gamma^{-1/2}$ is related to the confinement region of electrons in QD's.

H_{3D} and H_{2D} can be separated into center-of-mass and relative-motion terms. First, for the sake of convenience, H_{3D} is written as

$$H_{3D} = H_{3DR} + H_{3Dr}, \quad (3)$$

with

$$H_{3DR} = -\frac{\nabla_R^2}{2} + \frac{1}{2} \gamma^2 R^2, \quad (4)$$

and

$$H_{3Dr} = -2\nabla_r^2 + \frac{1}{8} \gamma^2 r^2 + \frac{2}{r}, \quad (5)$$

where $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$, $\nabla_R = \nabla_1 + \nabla_2$, $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$, and $\nabla_r = (\nabla_1 - \nabla_2)/2$. This separability and the spherical symmetry of the problem allow us to write the two-particle wave functions in spherical coordinates in the form $\Phi(\mathbf{R})\psi(r)\mathbf{Y}_{lm}(\vartheta, \varphi)$. The spatial part of the total wave function is symmetric (antisymmetric) with respect to particle permutation for even (odd) l . Since the Pauli exclusion principle requires the total wave function to be antisymmetric, we have spin singlet ($s=0$) and triplet ($s=1$) states for even and odd l , respectively. The energy eigenvalues of Eq. (4) are given by

$$E(N, L) = (2N + L + \frac{3}{2})\gamma, \quad (6)$$

with radial ($N=0, 1, 2, \dots$) and angular-momentum ($L=0, 1, 2, \dots$) quantum numbers. The eigenvalues of the relative motion excluding the electron-electron interaction are also given by

$$E_0(n, l) = (2n + l + \frac{3}{2})\gamma \quad (7)$$

with the corresponding radial and angular-momentum quantum numbers $n=0, 1, 2, \dots$ and $l=0, 1, 2, \dots$. However, we should solve the Schrödinger-like equation

$$H_{3Dr}[\psi(r)\mathbf{Y}_{lm}(\vartheta, \varphi)] = E(l)[\psi(r)\mathbf{Y}_{lm}(\vartheta, \varphi)] \quad (8)$$

to obtain the energy of the relative motion including the electron-electron interaction. Assuming that $\psi(r) = \sqrt{r}\phi(r)$, we find the equation satisfied by the function $\phi(r)$:

$$\frac{d^2\phi}{dr^2} + \frac{1}{r} \frac{d\phi}{dr} + \left(\frac{E(l)}{2} - \frac{1}{r} - \frac{(l+1/2)^2}{r^2} - \frac{1}{16} \gamma^2 r^2 \right) \phi = 0, \quad (9)$$

which will be solved by using the series expansion method shown in the next section.

It is similar for H_{2D} of Eq. (2), which can also be separated into center-of-mass and relative-motion terms. This separability and the cylindrical symmetry allow us to express the wave functions in plane polar coordinates in the form $\Phi(\mathbf{R})\phi(r)\exp(im\varphi)$. We therefore have spin singlet ($s=0$) and triplet ($s=1$) states for even and odd m , respectively. The eigenvalues of the center-of-mass term are as follows:

$$E(N, M) = (2N + |M| + 1)\gamma \quad (10)$$

with radial ($N=0, 1, 2, \dots$) and azimuthal ($M=0, \pm 1, \pm 2, \dots$) quantum numbers. The eigenvalues of the relative motion excluding the electron-electron interaction are easily obtained by

$$E_0(n, m) = (2n + |m| + 1)\gamma \quad (11)$$

with the corresponding radial and azimuthal numbers $n=0, 1, 2, \dots$ and $m=0, \pm 1, \pm 2, \dots$.

To obtain the eigenenergies of the relative motion including the electron-electron interaction, we should solve the Schrödinger-like equation

$$H_{2Dr}[\phi(r)\exp(im\varphi)] = E(m)[\phi(r)\exp(im\varphi)]. \quad (12)$$

It is easy to find the equation satisfied by the function $\phi(r)$:

$$\frac{d^2\phi}{dr^2} + \frac{1}{r} \frac{d\phi}{dr} + \left(\frac{E(m)}{2} - \frac{1}{r} - \frac{m^2}{r^2} - \frac{1}{16} \gamma^2 r^2 \right) \phi = 0, \quad (13)$$

which is similar to Eq. (9) and can be solved by the same method.

