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Electronic structure of three-dimensional quantum dots in tilted magnetic fields

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We investigate the electronic structure of three-dimensional quantum dots with two electrons in high magnetic fields and find that the combined effect of electron-electron interaction and hybrid-magnetoelectric quantization governs the whole energy spectrum. As a result, the spin transitions in the ground state of a quantum-dot disk can be quenched in the three-dimensional dot by relaxing the vertical confinement, or can be reentrant by tilting the magnetic field. We demonstrate that such behavior clearly appears in heat capacity and magnetization at sufficiently low temperatures.

Recent advances in nanostructure technologies make it possible to confine two-dimensional electrons at heterointerfaces into dots of radius l_0 comparable to the effective Bohr radius a^* of the host semiconductor.[1] Since the lateral size on the x-y plane is much larger than the extension of wave functions in the growth direction z, quantum dots in this limit often refer to quantum-dot disks.[2,3] Much attention has been paid to investigating the electronic structure of the quantum-dot disk, especially in the presence of magnetic fields.[3–7] A key factor in these studies is the role of electron-electron interactions. One remarkable observation in such a prospect is the change of the spin (or angular momentum) state[7] of the ground state with increasing of the magnetic field. These ground-state transitions are likely to occur at lower magnetic fields as the dot size l_0 increases because the relative strength of the Coulomb interaction to the confinement energy, which is estimated as l_0/a^*, increases accordingly. Thermodynamic quantities[4] such as heat capacity and magnetization were previously proposed as a sensitive probe to see the ground-state transitions, while far-infrared optical transitions are unaffected by the Coulomb interaction for parabolic confinements by virtue of the generalized Kohn's theorem.[8,9]

In this paper we study the electronic structure of three-dimensional (3D) quantum dots with two electrons in magnetic fields. The coupled motions on the lateral plane and along the vertical direction to the plane as well as the electron-electron interactions are fully taken into account. In contrast to quantum-dot disks, 3D quantum dots have comparable confinement energies in all three directions, in good agreement with recent studies.[2] The temperature and magnetic field (including its orientation and strength) dependences of heat capacity and magnetization are calculated and fully analyzed.

The most striking feature of our results is found to be a possible quenching of the ground-state transitions in 3D quantum dots despite the increase of the dot size, while this behavior occurs surely in quantum-dot disks.[4,7] In fact, we find that the important factor in governing the whole energy spectrum is the combined effect of the Coulomb interaction and the hybrid-magnetoelectric quantization, i.e., the coupling between the electric and magnetic confinements. The increase of the dot size in the radial direction about the magnetic field enhances the ground-state transitions while it suppresses the transitions along the direction parallel to the magnetic field over a certain range of magnetic fields. Thus, we suggest that the phase diagram[7] previously drawn for the spin state of quantum-dot disks should be modified by including the confinement-frequency ratio between the radial and parallel components. The main difference between the two kinds of quantum dots is manifested in the oscillating pattern of heat capacity and magnetization.

Our model for a parabolic 3D quantum dot is characterized by two different confinement frequencies, ω_p on the x-y plane ω_v along the z direction. Ignoring the Zeeman spin-splittings, the energy eigenvalues in the presence of tilted magnetic field, $\vec{B}=(B\sin\theta,0,B\cos\theta)$, are separated into two terms, the energies of the center-of-mass and relative motions. For $\theta=0°$, the former is given by[7]

$$E_{N,L,M}=\hbar\Omega(2N+|L|+1)+\frac{\hbar\omega_c}{2}L+\hbar\omega_v(M+\tfrac{1}{2}) , \quad (1)$$

where $\omega_c=eB/m^*$ is the cyclotron frequency and N $(=0,1,2,\ldots)$, L $(=0,\pm1,\pm2,\ldots)$, and M $(=0,1,2,\ldots)$ denote the radial, azimuthal, and vertical quantum numbers, respectively, for a cylindrical geometry. The frequency $\Omega=\sqrt{\omega_p^2+(\omega_c/2)^2}$ indicates the hybrid-magnetoelectric quantization on the x-y plane. The energies of the relative motion are specified by the corresponding quantum numbers n, l, and m, and they include electron-electron interaction energy obtained by an exact numerical diagonalization. Note that at $\theta=0°$ the quantum number l is conserved for the Coulomb interaction. However, for nonzero tilt angles, it is no longer a good quantum number because of the broken circular symmetry. To find eigenvalues, we expand the wave function in terms of eigenstates of a harmonic oscillator.

$$\psi_m = \sum C^m_{i,j,k} \phi_i(x)\phi_j(y)\phi_k(z) , \quad (2)$$

where $\phi_i(x)$ is the ith eigenstate of the harmonic oscillator and $C^m_{i,j,k}$ is determined by diagonalizing the Hamiltonian matrix. The calculated energies are accurate to within 10^{-5} meV, where the matrix size of the Hamiltonian is about 400.

