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Infrared optical versus transport spectroscopy for few-electron spherical quantum dots

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Abstract. The problem of detection of few-electron states has been studied for spherical quantum dots both with and without a donor centre. The confinement potential is assumed to be a three-dimensional spherical potential well of finite depth and range. The variational method has been applied to calculate bound states of one-, two-, and three-electron systems confined in donor-doped and undoped quantum dots. We have shown that the chemical potentials of the systems considered are significantly different, in contrast to the intraband optical dipole transition energies, which are almost indistinguishable. Therefore, the transport and capacitance spectroscopy techniques should yield clear evidence for the presence of a donor impurity in the quantum dot.

Modern nanotechnology enables one to fabricate nanometre-sized quantum dots (QDs) of different shapes. QDs of nearly spherical shape can be prepared by chemical methods [1]. These QDs are nanocrystals with almost spherical interfaces embedded in insulating matrices, e.g. Si nanocrystals in amorphous SiO₂ [2–6]. In the literature, one can find reports on QDs of spherical and ellipsoidal shape made of Ge in SiO₂ [7, 8] as well as GaAs [9] and InAs [10] in organic solvents.

Excess electrons introduced into the QD can form bound states if the confinement potential is strong enough and the dot is not too small [11–13]. The presence of a positively charged donor impurity centre in the QD creates an additional attractive Coulomb field, which leads to an enhancement of the binding [14, 15]. The bound-excess-electron states can be detected by either infrared optical spectroscopy [16, 17] or transport spectroscopy [18–21]. In optical experiments, the dipole-allowed intraband transitions are measured using far-infrared absorption; they are usually performed in a magnetic field [16, 17]. Infrared optical spectroscopy allows us to determine the energy differences between the lowest-energy states of the N -electron system confined in the QD.

In transport spectroscopy experiments, the current tunnelling through the dot is detected. Since the tunnelling current results from many single-electron tunnelling events, this technique is also called single-electron tunnelling spectroscopy [19]. The current flows between the emitter (source) and collector (drain) [19]. An additional gate electrode attached to the GaAs/InGaAs/AlGaAs vertical QD enabled the authors of [20] to carry out a very fine tuning of the nanodevice. The transport spectroscopy experiments performed on these QDs in [20] yielded clear evidence for the filling of electronic shells according to Hund's rule. The condition for single-electron tunnelling is formulated as follows [19]: the N th electron can tunnel from the emitter to the dot, which consists of the $(N - 1)$ excess electrons, if the electrochemical

potential μ_E of the emitter matches with the chemical potential μ_N of the N -electron system in the QD, i.e.

$$\mu_E = \mu_N. \quad (1)$$

The electrochemical potential $\mu_E = -eV_E + E_F$ is determined by the potential V_E of the emitter, which usually has a negative polarity, and its Fermi energy E_F . The chemical potential μ_N is defined as follows:

$$\mu_N = E_N - E_{N-1} \quad (2)$$

where E_N is the ground-state energy of N electrons confined in the QD. We note that the single electron can tunnel through the QD even for a negligibly small voltage between the leads. Then, μ_N is aligned with the Fermi energy E_F of either of the leads. According to equation (2), transport spectroscopy provides information about the ground states of both the N - and $(N - 1)$ -electron systems confined in the QD.

There exists another high-resolution spectroscopy technique applicable to QDs, which is based on the single-electron tunnelling. This is single-electron capacitance spectroscopy [21, 22], which detects changes of capacitance induced by the single-electron transitions from the lead to the dot and back. The condition for the corresponding single-electron charging of the QD is the same as that for the electron transport via the dot (cf. equation (1)).

The QD can contain a donor impurity centre [23], which leads to a modification of the few-electron energy spectra [14, 15]. The influence of the finite-size attractive impurity on the resonant tunnelling through a nanosystem was studied by the Green-function method [24]. It was shown [24] that the conductance of the doped nanostructure is strongly modified by the presence of the impurity.

