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2003 J. Phys.: Condens. Matter 15 1377

(http://iopscience.iop.org/0953-8984/15/9/301)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) 1377–1385 PII: S0953-8984(03)55415-7

Resonant photoionization absorption spectra of spherical quantum dots

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Received 31 October 2002
Published 24 February 2003
Online at stacks.iop.org/JPhysCM/15/1377

Abstract

We study theoretically the mid-infrared photon absorption spectra due to bound–free transitions of electrons in individual spherical quantum dots. It is established that change of the dot size in one or two atomic layers or/and number of electrons by one or two can change the peak value of the absorption spectra in orders of magnitude and energy of absorbed photons by tens of millielectronvolts. The reason for this is the formation of specific free states, called resonance states. Numerical calculations are performed for quantum dots (QDs) with radius varying up to 200 Å, and one to eight electrons occupying the two lowest bound states. It is supposed that realistic QD systems with resonance states would be of much advantage to design novel infrared QD photo-detectors.

1. Introduction

Quantum dots (QDs) have attracted much interest as a type of structure for optoelectronics due to their advantages over quantum wells [1]. One advantage is the capability to absorb and emit radiation of different polarizations, especially normal incidence radiation. On the other hand, it is known that electronic states in QDs can be very sensitive to the size quantization effect and modification of the effective potential in the dots that can affect dramatically optical features of QDs. This requires careful investigation for specific QD systems and stimulates seeking and engineering the most effective QD systems.

Photon absorption properties of QDs have been investigated experimentally in many works [2–11]. A number of theoretical works were devoted to the investigation of electronic structure of QDs and QD arrays [12–16], and optical transitions and absorption spectra of QDs due to electron transitions between bound states (bound–bound transitions) as well as

between bound and continuous (or free) states (bound–free or photo-ionization transitions) of electrons [17–25]. Usually the absorption spectra were calculated for a specific QD system with fixed shape and size of QDs and a set of specific parameters. However, it has still not been investigated how different the absorption spectra for QDs of different sizes and different electron occupations are. Obviously the levels in QDs are pushed up with decrease of the QD size. Besides, electron–electron interaction also pushes up the QD levels. The highest bound levels with negative eigenenergies can transform into free levels with positive energy. In fact, there is an important combination of the QD size and number of electrons in the QD. As a result of the combination effect the absorption spectra can be expected to experience essential modification, especially when the resonance is met [25]. Note that, in [26], the emission spectrum of a QD was found to be a sensitive function of the number of excitons in the excitonic artificial atoms (see also [10] and [21]). Lack of this knowledge can create difficulties in interpretation of experimental results. Recently obtained experimental data [7], which showed that the absorption grew with growing number of electrons in the dot, forced the authors to make a puzzling statement about the type of the confinement in the considered QD system: whether the confinement is of quantum wire type or QD type.

The aim of this work is to investigate in detail how the photo-ionization absorption spectrum is modified due to change of the QD size and the number of electrons in the QD. In this work we assume the number of electrons per QD to be a fixed parameter. In such a way we do not restrict ourselves to a specific QD–array–matrix system, and a specific way of loading the QDs with electrons. This allows us to simulate theoretically a lot of effects of the dot size and electron occupation on the absorption spectrum. The obtained results give a possibility for engineering appropriate QD systems to enhance absorption in QD photo-detectors.

As a model for calculations an individual spherical quantum dot (SQD) is utilized. From the theoretical point of view such a model allows us to avoid serious complications because of the specific geometry of QD systems, e.g. radiation polarization dependence. In addition, there is no depolarization effect in an individual SQD due to spherical symmetry: the optical transitions of electrons can only occur between two states whose angular momenta differ by unity, while the electron–electron interaction does not influence the angular momentum of the electron system. Thus, in an individual SQD the electron–electron interaction is represented fully by static interaction. No external applied electric field is considered. In our approach we consider bound–free optical transitions of electrons from the first and second lowest bound states of an SQD depending on the dot size and the number (one to eight) of localized electrons. Note that due to technological progress the model of SQDs has become important for describing realistic QD systems [3] (see also [24]).