The values of $E(n, l)$ and $E(n, m)$ can be obtained after Eqs. (9) and (13) are solved. For the sake of convenience, we define the electron-electron interaction energies $E_r(n, l)$ [$E_r(n, m)$] as the difference between $E(n, l)$ [$E(n, m)$] and $E_0(n, l)$ [$E_0(n, m)$], i.e.,

$$E_r(n, l) = E(n, l) - E_0(n, l), \quad (14)$$

and

$$E_r(n, m) = E(n, m) - E_0(n, m). \quad (15)$$

Then, the energy eigenvalues of H_{3D} (H_{2D}) are the sum of the center-of-mass energy and the energy of the relative motion as follows:

$$\begin{aligned} E(n, l; N, L) &= E(n, l) + E(N, L) \\ &= [2(N+n) + L + l + 3]\gamma + E_r(n, l) \end{aligned} \quad (16)$$

and

$$\begin{aligned} E(n, m; N, M) &= E(n, m) + E(N, M) \\ &= [2(N+n) + |M| + |m| + 2]\gamma + E_r(n, m). \end{aligned} \quad (17)$$

III. SERIES SOLUTIONS

Now, we are prevented from analytical exact solutions of the eigenvalue problem introduced in Sec. II because Eqs. (9) and (13) with suitable boundary conditions are beyond the analytical problem of confluent hypergeometric equations. However, we can use the method of series expansion¹⁵ to obtain exact series forms in different regions for Eqs. (9) and (13) and the exact values of $E(n, l)$ and $E(n, m)$ and, then, the exact solutions of two electrons in 3D and 2D QD's. Here we show the formulas only to solve Eq. (9) since they are very similar to those to solve Eq. (13).

It should be noted that zero and infinity are regular and irregular singular points of Eq. (9), respectively. In the region $0 < r$ we have a series solution, which has a finite value at $r=0$ as follows:

$$\phi(r) = Ar^l \sum_{n=0}^{\infty} a_n r^n, \quad (18)$$

where A is a constant and a_0 is equal to 1. Noting that a_n is equal to zero as n is equal to a negative integer, the other a_n can be determined by the following recurrence relation:

$$a_n = [2a_{n-1} - E(l)a_{n-2} + \frac{1}{8}\gamma^2 a_{n-4}] / (4l + 2n + 2)n$$

for $n = 1, 2, 3, \dots$ (19)

In the region $r < \infty$ we can obtain a normal solution. It approaches zero at $r = \infty$ and is found in the form

$$\phi(r) = B \exp(-\frac{1}{8}\gamma r^2) r^s \sum_{n=0}^N b_n r^{-n}, \quad (20)$$

where

$$s = E(l) / \gamma - 1, \quad (21)$$

$$b_0 = b_1 = 1,$$

$$b_n = 2b_{n-1} - [(s-n+2)^2 - (l+1/2)^2] b_{n-2}$$

for $n = 2, 3, 4, \dots$, (22)

and B is a constant. We should point out that Eq. (18) and Eq. (20) are suitable for numerical calculations in regions of small r and large r , respectively.

In order to match the solution of Eq. (18) with that of Eq. (20), we give T solutions around R_1, R_2, \dots and R_T , which are the proper points for solving Eq. (9) exactly. The solution of uniformly convergent Taylor series around R_i ($i = 1, 2, \dots, T$) is written as follows:

$$\phi(r) = C_i \sum_{n=0}^{\infty} c_{in} (r - R_i)^n + D_i \sum_{n=0}^{\infty} d_{in} (r - R_i)^n, \quad (23)$$

where C_i and D_i are constants, c_{i0} and d_{i1} are equal to 1, and c_{i1} and d_{i0} are equal to 0. The c_{in} and d_{in} can be determined by recurrence relations.

Using the matching conditions at $r = R_i$ ($i = 1, 2, \dots, T$), and the 2×2 transfer matrices, we can deduce the equation for eigenenergies $E(n, l)$ easily. The values of $E(n, l)$ and $\psi_{nl}(r) [\sqrt{r} \phi_{nl}(r)]$ are obtained numerically.

To close this section, it is interesting to point out that the method mentioned above is powerful compared with the perturbation theory, the WKB approximation, and so on because it allows us to obtain the exact eigenvalues in the whole region of γ .