In this paper, we present the results of variational calculations of the dipole transition energy and chemical potential for few-electron states in spherical QDs both with and without a donor impurity centre. These results will allow us to discuss the applicability of the optical, transport, and capacitance spectroscopy techniques to the investigations of electronic properties of QDs.

We approximate the confinement potential of the QD by a spherically symmetric potential well of finite depth and range. Therefore, the electron-confinement-potential energy $U(r)$ has the following form: $U(r) = -V_0$ for $r < R$ and $U(r) = 0$ otherwise, where V_0 is the depth of the potential well ($V_0 > 0$) and R is the radius of the QD. We denote by Ne the few-electron systems confined in the QD without the donor centre. In the present calculations, we put $N = 1, 2$, and 3 . The few-electron donor states considered, denoted by D^0 , D^- , and D^{2-} , are the systems consisting of the positive Coulomb centre at the central position in the QD and one, two, and three bound electrons, respectively.

In order to find the bound-state energy levels we have applied the variational method, which was presented in our previous papers [11, 12]. The excess electrons can form bound states in the QD if the depth and radius of the confinement-potential well are sufficiently large [11–13]. In the QD doped with the donor, the binding energy of electrons is additionally increased [14, 15]. In the bulk crystals, the D^- centre possesses exactly one bound state and the D^{2-} centre is unbound. We have demonstrated [14, 15] that—in the spherical QDs with a confinement potential of finite depth and range—the excited states of the D^- centre can also be bound [14] and the D^{2-} centre can form a bound ground state as well as bound excited states [15].

The energy levels calculated for the ground state and the first excited states enable us to determine the experimentally accessible energy ΔE of intraband dipole-allowed transitions. In this paper, we present the results for the transitions between the lowest-energy S-, P-, and D-type states, i.e. the states with the total-angular-momentum quantum number $L = 0, 1$,

and 2, respectively. Therefore, we consider the following states of the N -electron systems in the undoped and doped spherical QDs:

- (i) S-type states: 1s states for one-electron systems 1e and D^0 , $(1s)^2$ 1S states for two-electron systems 2e and D^- , and $(1s1p^2)$ 2S states for three-electron systems 3e and D^{2-} ;
- (ii) P-type states: 1p states for 1e and D^0 , $(1s1p)$ 1P states for 2e and D^- , and $(1s^21p)$ 2P states for 3e and D^{2-} ;
- (iii) D-type states: 1d states for 1e and D^0 , $(1s1d)$ 1D for 2e and D^- , and $(1s^21d)$ 2D for 3e and D^{2-} .

The calculated S–P and P–D transition energies are displayed in figures 1 and 2, respectively. Throughout the present paper, we use the effective donor rydberg R_D as the