Numerical calculations are performed for GaAs/$Al_{0.3}Ga_{0.7}$As SQDs. Since there is no strain in GaAs/$Al_{0.3}Ga_{0.7}$As QDs, it simplifies the model for calculations essentially. Besides, the numerical results for GaAs/$Al_{0.3}Ga_{0.7}$As SQDs are very illustrative because their free electronic states are very sensitive to the size quantization and to the influence of the electron–electron interaction. It should be added that good quality GaAs/AlGaAs QDs of different sizes can be manufactured by means of templates and wet etching (see, e.g., [1] and [27]).

2. The model

We consider a single SQD of radius a, which is a spherical potential well with depth U_0, embedded in a bulk medium (matrix) that serves as a barrier. The states of one electron in an SQD are described by the spherical eigenfunctions $\Psi_{n,l,m}(\boldsymbol{r}) = R_{n,l}(r)Y_{l,m}(\theta, \varphi)$ for the bound states (whose eigenenergy $E < 0$) and by $\Psi_{E,l,m}(\boldsymbol{r}) = R_{E,l}(r)Y_{l,m}(\theta, \varphi)$ for the free states ($E > 0$). Here n, l and m are the main, azimuth and magnetic quantum numbers,

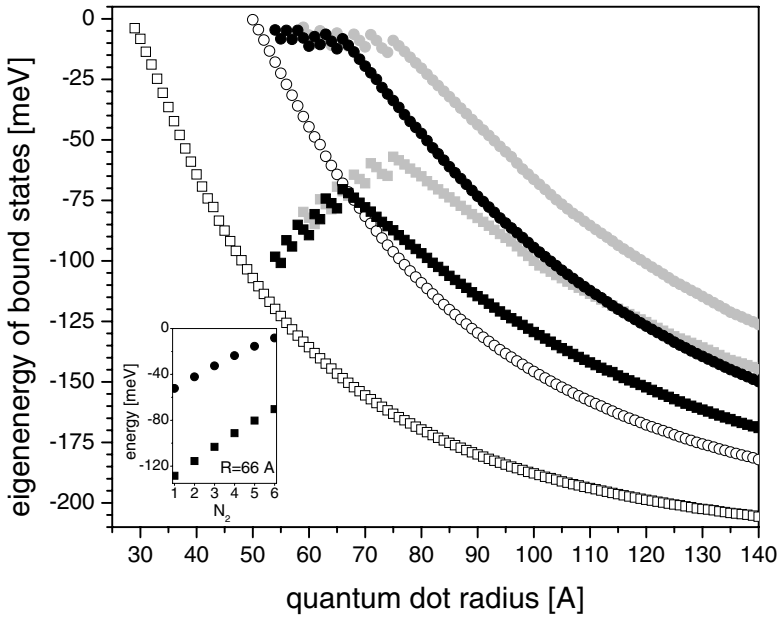

Figure 1. Radius dependence of the eigenenergies of the first (squares) and second (circles) lowest bound states of the GaAs/$Al_{0.3}Ga_{0.7}As$ SQD. Empty symbols represent the one-electron approximation, filled light grey ones for the Hartree approximation, filled black ones for the LDA. The eigenenergies of the states with two electrons in the dot are about 3 meV bigger than those for one electron. The inset presents the change of the eigenenergies of a 66 Å radius QD for different numbers of electrons, N_2, in the (1; 1) state within the LDA.

respectively, and $\boldsymbol{r}$ the radius vector from the centre of the SQD. In this work no magnetic field is considered and that is why the total spin of the system and the magnetic quantum number are conserved. Further we denote the bound states by $(n; l; m)$ and free states by $(E; l; m)$.

In the following we consider SQDs occupied by N electrons ($N > 1$). It should be emphasized that no other charges but the electrons in the dot are considered. The electron–electron interaction only affects the radial part of the eigenfunctions.

