# Stark shift in single and vertically coupled type-I and type-II quantum dots

K. L. Janssens,* B. Partoens,† and F. M. Peeters‡
*Departement Natuurkunde, Universiteit Antwerpen (UIA), Universiteitsplein 1, B-2610 Antwerpen, Belgium*
(Received 22 March 2002; published 23 May 2002)

We theoretically studied the Stark effect on an exciton in single and vertically coupled type-I and type-II quantum dots. Only for the single type-I dot does the exciton energy show a parabolic dependence on the applied field. A dipole moment is induced by the applied electric field for vertically coupled type-I dots and for type-II dots, leading to a linear dependence of the Stark shift on the electric field. Furthermore, we predict that spontaneous symmetry breaking can occur for vertically coupled type-II dots leading to a permanent dipole moment.

DOI: 10.1103/PhysRevB.65.233301 PACS number(s): 73.21.La, 78.67.Hc

In recent years self-assembled quantum dots (QD's) have attracted considerable interest because they can be regarded as ideal model systems for quasi-zero-dimensional systems.[1] Therefore, QD's are often called "artificial atoms." Self-assembled QD's are expected to have applications in optoelectronic devices. Many effects known from atomic physics were found in QD's, and of particular interest is the effect of an electric field on an exciton in a quantum dot, the so-called Stark shift. From an application point of view, optical nonlinear effects[2] and usefulness for quantum computing[3] have been predicted, whereas from a basic physics point of view, the Stark effect can provide detailed insight into the interdot charge distribution. Recent experimental results[4,5] have shown that the Stark shift exhibits an asymmetry in the presence of an electric field, due to a built-in dipole moment. Theoretical studies[6,7] have shown that the sign of this dipole moment in single dots depends on the composition gradient and/or strain effects, localizing the electron and hole in different regions of the quantum dot. Vasanelli *et al.*[8] studied the system of very strong multiple stacked dots and showed that the Stark tunability in this system is much larger than for the single dot structure. Very recently, Sheng and Leburton[9] have found an anomalous behavior of the Stark shift for stacked quantum dots. They predicted that vertically coupled type-I dots exhibit a strong nonparabolic Stark shift. They claimed that this anomalous dependence of the Stark shift is caused by the three-dimensional strain field distribution which influences drastically the hole states in the stacked QD structure. Where all mentioned works, both experimental and theoretical, were based on type-I quantum dots, as, e.g., InAs/GaAs dots, very little attention has been paid to the electric-field effect on type-II dots, such as InP/InGaP or GaSb/GaAs dots where only one of the charge carriers is confined inside the dot.

In the present work we investigate the effect of an electric field on single and vertically coupled quantum dots, both for the type-I and the type-II systems. Our quantum dots are modeled by quantum disks of finite height and our calculations are based on a Hartree-Fock mesh calculation, done within the two-band model of the effective-mass approximation, i.e., strain effects and band mixing are presently neglected. The advantage of this approach is that the intricacies of the Stark effect due to the interdot coupling can be shown much more clearly. For example, this will lead to a different interpretation of the nonparabolic Stark effect as found in Ref. 9. We are only interested in the overall functional behavior of the Stark effect and therefore do not take into account the built-in dipole moment due to strain and/or composition, as it will mainly lead to a shift in the electric-field axis.

The energy and wave functions are obtained by solving the following Schrödinger equation:

$$(H_e+H_h+V_c)\Psi(\mathbf{r}_e,\mathbf{r}_h)=E\Psi(\mathbf{r}_e,\mathbf{r}_h), \tag{1}$$

with $V_c$ the Coulomb interaction term, and

$$H_{e,h}=\frac{p_{e,h}^2}{2m_{e,h}}+V_{e,h}(\mathbf{r}_{e,h})\mp eFz_{e,h}, \tag{2}$$

where $H_{e,h}$ and $V_{e,h}$ are, respectively, the single electron (hole) Hamiltonian and confinement potentials, and $F$ is the applied electric field. Our method of solving this problem is explained in more detail in Ref.10.

