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Addition energies in semiconductor quantum dots: Role of electron–electron interaction

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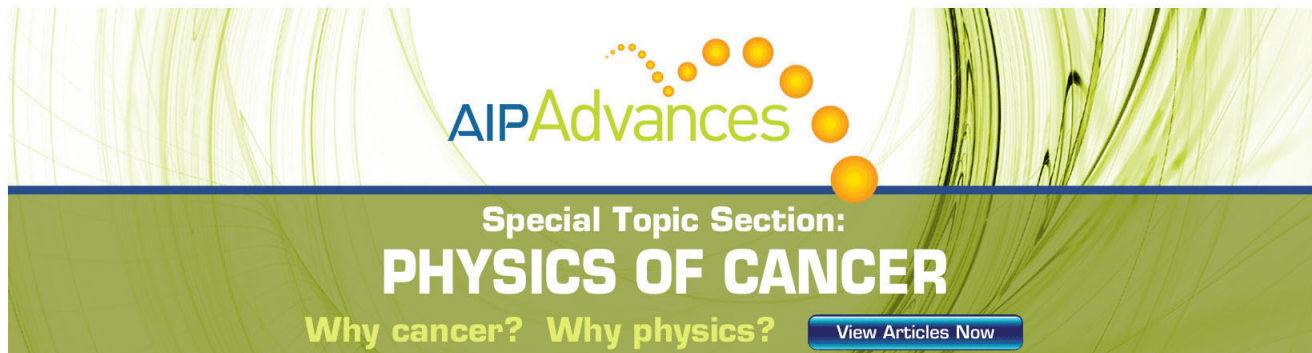
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Addition energies in semiconductor quantum dots: Role of electron–electron interaction

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We show that the addition spectra of semiconductor quantum dots in the presence of magnetic field can be studied through a theoretical scheme that allows an accurate and practical treatment of the single-particle states and electron–electron interaction up to large numbers of electrons. The calculated addition spectra exhibit the typical structures of Hund-like shell filling, and account for recent experimental findings. A full three-dimensional description of Coulomb interaction is found to be essential for predicting the conductance characteristics of few-electron semiconductor structures. © 1998 American Institute of Physics. [S0003-6951(98)03608-0]

In semiconductor quantum dots (QDs) Coulomb-correlation effects are expected to become more and more pronounced as their spatial confinement is increased. This has been so far one of the reasons of interest for such systems: they display the rich shell structure typical of atomic physics, but in addition they offer the possibility to tailor the confining potential and to vary the electron occupation by adjusting external parameters,¹ thus allowing direct investigation of fundamental properties of charge correlation.

At the same time, however, these results imply that many-body effects cannot be neglected in the theoretical description of QDs, and may be very relevant for predictions on their possible applications, for example, in semiconductor-based few-electron devices. While capacitance and tunneling spectroscopy experiments are becoming available for different dot structures,^{2–6} it is still an open and difficult problem to devise theoretical approaches allowing to include many-body effects in calculations for realistic systems. Exact calculations⁷ are necessarily limited to very few electrons; state-of-the-art QD structures instead involve several electrons (of the order of ten or hundred), therefore in general out of reach for these methods.

In this letter we propose an accurate and effective theoretical scheme that includes electron–electron interaction and can be applied to systems up to a large number of electrons. This approach can treat arbitrary three-dimensional (3D) confinement potentials, and hence deal with realistic QD structures also in the presence of external magnetic field. We apply this scheme to structures that were investigated in a recent experiment. The results are in good quantitative agreement with the observed spectra,⁵ and provide a transparent interpretation of their physical origin.

A key quantity that characterizes transport into a QD is the addition energy, i.e., the energy $A(N)$ required in order to place an extra electron into a dot that is initially occupied by $N-1$ particles. Such quantity, analogous to electron affinity in atomic physics, can be measured experimentally as a function of N . A systematic experiment on disk-shaped dots⁵ has shown that the voltage increment ΔA between successive

single-electron tunneling processes—i.e., between two successive maxima in the conductance—peaks at “magic” values of N ($N=2,6,12$), consistent with the filling of complete shells calculated⁸ for a dot of the same symmetry. Moreover, the experimental analysis showed that unusually large values of ΔA also occur for values of N corresponding to half-shell filling ($N=4,9$). The origin of these features, reminiscent of Hund’s rule in atomic physics,^{5,9,10} is intimately related to electron–electron interaction; therefore their quantitative evaluation is a challenge for any theoretical analysis of addition spectra in QDs.

