Excitonic trions in single and double quantum dots

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Excitonic trions in quantum dots with Gaussian confinement potential are studied by the variational method. We show that the photoluminescence line associated with the negative trion is always shifted towards lower energies with respect to the exciton line, and that this shift is larger for smaller dots. The qualitative behavior of the photoluminescence line of the positive trion is the same only in dots which resemble quantum wells or quantum wires. In other dots the size dependence of the positive-trion shift is more complex. In particular, we show that the order of the positive-trion line and the exciton line can be changed. The present approach has been generalized to the trion states in vertically coupled dots. We discuss the trion energy-level splitting induced by the coupling between the dots, as well as the relation between the photoluminescence-line shift with the binding energy of the trion in the double quantum dots.

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I. INTRODUCTION

The excitonic trions are charged exciton complexes composed of either two holes and one electron (positive trion X_2^+) or two electrons and one hole (negative trion X^-). These complexes have been a subject of an extensive theoretical[1–17] and experimental study.[18–24] In bulk semiconductors the excitonic trions are stable against dissociation into an exciton and a free carrier. However, their binding energies in bulk materials are very small. The confinement of the trions in two-dimensional quantum wells increases these binding energies by an order of magnitude.[2] The enhancement of the trion binding energy in quantum wells allows for experimental observation[18] of this complex. The excitonic trions confined in quantum dots (QD's) are observed in charge-tunable nanostructures.[23,24]

The first observations of QD-confined charged excitons were performed on ensembles of QD's.[23] The results of these experiments were perturbed by the inhomogenous broadening caused by the variation of the sizes of the dots. Recently, measurements of the photoluminescence (PL) spectra of charged excitons from a single self-assembled QD were reported.[24] This technique, which allows for selection of a single dot as a signal source, may be used in order to determine the dependence of the trion PL lines on the size and geometry of the dots. A theoretical study of this dependence will be useful in the interpretation of the experimental data. The aim of the present paper is to furnish this study. The present work is also motivated by the measurements of the exciton spectra in coupled self-assembled QD's.[25] The present results for the splitting of the trion-energy levels induced by the coupling between the dots should be useful for identification of the trion-related lines in the PL spectra of coupled dots. The present paper is a continuation of our previous research on neutral[26] and charged[15] excitons. In Ref. 15 we studied the exciton trions in spherical quantum dots with square-well confinement potential. Here, we generalize this study to more realistic cylindrical symmetry. In Ref. 26 we studied the neutral-exciton spectra in vertically coupled QD's. The present paper extends the previous work[26] to the problem of charged excitons.

The exciton PL peak, as well as peaks corresponding to exciton complexes are blueshifted (i.e., shifted towards higher energies) by the confinement in quantum wells, wires, and dots.[27] Here, we present a theoretical study of the excitonic trions confined in single and double QD's. In particular, we are interested in the confinement-induced shift of the excitonic trion PL line with respect to the exciton line. This shift is a basic quantity of experimental interest for the excitonic trions. The excitonic trion PL line is shifted with respect to the exciton PL line by[15]

$$S_{X_2^+}=h\nu_X-h\nu_{X_2^+}=E_X+E_h-E_{X_2^+} \tag{1}$$

for the positive trion and by

$$S_{X^-}=h\nu_X-h\nu_{X^-}=E_X+E_e-E_{X^-} \tag{2}$$

for the negative trion. If $S>0$, the energy of the trion PL line is smaller than the energy of the exciton line. Then, we speak about the redshift of the trion PL line with respect to the exciton line. If the energy difference is negative $(S<0)$, the trion line is blueshifted with respect to the exciton line. We will shortly refer to energy differences (1) and (2) as the positive-trion energy shift $S_{X_2^+}$ and the negative-trion energy shift S_{X^-}. In bulk materials, quantum wells and quantum wires, energy shifts (1) and (2) can be identified with the binding energy of the trions. However, it is not the case for QD's[15] (cf. a discussion of the binding energy of D^- center in the QD[28]).

This paper is organized as follows. In Sec. II we present the model of the single QD, introduce the variational wave functions for the exciton and excitonic trions, and discuss the influence of the shape and geometry of the QD on the trion PL line shifts. In Sec. III we generalize the approach of Sec. II to the trions in double QD's. Section IV contains conclusions and the summary.

II. EXCITONIC TRIONS IN A SINGLE QUANTUM DOT

A. Confinement potential and variational wave functions

In the present paper we adopt the Gaussian model of the confinement potential,[29] which was successfully applied[26] to a quantitative interpretation of the exciton spectra[25] in $In_xGa_{1-x}As$ self-assembled QD's. The confinement potential in $In_xGa_{1-x}As$ QD's embedded in the GaAs matrix can be derived from spatial distribution of indium concentration within the QD's.[30] We assume that this distribution can be described by a cylindrically symmetric Gaussian function[26]

$$X(\rho,z;R,Z)=X_0\exp(-\rho^2/R^2-z^2/Z^2), \tag{3}$$

where $\rho^2=x^2+y^2$, R is the radius of the QD, Z is half of its height, and X_0 is the concentration of indium at the center of the QD. In accordance with Eq. (3), we take the confinement potential for electrons

$$V_e(\rho,z;R,Z)=-0.7\Delta E_g X(\rho,z;R,Z) \tag{4}$$

and for holes

$$V_h(\rho,z;R,Z)=-0.3\Delta E_g X(\rho,z;R,Z), \tag{5}$$

where ΔE_g is the energy-gap difference between GaAs and InAs. We assume that the band offset ratio is 70/30.[31] Throughout the paper, we take the conduction-band minimum of the barrier material as the reference energy level for the electrons and the barrier valence-band maximum as the reference energy level for the holes. The calculations have been performed for $\Delta E_g=1.11$ eV[26] and the material parameters of GaAs, i.e., the static dielectric constant $\varepsilon=12.5$, the band mass of the electron $m_e=0.0667$, and the band mass of the hole $m_h=0.5$.