In the case of many electrons, $N \gg 1$, the self-consistent mean field approach is applied to take into account the electron–electron interaction that modifies the potential profile in the SQD and, hence, the eigenstates (see, e.g. [17–19]). This approach is based on the assumption that the contribution of one electron is small. Note that it is the value of the confining potential, mainly U_0, that determines the accuracy of the obtained results. Even for small N the mean field approach can provide rather good accuracy at rather big U_0. It is the sufficiently big value of U_0 that provides the spherical symmetry of the wavefunctions of electrons in SQDs. We assume that the SQDs meet these conditions. (Our numerical results support this assumption: the empty-square curve and the black-square curve in figure 1 match each other rather well at dot radii of about 55 and 66 Å.) This simplifies calculations and lets us get approximate results for systems with $N > 2$ within the approach for $N \gg 1$. The exchange–correlation interaction effect becomes essential for QDs with small numbers of electrons. We take this effect into account within the local density approximation (LDA) exploiting a standard parametrized form (originally by [28]) for the exchange–correlation potential energy (see, e.g., [29]). Furthermore the approach taking into account both the direct Coulomb interaction and exchange–correlation interaction is called the LDA, while the approach taking into account only the direct Coulomb interaction is referred to as the Hartree one.

When there are only two electrons, $N = 2$, in an SQD a different approach is utilized. The initial state of two electrons in the lowest bound state is described in a standard way including the exchange–correlation effect (see, e.g., [30]). After having absorbed a photon the two-electron system is in its final state: one of the electrons remains in the lowest bound state in the dot while another electron is in a free state. To find the wavefunction and the energy of the final state we use the following reasonable approach to take into account the effect of the Coulomb interaction between the localized and free electrons. The interaction effect on the bound state is assumed to be negligible due to strong localization in the dot. Thus, the bound state is just the state of one electron in the SQD governed by the one-electron Hamiltonian. However, the free electron is assumed to experience the potential of the SQD with one electron in it. The advantage of this approach is that it allows us to separate the Hamiltonians describing the bound and the free electrons in the Hamiltonian driving the two-electron system. The dipole moment of the two-electron system is $\boldsymbol{d} = -e(\boldsymbol{r}_1 + \boldsymbol{r}_2)$, where $\boldsymbol{r}_i$ describes the ith electron.

Developing in a standard way we get the cross section for photon absorption due to electron transitions from the ith bound state, $(n_i; l_i; m_i)$, with energy E_i to any free state, $(E_f, l_f; m_f)$, with energy E_f in an SQD as the following:

$$\sigma_i(\omega) = \frac{8\pi e^2 \omega}{nc} \int \frac{\mathrm{d}E_f \, |z_{if}|^2 \Gamma_{if} N_{if}}{(E_f - E_i - \hbar\omega)^2 + \Gamma_{if}^2}, \tag{1}$$

where $|z_{if}|^2 = |\int \mathrm{d}\boldsymbol{r}\, \Psi^*_{E_f,l_f,m_f}(\boldsymbol{r}) z \Psi_{n_i,l_i,m_i}(\boldsymbol{r})|^2$ with $l_f = l_i \pm 1$ and $m_f = m_i$. For simplicity in our approximation we reasonably neglect a difference between the values of $|z_{if}|^2$ for the transitions from states (1; 1; 0) and (1; 1; $\pm$1) due to different θ-dependence of their wavefunctions. Further we neglect the magnetic quantum number and denote the states by $(n; l)$ and (E, l). $N_{if} = N_i - N_f$ where N_i and (N_f) are the numbers of electrons in the initial and final states respectively. We make a reasonable assumption that the final state is non-occupied and $N_{if} = N_i$. The broadening of the eigenenergy of the ith bound state induced by a finite time for an electron to be in the state is represented by Γ_{if}. In this work we treat Γ_{if} as a phenomenological parameter. For our numerical calculations we set $\Gamma_{if} = \Gamma =$ constant and take a reasonable value for Γ. n is the refractive index of the medium surrounding the dot and c is the light speed in vacuum. Note that $\sigma_i(\omega)$ is proportional to the number of electrons in the initial state, N_i. The cross section of a QD is $\sigma(\omega) = \sum_i \sigma_i(\omega)$.