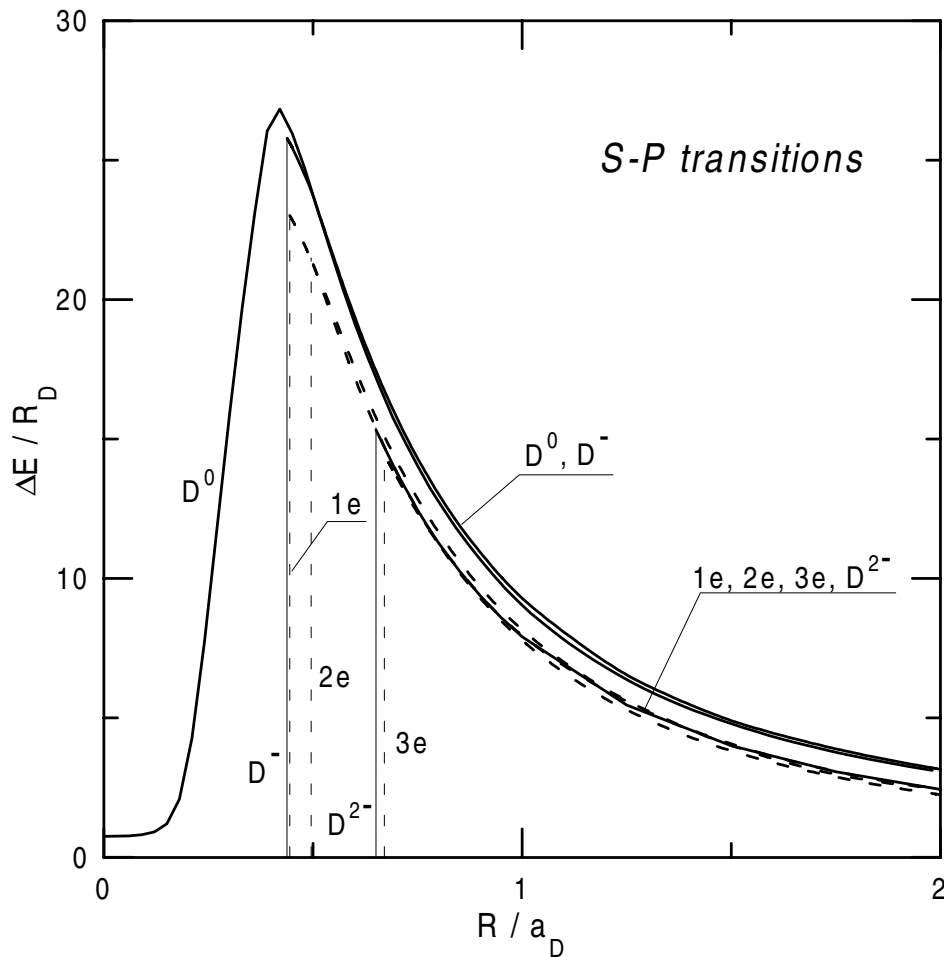


Figure 1. The energy difference ΔE between the energy levels of the P- and S-type states, listed in the text, as a function of the QD radius R . Thin vertical lines correspond to the critical values $R = R_c$: for $R < R_c$ the P-type state is unbound. Solid lines display the results for donor centres D^0 , D^- , and D^{2-} and dashed lines display those for few-electron systems 1e, 2e, and 3e confined in the QD. The effective donor rydberg R_D is the unit of energy and the effective donor Bohr radius a_D is the unit of length. The potential-well depth $V_0 = 50 R_D$ corresponds to the QD made of GaAs in the $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ matrix.