In Figs. 1(a) and 1(b), the calculated energy dispersions are plotted as a function of magnetic field for $\hbar\omega_v = 30$ and 1.5 meV, respectively, with keeping $\hbar\omega_p = 3$ meV and $\theta = 0°$. In this case, we choose the dielectric constant $\varepsilon = 12.4$ and the effective mass $m^* = 0.0665$, which are relevant to GaAs. The electronic structure in Fig. 1(a) is found to be very similar to that of a quantum-dot disk[3] while the quantum dot in Fig. 1(b) can be considered as a football-like dot. One of the major differences between the two figures, except for the shift of the zero point energy, is the ground state. As the magnetic field increases, the ground state of the disklike dot has a sequence of l values ($l = 0, -1, -2, \ldots$) with the other quantum numbers zero, as viewed easily by the level crossings in Fig. 1(a), while the l value of the ground state in the football-like dot is zero for the magnetic fields considered here. Since the total spin of the two electrons is $S = [1-(-1)^l]/2$, the ground state in Fig. 1(a) entails an alternating sequence of the singlet and triplet states.[7] However, the feature in Fig. 1(b) is rather unexpected. To investigate the origin of the ground-state transitions, we consider two lowest energy levels, i.e., $l = 0$ and -1 states. The energy difference ΔE between these two states can be separated into the hybrid and Coulomb contributions[10] to first-order approximation by neglecting small couplings,

$$\Delta E \simeq \Delta E_{\text{hybrid}} + \Delta E_{\text{Coul}} , \quad (3)$$

$$\Delta E_{\text{hybrid}} = \hbar\Omega - \frac{\hbar\omega_c}{2} = \hbar\omega_p[\sqrt{1+t^2} - t] , \quad (4)$$

where $t = \omega_c / 2\omega_p$. As the hybrid effect represented by t increases, ΔE_{hybrid} in Eq. (4) becomes smaller. For the quantum-dot disk, ΔE_{Coul} is roughly estimated as

$$\Delta E_{\text{Coul}} \simeq \frac{\hbar^2}{a^* m^*}\left[\frac{1}{R_1} - \frac{1}{R_0}\right] \simeq -0.29\hbar\omega_p \frac{l_0}{a^*}(1+t^2)^{1/4} , \quad (5)$$

where $R_1 = \sqrt{2}R_0$ and $R_0 = l_0 = \sqrt{\hbar/m^*\omega_p}$ is the lateral size of the disk. Then, the ground-state transition ($\Delta E = 0$) can occur at a certain magnetic field B_c, which depends on the dot size l_0/a^*. From Eqs. (3)–(5), a rough estimate for B_c is found to be accurate to within a factor of 2 compared with the exact value. However, for the football-like quantum dot in Fig. 1(b), ΔE_{hybrid} is almost the same as that of the disk-like dot, while the absolute value of ΔE_{Coul} is smaller since the dot size along the field direction is larger than that of the dot in Fig. 1(a). Thus, the combined effect of the hybrid and Coulomb contributions is much reduced, and the ground-state transitions are suppressed in the football-like dot. However, we note that the football-like quantum dot can undergo the ground-state transition by tilting magnetic field due to the enhanced hybrid-magnetoelectric effect. Figure 2 shows the phase digram for the ground state for $\hbar\omega_p = 3$ meV. We find that it requires three scaled variables ω_v/ω_p, ω_c/ω_p, and l_0/a^* to complete such a phase diagram.

From the calculated eigenvalues, the equilibrium thermodynamic quantities, heat capacity, and magnetization are calculated and plotted as a function of magnetic field in Figs. 3 and 4, respectively. The heat capacity is given by[11]

$$C_v = \frac{dE}{dT} = k_B\left[L_2 - \frac{L_1^2}{L_0}\right] , \quad (6)$$

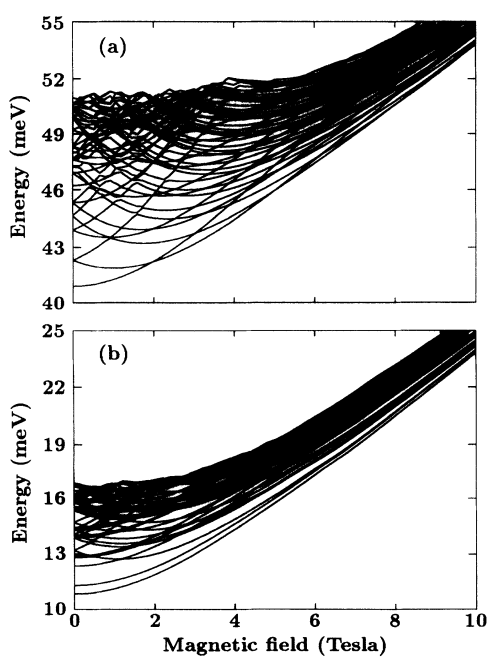

FIG. 1. Energy dispersions are plotted as a function of magnetic field in the z direction for (a) $\hbar\omega_v = 30$ meV and (b) $\hbar\omega_v = 1.5$ meV for two quantum dots with a lateral confinement $\hbar\omega_p = 3$ meV.

