Anomalous Stark effect in semiconductor quantum dots

Sandra Ritter,[1] Paul Gartner,[1,2] Norman Baer,[1] and Frank Jahnke[1]
[1]*Institute for Theoretical Physics, University Bremen, P.O. Box 330 440, 28334 Bremen, Germany*
[2]*National Institute of Materials Physics, P.O. Box MG-7, Bucharest-Magurele, Romania*
(Received 24 May 2007; revised manuscript received 1 August 2007; published 3 October 2007)

The effect of a lateral electric field on the ground-state energy of an electron-hole pair confined in a quantum dot is investigated on different levels of sophistication. Two different regimes occur: a Coulomb dominated one, where the influence of the field is weak, and a regime dominated by field induced effects. Depending on the strength of the Coulomb interaction relative to the single-particle energies, which varies with the quantum dot parameters, the transition between these regimes is either smooth or rather abrupt.

DOI: 10.1103/PhysRevB.76.165302 PACS number(s): 78.67.Hc, 73.21.La

I. INTRODUCTION

Nanostructures in general and their zero-dimensional realizations known as quantum dots (QDs) have developed over the last few years into one of the major research topics of condensed matter physics. Electronic, optical, and structural properties of QDs have received a lot of attention both experimentally and theoretically. One of the driving forces in this research field is the wealth of possible applications associated with these new structures. For a great number of applications it is desirable to (fine) tune, for example, the excitonic transition energy as needed. While many studies have, to this end, relied on ultrahigh magnetic fields and provided deeper insight into the optical and electronic properties, this approach will fail for many practical applications due to the large experimental effort involved. In view of an ongoing need for miniaturization the utilization of an electric field for a desired tuning seems to be clearly advantageous due to its relative ease of implementation.

The quantum-confined Stark effect (QCSE) appears when an electric field is applied on a system in which the carriers are confined in one or several directions, including the direction of the field. Initially the effect was studied in quantum wells[1–3] and quantum wires,[4,5] but more recently extensive attention was devoted to the QCSE in QDs (Refs. 6–20) and QD molecules.[21–26] In all these cases one observes a shift of the excitonic peak in absorption or photoluminescence (PL) spectra which, for small fields F, has the form $\Delta E(F) = E(F) - E(0) \approx pF - \alpha F^2$, where p and α are, respectively, the components of the permanent dipole moment and the polarizability in the direction of the applied electric field. If the symmetry of the problem rules out a preferential direction, the Stark effect is an even function of F and the expansion begins with the polarizability term ($p=0$).

The sign of this term was the object of some discussion in the literature. Two conflicting effects of the electric field have been identified: On the one hand, the field is pushing the electrons and holes in opposite directions, as confirmed by the decrease in the excitonic oscillator strength. The lower one-particle energies of the carriers in their new positions give rise to a redshift ($\alpha > 0$) of the excitonic line. On the other hand, the Coulomb attraction is diminished by this carrier separation, and this increases the electron-hole pair energy. It was argued[3–5,7] that the last effect may be strong enough to prevail over the first, therefore leading to an overall blueshifted QCSE ($\alpha < 0$).

However, this conclusion runs against a general quantum-mechanical theorem[27] which states that the ground-state energy is a concave function of any parameter that enters linearly in the Hamiltonian. This is a quite general result, which does not depend on the model Hamiltonian used. It is also not dependent on the nature of the spectrum, being valid both for a discrete ground state (as in QDs) and for the bottom of a continuum (as in quantum wells and quantum wires). The concavity of the ground-state energy implies that its second derivative with respect to the field is negative, not only around $F=0$ but in all the points of the curve $E(F)$. In particular, the polarizability α cannot become negative and, in the absence of a permanent dipole, this leads to a redshifted QCSE. The last point was clarified as early as 1985,[2] by invoking a special case of the above-mentioned theorem. Nevertheless, papers claiming a blueshifted QCSE, both in experiment and in theory, kept appearing[3–5,7,18] without reference to this result.

Most of the theoretical calculations of the QCSE use the effective mass approximation with a steplike or infinite barrier confinement potential.[2–4,6,15,18,28] The electron-hole interaction is treated in many cases in a self-consistent Hartree scheme.[4,18,28] More elaborated approaches to this few-body problem involve expansion in the one-particle eigenfunctions,[6] inclusion of correlations in a self-consistent Kohn-Sham scheme,[15] or variational procedures.[2,3] Simpler treatments use perturbation theory with respect to the Coulomb interaction.[4]

In the present paper we theoretically investigate the effect of a lateral field on the excitonic ground state of a lens-shaped QD. It was shown[29] that the confinement in such a system is accurately described by a parabolic potential. Then the one-particle states in an external field are given by displaced oscillator states, with the energies reduced by an amount proportional to F^2. If only this redshift is considered one obtains, with physically reasonable QD parameters, a value of the polarizability $\alpha \sim 10^{-2}$ meV cm^2/kV^2. The experimental values[10,13,16,19,20] in such systems are at most of the order of $\sim 10^{-3}$ meV cm^2/kV^2 showing that the reduction of the exciton binding energy due to the charge separation plays an important role.[32] Several methods of including the Coulomb interaction are considered and compared in the following.

As the simplest attempt we analyze the perturbative approach. This leads to a reduction of the polarizability and, for strong Coulomb couplings, even to a negative α. As discussed before, this result contradicts the concavity theorem, and therefore it should be taken as a sign of the breakdown of the perturbation approach rather than as physically meaningful.