In order to determine the trion-energy shifts we need to know the trion ground-state energy, as well as the ground-state energies of the exciton, electron, and hole confined in the QD. For this purpose we use the confinement potentials (4) and (5) and assume the effective-mass approximation for electrons and holes. The ground-state energy of a single electron and a single hole confined in a QD with radius R and height $2Z$ is determined by the variational method with a Gaussian trial-wave-function

$$\phi_{e(h)}(\mathbf{r}_{e(h)};R,Z)=\sum_{i=1}^{N_{e(h)}}\sum_{j=1}^{M_{e(h)}}c_{ij}\exp(-\alpha_i^{e(h)}\rho^2-\beta_j^{e(h)}z^2), \tag{6}$$

where c_{ij} are the linear variational parameters, $\alpha_i^{e(h)}$ and $\beta_j^{e(h)}$ are the nonlinear variational parameters, which describe the localization of the particles radial and vertical directions, respectively. In this paper, we take $M_h=1$, $N_h=2$, $M_e=2$, and $N_e=4$, which ensures that the one-particle energies are determined with a precision of 0.1 meV.

The Hamiltonian of the exciton confined in the QD has the form

TABLE I. Ground-state energy E_X (in meV) of the exciton confined in the single QD calculated with wave function (8) with $M_e=2$, $N_e=4$, and $N_h=2$ quoted for several numbers of exponents describing the relative electron-hole position in x-y plane M_{eh} and in z direction N_{eh}. The parameters of the QD are $X_0=0.67$, $R=24.9$ nm, and $Z=0.92$ nm. The total number of basis elements ($N=M_e\times N_e\times N_h\times M_{eh}\times N_{eh}$) is listed in the third column.

M_{eh}	N_{eh}	N	E_X
1	1	16	−261.47
2	1	32	−262.19
2	2	64	−262.20
3	2	98	−262.23
3	3	144	−262.25

$$H_X=-\frac{\hbar^2}{2m_e}\nabla_e^2-\frac{\hbar^2}{2m_h}\nabla_h^2-\frac{1}{4\pi\varepsilon_0\varepsilon r_{eh}}+V_e(\rho_e,z_e;R,Z)+V_h(\rho_h,z_h;R,Z), \tag{7}$$

where $r_{eh}=|\mathbf{r}_e-\mathbf{r}_h|$. The ground-state energy of the exciton confined in the QD can be determined with the following variational wave function

$$\Psi_X^{(1)}(\mathbf{r}_e,\mathbf{r}_h)=\sum_{i_e=1}^{M_e}\sum_{j_e=1}^{N_e}\sum_{j_h=1}^{N_h}\sum_{i_{eh}=1}^{M_{eh}}\sum_{j_{eh}=1}^{N_{eh}}c_{i_e j_e j_h i_{eh} j_{eh}}\times\exp(-\alpha_{i_e}^e\rho_e^2-\beta_{j_e}^e z_e^2-\alpha^h\rho_h^2-\beta_{j_h}^h z_h^2-\alpha_{i_{eh}}^{eh}\rho_{eh}^2-\beta_{j_{eh}}^{eh}z_{eh}^2), \tag{8}$$

where $\rho_{eh}^2=(x_e-x_h)^2+(y_e-y_h)^2$, $z_{eh}=z_e-z_h$, $\alpha_{i_{eh}}^{eh}$ and $\beta_{j_{eh}}^{eh}$ are the variational parameters, which describe the relative position of the electron and the hole in x-y plane and z direction, respectively. The other variational parameters in Eq. (8) play the same role as in wave function (6). The convergence of variational basis (8) with respect to the number of the Gaussians applied was verified for the values[26] of parameters corresponding to $In_xGa_{1-x}As$ self-assembled QD's,[25] i.e., $X_0=0.67$, $R=24.9$ nm, $Z=0.92$ nm. The variational estimates of the exciton ground-state energy obtained with various numbers of terms in Eq. (8) are listed in Table I. We note, that the convergence of these estimates is very fast. The results are not significantly improved if one introduces a second β^{eh} exponent or a third α^{eh} parameter. The electron and hole wave functions are stiffened in z direction due to the strong confinement and react only weakly to the mutual Coulomb interaction. The change of the one-particle wave functions under the influence of the interaction is more pronounced in x-y plane, where the confinement is weaker. In this paper we consider not only the QD's in form of flat disks, but also QD's of different shape and size. Therefore, we have taken $M_{eh}=N_{eh}=3$ in the following calculations.

The results of Table I show, that variational wave function (8) is an effective tool in the calculations for the confined exciton ground state. However, this wave function is not suitable for a direct generalization to the problem of trions, since the number of basis elements grows very fast with the num-

TABLE II. Ground-state energy E_X (in meV) of the exciton confined in the single QD calculated with trial function (9). The parameters of the QD are the same as in Table I. In the first two columns, the number of elements taken in sum (9) is listed. In the third column, the energy E_X' calculated with the fixed variational parameters $R_{e(h)}=R$ and $Z_{e(h)}=Z$ is quoted. The number of basis elements is equal to $M_{eh}\times N_{eh}$. The results in the fourth column are obtained with optimized parameters $R_{e(h)}$ and $Z_{e(h)}$.