First we investigated the influence of the electric field on an exciton in a single type-I disk, where we took material parameters of InAs/GaAs dots,[11] $V_e=450$ meV, $V_h=316$ meV, $m_e=0.04m_0$, and $m_h=0.34m_0$. We model the quantum dot by a quantum disk, with radius $R=9.81$ nm and thickness $d=3.6$ nm, which are comparable to the dots studied by Sheng and Leburton[9].

Figure 1 depicts the result for the exciton energy as a function of the applied electric field, showing a parabolic dependence. Generally, the Stark shift varies quadratically with the applied field as

$$E(F)=E(F_0)-p(F-F_0)-\beta(F-F_0)^2, \tag{3}$$

which follows from a second-order perturbation theory with $p$ the dipole moment and $\beta$ the polarizability of the electron-hole system.[6] As both electron and hole are strongly confined inside the type-I disk, the electric field has only a very small effect on the wave functions (inset of Fig. 1, for $F=100$ kV/cm), leading to a dipole moment $p\approx 0$ at all field values. In the inset of Fig. 1, we plotted the density along the $z$ direction at $r=0$, i.e., $|\Psi_{e,h}(0,z)|^2$. As a consequence the second-order term, quadratic in the applied field, will be the most important, leading to the parabolic behavior of the Stark shift. Note that the Stark shift is symmetric around $F$

 

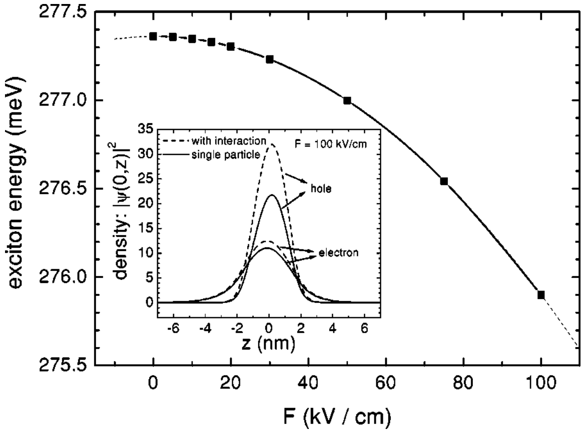


FIG. 1. The exciton energy as a function of the electric field for a single type-I disk. The dashed line indicates a quadratic fit. Inset: The electron and hole densities at $F=100$ kV/cm along the $z$ direction for $r=0$. The solid curves denote the single-particle densities, whereas the dashed curves show the results with interaction.

$=0$ kV/cm, as we have no built-in dipole moment. In order to obtain the parameters $p$ and $\beta$, we fitted our result to Eq. (3) with $p=0$ (dashed line in Fig. 1) and found $\beta=1.47\times10^{-3}$ $e$nm/(kV/cm).

We then extended our study to a system of two vertically coupled type-I disks. The same material and disk parameters were used as for the single disk, but we have now one extra parameter, the interdot distance $d_z$, which we took $d_z=1.8$ nm, as in Ref. 9. For this system, the Stark shift appears to be no longer quadratic (Fig. 2). In addition, due to the linear behavior, the stacked structure exhibits a Stark shift which is one order-of-magnitude larger than for the single dot! For the single type-I disk of Fig. 1 we found a Stark shift of $\Delta E=1.5$ meV at $F=100$ kV/cm which compares to $\Delta E=35$ meV for the two coupled disks system of Fig. 2. As a comparison we have (for $F=100$ kV/cm) $\Delta E=1.65$ meV for a polar CsF molecule, $\Delta E=2.06\times10^{-3}$ meV for a neutral Cs atom,[12] and $\Delta E\simeq20$ meV for 100-Å wide GaAs/AlGaAs quantum well structures.[13]