The next method employed is a variational calculation in which the trial function is factorized as $\psi(\mathbf{r}_e,\mathbf{r}_h)=\phi^e(\mathbf{r}_e)\phi^h(\mathbf{r}_h)$. The concavity theorem is of variational nature, therefore it is guaranteed to hold in any variational approach. Indeed, our results show that the higher the Coulomb strength the smaller the values of α, but without a change of sign. One also observes that such a flat parabolic dependence around $F=0$ is followed by a much steeper redshift at larger fields. This change of regime corresponds to a sufficiently large electron-hole separation so that the Coulomb binding energy becomes negligible and the pair behaves as independent carriers. The transition between the two regimes is sharper for strong Coulomb interaction.

Since both the Coulomb and the electric field term in the Hamiltonian depend only on the relative coordinate, it is natural to try a factorization procedure involving this coordinate. Such a factorization is exact in the particular case of equal electron and hole oscillator frequencies in the assumed harmonic in-plane confinement potential. Then the center of mass motion is trivial and field independent, and the QCSE is fully contained in the relative motion part. The latter is a one-particle problem, with a two-dimensional potential consisting of a Coulomb attractive center and a field-shifted parabolic confinement. This problem can be exactly solved to numerical accuracy and it shows clearly the two-regime behavior produced by the competition between these two attractive potentials.

In the more general case of different electron and hole oscillator frequencies the separation into the center-of-mass and relative motion is no longer exact, but a variational factorization scheme involving a suitably chosen central coordinate and the relative coordinate can be used. It turns out that the functional to be minimized is closely related to the ground-state energy found by the previous procedure for the special case of equal frequencies.

II. MODEL

In order to describe the interplay between the confinement of the carriers due to the QD, the external electric field, and the attractive Coulomb interaction between the electron and the hole, we use the following Hamiltonian in effective mass approximation

$$H=-\frac{\hbar^2}{2m_e}\Delta_e+\frac{m_e\omega_e^2}{2}\mathbf{r}_e^2+eFx_e - \frac{\hbar^2}{2m_h}\Delta_h+\frac{m_h\omega_h^2}{2}\mathbf{r}_h^2-eFx_h-\frac{e^2}{4\pi\epsilon_0\epsilon_r|\mathbf{r}_e-\mathbf{r}_h|}. \quad (1)$$

The first two terms of Eq. (1) describe the motion of an electron of mass m_e in a harmonic confinement potential with frequency ω_e. For flat lens-shaped cylindrically symmetric QDs the in-plane part of the wave function can be approximately decoupled from the motion in the growth direction. Furthermore, it has been shown that in this case the in-plane potential is well described by a harmonic oscillator potential.[29] Due to the strong confinement in the z direction and the fact that only lateral electric fields are investigated, we discard in the following, for clarity, the z degree of freedom. Therefore $\mathbf{r}_e=(x_e,y_e)$ denotes the two-dimensional in-plane vector and Δ_e the corresponding Laplacian. The third term of Eq. (1) incorporates the coupling of a homogeneous, constant electric field F to the electronic degree of freedom with charge $-e$. Due to the cylindrical symmetry one can assume, without loss of generality, that the field is directed along the x axis, $\mathbf{F}=(F,0)$. The second line of Eq. (1) contains the corresponding terms for the hole with mass m_h, frequency ω_h, and charge e. Finally, the attractive coupling between the electron and the hole via Coulomb interaction is given by the third line of Eq. (1), where ϵ_r is the background dielectric constant.

In the absence of Coulomb interaction the two-body problem is decoupled and can readily be solved by completing the square for each degree of freedom. For the single-particle ground-state energy one finds

$$E^i=\hbar\omega_i-\frac{(eF)^2}{2m_i\omega_i^2} \quad (2)$$

with the corresponding wave function

$$\phi^i(\mathbf{r})\sim e^{-(1/2l_i^2)(\mathbf{r}-\mathbf{c}_i)^2}, \quad (3)$$

where $i\in\{e,h\}$ refers to either the electron or the hole, $l_i=\sqrt{\hbar/m_i\omega_i}$ is the oscillator length, and $\mathbf{c}_i=\mp\frac{e\mathbf{F}}{m_i\omega_i^2}$ is the center of the wave function in the presence of the field. In the absence of the Coulomb interaction the center is shifted linearly with the electric field. The above equation shows a quadratic dependence of the single-particle ground-state energy on F.

As discussed in the Introduction, it is rather obvious that the ground-state energy of the electron-hole pair is reduced by the attractive Coulomb interaction and that this attraction becomes weaker as the electric field is increased. This happens because the electron and the hole are pulled in opposite directions by the electric field, thus reducing the exciton binding energy. Therefore one has two competing trends: the pure single-particle effect, which lowers the ground-state energy as the field strength is increased and is responsible for a redshift, and a decrease of the binding energy that has the opposite effect. Their interplay is discussed in the following subsections on different levels of refinement.