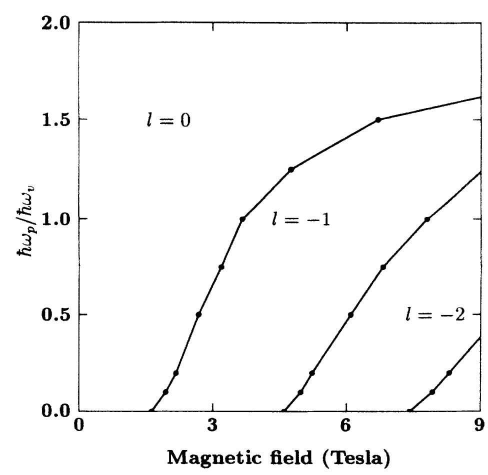

FIG. 2. The angular momentum phase diagram for the ground state of the 3D quantum dot is shown.

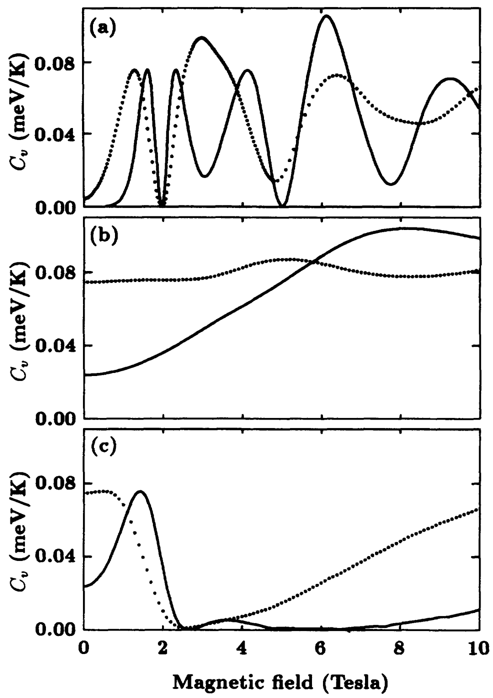

FIG. 3. Heat capacities vs magnetic field at $T=0.5$ K (solid line) and $T=1$ K (dotted line) for quantum dots with the confinement frequency $\hbar\omega_p=3$ MeV; (a) $\hbar\omega_v=30$ meV and $\theta=0°$, (b) $\hbar\omega_v=1.5$ meV and $\theta=0°$, and (c) $\hbar\omega_v=1.5$ meV and $\theta=55°$.

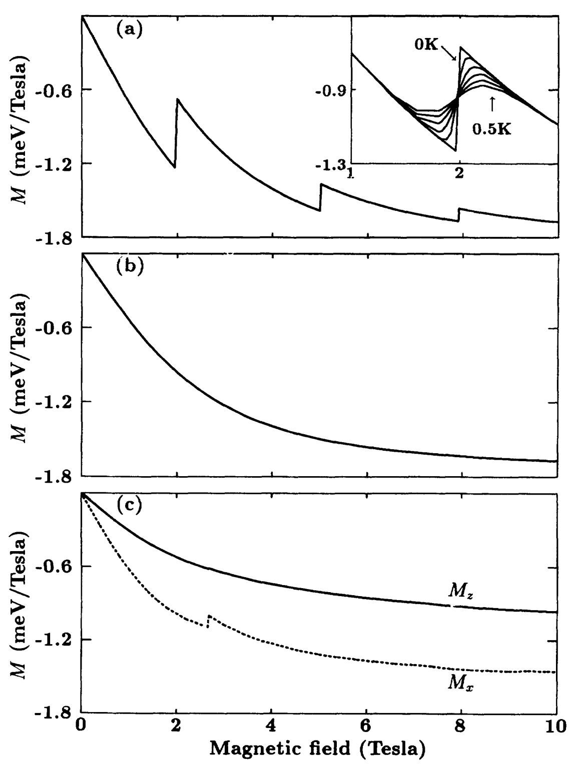

FIG. 4. Magnetizations vs magnetic field at $T=0$ K for the quantum dots shown in Fig. 3. In (c), dotted and solid lines denote M_x and M_z, respectively. The inset in (a) enlarges the temperature dependence of magnetization near the ground-state transition point.