IV. RESULTS AND DISCUSSION

Before the present results are shown and discussed, it is useful to specify labeling of quantum levels of two electrons in 3D and 2D QD's. As shown in Eqs. (17) and (16), the levels $E(n, m; N, M)$ and $E(n, l; N, L)$ can be labeled by four symbols n, m, N, M and n, l, N, L , respectively. The even and odd m (l) correspond to the spin singlet ($s=0$) and triplet ($s=1$) states because of the Pauli exclusion principle. We have states $1s, 2p, 2s, 3d, 3p$ and $1S, 2P, 2S, 3D, 3P$ and so on if the principal quantum numbers $n + |m| + 1$ ($n + l + 1$) and $N + |M| + 1$ ($N + L + 1$) are used instead of n and N and the notations s, p, d, \dots and S, P, D, \dots are used for $|m|(l)$ and $|M|(L) = 0, 1, 2, \dots$.

TABLE I. Exact quantum levels of two electrons in 2D QD's with different γ ($\gamma^{-1/2}$). The level sequences are in order of increasing magnitude. For the sake of convenience, the short notation, i.e., a, b, c, etc., is used to indicate the quantum numbers ($n, m; N, M; s$) and to show the changes of the level order. The energy unit is Ry*.

γ ($\gamma^{-1/2}$)	1.0(1.0)	0.05(4.4721)
a: (0,0;0,0;0)	(a) 3.3196	(a) 0.2962
b: (0,1;0,0;1)	(b) 3.8278	(b) 0.3062
c: (0,0;0,1;0)	(c) 4.3196	(d) 0.3310
d: (0,2;0,0;0)	(d) 4.6436	(c) 0.3462
e: (0,1;0,1;1)	(e) 4.8278	(h) 0.3476
f: (1,0;0,0;0)	(f) 5.1472	(e) 0.3562
g: (0,0;1,0;0)	(g) 5.3196	(i) 0.3810
h: (0,3;0,0;1)	(h) 5.5174	(f) 0.3854
i: (0,2;0,1;0)	(i) 5.6436	(g) 0.3962
j: (1,1;0,0;1)	(j) 5.7438	(j) 0.3968
k: (0,1;1,0;1)	(k) 5.8278	(k) 0.4062
l: (1,0;0,1;0)	(l) 6.1472	(n) 0.4066
m: (0,0;1,1;0)	(m) 6.3196	(o) 0.4240
n: (0,4;0,0;0)	(n) 6.4693	(p) 0.4310
o: (1,2;0,0;0)	(o) 6.5956	(l) 0.4354
p: (0,2;1,0;0)	(p) 6.6436	(m) 0.4462

We have performed numerical calculations for energy levels of two electrons in 2D and 3D QD's with γ between 0.05 and 10. As shown in Table I, the two-electron spectra of 2D QD's vary not only in the values but also in the level ordering as γ changes from 0.05 to 1. In order to better show the quantum-size effects and compare with others, we have plotted most of them normalized by γ as functions of $\gamma^{-1/2}$ in Fig. 1. It is readily seen that the four low levels are in good agreement with those in Refs. 7 and 9, which we can find to compare our results with, and that the energy-level structure is dramatically changed as the $\gamma^{-1/2}$ changes from 0 to 4. As illustrated in the figure and Table I, an important aspect of the quantum-size effects is the changes of the level ordering and the level differences and then the crossover of two levels with the same or different spins can appear as $\gamma^{-1/2}$ (γ) is larger (less) than 1. The crossover between singlet and triplet levels is related to the magnetic properties of confinement systems such as metal clusters, which will be discussed below.

In order to compare the spectra of two electrons in 3D QD's with those in 2D QD's shown in Fig. 1, we have plotted the corresponding levels ($l = |m|, L = |M|$) normalized by γ as functions of $\gamma^{-1/2}$ in Fig. 2. The figure also shows that the level ordering and the level differences vary with the size $\gamma^{-1/2}$, and the level crossover appears as $\gamma^{-1/2} \gg 1$. It is obvious, however, that the changes of energy levels with $\gamma^{-1/2}$ are smaller for 3D QD's than for 2D QD's, and then the crossover points of two levels correspond to much larger values of $\gamma^{-1/2}$ for 3D QD's than those for 2D QD's. It means that the spectra and related properties depend not only on the dot sizes but also on the dot shapes.