A. Perturbation theory

In this most simple approach the first two lines of Eq. (1) are used as the unperturbed Hamiltonian H_0, which is exactly solvable, and the Coulomb interaction in the last line is treated as the perturbation. The first order correction is then given by the direct Coulomb matrix element

$$D^{eh} = -\frac{e^2}{4\pi\epsilon_0\epsilon_r}\int d^2r_e\, d^2r_h \frac{|\phi^e(\mathbf{r}_e)|^2|\phi^h(\mathbf{r}_h)|^2}{|\mathbf{r}_e - \mathbf{r}_h|} \tag{4}$$

that describes the electrostatic interaction between the charge distributions of the electron and the hole in their respective unperturbed ground states given by Eq. (3). This matrix element can be evaluated analytically in the present situation and yields

$$D^{eh} = -\frac{e^2}{4\pi\epsilon_0\epsilon_r l}\sqrt{\pi}e^{-(1/2l^2)\mathbf{c}_{he}^2} I_0\left(\frac{1}{2l^2}\mathbf{c}_{he}^2\right). \tag{5}$$

Here $I_0(x)$ is the modified Bessel function of the first kind and index 0, l is defined by $\sqrt{l_e^2+l_h^2}$, and $\mathbf{c}_{he}=\mathbf{c}_h-\mathbf{c}_e$ measures the electron-hole separation, which is linear in the applied electric field.

By means of a Taylor expansion of Eq. (5) in the electric field F one obtains for the energy shift of the ground state of the electron hole pair the expression

$$\Delta E(F) = -(\alpha_f - \alpha_C)F^2 - \beta_C F^4 + \cdots, \tag{6}$$

where $\alpha_f = \frac{e^2}{2}\left(\frac{1}{m_e\omega_e^2}+\frac{1}{m_h\omega_h^2}\right)$ provides the field dependence of the free-particle energy and α_C and β_C are the coefficients of the second and fourth order term in the expansion of the Coulomb matrix element, respectively. It is clear that the sign of the polarizability is controlled by the ratio α_C/α_f which, at a closer examination, turns out to be proportional to the Coulomb interaction energy for the charges at a distance l measured in units of $\hbar\omega$, with ω a suitable average between the electron and hole oscillator frequencies. Therefore the effective strength of the Coulomb interaction is a function of the QD geometry and material parameters. With increasing Coulomb strength the polarizability in Eq. (6) can turn from positive to negative. As discussed above, this is in contradiction to the concavity theorem and shows the limits of the perturbative approach. The failure is mainly due to the fact that in the case of strong Coulomb interaction the true wave functions deviate considerably from the wave functions given by Eq. (3), which was used to evaluate the perturbation. In other words, the method lacks consistency in the sense that it computes the expectation value of the full Hamiltonian H on the states that minimize the expectation value of H_0. This shortcoming is addressed in the next subsection.

B. Variational ansatz I

In this variational approach we consider the minimum of the expectation value $\langle\psi|H|\psi\rangle$ of the full Hamiltonian Eq. (1) taken on the class of trial functions given by (normalized) products of Gauss functions

$$\psi(\mathbf{r}_e,\mathbf{r}_h) \sim e^{-(x_e-c_e)^2/2(\sigma_x^e)^2-(y_e)^2/2(\sigma_y^e)^2} e^{-(x_h-c_h)^2/2(\sigma_x^h)^2-(y_h)^2/2(\sigma_y^h)^2}. \tag{7}$$

The extension σ_x^i and σ_y^i of the single-particle wave functions in x and y directions as well as their center in the field direction are now left as variational parameters. This ansatz for the wave function is in part motivated by the fact that the exact solution in the absence of Coulomb interaction is of this form, see Eq. (3). However, the separation between the electron and the hole as well as their spatial extension are not prescribed, as in the perturbative case, but can adjust themselves to the applied field. For the numerical evaluation, the energy functional can be calculated analytically, up to a single one-dimensional integral. The minimum in the six-dimensional parameter space is then found using a simplex search algorithm.[30]

Results are presented in Sec. II D in comparison with other approaches. Let us mention here only the fact that the concavity condition is automatically met due to the variational nature of the method employed.

C. Decomposition in relative and center coordinates

Noting that both the field and the Coulomb interaction act only on the relative coordinate $\mathbf{r}=\mathbf{r}_h-\mathbf{r}_e$ it is reasonable to rewrite the problem in terms of this coordinate and a center coordinate $\mathbf{R}$, which is a suitable chosen average of the two carrier positions

$$\mathbf{R} = \lambda_e\mathbf{r}_e + \lambda_h\mathbf{r}_h, \quad \text{with } \lambda_e+\lambda_h = 1,\ \lambda_e,\lambda_h > 0, \tag{8}$$

$$\mathbf{r} = \mathbf{r}_h - \mathbf{r}_e. \tag{9}$$

The usual choice of $\lambda_e=m_e/(m_e+m_h)$ and $\lambda_h=m_h/(m_e+m_h)$ leads to $\mathbf{R}$ being the center of mass (c.m.). In the case of equal confinement energies for the electrons and holes ($\omega_e=\omega_h$) this choice provides a decoupling of the Hamiltonian into one part containing solely the c.m. and another part describing the relative motion (RM), $H(\mathbf{r}_e,\mathbf{r}_h) = H_{\text{c.m.}}(\mathbf{R})+H_{\text{RM}}(\mathbf{r})$, and the eigenstates are of the form $\psi(\mathbf{R},\mathbf{r})=\Phi(\mathbf{R})\varphi(\mathbf{r})$. This case will be presented in full detail below.