M_{eh}	N_{eh}	E_X'	E_X
1	1	−261.17	−261.34
1	2	−261.17	−261.36
2	1	−261.93	−262.19
3	1	−262.03	−262.22

ber of particles. Therefore, we have elaborated another approach to the problem of the QD-confined trions, which we will first demonstrate on the example of the confined electron-hole pair. The dependence of the wave function on the relative interparticle positions will be referred to as the "correlation between the particles." In wave function (8), this correlation is directly described by the exponents with α^{eh} and β^{eh}. However, even if the parameters α^{eh} and β^{eh} are set equal to zero, wave function (8) cannot be separated into a product of one-particle functions. Therefore, even without α^{eh} and β^{eh} exponents, a part of correlation is indirectly included in wave function (8). Now, we introduce another trial wave function for the QD-confined exciton, which takes into account the correlation between the particles in the direct way only,

$$\Psi_X^{(2)}(\mathbf{r}_e,\mathbf{r}_h)=\phi_e(\mathbf{r}_e;R_e,Z_e)\phi_h(\mathbf{r}_h;R_h,Z_h)\times\sum_{i_{eh}=1}^{M_{eh}}\sum_{j_{eh}=1}^{N_{eh}}c_{i_{eh}j_{eh}}\exp(-\alpha_{i_{eh}}^{eh}\rho_{eh}^2-\beta_{j_{eh}}^{eh}z_{eh}^2). \tag{9}$$

Trial wave function (9) is applied to the exciton confined in the QD with radius R and height $2Z$. It is composed of the ground-state wave functions of the electron (ϕ_e) confined in potential (4) with effective parameters R_e and Z_e and the hole (ϕ_h) confined in potential (5) with effective parameters R_h and Z_h. The double sum in Eq. (9) describes the electron-hole correlation. The parameters R_e, Z_e, R_h, and Z_h are treated as variational parameters. In this way, the one-particle wave functions ϕ_e and ϕ_h are allowed to change their spatial extension under influence of the Coulomb interaction between the particles.

The results obtained with wave function (9) are listed in Table II, which shows that the energy estimates obtained with the values of the variational parameters $R_{e(h)}$, $Z_{e(h)}$ fixed at the physical values of the QD size $R_{e(h)}=R$ and $Z_{e(h)}=Z$ converge to a value, which is larger by 0.2 meV than the energy obtained with basis (8) (cf. Table I). However, if we perform the optimization with respect to $R_{e(h)}$ and $Z_{e(h)}$, we obtain results equivalent to those obtained with wave function (8). Although wave function (9) does not require less numerical effort than function (8), the number of basis elements is much smaller and less memory consuming, so, it can be easily generalized to the problem of excitonic trions.

The Hamiltonian for the negatively charged trion X^- confined in a QD has the form

$$H_{X^-}=-\frac{\hbar^2}{2m_e}\nabla_1^2-\frac{\hbar^2}{2m_e}\nabla_2^2-\frac{\hbar^2}{2m_h}\nabla_h^2+\frac{1}{4\pi\varepsilon_0\varepsilon}\left(-\frac{1}{r_{1h}}-\frac{1}{r_{2h}}+\frac{1}{r_{12}}\right)+V_e(\mathbf{r}_1;R,Z)+V_e(\mathbf{r}_2;R,Z)+V_h(\mathbf{r}_h;R,Z), \tag{10}$$

where $\mathbf{r}_1$ and $\mathbf{r}_2$ are the position vectors of the electrons, $\mathbf{r}_h$ determines the position of the hole, $r_{12}=|\mathbf{r}_1-\mathbf{r}_2|$, $r_{1h}=|\mathbf{r}_1-\mathbf{r}_h|$, and $r_{2h}=|\mathbf{r}_2-\mathbf{r}_h|$. The Hamiltonian for the positive trion X_2^+ can be obtained from Eq. (10) by interchanging the indices $e\leftrightarrow h$ and ascribing the indices 1 and 2 to the holes.

In the negative-(positive) trion ground state, the electron (hole) subsystem is a spin singlet. This means that the ground-state spatial wave function of the negative (positive) trion is symmetric with respect to the interchange of the position vectors of the electrons (holes). Therefore, we apply the following trial wave function for the negative trion

$$\begin{aligned}\Psi_{X^-}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_h)&=\phi_e(\mathbf{r}_1;R_e,Z_e)\phi_e(\mathbf{r}_2;R_e,Z_e)\phi_h(\mathbf{r}_h;R_h,Z_h)\\&\times\sum_{i_1i_2i_{12}}\sum_{j_1j_2j_{12}}c_{i_1i_2i_{12}j_1j_2j_{12}}(1+P_{12})\\&\times\exp(-\alpha_{i_1}^{eh}\rho_{1h}^2-\alpha_{i_2}^{eh}\rho_{2h}^2-\alpha_{i_{12}}^{ee}\rho_{12}^2)\\&\times\exp(-\beta_{j_1}^{eh}z_{1h}^2-\beta_{j_2}^{eh}z_{2h}^2-\beta_{j_{12}}^{ee}z_{12}^2).\end{aligned} \tag{11}$$

The presence of the sum in wave function (11) introduces the correlation for the three particles. In Eq. (11), the summations start from 1 and run to M_{eh} over indices i_{1h} and i_{2h}, to N_{eh} over j_{1h} and j_{2h}, and to M_{12} and N_{12} over i_{12} and j_{12}. P_{12} is the operator which exchanges the coordinates of the two electrons. The factor $(1+P_{12})$ ensures, that the electron subsystem is in the symmetric spatial state and enables us to reduce the number of the basis elements. Namely all the basis elements, for which $j_1=j_2$ and $i_2>i_1$ are omitted in the summation (11). R_e, Z_e, R_h, and Z_h are treated as variational parameters like in wave function (9) for the exciton. For the positive trion we apply an analogous trial wave function. Similarly as in the case of the exciton confined in the flat QD, a single Gaussian is sufficient for the description of the correlations in z direction. The convergence of the variational results with the increasing number of Gaussians describing the in-plane correlation is displayed in Table III. In the following section we discuss the influence of the size and shape of the dot on the relative shifts of trion PL peaks. This discussion goes beyond the flat QD geometry, so the correlation in z direction should be described with the same precision as the in-plane correlation. The results presented in the following section have been obtained with 90-element basis

TABLE III. Ground-state energy E_{X^-} (in meV) of the negative excitonic trion calculated with trial wave function (11). The first column shows the number of Gaussians taken for the description of the electron-hole correlation in *x*-*y* plane. The number of Gaussians describing the electron-electron in-plane correlation is listed in the second column. In these calculations, a single Gaussian describing the correlation in *z* direction was applied. The third column shows the total number of basis elements. The parameters of the QD are the same as in Table I.