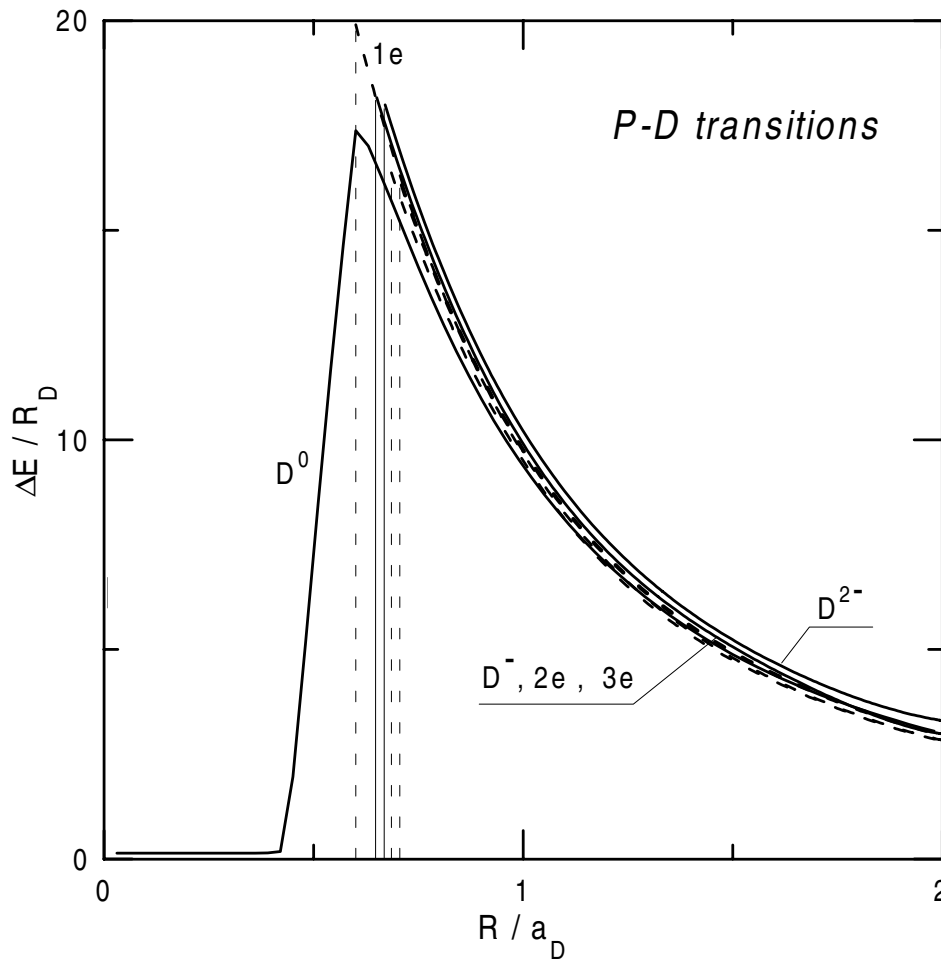


Figure 2. The energy difference ΔE between the energy levels of the D- and P-type states, listed in the text, as a function of the QD radius R . Thin vertical lines correspond to the critical values $R = R_c$: for $R < R_c$ the D-type state is unbound. Solid (dashed) lines show the results for the donor-doped (undoped) QDs. The units and the value of V_0 are the same as for figure 1.

unit of energy and the effective donor Bohr radius a_D as the unit of length, and take the values of material parameters typical for the GaAs QD in the $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ matrix, i.e. $R_D = 5.23$ meV, $a_D = 10.4$ nm, and $V_0 = 50 R_D$. Figure 1 shows that the S–P transition energies are almost indistinguishable for the following two groups of systems: $\{D^0, D^-\}$ and $\{1e, 2e, 3e, D^{2-}\}$. Figure 2 shows that the P–D transition energies are very close to each other for all the systems considered. For example, for $R = a_D$ the S–P energy differences do not exceed $0.2 R_D$, i.e. ~ 1 meV, for $\{D^0, D^-\}$ and $0.4 R_D$, i.e. ~ 2 meV, for $\{1e, 2e, 3e, D^{2-}\}$ (cf. figure 1). For $R = a_D$ the six P–D transition energies take on the values from the interval of the width of $\sim 0.8 R_D$, i.e. ~ 4 meV (cf. figure 2). If the QD radius decreases, both the S–P and P–D transition energies increase and take on the largest values at the QD radius, at which the higher-energy state becomes unbound (cf. thin vertical lines in figures 1 and 2). The D^0 centre is the only system for which the excited states are bound for all values of the QD parameters. The S–P and P–D transition energies for the D^0 exhibit a typical behaviour for the quantum-confined

systems, i.e. they reach the sharp maxima for the QDs of small size and tend to the corresponding bulk-crystal values if $R \rightarrow 0$.

The results reported in figures 1 and 2 suggest that—for the few-electron systems confined in the spherical QDs—unambiguous interpretation of the infrared optical spectroscopy data is rather difficult and may not even be possible. In figure 1, the transition energy differences between one-electron systems $1e$ and D^0 as well as between two-electron systems $2e$ and D^- are of the order of R_D . This means that the infrared optical spectroscopy could provide evidence for a presence of the donor impurity in the QD. However, the D^0 and D^- centres will be hardly distinguishable from each other in the intraband transition measurements.