Moreover, this suggests a factorization scheme as a variational ansatz for the more general situation with different electron and hole confinement energies, when an exact factorization is no longer possible. The idea is supported by another exactly soluble case, that of vanishing Coulomb interaction, but arbitrary confinement energies for the electron and hole. In this case the ground state consists of a product of displaced Gaussians

$$\psi(\mathbf{r}_e,\mathbf{r}_h) \sim e^{-(1/2l_e^2)(\mathbf{r}_e-\mathbf{c}_e)^2} e^{-(1/2l_h^2)(\mathbf{r}_h-\mathbf{c}_h)^2}, \tag{10}$$

which keeps the factorized form in terms of $\mathbf{R},\mathbf{r}$ too,

$$\psi(\mathbf{R},\mathbf{r}) \sim e^{-(1/2L^2)(\mathbf{R}-\mathbf{C})^2} e^{-(1/2l^2)(\mathbf{r}-\mathbf{c})^2}, \tag{11}$$

provided $\mathbf{R}$ and $\mathbf{r}$ are given by Eq. (8) and the weighting factors are proportional to the inverse square of the oscillator lengths $\lambda_i \sim 1/l_i^2 \sim m_i\omega_i$. More precisely

$$\lambda_i = \frac{m_i\omega_i}{m_e\omega_e + m_h\omega_h}, \quad i \in \{e,h\}, \tag{12}$$

and $l^2=l_e^2+l_h^2$, $1/L^2=1/l_e^2+1/l_h^2$. $\mathbf{C}$ and $\mathbf{c}$ are obtained from $\mathbf{c}_e$ and $\mathbf{c}_h$ in the same way as $\mathbf{R},\mathbf{r}$ are determined by $\mathbf{r}_e,\mathbf{r}_h$.

In the new coordinates given by Eq. (8), with the weighting factors of Eq. (12), the Hamiltonian Eq. (1) takes the form

$$H = \frac{1}{2M}\mathbf{P}^2 + \frac{1}{2}M\Omega^2\mathbf{R}^2 + \frac{1}{2\mu}\mathbf{p}^2 + \frac{1}{2}\mu\omega^2\mathbf{r}^2 - e\mathbf{F}\cdot\mathbf{r} + V_C(r) - (\omega_e - \omega_h)\left[\frac{1}{M\Omega}\mathbf{P}\cdot\mathbf{p} + \mu\omega\mathbf{R}\cdot\mathbf{r}\right]. \quad (13)$$

The Coulomb interaction V_C is given by the last term in Eq. (1), the momenta $\mathbf{P}$ and $\mathbf{p}$ are conjugated to the coordinates $\mathbf{R}$ and $\mathbf{r}$, and the frequencies Ω and ω for the central and relative motion, respectively, are weighted means of ω_e and ω_h:

$$\Omega = \frac{l_h^2}{l^2}\omega_e + \frac{l_e^2}{l^2}\omega_h, \quad \omega = \frac{l_e^2}{l^2}\omega_e + \frac{l_h^2}{l^2}\omega_h. \quad (14)$$

The two masses are defined via $M\Omega/\hbar = 1/L^2$ and $\mu\omega/\hbar = 1/l^2$. Note that μ is the reduced mass but in general M is not the total mass (actually $M < m_e + m_h$ for $\omega_e \neq \omega_h$).

1. Exact solution for $\omega_e = \omega_h$

The last line of Eq. (13), which contains the term mixing the central and the relative motions, vanishes for $\omega_e = \omega_h = \omega$. In such circumstances the separation of the two motions becomes exact. Note also that in this case $\mathbf{R}$ corresponds to the c.m. The c.m. motion is defined by a simple oscillator Hamiltonian, and contributes to the ground-state energy with $\hbar\Omega$ which, in this case, is equal to the common energy $\hbar\omega$.

The Coulomb interaction and the external field are present only in the RM Hamiltonian, given by the second row of Eq. (13), which contains the field-displaced oscillator potential and a Coulomb attractive center. For this one-particle problem a full solution, exact to the numerical accuracy but otherwise not containing further approximations, is accessible. We have obtained the ground-state energy and the corresponding wave function by using a relaxation method.[30,31]

2. Variational ansatz II

Equation (11) suggests a variational approach in which the trial function has the form

$$\psi(\mathbf{R},\mathbf{r}) = \Phi(\mathbf{R})\varphi(\mathbf{r}), \quad (15)$$

with the central motion described by a shifted Gaussian

$$\Phi(\mathbf{R}) = \frac{1}{L\sqrt{\pi}}e^{-(1/2L^2)(\mathbf{R}-\mathbf{R}_0)^2}. \quad (16)$$

The spatial displacement follows the direction of the field $\mathbf{R}_0 = (X_0, 0)$ by an amount X_0, which is a variational parameter. We leave the RM function $\varphi(\mathbf{r})$ completely arbitrary.

The minimization problem for the expectation value $\langle\psi|H|\psi\rangle$ with respect to $\varphi(\mathbf{r})$ is equivalent to finding the ground-state energy of the following RM effective Hamiltonian obtained by "averaging out" the central coordinate

$$H_{\text{eff}}(\mathbf{r},\mathbf{R}_0) = \langle\Phi(\mathbf{R})|H|\Phi(\mathbf{R})\rangle = \hbar\Omega + \frac{1}{2}M\Omega^2\mathbf{R}_0^2 + \frac{\mathbf{p}^2}{2\mu} + \frac{\mu\omega^2}{2}\mathbf{r}^2 - [e\mathbf{F} + (\omega_e - \omega_h)\mu\omega\mathbf{R}_0]\mathbf{r} + V_C(r). \quad (17)$$

One sees that the central motion contributes to the energy with a term which is quadratic in its displacement, but also provides an additional field term in the RM problem. Except for these changes, H_{eff} is of the same form as the RM Hamiltonian discussed in the case of equal frequencies. Therefore, the exact solution described in the previous subsection can be used here. More precisely, if one denotes the ground state energy found for equal frequencies by $\mathcal{E}(F)$, then the minimum of the variational problem for arbitrary frequencies is given by

$$E(F) = \hbar\Omega + \min_{X_0}\left\{\mathcal{E}\left(F + \frac{\omega_e - \omega_h}{e}\mu\omega X_0\right) + \frac{M\Omega^2}{2}X_0^2\right\}. \quad (18)$$

Up to trivial terms, the functional to be minimized over X_0 is given by the ground-state energy of the equal frequency case. Therefore the latter is not only a particular case of our problem, but is also instrumental in solving the general case.