M_{eh}	M_{12}	N	E_X^-
1	1	1	−396.45
2	1	3	−397.91
2	2	6	−398.48
2	3	9	−398.59
3	3	12	−398.70
4	4	40	−398.71

generated by $M_{eh}=N_{eh}=2$ and $M_{12}=N_{12}=3$. We estimate that the trion ground-state energies are determined with precision of 0.2–0.3 meV.

B. Results

Figure 1 displays the energy shifts of the PL lines for the trions confined in a spherically symmetric ($R=Z$) QD with respect to the line of confined exciton. In bulk GaAs, the trion PL peaks are only slightly shifted with respect to the exciton PL line ($S_{X_2^+}\approx 0.4$ meV and $S_{X^-}\approx 0.25$ meV[4,15]). In large QD's (with radius R larger than ~ 100 nm), the shift of the positive-trion line is not significantly changed with respect to the bulk-limit value. If the radius of the dot decreases below 80 nm, X_2^+ PL line approaches the exciton line, i.e., $S_{X_2^+}$ decreases to 0. For $R\simeq 40$ nm the recombination of the electron-hole pair in the positive trion releases the same amount of energy as the exciton recombination. For $R<40$ nm $S_{X_2^+}$ takes on negative values; so, the order of the exciton and X_2^+ PL lines changes. The behavior of the negative-trion line is just opposite. The quantum confinement shifts X^- line deep below the exciton line on the energy scale. The opposite behavior of the positive- and negative-trion energy shifts was reported in our previous paper[15] on spherical quantum dots with the square-well confinement potential. In the strong confinement limit this effect can be explained in the framework of the perturbation theory. In this approach,[15] the trion-energy shifts are expressed in terms of the energies of the Coulomb interaction between the particles forming the trion complexes, as follows: $S_{X_2^+}=V_{eh}-V_{hh}$, and $S_{X^-}=V_{eh}-V_{ee}$, where V_{eh}, V_{ee}, and V_{hh} are the electron-hole, electron-electron, and hole-hole interaction energies, respectively. The quantum confinement in the QD's leads to the localization of the charge carriers, which is much stronger for the holes. Therefore, the absolute value of the Coulomb interaction energy between the holes increases more than the electron-hole and electron-electron interaction energies, i.e., $V_{hh}>V_{eh}>V_{ee}$, which explains the qualitative difference in the confinement-induced changes of X^- and

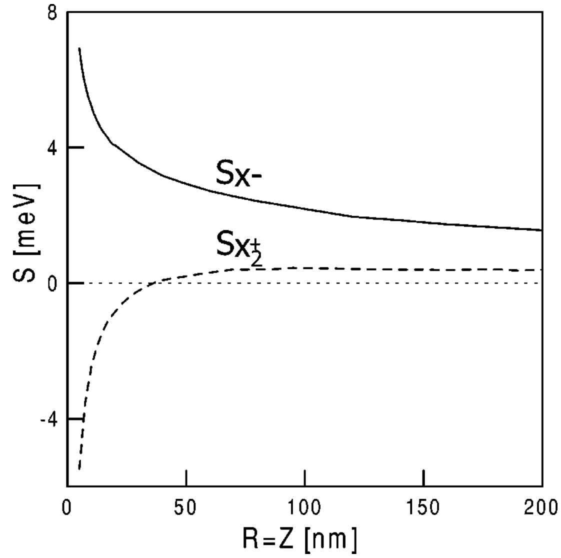

FIG. 1. Energy shifts S of the trion PL line with respect to the exciton PL line calculated for the spherical QD as a function of radius $R=Z$ for $X_0=0.67$. The solid (dashed) curve corresponds to the negative-(positive-) trion-energy shift ($S_{X^-}=E_X+E_e-E_{X^-}$ and $S_{X_2^+}=E_X+E_h-E_{X_2^+}$).

X_2^+ energy lines in spherical QD's ($S_{X_2^+}<0<S_{X^-}$). The results of the present calculations show, that the same effect occurs also for spherical quantum dots with the Gaussian potential profile.

The problem is more complex if the QD is anisotropic. Then, the strength and range of the confinement potential in *x*-*y* plane and the growth direction are different. We have performed the calculations of the trion-energy shifts for different values of R and Z. Figure 2 shows the calculated trion-energy shifts as functions of Z (half of the QD height) for several values of the QD radius R. We have considered QD's with very different height-to-radius ratios. The left (right) end of horizontal axis of Fig. 2 corresponds to the QD in a form of a flat disk (elongated cylinder). Each of the curves in Fig. 2 passes through the point, for which the QD has spherical symmetry. These points are marked by circles. The curves for the negative-trion exhibit the following simple regularity: the smaller is the QD, the stronger is the redshift of X^- PL line with respect to the exciton line. The dependence of the positive-trion energy shift is more complex. In QD's of large height, the movement of the confined charge carriers in z direction is nearly free. We can say that these QD's resemble the quantum wires. We note, that the PL peaks of both the negative and the positive trions are redshifted with respect to the exciton PL line if the radius of the "wire-like" dot decreases. For $R=100$ nm, the positive-trion line is monotonously redshifted with the decreasing height of the QD. This dependence on the height is qualitatively the same as in the case of the two-dimensional quantum wells, in which both the positive- and negative-trion PL peaks are redshifted with respect to the exciton peak, if the width of the quantum well decreases.[16] In contrast to $S_{X_2^+}$ shift for $R=100$ nm, the curve for $R=50$ nm is nonmonotonous. When Z decreases below 120 nm the X_2^+ PL line ap-