3. Numerical results and discussion

Numerical calculations are performed for GaAs/$Al_{0.3}Ga_{0.7}$As SQDs. The parameters used are $U_0 = 227.9$ meV, $m_d = 0.066\, m_0$, $m_b = 0.092\, m_0$, m_0 is the free electron mass, $\epsilon_d = 13.18$, $\epsilon_b = 12.24$, the refractive index of the matrix $n = 3.25$ and $\Gamma = 4$ meV.

The important peculiarities of the ionization absorption spectra of SQDs presented in this work are based on the features of the energy structure of the dot, and first of all the formation of the resonance states among the free states. A resonance state is the state which combines features of the bound and free states.

(a) The most valuable part of its wavefunction remains in the region of the dot and resembles the eigenfunction of a certain bound state.
(b) A resonant state belongs to the continuum spectrum of states with positive energy, i.e. it extends to infinity.

We call such states resonance ones because the photon absorption due to transitions from the bound states to these states experiences peak values (see the Fano resonance [25]). The resonance states of a dot exist for any number of electrons in the dot. The resonance states are

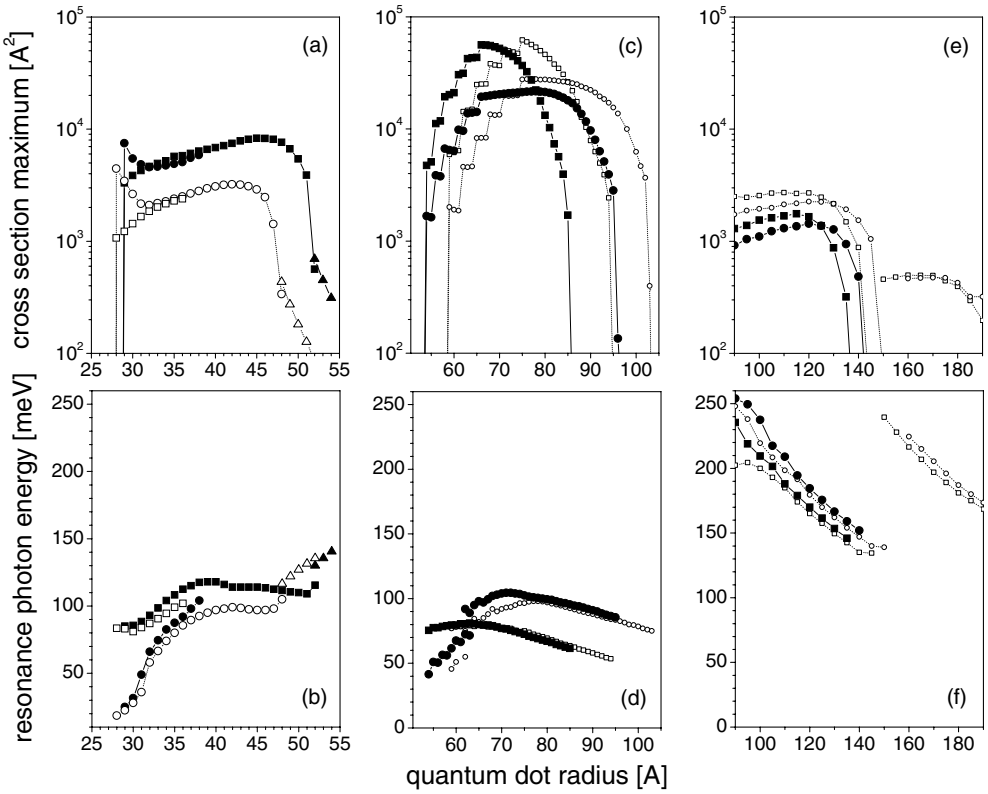

Figure 2. Maximum values of the cross section (a), (c), (e) and the resonance photon energies (b), (d), (f) of the spectra as functions of the QD radius. (a) and (b) describe the transitions from the (1; 0) state. Squares, circles and triangles represent different peaks due to transitions to different final resonance states. Empty symbols stand for the one-electron approach, filled black ones for the two-electron approach. (c) and (d) describe $(1;1) \rightarrow (1;2)_{\mathrm{res}}$ (squares) and $(1;1) \rightarrow (2;0)_{\mathrm{res}}$ (circles) transitions. Filled black symbols represent the LDA, empty ones for the Hartree approximation. As many electrons as possible occupy the (1; 1) state. (e) and (f) describe $(1;1) \rightarrow (2;2)_{\mathrm{res}}$ (squares) and $(1;1) \rightarrow (3;0)_{\mathrm{res}}$ (circles) transitions. Filled black symbols stand for the LDA, empty ones for the Hartree approximation. $N_2 = 6$. This kind of absorption is effective due to the electron–electron interaction.