Of course, in the particular case when $\omega_e = \omega_h$ the solution of the variational problem Eq. (18) is $X_0 = 0$ and $E(F) = \mathcal{E}(F)$. Another situation in which the present variational approach recovers the exact result is the noninteracting case $V_C = 0$.

D. Results and discussion

For the purpose of comparing the different approaches, we consider first equal confinement energies $\hbar\omega_e = \hbar\omega_h$. Later we give an example involving different confinement energies and discuss how this affects the results. All energies are given in units of $\hbar\omega$, as defined in Eq. (14) and for the length unit we use $l_0 = \sqrt{\hbar/2\mu\omega}$. Then we have the dimensionless quantities $E_C = \frac{1}{\hbar\omega}\frac{e^2}{4\pi\epsilon_0\epsilon_r l_0}$ to characterize the strength of the Coulomb coupling and $\eta = \frac{eFl_0}{\hbar\omega}$ for the electric field. Typical results of different approaches for the energy versus electric field curve for $E_C = 2$ are shown in Fig. 1.

In the absence of the Coulomb interaction, one finds for the ground-state energy a quadratic dependence on the electric field $E(\eta) = 2 - \eta^2$. Due to the attraction between the electron and the hole the ground-state energy is lowered with respect to the noninteracting solution. The failure of the perturbative approach is made obvious by the nonconcavity of the ground-state energy as a function of η. As discussed earlier such a behavior is unphysical.

The variational procedure based on a product of Gaussians in the electron and hole coordinates as described in Sec. II B yields an even lower ground-state energy than the perturbative result and is concave everywhere. In contrast to the behavior of the noninteracting electron-hole pair energy, one finds in this approach a plateau for small η. In this region the energy varies only slightly with the applied field. For larger field strengths a transition occurs after which the energy follows closely the noninteracting result $2 - \eta^2$.

The numerically exact result, obtained using the procedure of Sec. II C 1, shows qualitatively the same behavior as the variational procedure, but is shifted to lower energies and the transition point occurs only at higher fields. That the plateau region cannot exist for arbitrarily large fields is sim-

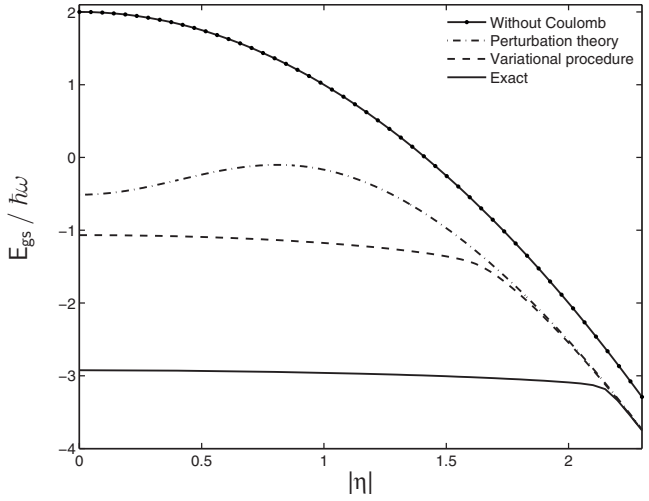

FIG. 1. Ground-state energy versus dimensionless electric field η, in the equal confinement energy case, calculated by the methods described in the text. For reference, the noninteracting result is also given.

ply due to the fact that the true energy must always be smaller than the noninteracting energy, because of the attractive interaction between the electron and the hole. In Fig. 2 the field dependence of excitonic ground-state energy obtained by the full numerical solution of Sec. II C 1 is shown for various values of the Coulomb interaction E_C. As the Coulomb interaction is increased, the ground state is shifted to lower energies. Correspondingly the transition point occurs only for higher field strength. Furthermore the transition becomes sharper as E_C is increased.

The transition between the Coulomb-dominated regime and the quasinoninteracting behavior can be directly seen in the electron-hole separation

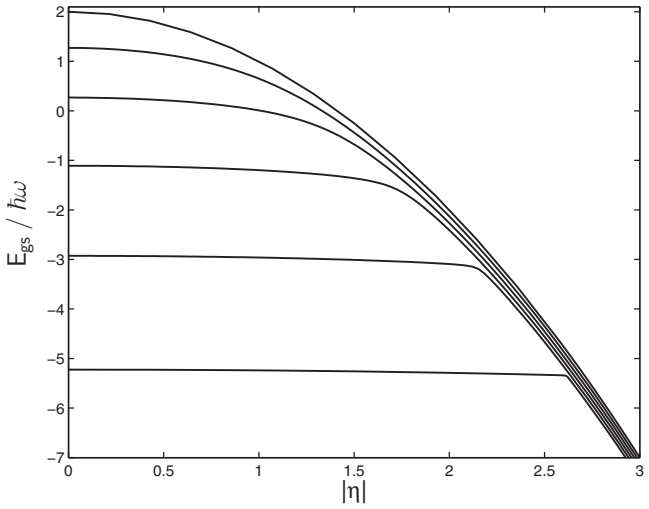

FIG. 2. Ground-state energy as a function of the dimensionless electric field η, for various values of the Coulomb interaction E_C, calculated as the solid line of Fig. 1. From top to bottom E_C varies from 0 to 2.5 in steps of 0.5. For larger E_C one can more clearly distinguish a plateau (due to dominating Coulomb interaction) from the nearly free regime.