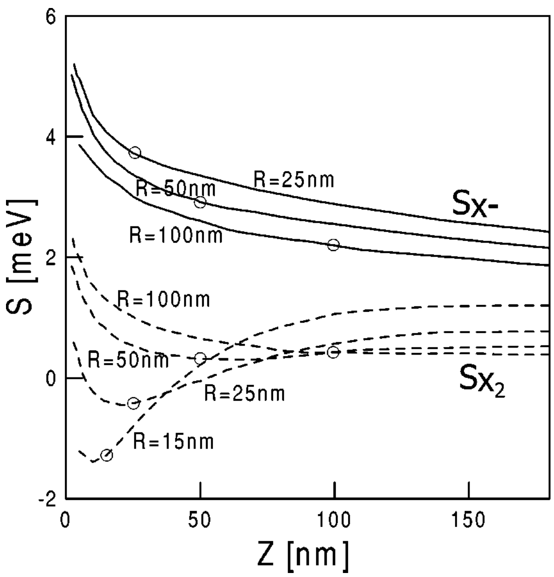

FIG. 2. Trion-energy shifts S with respect to the exciton PL line calculated as a function of Z (half of the QD height) for several values of the QD radius R. The solid curves show the results for the negative trion, and the dashed curves show the results for the positive trion. Open circles correspond to spherical symmetry of QD's.

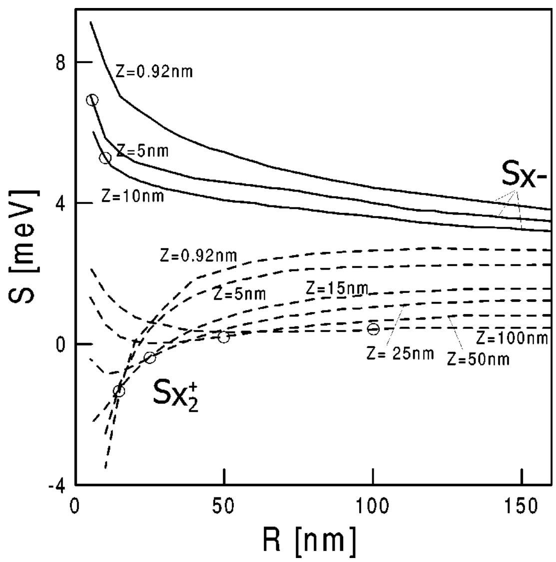

FIG. 3. Trion-energy shifts S calculated with respect to the exciton PL line as a function of radius R of the QD for several values of Z (half of the QD height). The solid (dashed) curves correspond to the negative (positive) trion. Open circles correspond to spherical QD's.

proaches the exciton PL peak. The curve for $R=50$ nm exhibits a flat minimum for Z between 50 and 80 nm. The minima of the curves for $R=25$ and 15 nm are distinctly more pronounced and appear at $Z=20$ and $Z=10$ nm, respectively. We note, that these minima correspond to quantum dots with nearly spherical shapes. The PL line of the positive-trion confined in the QD with radius $R=50$ nm has smaller energy than the exciton line regardless of Z. In other words, $S_{X_2^+}$ is always positive for $R=50$ nm. This is not the case of QD's with smaller radii, for which the value of the shift can be negative. The PL peak of the positive-trion confined in the dots with radius 25 or 10 nm can be blue shifted or redshifted with respect to the exciton line depending on the height of the QD.

Figure 3 shows the shifts of the trion PL lines with respect to the exciton line as functions of the radius of the dot for fixed values of its height. The points for which the QD potential is spherically symmetric are marked by circles. Again, the negative trion is redshifted more strongly for smaller QD's, while the dependence of the positive-trion shift is more complex. In Fig. 3, the QD with the radius $R\sim150$ nm can be treated as a quasi-two-dimensional quantum well. In this QD, the redshift of the PL lines is largest for the lowest value of the height of the well. On the other hand, the QD with the largest value of the height ($Z=100$ nm) looks like a quantum wire and the redshift of the PL lines grows, when the radius of this "wire-like" QD decreases. The curve for $Z=50$ nm shows a flat minimum at $R=30$ nm. This minimum is more pronounced for QD's with smaller height. We note, that the minima of $S_{X_2^+}$ can correspond to negative values of the shift.

C. Conclusions

In this section, we have studied the dependence of the energy shifts of the trion PL peaks with respect to the exciton peak on the size and geometry of the QD's. The obtained results indicate, that the negative-trion PL peak is always redshifted with respect to the exciton line. The stronger is the confinement (the smaller is the QD) the larger is this redshift. The positive-trion line behaves qualitatively in the same manner only in the cases, where the QD geometry resembles a quantum well or a quantum wire. For the "well-like" QD's the redshift of X_2^+ PL line is larger for the small height of the quantum well, while for the "wire-like" QD's the redshift is more pronounced for the small radius of the "wire." For the QD's with the diameter $2R$ comparable to the height $2Z$, the size dependence of the positive-trion energy shift is more complex. This energy shift plotted as a function of the radius or the height of the QD exhibits minima near $R/Z=1$, i.e., close to the QD's with the spherical symmetry. If the values of R or Z are of order of 25 nm or smaller, these minima correspond to negative values of the energy shift. Then, the order of exciton and positive-trion PL lines is opposite that in the bulk limit. The results presented in this section are in a qualitative agreement with the present knowledge on the trions in quantum wells,[4,16] and with the previous study of the trions in spherically symmetric QD's.[15] Moreover, based on the present results for the "wire-like" QD's, we can predict that the PL lines for both the negative and the positive trions in quantum wires should be redshifted with respect to the exciton line if the quantum-wire radius is decreased.