$$L_r=\int_0^\infty \frac{\exp(z)z^r}{[1+\exp(z)]^2}g(\epsilon)d\epsilon\ , \tag{7}$$

where E is the total energy of electrons, $g(\epsilon)$ the density of states, and $z=(\epsilon-\mu)/k_BT$ with the chemical potential μ. At sufficiently low temperatures, the thermodynamic quantities are mainly contributed from the two lowest energy levels with the chemical potential lying in the middle of these two states. In this case, C_v can be approximated as

$$C_v\simeq\frac{\exp(z')z'^2}{[1+\exp(z')]^2}+\frac{\exp(-z')z'^2}{1+\exp(-z')]^2}\ , \tag{8}$$

where $z'=\Delta E/2k_BT$. Then, C_v has a maximum at $\Delta E/k_BT=4.8$ and becomes zero when $\Delta E=0$ or $\Delta E\to\infty$. Since ΔE oscillates as a function of B due to competition between the electron-electron interaction and the hybrid-magnetoelectric effect, C_v also varies with B as shown in Fig. 3. Thus, the ground-state transition $(\Delta E=0)$ is signaled by the minimum in heat capacity, $C_v\simeq 0$, accompanied by double peaks at sufficiently low temperatures. With the two-level approximation, a considerable difference in the heat capacities of the disklike [Fig. 3(a)] and football-like [Fig. 3(b)] dots can be explained. We find a strongly oscillating pattern in C_v of the disklike dot, while C_v of the football-like dot varies smoothly with magnetic field because of the absence of the ground-state transitions. As temperature increases, the two-level approximation given by Eq. (8) fails because thermal excitations of electrons to higher-energy levels become significant. Therefore, the heat capacity minimum which occurs at the ground-state transition does not completely drop to zero. Figure 3(c) shows the variation of C_v with the magnetic field for the football-like dot at a tilt angle $\theta=55°$. We find a ground-state transition at a magnetic field of $2<B<3$ T, and the energy spectra are almost degenerate for magnetic fields above B_c. This electronic structure is also confirmed by C_v, which has small values (above B_c) at $T=0.5$ K.

Next, we consider the magnetization and the results are shown in Fig. 4. Since $M=-\partial E/\partial B$ at $T=0$ K, the magnetization at very low temperatures is closely related to the slope of the ground-state energy. Similarly to the heat capacity, we find a considerable difference between the two quantum dots at $\theta=0°$. The magnetization of the disklike dot [Fig. 4(a)] has sharp jumps at the ground-state transition points, while the football-like dot [Fig. 4(b)] shows no peaks. At $T=0$ K, a sharp discontinuity in magnetization is expected at the transition point because the slope of the ground-state energy is discontinuous. However, at finite but very low temperatures, as shown in the inset of Fig. 4(a), the sharp peaks are broadened and its maximum is shifted to higher magnetic field as temperature increases. The amount of the shift is approximately proportional to temperature as expected from the following two-level analysis. At the transition field B_c, the two levels E_1 and E_2 cross each other, and they can be approximated as $E_1=k_1(B-B_c)$ and $E_2=-k_2(B-B_c)$, where k_1 and k_2 $(k_1>k_2>0)$ are the absolute values of the slopes of the two levels at $B=B_c$. Then, the total energy E and the magnetization M near

B_c are given by

$$E=(1-x)E_1+xE_2 , \quad (9)$$

$$M=-\frac{\partial E}{\partial B}=-k_1+(k_1+k_2)\frac{(\Delta+1)e^{-\Delta}+1}{(1+e^{-\Delta})^2} , \quad (10)$$

where $x=(1+e^{-\Delta})^{-1}$ and $\Delta=(k_1+k_2)(B-B_c)/k_BT$. Here, the variable x represents the probability of occupying the E_2 state near $B=B_c$, Δ varies very sensitively with magnetic field at very low temperatures. The magnetization in Eq. (10) has a maximum at $\Delta=2.4$, implying that the shift of the peak position from B_c $[=2.4k_BT/(k_1+k_2)]$ is proportional to temperatures. Since the difference of the two slopes, (k_1+k_2), corresponds to the amount of the magnetization jump at B_c and $T=0$ K, the peaks at the higher-order transitions, i.e., larger negative l value, are more rapidly rounded off with increasing temperature. In Fig. 4(c), the magnetizations, both the x and z components, in the football-like dot are shown in the presence of a tilted magnetic field, i.e., at $\theta=55°$. We find that M_z exhibits a smooth monotonic diamagnetism while M_x shows a discontinuous jump at $B=2.7$ T. Thus, as in case of heat capacity, this result indicates again the ground-state transition, which is caused by the enhanced hybrid-magnetoelectric effect due to the x component of the magnetic field.

In conclusion, we have investigated the electronic structure of three-dimensional quantum dots and find that their energy spectra are different from those of usual quantum-dot disks, especially in the ground state. We demonstrate that such a difference is clearly seen in heat capacity and magnetization.

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