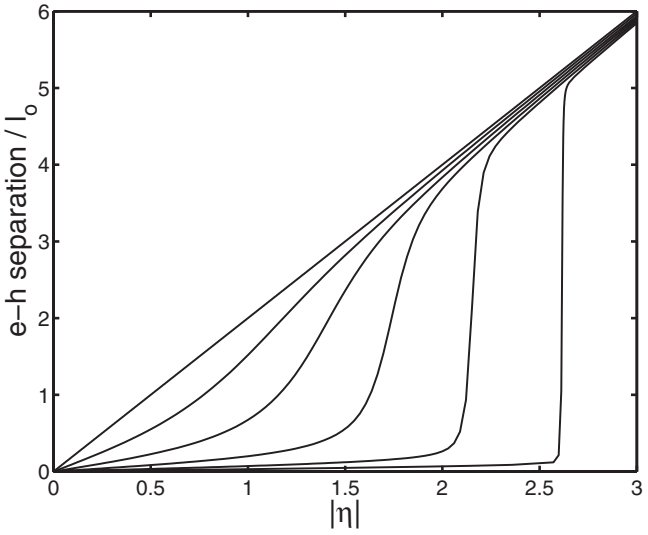

FIG. 3. Separation of electron and hole as a function of the dimensionless applied field η for different values of the relative strength of the Coulomb interaction E_C. From left to right: E_C varies from 0 to 2.5 in steps of 0.5. For larger values of E_C the transition point occurs only for higher electric fields and the transition becomes more abrupt.

$$d_{he} = \int d^2r\, x|\varphi(\mathbf{r})|^2 \tag{19}$$

as a function of the applied electric field. Results for different values of E_C are shown in Fig. 3. In the absence of the Coulomb interaction this quantity would be strictly proportional to the field, $d_{he}=c_{he}=2\eta$. With the Coulomb attraction included, however, the distance d_{he} is kept at much lower values in the small field region. As the field increases the carriers become sufficiently separated for their interaction to become negligible and the noninteracting result is recovered. The stronger the Coulomb coupling the sharper the transition and the later it takes place.

An intuitive picture of the two-regime behavior can be obtained if one has in mind the competition between the two attractive potentials present in the RM Hamiltonian: the Coulomb term centered at $\mathbf{r}=0$ and the field-displaced parabolic potential. In Fig. 4 they are shown for various field values. If, for the sake of the argument, we consider them separately, each provides a ground-state energy indicated by a horizontal bar. For weak fields the Coulomb potential yields a lower ground state and is preferred, but as the field increases the oscillator ground state gradually decreases and eventually the system jumps into this state. If the Coulomb coupling E_C is large this transition takes place when the two competing states are sufficiently far apart to be practically orthogonal, and then the change of regime is in fact an anticrossing of two noninteracting levels, and therefore quite sharp. For lower values of E_C the admixture of the two states is smoothening the transition.

In Fig. 5 a cross section of the wave function is shown for $E_C=1$. For low fields the strong Coulomb character of the wave function is seen in the cusp produced by the Coulomb

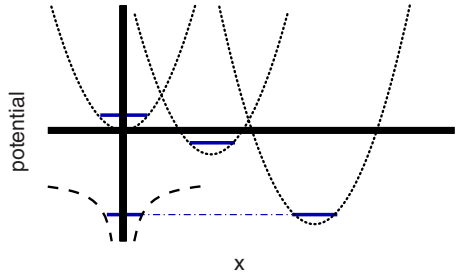

FIG. 4. (Color online) Sketch of the Coulomb potential together with a series of harmonic confinement potentials shifted by the electric field. The ground states provided separately by each potential are indicated by the horizontal bars. For weaker fields the Coulomb potential yields a lower ground-state energy than the shifted oscillator potential. For a certain field strength the two energies are at level. This roughly marks the transition point between the two regimes discussed in the text. For even higher fields the shifted oscillator yields the lowest energy.

singularity and in the exponential decay away from it. In the large field limit a Gaussian function is obtained, as expected. The transition between these limiting cases is gradual and involves a strong mixing of the corresponding solutions.

Turning now to the more general problem of different confinement energies we consider, for the sake of illustration, the case of equal electron and hole envelopes $l_e=l_h=l_0$. Then the frequency ratio is given by $\omega_e/\omega_h=m_h/m_e$. In our example we take $m_e=0.065m_0$ and $m_h=0.17m_0$, which are typical values for the InGaAs system. The results are shown in Fig. 6 for several values of E_C. It is seen that qualitatively the behavior remains unchanged, the main effect of having different confinement energies being that the transition point occurs at lower field strengths. This can be explained by the fact that the parabola of the noninteracting system is steeper in this case.

The plateau corresponding to the low-field regime is characterized by a reduction of the polarizability from its noninteracting value by one and even two orders of magnitude when increasing the E_C value as in Fig. 6. If the noninteracting polarizability has typical values of 10^{-2} meV cm^2/kV^2 then one obtains polarizabilities in the range of 10^{-3} to 10^{-4} meV cm^2/kV^2. Such low values of the QCSE have been observed in experiments and were sometimes attributed to the weak potential drop felt by the QD in the case of Schottky contacts[20] but our results show that a small Stark shift can also be an intrinsic effect.