III. EXCITONIC TRIONS IN VERTICALLY COUPLED QUANTUM DOTS

A. Theory

The energy shifts S_{X^-} and $S_{X_2^+}$ for the trions confined in a single quantum dot cannot be identified with the trion binding energy.[15] However, if the QD is not single, i.e., if there is another identical QD at a distance large enough to exclude the coupling between the dots, the trion-energy shifts are exactly equal to the energy needed to transfer one electron (for X^-) or one hole (for X_2^+) from the QD occupied by the three charge carriers to the other empty QD. Let us assume, that the charge carriers have at their disposal two identical, remote QD's. Since in a single QD S_{X^-} is always positive, the ground state of a system composed of two electrons and one hole will always correspond to a state, in which all the particles are confined within the same QD (this is the confined-trion state). However, this is not the case for X_2^+ trion. For QD's in which the energy shift $S_{X_2^+}$ is negative, the ground-state corresponds to the electron-hole pair confined in one dot (confined exciton) and one hole confined in the other QD. If the QD's are closer, the coupling between them can essentially change the energies of the exciton complexes and the type of the localization of particles.[26] The coupling between vertically stacked $In_xGa_{1-x}As$ self-assembled QD's was observed in the PL spectroscopy[25] in the shifts of the PL peaks as functions of the thickness of the interdot barrier.

In this section we study the effect of the vertical coupling between the QD's on the trion states. We assume that the coupled dots have identical shapes and sizes, and possess a common axis of the rotational symmetry. We apply the following confinement potentials for the electrons and the holes:

$$V^c_{e(h)}(\rho_{e(h)},z_{e(h)})=V_{e(h)}(\rho_{e(h)},z_{e(h)}-a/2;R,Z)+V_{e(h)}(\rho_{e(h)},z_{e(h)}+a/2;R,Z), \tag{12}$$

where V_e and V_h are the given by Eqs. (4) and (5), respectively, and a is the distance between the centers of the QD's. The thickness of the barrier between the QD's can be expressed in terms of the distance between the QD centers and the height of the QD as follows: $t=a-2Z$. We adopt the values of the QD parameters corresponding to $In_xGa_{1-x}As$ self-assembled QD's: $X_0=0.67$, $R=24.9$ nm, $Z=0.92$ nm (same as in the test calculations of Sec. II). The ground-state wave function of a single-particle confined in potential (12) possesses an even parity with respect to the change of sign of z coordinate. We calculate the one-particle ground-state energy using the wave function

$$\phi^c_{e(h)}(\rho_{e(h)},z_{e(h)})=\phi_{e(h)}(\rho_{e(h)},z_{e(h)}-a/2;R,Z)+\phi_{e(h)}(\rho_{e(h)},z_{e(h)}+a/2;R,Z), \tag{13}$$

where $\phi_{e(h)}$ is wave function (6) of the ground state of the electron (hole) confined in the single isolated QD. Wave function (13) is a good approximation of the exact ground-state wave function for the studied range of the barrier thickness between the dots, i.e., for $t=a-2Z>2$ nm. The ground-state energy of the electron-hole pair in the coupled dots is determined variationally with the following trial wave function:

$$\begin{aligned}\Psi^c_X(\mathbf{r}_e,\mathbf{r}_h)=\sum_{k_e,k_h=0}^{1}\sum_{i_{eh}=1}^{M_{eh}}c_{i_{eh}k_ek_h}\phi_e&[\rho_e,z_e+(-1)^{k_e}(a/2);R_e,Z_e]\phi_h\\&[\rho_h,z_h+(-1)^{k_h}(a/2);R_h,Z_h]\\&\times\exp(-\alpha^{eh}_{i_{eh}}\rho^2_{eh}-\beta^{eh}z^2_{eh}),\end{aligned} \tag{14}$$

where $c_{i_{eh}k_ek_h}$ are the linear and $R_e,Z_e,R_h,Z_h,\alpha^{eh}_{i_{eh}},\beta^{eh}$ are the nonlinear variational parameters. This function is a direct generalization of trial wave function (9) for the exciton confined in an isolated QD with $N_{eh}=1$. In Eq. (14) the summations over k_e and k_h take into account all the possible distributions of the charge carriers over the two QD's. The z direction correlation between the particles confined in the same self-assembled QD is weak (cf. Tables I–II); so we neglect it almost totally in wave function (14). The interdot correlations are introduced via the two-center localization of the products of the one-particle functions $\phi_{e(h)}$.

We have calculated the lowest-energy levels of the negative-(positive) trion assuming that the electron (hole) subsystem is the spin singlet. For the negative trion we use the following wave function:

$$\begin{aligned}\Psi^c_{X^-}&(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_h)\\&=\sum_{k_1k_2k_h}\sum_{i_1i_2i_{12}}c_{i_1i_2i_{12}k_1k_2k_h}(1+P_{12})\phi_h\\&\quad[\rho_h,z_h+(-1)^{k_h}(a/2);R_h,Z_h]\\&\quad\phi_e[\rho_1,z_1+(-1)^{k_1}(a/2);R_e,Z_e]\phi_e\\&\quad[\rho_2,z_2+(-1)^{k_2}(a/2);R_e,Z_e]\\&\quad\times\exp(-\alpha^{eh}_{i_1}\rho^2_{1h}-\alpha^{eh}_{i_2}\rho^2_{2h})\\&\quad\times\exp(-\alpha^{ee}_{i_{12}}\rho^2_{12}-\beta^{eh}z^2_{1h}-\beta^{eh}z^2_{2h}),\end{aligned} \tag{15}$$

where the summations over k_1 and k_h run from 0 to 1, over k_2 from 0 to k_1, over i_{12} from 1 to M_{ee}, and over i_1 and i_2 from 1 to M_{eh}. The terms with $k_1=k_2$ and $i_2>i_1$ are excluded from the sum, because of the symmetrization of the basis elements $(1+P_{12})$. Basis (15) takes into account all the possible distributions of the three particles between the two QD's. In the calculations, we take $M_{ee}=3$ and $M_{eh}=2$, like in the case of the trion in the single QD. In consequence, basis (15) consists of 60 elements. The trial wave function for the positive trion has been chosen in the same way.