Figure 3 displays the results of the present calculations of the chemical potential for the few-electron systems confined in the spherical QD with and without the donor centre. As the negative value of the chemical potential corresponds to a formation of a bound state of the N -electron system [13], we see from figure 3 that systems $1e$, $2e$, $3e$, and D^{2-} become bound if the QD radius is sufficiently large [11, 15]. For each number of electrons the chemical potential takes on lower values for the dot with the donor centre than for the undoped QD, which demonstrates the enhancement of the electron binding by the positive donor centre. Moreover, we observe clear differences between the chemical potentials for the systems considered. In particular, for each N -electron system the chemical potential for the QD with the donor

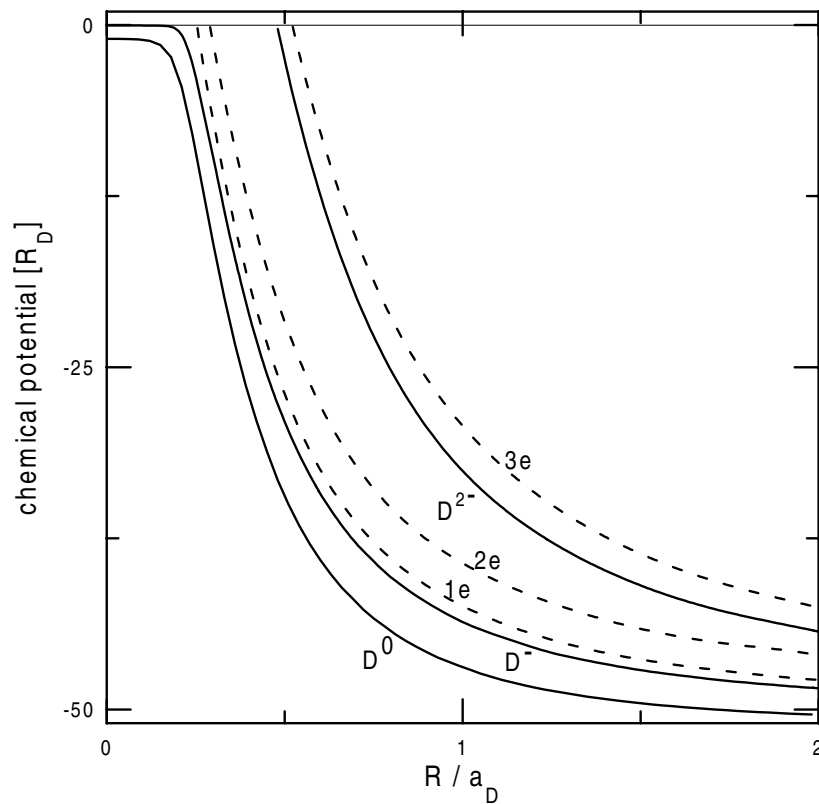


Figure 3. The chemical potential for the few-electron systems confined in the QD, with (solid curves) and without (dashed curves) the donor centre, as a function of radius R for $V_0 = 50 R_D$. The units are the same as for figure 1.

impurity considerably differs from that for the QD without the donor. The corresponding differences are of the order of $2\text{--}4 R_D$, i.e. $10\text{--}20$ meV. According to equation (1), the chemical potential determines the condition for the single-electron tunnelling via the QD. Due to the pronounced differences of the chemical potentials, the single-electron tunnelling will occur at clearly different energies. This leads to the conclusion that the spectroscopy techniques based on the single-electron tunnelling should enable the detection of the presence of the donor centre in the QD as well as determination of the number of confined electrons. We note that the D^- centre has been found in doped quasi-two-dimensional GaAs/AlGaAs quantum wells in resonant tunnelling experiments [25]. In spherical Si/SiO₂ QDs, resonant tunnelling via bound-electron states was observed [2, 4, 5].