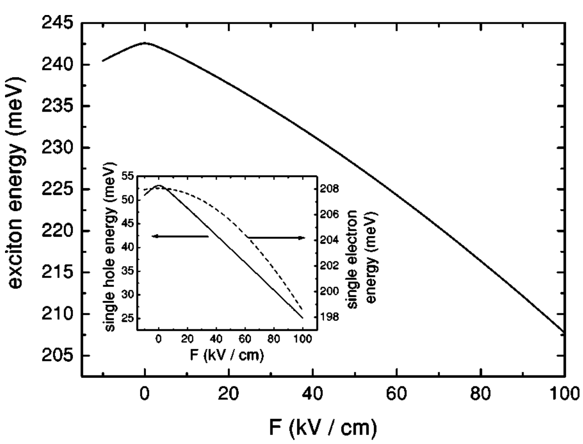


FIG. 2. (a) The exciton energy as a function of an electric field along the $z$ direction for two vertically coupled type-I disks. Inset: The single-particle energies for hole and electron as a function of the electric field. Note the linear behavior of the hole energy in contrast to the parabolic behavior of the electron energy.

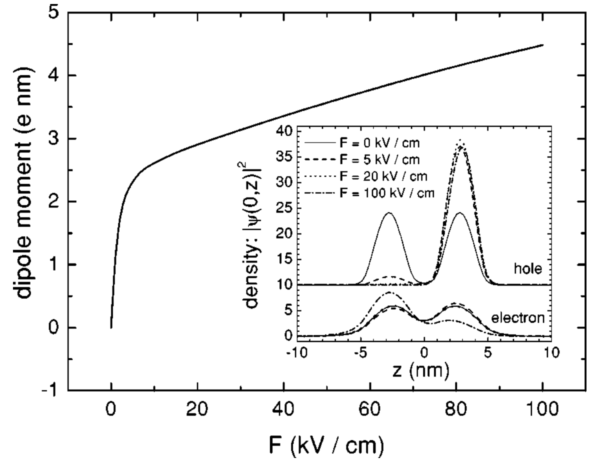


FIG. 3. The dipole moment as a function of the electric field for two coupled type-I disks. Inset: The electron and hole densities along the $z$ axis for different values of the electric field.

The reason for the nonparabolic behavior can be traced back to the strong effect of the electric field on the hole. For $F=0$ kV/cm, both electron and hole are equally spread over the two disks. The electric field draws the two particles apart. As the hole is a rather heavy particle, it is almost completely pushed towards the upper (lower) disk for positive (negative) electric field.

The electron is much less affected by the field, because it is much lighter and is therefore more tightly bound to the quantum disks. This can also be seen from the electric field dependence of the single hole and single electron energies (inset of Fig. 2). For noninteracting particles, the effect of the electric-field on the energy is described by $E_{e,h}(F)=E_{e,h}(0)+eF\langle z_{e,h}\rangle+\beta_{e,h}F^2$, with $\langle z_{e,h}\rangle$ the mean electron (hole) position along the $z$ axis,[6] which is zero for $F=0$. Already for small electric-field values, the hole has moved to one of the dots (see the inset of Fig. 3). Therefore, $\langle z_h\rangle$ has a finite value, and the first-order contribution will be most important, leading to an almost linear behavior (inset of Fig. 2). The electron, however, is only slightly influenced by the electric field. Therefore, the second-order term will be more important in this case, leading to the parabolic behavior of the electron energy with the electric field. We found that for very small fields, the electron tends to follow the hole as a consequence of the Coulomb attraction. For larger fields, however, i.e., $F\geqslant20$ kV/cm, the electric field overcomes the Coulomb attraction and the electron moves from the upper into the lower disk, which is illustrated in the inset of Fig. 3. Notice the changed position of the electron for different electric-field values. A direct consequence of the more pronounced effect of the electric field is the creation of a dipole moment, which was absent in the single disk case. The presence of this dipole moment implies that the first-order term, i.e., the term linear in $F$, is now the most important, leading to a linear behavior of the Stark shift. Only for very small electric fields, i.e., $F\leqslant2$ kV/cm, is the dipole moment negligible and the Stark shift still parabolic. Although the dipole