formed from the bound states when the bound states are pushed out of the discrete spectrum of the dot eigenstates to the continuum energy spectrum with decreasing dot size and/or increasing electron–electron interaction in the dot. It is convenient to denote the resonance state as $(n, l)_{\mathrm{res}}$ if it is originated from bound state (n, l). Both $(n, l)_{\mathrm{res}}$ and (n, l) possess the same symmetry. The sequence of the resonance states, $(n, l)_{\mathrm{res}}$, is the same as the sequence of the corresponding bound states, (n, l) (see figure 2 in [17]).

We consider the cases when only one or two lowest bound states, (1; 0) and (1; 1), of an SQD are occupied by one to eight electrons. The location of states (1; 0) and (1; 1) in SQDs with different radii, R, and different numbers of electrons is presented in figure 1. State (1; 1) can only be occupied by one electron at $R = 54$ Å, and only at $R \geqslant 66$ Å can the state be occupied by a maximum $N_2 = 6$ electrons. The reason is that loading each additional electron in the dot induces a sharp increase of the potential energy of the electrons. This is why the dependence of the eigenenergy upon R is sawlike (and the dependences in figures 2(c) and (d)

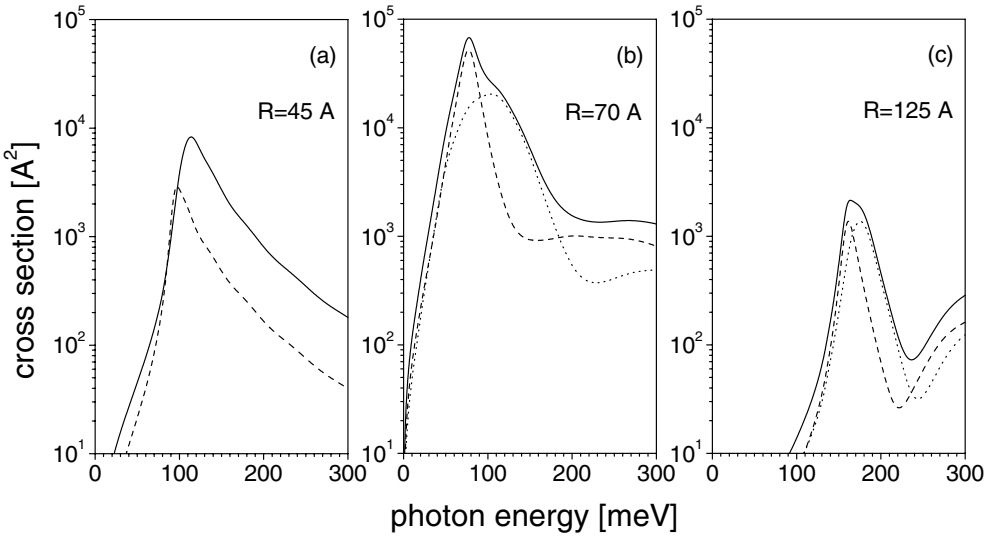

Figure 3. Spectra for the QDs of different radii. (a) Transitions $(1; 0) \rightarrow (1; 1)_{\text{res}}$. The solid (dashed) curve represents the two- (one-) electron case. (b) The dashed (dotted) curve represents $(1; 1) \rightarrow (1; 2)_{\text{res}}$ ($(1; 1) \rightarrow (2; 0)_{\text{res}}$) transitions. The solid curve stands for their sum. (c) The dashed (dotted) curve represents $(1; 1) \rightarrow (2; 2)_{\text{res}}$ ($(1; 1) \rightarrow (3; 0)_{\text{res}}$) transitions. The solid curve stands for their sum.

are steplike). Each step of the saw corresponds to loading one more electron. Numerical calculations show that an electron can occupy the third bound state in the dot if $R \gtrsim 95$ Å.