Our description is based on an accurate treatment of the single-particle Hamiltonian and on the inclusion of Coulomb correlation according to first-order perturbation theory: assuming the many-particle ground state to be described by a single Slater determinant, the total energy of the full interacting Hamiltonian is

$$E(N) = \sum_{\alpha\sigma} \epsilon_{\alpha} \langle \hat{n}_{\alpha\sigma} \rangle + \frac{1}{2} \sum_{\alpha\beta\sigma} [U_{\alpha\beta} \langle \hat{n}_{\beta-\sigma} \rangle + (U_{\alpha\beta} - J_{\alpha\beta}) \langle \hat{n}_{\beta\sigma} \rangle] \langle \hat{n}_{\alpha\sigma} \rangle, \quad (1)$$

where $\langle \hat{n}_{\alpha\sigma} \rangle$ is the orbital occupation number, α denoting the set of radial and angular quantum numbers (n, m), and σ the spin value. According to Eq. (1), the evaluation of the energies for the various many-electron states in the dot requires only the knowledge of the Coulomb and exchange matrix elements

$$U_{\alpha\beta} = e^2 \iint \frac{|\phi_{\alpha}(\mathbf{r})|^2 |\phi_{\beta}(\mathbf{r}')|^2}{\kappa |\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}', \quad (2)$$

$$J_{\alpha\beta} = e^2 \iint \frac{\phi_{\alpha}^*(\mathbf{r}) \phi_{\beta}^*(\mathbf{r}') \phi_{\alpha}(\mathbf{r}') \phi_{\beta}(\mathbf{r})}{\kappa |\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}'. \quad (3)$$

Here κ is the dielectric constant and ϕ_{α} are single-particle real-space eigenstates of the dot in the presence of the applied magnetic field.

The important feature of the present scheme is that $U_{\alpha\beta}$, $J_{\alpha\beta}$ are calculated directly, contrary to other common approaches where Coulomb and exchange integrals are taken as input parameters.¹¹ Thus, many-body effects are taken into

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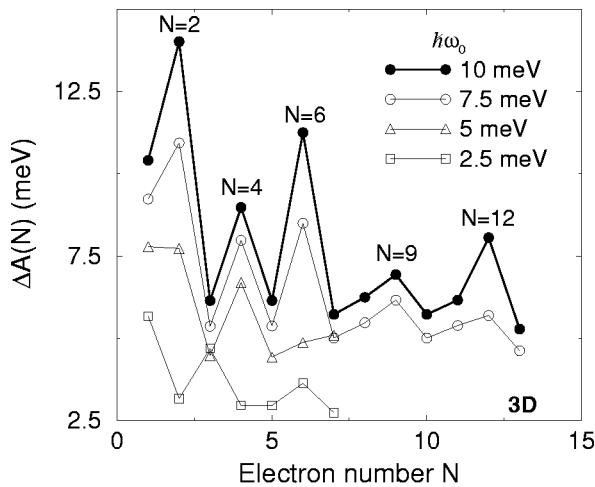


FIG. 1. Calculated addition-energy increment $\Delta A(N)$ vs electron number N for different QD structures characterized by a parabolic potential in the xy plane (confining energy $\hbar\omega_0$) and by a finite-barrier quantum-well potential along the z direction.

account through (1), and quantum-confinement effects through the single-particle ingredients, energies ϵ_α and wave functions ϕ_α ; the latter enter only in the calculation of Coulomb and exchange matrix elements. This has the advantage that realistic confining potentials (e.g., extracted from high-resolution transmission electron microscope (TEM) micrographs) and an external magnetic field can be included quite easily by choosing the appropriate single-particle Hamiltonian, as is now commonly done for the calculation of optical properties.¹²

Note that previous approaches have modeled the QD confinement in terms of a purely two-dimensional (2D) coordinate space.^{7,9,10} As a matter of fact, in most state-of-the-art QD structures the confinement is far from being 2D: for example, in the gated dots of Ref. 5 the typical confinement width in the growth direction z (of the order of 10 nm) is comparable with the typical extension of the carrier ground state in the lateral-confinement (xy) plane. Therefore, in our calculations we fully retain the 3D nature of the problem and mimic the dots of Ref. 5 by assuming a finite-barrier quantum-well potential along z and a parabolic potential in (xy). Note that the assumption of parabolic potentials reflects the experimental indications due to the specific sample structure and is not required by our model. Within the usual envelope-function approximation, the only input parameters are the electron effective masses ($m^*=0.065m_0$ in the dot and $m^*=0.079m_0$ in the barrier), the dielectric constant in the dot ($\kappa=12.98$), and the quantum-well height and width ($V_0=200$ meV and $L=12$ nm).

Our results for the addition-energy variations, $\Delta A(N) = A(N+1) - A(N)$, are displayed in Fig. 1 as a function of the electron number N for different dots, characterized by different in-plane confinement energies $\hbar\omega_0$. Here, $A(N)$ is obtained as $E(N) - E(N-1)$, where $E(N)$ is the ground-state energy in Eq. (1). As we can see, $\Delta A(N)$ exhibits peaks corresponding both to complete and half-shell filling, and is in excellent agreement with experiments in Ref. 5.