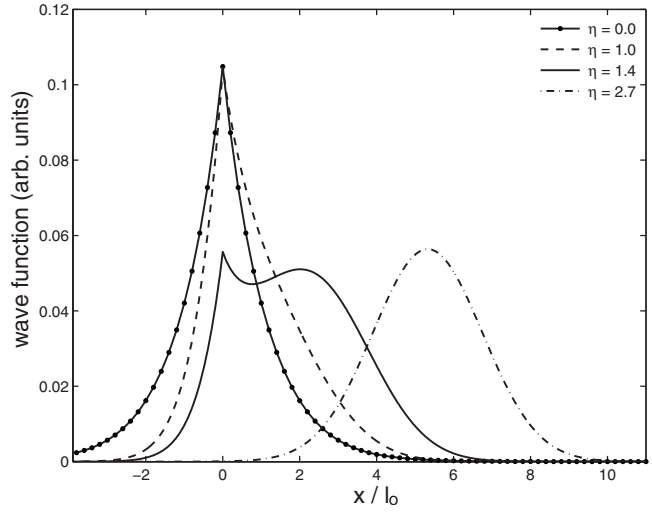

FIG. 5. Cross section of the ground-state wave function for $E_C=1$ and different values of the applied bias. A transition from the Coulomb dominated state for small fields to the shifted harmonic oscillator state for large fields can be observed.

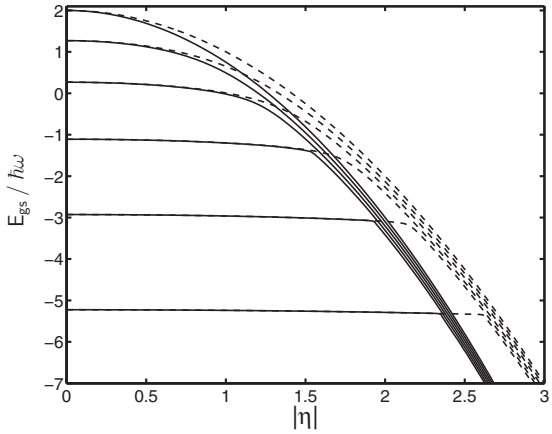

FIG. 6. Ground-state energy as a function of the dimensionless electric field η for different confinement energies (solid) and equal confinement energies (dashed). From top to bottom the relative strength of the Coulomb interaction E_C varies from 0 to 2.5 in steps of 0.5. For different confinement energies the transition point occurs at smaller fields.

III. SUMMARY AND CONCLUSION

The problem of the excitonic ground state in a QD with parabolic confinement and a lateral external electric field was considered. The interplay between these potentials on the one hand, and the Coulomb attraction on the other, turns out to be a nontrivial problem. This was illustrated by showing that simple approaches can be too crude to be accurate.

The inclusion of the Coulomb interaction at the perturbative level can lead to wrong results, and even change the sign of the Stark effect. This is due to an inconsistency in the procedure: the expectation value of the total Hamiltonian is evaluated for states which minimize the expectation value of the noninteracting one. In this picture the electron and the hole are described as following faithfully the displacement of their oscillator center, while in reality they lag behind, being kept close together by the Coulomb attraction (see Fig. 3). As a consequence, the perturbative approach may lead to negative polarizabilities, in contradiction with the concavity theorem concerning the bias dependence of the ground state. This casts a doubt on the perturbative result for the polarizability even when still positive. One may also consider in this problem a configuration interaction scheme, using a limited number of displaced oscillator states. This may improve the picture but, by the same token, is not guaranteed to meet the concavity requirement. Such procedures, albeit widely

employed for calculating QD emission and absorption spectra, should be used with caution in this case.

The concavity requirement is automatically met by consistent variational procedures (with bias-independent variational reservoirs), as illustrated by our variational ansatz I. Yet the ground-state energies obtained in this scheme are not quite satisfactory, due to the choice of the trial functions. Simple Gaussians are not good approximates for wave functions such as those of Fig. 5, especially at low fields.

A more accurate variational procedure (ansatz II) is proposed instead. It is still numerically simple and is exact in two limiting cases: in the absence of the Coulomb interaction and in the case of equal electron and hole single-particle energies. The method predicts a nontrivial, two-regime behavior. For low fields the ground state is dominated by the Coulomb attraction, while for large fields it is oscillatorlike. The transition between the two can be sharper or smoother, depending on the relative strength of the Coulomb interaction with respect to the one-particle energies.

ACKNOWLEDGMENTS

This work has been financially supported through the Deutsche Forschungsgemeinschaft. A grant for CPU time at the Forschungszentrum Jülich is gratefully acknowledged. The authors thank Peter Michler, Sven M. Ulrich, and Moritz Vogel for fruitful discussions.