For a better understanding of the size and shape effects, it is interesting to study the electron-electron interaction energies $E_r(n, m)$ and $E_r(n, l)$ defined by Eqs. (15) and (14). In

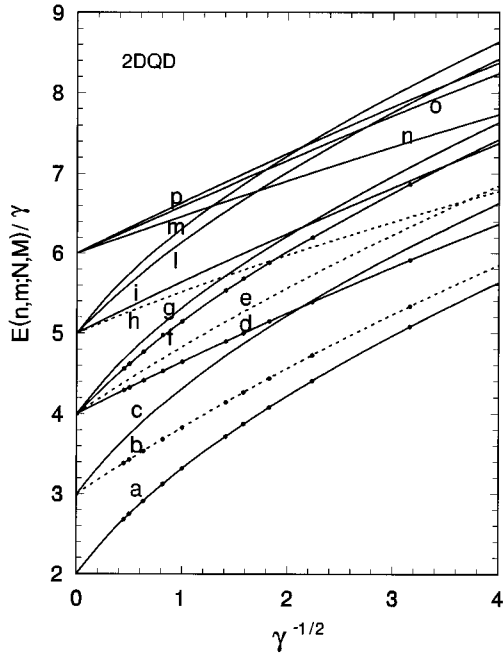


FIG. 1. $E(n,m;N,M)$ normalized by γ vs $\gamma^{-1/2}$ for a, b, c, d, e, f, g, h, i, l, m, n, o, and p states with the quantum numbers labeled in Table I. The solid and dashed lines represent, respectively, singlet and triplet states, and the solid circles those obtained by using the “exact” diagonalization method.

Fig. 3, the $E_r(n,m)$ and $E_r(n,l)$ are plotted as functions of γ . As shown in the figure, the $E_r(n,m)$ increases with γ and the ordering is as follows: $E_r(0,0) > E_r(1,0) > E_r(2,0) > E_r(0,1) > E_r(1,1) > E_r(0,2) > E_r(1,2) \dots$. The order is the same for $E_r(n,l)$ of 3D QD’s but the value of the 1s state and those of 2p and 3d states are, respectively, much less and less than the corresponding ones of 2D QD’s shown in the figure.

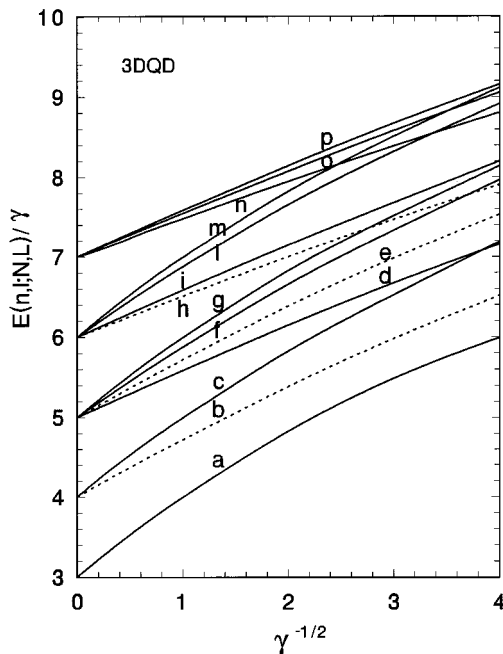


FIG. 2. Same as in Fig. 1 except for $E(n,l;N,L)$ of 3D QD’s.

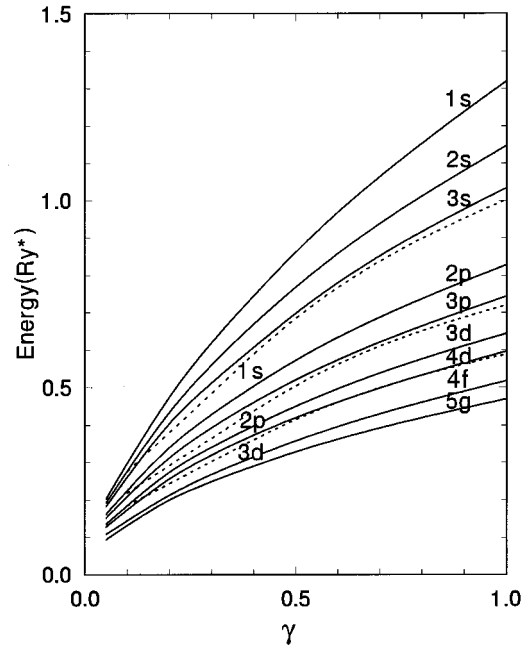


FIG. 3. $E_r(n,m)$ and $E_r(n,l)$ vs γ for 1s, 2s, 3s, 2p, 3p, 3d, 4d, 4f, and 5g states (solid lines) of 2D QD’s and for 1s, 2p, and 3d states (dashed lines) of 3D QD’s.