The origin of this result is in the delicate interplay between single-particle contributions and electron-electron repulsion, which according to Eq. (1) defines the ground-state

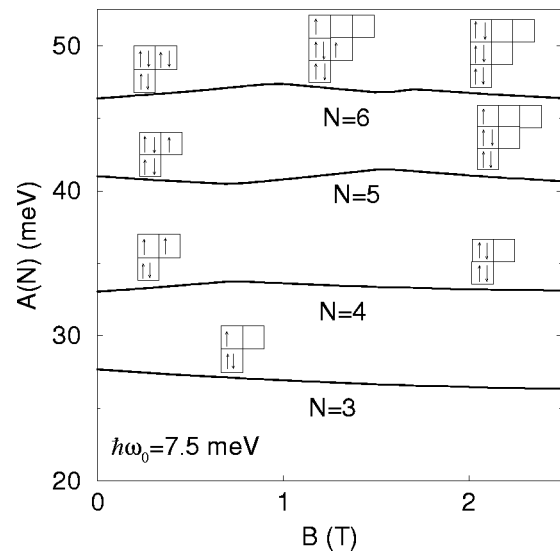


FIG. 2. Addition energy $A(N)$ vs magnetic field calculated for a realistic (3D) QD structure with confinement energy $\hbar\omega_0 = 7.5$ meV and for different electron numbers. Ground-state configurations, varying with B , are also shown.

configuration: the single-particle term favors complete shell filling, while the repulsion among parallel-spin electrons, smaller than the repulsion among opposite-spin ones, makes the configurations with maximum total spin energetically favored. Hund's rule is therefore already contained in Eq. (1) and clearly explains the physical origin of the half-shell-filling structures. In fact, adding an electron to a half-filled shell forces the double occupancy of a level: consequently, ΔA is raised by the dominant Coulomb repulsion $U_{\alpha\alpha}$ between opposite-spin electrons on the same level.

Ground-state configurations and filling rules change when a magnetic field is applied. It affects both single-particle energies and U and J integrals through the induced changes in the wave function localization. For sufficiently large values, the magnetic field energetically favors configurations with higher total angular quantum number m . This is the physical origin of the wiggles in the $A(N)$ vs B plot shown in Fig. 2 and observed in the experiments of Ref. 5.

Figure 3 shows the Coulomb and exchange integrals vs B for the first states, obtained for $\hbar\omega_0 = 7.5$ meV. For comparison, we also show the corresponding quantities calculated within the simplified 2D model. We clearly see that U integrals describing the interaction between opposite-spin electrons are smaller by a few meV in the case of 3D confinement, while the differences in the interaction between parallel-spin electrons are much smaller. This affects dramatically the energy balance which determines ground-state configurations, thus clearly showing the inadequacy of a pure 2D description of state-of-the-art QD structures.

The proposed approach shares with the Hartree-Fock (HF) method the *ansatz* for total energy, expressed as the average of the exact Hamiltonian over a single Slater determinant; the variational prescription—allowing the construction of optimal single-particle orbitals through self-consistency—is not contained in our approach. We notice, however, that in the present case the results of perturbation theory are already much closer to the *exact* results than in previous 2D calculations:¹³ this is due to the fact that Cou-

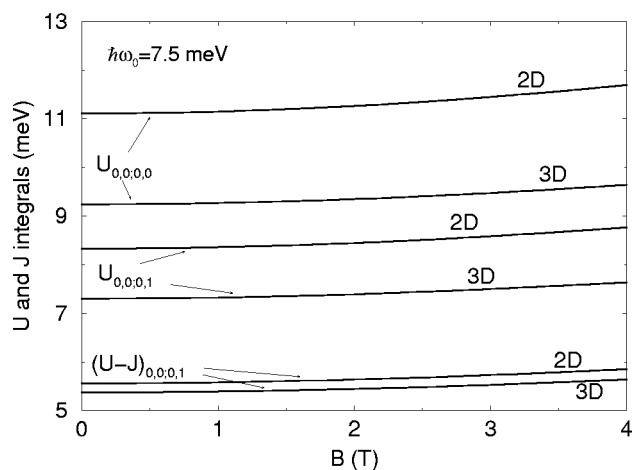


FIG. 3. Comparison between Coulomb and exchange integrals $U_{\alpha\beta}$ and $J_{\alpha\beta}$ calculated assuming a simple two-dimensional parabolic model (2D) and the corresponding three-dimensional model (3D). α and β denote the sets of radial and angular quantum numbers (n, m) for the two single-particle states involved in the interaction.

lomb integrals are artificially enhanced in 2D with respect to the realistic 3D description. Hence the role of self-consistency and the approximations of HF theory are much less relevant in 3D. Finally, we point out that the present formalism can be reduced to a Hubbard model description: expression (1) is in fact formally equivalent to the exact solution of a single-site Hubbard Hamiltonian. This is important in view of a direct extension of this approach to coupled dots.

In summary, we have proposed an effective theoretical approach for the analysis of addition spectra of quantum dots by combining a careful 3D description of electron–electron interaction with the realistic treatment of single-particle properties. By construction, the proposed scheme is ideally

suited for applications to dots with confinement potentials of arbitrary shapes and symmetries. A full 3D description of Coulomb interaction is found to be crucial in determining single-electron conductance properties of realistic nanostructures.

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