The most considerable cross section is expected, and our numerical calculations prove it, for the transitions from a bound state $(n; l)$ to its nearest (in the energy scale) resonance states. This determines the radius interval for the biggest absorption associated with the transitions from $(n; l)$. The smallest R of this interval corresponds to the condition of the formation of $(n; l)$; for smaller R there is no absorption of this type at all. The biggest R corresponds to the condition of disappearance of the nearest resonance states, i.e. the formation of the corresponding bound states. For bigger R the absorption happens due to the transitions to further free (resonance) states and, thus, is supposed to be considerably weaker, as follows from the oscillator strength sum rule. The oscillator strength for transitions from $(n; l)$ to its nearest resonance states is considerably larger than that for the transitions to further resonance states. (See also figure 3 in [17] which represents well the sum rule for the bound–bound transitions; the case of the bound–resonance state transitions is similar to this.)

The electrons in the first bound state $(1; 0)$ are allowed to absorb photons and transit to free states $(E_f, 1)$, with the most considerable cross section expected for $(1; 0) \rightarrow (1; 1)_{\text{res}}$ transitions. Figure 2(a) shows that SQDs with one or two electrons eventually absorb photons only if their radius lies in the small interval from about 30 to 55 Å. Figure 2(b) displays that the resonance photon energy also varies considerably, however being mainly around 110 meV. The calculations show that the highest and narrowest absorption peak occurs for the dot radius near 45 Å, with the maximum cross section of almost 10^4 Å^2; its spectrum is displayed in figure 3(a). For 30 Å $< R <$ 38 Å an SQD actually absorbs photons of about 130 meV energy interval, with two peaks distinguished on the spectra (see figures 2(a) and (b)).

The electrons in the second bound state $(1; 1)$ can transit with photon absorption to free states $(E_f, 2)$ and $(E_f, 0)$. Figures 2(c)–(f) show that an SQD with one to six electrons in the

second bound state (and with two electrons at the first state) can absorb photons effectively if the dot radius is in the interval from about 55 to 150 Å. The absorptions differ drastically in two sequential parts of that radius interval which are approximately 55–100 Å (the effective transitions $(1;1) \rightarrow (1;2)_{\mathrm{res}}$, $(1;1) \rightarrow (2;0)_{\mathrm{res}}$, $(1;1) \rightarrow (2;2)_{\mathrm{res}}$ and $(1;1) \rightarrow (3;0)_{\mathrm{res}}$) and 100 to 150 Å (the effective transitions $(1;1) \rightarrow (2;2)_{\mathrm{res}}$ and $(1;1) \rightarrow (3;0)_{\mathrm{res}}$). (It should be noted that the effectiveness of the absorption for 100 Å $< R <$ 150 Å is caused by the static electron–electron interaction. In the one-electron approximation there is no absorption of this kind.) The resonance energy lies in completely different intervals for the first radius interval and for the other one: approximately 60–100 and 140–240 meV, respectively. The spectra of SQDs are a composition of two peaks which can be distinguished when 60 Å $< R <$ 100 Å, and cannot be distinguished for $R >$ 100 Å due to their rather close resonance energies. The maximum cross section is also radically different in the different radius intervals: almost 10^5 Å^2 if $R \approx$ 75 Å, and about 10^3 Å^2 if $R \approx$ 130 Å. The most pronounced spectra are displayed in figures 3(b) and (c). It should be noted that an SQD with 70 Å $< R <$ 100 Å can absorb photons of any energy from the interval 170 to more than 300 meV with approximately the same cross section of about 10^3 Å^2 due to $(1;1) \rightarrow (2;2)_{\mathrm{res}}$ and $(1;1) \rightarrow (3;0)_{\mathrm{res}}$ transitions as in figure 3(b), which is merely due to the static electron–electron interaction (compare figures 3(b) and (c)).