It is accurate enough to calculate the $E_r(n,m)$ and $E_r(n,l)$ with the use of the first-order perturbation as γ is sufficiently large compared with electron-electron interaction. Then, $E_r(n,m)$ and $E_r(n,l)$ are, respectively, given by

$$E_r(n,m) = \left\langle \phi_{nm}(r) \left| \frac{2}{r} \right| \phi_{nm}(r) \right\rangle \quad (24)$$

and

$$E_r(n,l) = \left\langle \psi_{nl}(r) \left| \frac{2}{r} \right| \psi_{nl}(r) \right\rangle, \quad (25)$$

where $\phi_{nm}(r)$ and $\psi_{nl}(r)$ are normalized radial wave functions of Eqs. (13) and (9) without the electron-electron interaction terms. Using Eqs. (24) and (25), we can easily find the ordering mentioned above. The values are proportional to $\gamma^{1/2}$. However, both $E(N,M)$ ($E(N,L)$) and $E_0(n,m)$ ($E_0(n,l)$) are proportional to γ . As $\gamma^{-1/2}$ (γ) is less (larger) than 1, the level ordering is mainly determined by the sum of $E_0(n,m)$ ($E_0(n,l)$) and $E(N,M)$ ($E(N,L)$). It can be strongly changed by $E_r(n,m)$ ($E_r(n,l)$) if $\gamma^{-1/2}$ (γ) is much larger (less) than 1. This is the reason the quantum-size effects appear. Furthermore, for fixed $\gamma^{-1/2}$ and $l=|m|$, the $E_r(n,l)$ is less than $E_r(n,m)$ and larger than $E_r(n,|m|+1)$ as shown in Fig. 3. The same picture can be shown by using Eqs. (24) and (25). It is easily understood if the difference between Eqs. (9) and (13) and then the extension of wave functions $\phi_{nm}(r)$ and $\psi_{nl}(r)$ are noted. This is the reason for the spectrum difference between 2D and 3D QD’s shown in Figs. 1 and 2.

It is interesting to note that the crossover point and separation of singlet and triplet levels are related to the difference of electron-electron interaction energies between the even and odd $l(m)$ levels and strongly dependent on the sizes and

shapes of QD's. In Ref. 14, it has been shown that the shape and size effects of QD's and the role of an ion in the QD's are very important for the single-electron spectra. It is reasonable that the ion-electron interaction can also influence the two-electron spectra in QD's because the exchange interaction in the QD's is changed in the presence of the electron-ion interaction. For example, the energy difference between the single and triplet P states ($1s2P$ and $2p1S$ notations used here) in 2D QD's without a positive ion center obtained here is 0.0748, 0.4916, 1.0620, 1.4793, and 2.2679 Ry* and that with the ion center 0.0048, 0.2324, 0.7828, 1.2333, and 2.0472 Ry* (Ref. 16) for $\gamma=0.1, 1, 3, 5,$ and 10, respectively. It is obviously shown that the ion-electron interaction changes the exchange energy. What are mentioned above are useful for understanding why the magnetic moment per atom of metal clusters is very sensitive to the size and symmetry of the clusters^{17,18} and doping. It will be studied further and reported elsewhere.

V. SUMMARY

We have used different series solutions in different regions for the radial equation of the relative motion of two electrons in 2D and 3D QD's with parabolic potentials to obtain the exact solutions. Numerical results of 2D QD's are in good agreement with those obtained by using the "exact" diagonalization method.^{7,9} The quantum-size effects of 2D and 3D QD's on the spectra are clearly shown. The impor-

tant aspect of the effects is the crossover of two levels with the same or different spins. It is found that the variation of energy levels with dot size is smaller for 3D QD's than that for 2D QD's, and the positions of crossover points of two levels with the same or different spins are also different between 2D and 3D QD's. It is explained on the basis of analyzing the characteristics of electron-electron interaction energies in 2D and 3D QD's.

The present results are useful to understand the optical and magnetic properties of quantum-dot materials since the shape of realistic QD's is usually between spherical and circular ones. The size and shape effects predict a possibility to observe phenomena related to electron-electron interactions in QD's. Finally, it can be expected that the proper electronic structures of QD's and then the related properties will be obtained if the sizes and shapes of QD's including doping are better controlled.

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