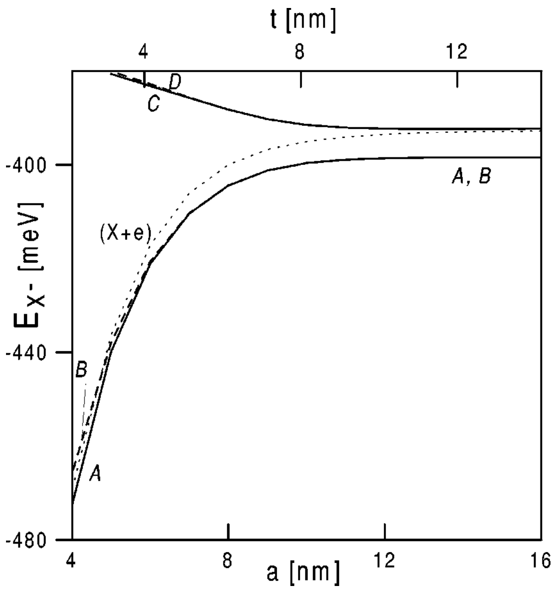

FIG. 4. Lowest-energy levels of the negative excitonic trion in the double QD as functions of the distance a between the centers of the QD's (thickness t of the barrier between the QD's). Solid (dashed) lines correspond to the energy levels of the even-(odd-) parity states. Thin dotted $(X+e)$ line shows the sum of the ground-state energies of the exciton and the single-electron confined in the double QD. The meaning of symbols A,B,C, and D is explained in the text.

B. Results

Figure 4 shows the four lowest-energy levels for X^- states, in which the electron subsystem is the spin singlet. The corresponding barrier thickness t is marked on the upper horizontal axis. Since the confinement potential of the coupled QD's (12) is invariant with respect to the reflection through $z=0$ plane, the wave functions of the three-particle complex possess a definite (even- or odd-) parity symmetry with respect to the operation of a simultaneous change of signs of z coordinates for all the particles. The parity properties of considered states result from the symmetries of the calculated three-particle wave functions, cf. the detailed discussion of the parity symmetry for the electron-hole pair in vertically coupled self-assembled QD's given in Ref. 26. The wave functions associated with the energy levels A and C (solid lines) possess even parity, while the energy levels B and D (dashed lines) correspond to the wave functions with odd parity. For $a>$ 14 nm all the energy levels are independent of the interdot distance. Then, the QD's can be treated as separate (uncoupled). In this limit, the distribution of the charge carriers between the QD's is a definite property of all the eigenstates, i.e., in all the eigenstates, the charge carriers occupy with a 100% probability either the same QD or different QD's.[26] At large interdot distance, the degenerate states A and B correspond to the trion localized in one of the QD's. In these states all the charge carriers are confined within the same QD. The energy of these states is equal to the energy of the negative-trion confined in the single isolated QD (cf. Table III). The ground state is twofold degenerate, because the trion can be located in one of the two QD's.

For $a<14$ nm both the degenerate A and B energy levels start to decrease, which is a signature of the tunnel coupling between the QD's. The presence of the tunnel coupling means, that in states A and B there is a nonvanishing probability of finding one of the particles in the other QD than the remaining two charge carriers. However, the tendency of all the particles to occupy the same QD is still visible in these states.[26] The degeneracy of the energy levels A and B is lifted for $a<6$ nm. For $a=4$ nm ($t\approx 2$ nm) the QD's are strongly coupled. In the strong coupling limit the trion wave functions exhibit an approximate one-particle parity, i.e., parity with respect to the change of the sign of z coordinate of each of the particles separately.[26] Due to the difference of the electron and hole masses, the effective height of the barrier between the dots is much larger for the hole than for the electron. In consequence, the even-odd energy-level splitting for the electron is considerably larger than the splitting for the hole. Both A and B states correspond to an approximate even parity of both the electrons. Moreover in state A, the hole is in the even-parity state, whereas in state B, the hole is in the state with an approximate odd parity. In both the excited states C and D, one of the electrons is in the even-parity state and the other is in the odd-parity state. The corresponding C and D energy levels split for $a<6$ nm. Similarly as states A and B, states C and D differ by the parity of the hole, which is even in C state and odd in D state. In the limit of the separate QD's, C and D states have the same energy and correspond to the electron-hole pair confined in one of QD's, while the second electron is confined in the other QD.

The thin dotted line marked by $(X+e)$ in Fig. 4, shows the sum of the ground-state energies of the following two systems: the electron-hole pair and a single electron in the double QD structure. In the limit of large a, this sum coincides with the degenerate C and D energy levels. The difference between the sum of energies $(X+e)$ and the ground-state energy of the trion is equal to the shift of X^- PL peak with respect to the exciton line, i.e., to S_{X^-}. In the limit of separate QD's, the energy shift S_{X^-} becomes identical with the difference of the energies between the trion ground state (A,B energy levels) and the excited state (C,D energy levels). Then, this difference can be interpreted as the binding energy of the trion in the double QD structure, i.e., as the amount of energy needed to remove one of the electrons from the QD occupied by the trion and transfer it to the other QD. On the other hand, in the strong coupling limit, the double QD can be treated as a single QD with enlarged height (larger vertical extension). The trion energy shift S_{X^-} decreases with the decreasing distance between the QD's. This effect is consistent with the results of the preceding section, which show that the negative-trion energy shift is larger for smaller QD's.

The results for the lowest-energy states of X_2^+ trion are displayed in Fig. 5. We restrict our study to the singlet states of the hole subsystem. The energy levels corresponding to the states with even (odd) total parity are plotted by the solid (dashed) lines. In the limit of separate dots (large a), the ground state is twofold degenerate (cf. degenerate energy-levels marked by A and B). In the ground-state all the three particles occupy the same QD, while the other dot is empty.