The near independence of the intraband dipole transition energy of the number of electrons confined in the QD resembles a similar property resulting from the generalized Kohn theorem [26–28]. It was shown [26–28] that the dipole transition energy for the N -electron system confined in the QD is independent of the number of electrons, i.e. the electron–electron interaction, if the confinement potential is parabolic. In this case, the N -electron Hamiltonian can be exactly separated in the centre-of-mass and relative coordinates. Therefore, in the dipole approximation for the electron–photon interaction, the long-wavelength radiation is only coupled to the centre of mass, for which the cyclotron-resonance-type transitions occur with equal energies. The indistinguishability of the dipole transitions, obtained in the present paper, results from an approximate cancellation of the Coulomb interaction contributions to the lowest energy levels for the QDs [11]. Therefore, the dipole transition energies take on nearly the same values. The influence of the donor centre shifts the S–P transition energies for the D^0 and D^- states to higher values. However, the S–P transition energies for both of these centres still take on very close values, which prevents us from distinguishing between the presence of the D^0 and D^- centres when using infrared optical spectroscopy. We note that the near independence of the number of electrons obtained in this paper for the intraband transitions results from the almost equal separations between the energy levels, i.e. this property is also valid beyond the dipole approximation for the electron–photon interaction.

Condition (1) for the single-electron tunnelling is determined by the energy needed to add a single electron to the dot, i.e. the chemical potential. We have shown (cf. figure 3) that the chemical potentials for the few-electron systems considered differ considerably from each other. So, we expect the transport (capacitance) spectroscopy technique to provide pronounced, clearly separated, current (capacitance) peaks. Therefore, these techniques should be well suited for performing spectroscopic studies of the QDs. Additionally, they are sensitive to the presence of the donor impurity centre in the QD.

The present conclusion regarding the electron-number independence of the dipole transition energy is valid for the donor located in the central region of the spherical QD. There arises a question of how much the present results depend on the position of the donor in the QD. In general, this is a rather difficult problem, which cannot be solved with the use of the Slater-type orbitals [11, 12]. On the basis of the present results, we can however discuss this problem. According to figures 1 and 2, the dipole transition energies for the few-electron systems confined in the QDs without the donor are approximately equal to each other for a QD of arbitrary size. The donor impurity introduced into the QD at the central position has the largest influence on the transition energies, since the difference in electron probability density between the S and P (P and D) states is largest at this position. The present results show that the presence of the on-centre donor leads to a small perturbation of the transition energies (cf. the small differences between the solid and dashed curves in figures 1 and 2). This perturbation is weak, since—in the strong- and intermediate-confinement regimes—the effect of the electron-number independence of the dipole transition energy is mainly caused

by the confinement potential of the QD. Moving the donor from the dot centre towards the QD boundary results in a decreasing difference in electron density at the donor site between the states involved in the transition. Therefore, for all the off-centre positions of the donor the influence of the donor on the dipole transition energy should be smaller than at the on-centre position considered in the present paper. We note that the calculated dipole transition energies exceed several times the effective donor rydberg, R_D , which can be treated as a measure of the influence of the donor on the spectra. We cannot however exclude the possibility of some noticeable effect of the donor position for the QDs with weak confinement potentials.

We have presented numerical results for $V_0 = 50 R_D$, which corresponds to the GaAs/Al_{0.3}Ga_{0.7}As QDs. Due to the use of effective donor units, the same conclusions are also valid for the Si/SiO₂ QDs, for which the effect of the electron confinement is much stronger ($V_0 \simeq 100 R_D$).

The results of the present paper allow us to conclude that the donor impurity, which can be unintentionally introduced into the nanostructure, does not disturb the intraband optical spectrum of the spherical QD. However, the transport data should be strongly modified in comparison to those for the undoped QDs. This leads to the possibility of detection of residual donors in the QDs by the transport spectroscopy technique.

In summary, we have studied the conditions for performing successful spectroscopic measurements on the QDs. We have found that the intraband optical dipole transitions are hardly distinguishable for the few-electron QDs both with and without the donor centre, which limits the applicability of infrared optical spectroscopy to studying the QDs. In contrast, distinct differences in the chemical potentials between the various few-electron systems in the QDs should enable us both to detect the donor states in the QD and to ascribe each of the tunnelling current peaks to different few-electron systems. Therefore, unlike the case for the infrared optical spectroscopy, the interpretation of the transport and capacitance spectroscopy data should be unambiguous.

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