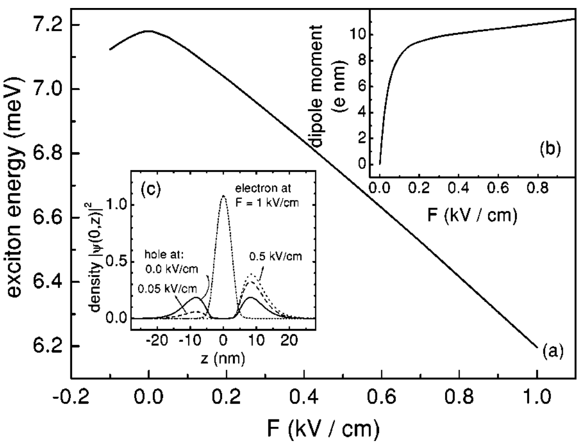

FIG. 4. The exciton energy (a) and dipole moment (b) as a function of the electric field for a single type-II disk. (c) The electron and hole densities along the $z$ axis for $r=0$. The electron density is shown only for $F=1$ kV/cm, since it changes very little with electric field.

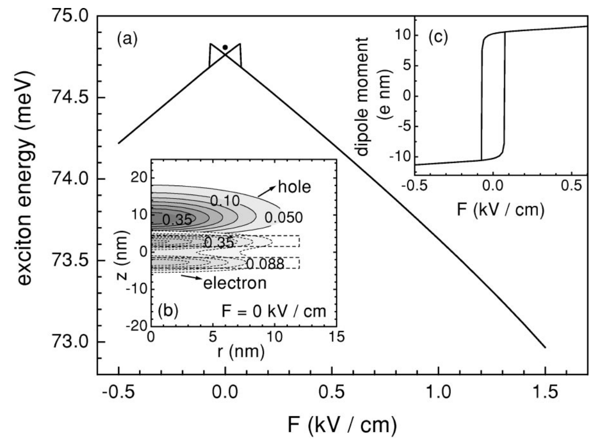

FIG. 5. (a) The exciton energy as a function of the electric field for two vertically coupled type-II disks. The dot at $F=100$ kV/cm is the exciton energy for a symmetric hole wave function. (b) The electron (dashed contour lines) and hole (solid contour lines) densities for $F=0$ kV/cm. Notice the asymmetry of both densities, although there is no electric field present. (c) The dipole moment as a function of the electric field.

moment depends on the applied field strength, an estimate of its average value can be obtained from a fit of the linear part of the Stark shift, i.e., $20\ \text{kV/cm} \leq F \leq 100\ \text{kV/cm}$, which gives $p=3.5\ e$ nm.

The dipole moment is connected with the mean electron-hole distance along the $z$ direction, i.e., $p=e(\langle z_h\rangle - \langle z_e\rangle)$. In Fig. 3 the dipole moment is depicted as a function of the electric field, showing a fast increase already for small electric fields. Remark that our calculated dipole moment agrees well with the one obtained by fitting the linear part of the exciton energy.

In Ref. 9, the different behavior of the Stark shift for single and coupled type-I disks was attributed to a difference in strain fields. However, we show here clearly that its origin lies in the tunneling between the two coupled dots which allows for the creation of a strong dipole moment which is not possible in the single dot case. Although strain fields are important in order to find more exact values of the exciton energy, they are only of secondary importance when explaining the origin of this Stark shift. This assertion is corroborated due to the fact that we found Stark shifts which are up to a factor-of-2 the same as found by Sheng and Leburton. For example, for $F=100$ kV/cm Sheng and Leburton found $\Delta E \simeq 40$ meV(3 meV) which compares to our result $\Delta E = 35$ meV(1.5 meV) for single (double) quantum dots.

After having discussed the type-I dots, we now turn to type-II dots, where we use parameters for the InP/InGaP dots, where the electron is confined inside the dot and the hole is located in the barrier. First we consider a single disk. Material and disk parameters are[14] $V_e=250$ meV, $V_h=-50$ meV, $m_e=0.077m_0$, $m_h=0.60m_0$, $R=10$ nm, and $d=8$ nm. Figure 4(a) shows that the Stark shift for this system also exhibits a nonparabolic behavior, in contrast to the previous result for the type-I disk. The origin of it can again be traced back to the existence of a dipole moment for larger fields. Note that the electric-field range for this type-II system is much smaller than for the type-I system, because the hole is only bound by the Coulomb interaction.