The results show that the shape of the peaks, being very different for the considered compositions of radius and number of electrons, has a common feature: the bigger the radius is the narrower the peaks become (within the considered radius intervals).

Our calculations show that static electron–electron interaction changes absorption spectra drastically. It is the direct Coulomb interaction that (i) increases the peak heights, (ii) shifts the position of the peaks to higher energy, (iii) broadens the peaks and (iv) makes the peaks with higher resonance photon energy more effective. Note that in part these features manifest themselves in the experimental results in [5]. Besides, the exchange–correlation interaction affects the photon absorption considerably by decreasing the effect of direct Coulomb interaction (see figure 2, especially (e) and (f) which show that the exchange–correlation interaction completely suppresses the absorption for $R >$ 150 Å due to $(1;1) \rightarrow (3;2)_{\mathrm{res}}$ and $(1;1) \rightarrow (4;0)_{\mathrm{res}}$ transitions). It should be emphasized that it is because of the static electron–electron interaction that an SQD with $R >$ 60 Å and electrons in the second bound state can absorb effectively photons of energy more than 140 meV.

According to equation (1) the peak value should decrease if Γ increases. This decrease is different for different widths of the peaks. For narrow peaks the decrease is almost as Γ^{-1}, while for wide peaks it is much weaker than that, which is confirmed by the numerical calculation of spectra. An increase of N is found to result in the broadening of the absorption peak, that is why the effect of N and the effect of Γ on the spectra are not independent. Every specific case needs a detailed theoretical investigation of these effects to correctly interpret experimental data like those in [9].

Some remarks should be made on comparison of our numerical results with those known in the literature. First of all, our results are consistent with those on the Fano resonance in QDs [25]. The focus of this work is the effect of size quantization and electron–electron interaction on the wavefunctions and ultimately on the inter-state coupling for optical transitions. Thus, it is the case that was avoided in [20] and where the cross section was calculated as a few Å^2. On the other hand, we found the coupling strength between the bound and resonance states to be similar to bound–bound coupling. At this end our values of the cross section match very well with the results of [17]. The present results explain the reason for the small value for cross section in [19]. (Note that we utilize the same SQDs as in [17] and [19].) In [24] the calculated bound–bound absorption matrix elements were found to vary

by a few orders of magnitude when the wavefunctions of the electrons in SQDs were changed by modelling the external parabolic potential. The experimentally obtained values for the cross section lie in a wide interval, being in many works a few hundred Å^2 and reaching a few thousand Å^2 (e.g., [10]). It is also worth mentioning the results of Sheng and Leburton [22] where the coupling between the bound states changed by almost an order of magnitude when the wavefunctions of the states were modified by interdot interaction and a strong external electric field.

In conclusion, in this work we present an investigation of the features of the photon absorption due to bound–free transitions in an *individual* GaAs/$Al_{0.3}Ga_{0.7}$As SQD of different radii (up to 200 Å) and different electron occupations (up to eight electrons per dot). It is shown that the combination of the QD radius and number of electrons in the dot is a driving factor determining the strength and shape of the absorption spectra. The most exciting result is that change of QD size by one or two atomic layers or/and change of the number of electrons in the dot by one or two can result in a change of absorption cross section by orders of magnitude and a change of absorption energy by tens of millielectronvolts. The reason for this is found to be the formation of the resonance states.

The obtained results show that detailed calculations of realistic QD systems are required to design photo-detection devices based on QDs. Moreover, we suppose that realistic QD systems with resonance states would be of much advantage to design novel infrared QD photo-detectors. We can also expect the resonant absorption to be more pronounced for systems of QDs of symmetrical shapes. It is worth mentioning the importance of designing a QD itself as well as its surroundings to get the most effective absorption.

Acknowledgment

One of the authors (VB) wishes to thank M Załużny for fruitful discussions.

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