[1] D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. Lett. **53**, 2173 (1984).
[2] D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. B **32**, 1043 (1985).
[3] Y. Miyake, J. Y. Kim, Y. Shiraki, and S. Fukatsu, Appl. Phys. Lett. **68**, 2097 (1996).
[4] S. Benner and H. Haug, Phys. Rev. B **47**, 15750 (1993).
[5] T. Arakawa, Y. Kato, F. Sogawa, and Y. Arakawa, Appl. Phys. Lett. **70**, 646 (1997).
[6] G. W. Wen, J. Y. Lin, H. X. Jiang, and Z. Chen, Phys. Rev. B **52**, 5913 (1995).
[7] W. Heller, U. Bockelmann, and G. Abstreiter, Phys. Rev. B **57**, 6270 (1998).
[8] P. W. Fry, I. E. Itskevich, D. J. Mowbray, M. S. Skolnick, J. J. Finley, J. A. Barker, E. P. O'Reilly, L. R. Wilson, I. A. Larkin, P. A. Maksym, M. Hopkinson, M. Al-Khafaji, J. P. R. David, A. G. Cullis, G. Hill, and J. C. Clark, Phys. Rev. Lett. **84**, 733 (2000).
[9] P. W. Fry, I. E. Itskevich, S. R. Parnell, J. J. Finley, L. R. Wilson, K. L. Schumacher, D. J. Mowbray, M. S. Skolnick, M. Al-Khafaji, A. G. Cullis, M. Hopkinson, J. C. Clark, and G. Hill, Phys. Rev. B **62**, 16784 (2000).
[10] J. Seufert, M. Obert, M. Scheibner, N. A. Gippius, G. Bacher, A. Forchel, T. Passow, K. Leonardi, and D. Hommel, Appl. Phys. Lett. **79**, 1033 (2001).
[11] R. Rinaldi, M. DeGiorgi, M. DeVittorio, A. Melcarne, P. Visconti, R. Cingolani, H. Lipsanen, M. Sopanen, T. Drufva, and J. Tulkki, Jpn. J. Appl. Phys., Part 1 **40**, 2002 (2001).
[12] A. Rodrguez and C. Trallero-Giner, Phys. Status Solidi B **230**, 463 (2002).
[13] G. Bacher, H. Schmig, J. Seufert, M. Rambach, A. Forchel, A. A. Maksimov, V. D. Kulakovskii, T. Passow, and D. Hommel, Phys. Status Solidi B **229**, 415 (2002).
[14] R. Oulton, J. J. Finley, A. D. Ashmore, I. S. Gregory, D. J. Mowbray, M. S. Skolnick, M. J. Steer, S.-L. Liew, M. A. Migliorato, and A. J. Cullis, Phys. Rev. B **66**, 045313 (2002).
[15] J. J. Finley, M. Sabathil, P. Vogl, G. Abstreiter, R. Oulton, A. I. Tartakovskii, D. J. Mowbray, M. S. Skolnick, S. L. Liew, A. G. Cullis, and M. Hopkinson, Phys. Rev. B **70**, 201308(R) (2004).
[16] J. W. Robinson, J. H. Rice, K. H. Lee, J. H. Na, R. A. Taylor, D. G. Hasko, R. A. Oliver, M. J. Kappers, C. J. Humphreys, and G. A. D. Briggs, Appl. Phys. Lett. **86**, 213103 (2005).
[17] M. Ohmori, K. Torii, and H. Sakaki, Phys. Status Solidi C **3**, 512 (2006).
[18] T. Nakaoka, S. Kako, and Y. Arakawa, Phys. Rev. B **73**, 121305(R) (2006).
[19] B. D. Gerardot, S. Seidl, P. A. Dalgarno, R. J. Warburton, D. Granados, J. M. Garcia, K. Kowalik, O. Krebs, K. Karrai, A. Badolato, and P. M. Petroff, Appl. Phys. Lett. **90**, 041101 (2007).
[20] K. Kowalik, O. Krebs, A. Lemaitre, S. Laurent, P. Senellart, P. Voisin, and J. A. Gaj, Appl. Phys. Lett. **86**, 041907 (2005).
[21] G. Ortner, M. Bayer, Y. Lyanda-Geller, T. L. Reinecke, A. Kress, J. P. Reithmaier, and A. Forchel, Phys. Rev. Lett. **94**, 157401 (2005).
[22] G. Bester and A. Zunger, Phys. Rev. B **72**, 165334 (2005).
[23] H. J. Krenner, M. Sabathil, E. C. Clark, A. Kress, D. Schuh, M. Bichler, G. Abstreiter, and J. J. Finley, Phys. Rev. Lett. **94**, 057402 (2005).
[24] H. J. Krenner, E. C. Clark, T. Nakaoka, M. Bichler, C. Scheurer, G. Abstreiter, and J. J. Finley, Phys. Rev. Lett. **97**, 076403 (2006).
[25] M. F. Doty, M. Scheibner, I. V. Ponomarev, E. A. Stinaff, A. S. Bracker, V. L. Korenev, T. L. Reinecke, and D. Gammon, Phys. Rev. Lett. **97**, 197202 (2006).
[26] G. J. Beirne, C. Hermannstadter, L. Wang, A. Rastelli, O. G. Schmidt, and P. Michler, Phys. Rev. Lett. **96**, 137401 (2006).
[27] W. Thirring, *Quantum Mathematical Physics* (Springer, Heidelberg, 2002).
[28] D. M.-T. Kuo, Jpn. J. Appl. Phys., Part 1 **45**, 3927 (2006).
[29] A. Wojs, P. Hawrylak, S. Fafard, and L. Jacak, Phys. Rev. B **54**, 5604 (1996).
[30] W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes: The Art of Scientific Computing* (Cambridge University Press, Cambridge, 1992).
[31] C. Presilla and U. Tambini, Phys. Rev. E **52**, 4495 (1995).
[32] These values are only indicative since the precise values of the field at the QD are difficult to assess (Refs. 11 and 20).