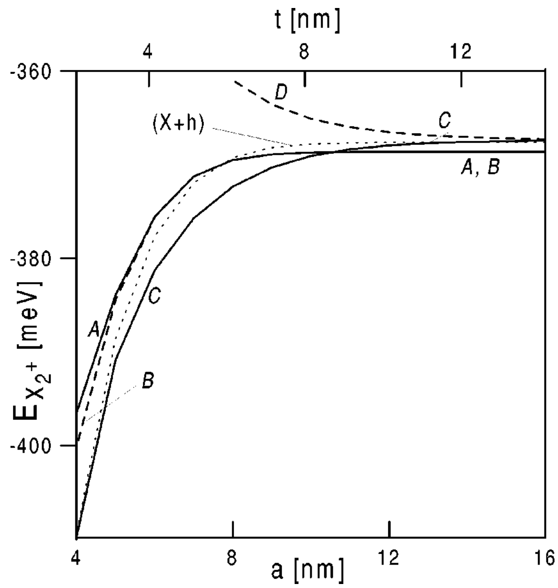

FIG. 5. Lowest-energy levels of the positive excitonic trion in the double QD as functions of the distance a between the centers of the QD's. Solid (dashed) lines present the energy levels of the states with the even (odd) parity. Thin dotted $(X+h)$ line shows the sum of the ground-state energies of the exciton and the single hole confined in the double QD. The meaning of symbols A,B,C, and D is explained in the text.

In other words, for separate QD's the ground state corresponds to the positive trion X_2^+ localized in one of the QD's. In the limit of the separate QD's, the first-excited state energy level (C,D) is also degenerate. In the states C and D, an electron-hole pair is confined in one QD and the second hole is confined in the other QD. For comparison, the sum of the ground-state energies $(X+h)$ of the exciton and hole is plotted by the thin dotted line. For smaller values of the barrier thickness, the relative distribution of the charge carriers between the QD's is uncertain as a consequence of the coupling. Nevertheless, even for the coupled QD's, in A and B states, all the particles exhibit a tendency to occupy the same dot, while in the excited C and D states, the holes exhibit a tendency to occupy different QD's.

The degenerate excited-state energy level (C, D) splits for larger values of the interdot distance than (A,B) level. The electron in A, B, and C states possesses an approximate even parity, therefore the corresponding energy levels decrease when the barrier thickness decreases. In D state the electron has an approximate odd parity, and in contrast to states A,B and C, its energy grows when the barrier thickness decreases. The state C corresponds to both the holes in even-parity states. In the state A (B) one hole (two holes) occupies the odd-parity state. The degeneracy of A,B energy levels is lifted for $a \simeq 6$ nm as the result of the splitting between the even- and odd-parity energy levels of the hole. The most interesting feature of the positive-trion spectrum is the fact, that the energy of C state passes below that of A and B states for $a < \sim 10$ nm. This change of order of energy levels can be understood in context of Fig. 2. In the single QD, the energy shift $S_{X_2^+}$ for $R=25$ nm and $Z=0.92$ nm is positive but decreases if the height of the QD increases. In particular, for $10 \leqslant Z \leqslant 40$ nm the energy shift $S_{X_2^+}$ is negative (cf. Fig. 2). As we have noted before, the coupled QD's correspond to a single dot with a larger height. This explains why the state C, in which the holes tend to occupy different QD's, becomes the ground state of X_2^+ trion under influence of the interdot coupling. We note, that the change of order of A, B, and C energy-levels appears for the specific value of the height of the dot. In the limit of separate dots, C energy-level should correspond to the ground state for these values of the height for which the positive-trion shift $S_{X_2^+}$ is negative (cf. Fig. 2).

IV. CONCLUSIONS AND SUMMARY

We have presented the results of variational calculations for the excitonic trions confined in the single and vertically coupled QD's with the Gaussian confinement potential. We have proposed the trial wave function expanded in the Gaussian basis, which takes into account both the single-particle confinement effects and interparticle correlations. We have studied the influence of the shape and geometry of the confinement potential on the shifts of the trion PL lines with respect to the exciton line. We have considered all the geometries of the cylindrically symmetric QD from a very flat QD, which resembles the two-dimensional quantum well to an elongated wire-like QD. We have shown, that the negative-trion PL line is always redshifted with respect to the exciton line and that this shift is larger for smaller QD's. The positive-trion exhibits qualitatively the same behavior only in these QD's, which resemble quasi-one-dimensional quantum wire or quasi-two-dimensional quantum well. However, in the nanostructures with similar height and diameter, the positive-trion line can be blueshifted with respect to the exciton line, if the linear size of the QD is sufficiently small. The limit cases of the present results are in a qualitative agreement with the results of previous studies of the trion binding energy in quantum wells,[4,16] and also with the shifts of the trion PL lines found for the spherical QD's.[15] Based on these results we can also formulate predictions about the binding energy of the trions in quantum wires. Namely, the present results indicate that the binding energy of both the negative and the positive trions should increase when the radius of the quantum wire is decreased.

Moreover, we have extended our study to the excitonic trions in the vertically coupled $In_xGa_{1-x}As$ self-assembled QD's. We have considered the low-energy spectrum of the system of two electrons and one hole, as well as the system of two holes and one electron in a couple of vertically stacked QD's. We have shown, that the shifts of the trion PL lines with respect to the exciton line obtained for a single isolated QD can be identified with the binding energy of the trion complexes for a pair of identical remote QD's. We have studied the splitting of the trion-energy levels under influence of the coupling between the QD's. We have found, that the interdot coupling decreases the redshift of the negative-trion PL line with respect to the exciton line. Moreover, we have shown that the coupling may induce a redistribution of the charge carriers between the dots in the positive-trion ground state. These effects have been explained on the basis of our studies for the single QD, since—in the strong-coupling limit—-the coupled QD's can be treated as a single QD with an enlarged extension in the vertical direction.

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