At $F=0$ kV/cm, the hole is sitting symmetrically above and below the disk, and consequently the dipole moment is zero at this point. Therefore the Stark shift in a small region around $F=0$ kV/cm will have a parabolical electric-field dependence. For larger fields, however, i.e., $F \geq 0.05$ kV/cm, the hole wave function is clearly asymmetric along $z$, and an appreciable dipole moment appears, as shown in Fig. 4(b). A fit of the linear part of the Stark shift, i.e., $0.2\ \text{kV/cm} \leq F \leq 1.0\ \text{kV/cm}$, gives us an estimate of the dipole moment, namely, $p=10.55\ e$ nm, or an electron-hole distance of $\langle z_h\rangle - \langle z_e\rangle = p/e = 10.55$ nm, which agrees well with the calculated dipole moment [see Fig. 4(b)] and the electron and hole densities [Fig. 4(c)].

We also investigated thinner type-II disks for which a smaller induced dipole moment is expected. We found indeed that the Stark shift became less linear.

Next we investigate the system of two vertically coupled type-II dots. We consider the same material parameters as discussed above, for disks with $R=12$ nm, $d=3$ nm, and an interdot distance $d_z=3$ nm. The exciton energy as a function of the applied electric field is depicted in Fig. 5(a), where one feature immediately catches the eye, namely, the appearance of hysteresis. This hysteresis is due to a *spontaneous symmetry breaking*, as we found recently in Ref. 15. The state with the asymmetric wave function has a lower energy than the symmetric state [dot in Fig. 5(a)]. This is a consequence of an enhancement of the Coulomb attraction for the asymmetric case [see Ref. 15]. When we increase the electric field very slowly, the system is trapped in this energy minimum and as long as the system remains trapped, the exciton energy increases. The effect on the wave function is shown in the inset of Fig. 5(b), where one can see that even for $F=0$ kV/cm the hole wave function (solid contour lines) is strongly asymmetric, and that the electron (dashed contour lines) tends to follow the hole. As a consequence, this leads to a permanent dipole moment. Without symmetry breaking the dipole moment would be zero for $F=0$ kV/cm. The spontaneous symmetry broken state is for $F=0$ kV/cm doubly degenerate [see Fig. 5(a)]: the hole can sit above or below the stack of vertically coupled dots. This leads to a hysteretic behavior for the dipole moment as a function of the electric field [Fig. 5(c)]. Starting from positive electric-field values we find a positive value for the dipole moment $p=e(\langle z_h\rangle-\langle z_e\rangle)$, i.e., the hole is located on top of the stacked disk system. Decreasing the electric field leads to a slow decrease of the dipole moment, as the relative distance between the electron and the hole decreases. At $F=0$ kV/cm, the dipole moment is still strongly positive, as the main part of the hole wave function is still located on top of the system. When further decreasing the electric field, as long as the system remains trapped in the energy minimum, the wave function remains at this position, although one would expect the negative field to attract the hole towards the bottom of the stacked disk system. Only when the electric field is large enough to pull the system out of the trap does the hole suddenly jump to its expected place, and the dipole moment suddenly reverses its sign, i.e., becomes negative. When we start from negative electric-field values and increase the field, we find similar behavior, leading to the observed hysteresis in Fig. 5(a). This behavior of the dipole moment is of particular interest as it should be measurable experimentally.

To conclude, we find that only for the single type-I quantum dot does the Stark shift of the exciton energy exhibit a quadratic dependence on the electric field, whereas for vertically coupled type-I dots and both for single and vertically coupled type-II dots is a strongly linear component in the Stark shift predicted. We attribute this linear dependence to the appearance of a strong dipole moment. This is a consequence of the electric field which leads to a tunneling of the hole to one side of the dot structure inducing a dipole moment.

K. L. J. is supported by the Instituut voor de Aanmoediging van Innovatie door Wetenschap en Technologie in Vlaanderen (IWT-Vl) and B. P. is a post-doctoral researcher with the Flemish Science Foundation (FWO-Vl). Part of this work was supported by the FWO-Vl, The Belgian Interuniversity Attraction Poles (IUAP), the Flemish Concerted Action (GOA) Program, the University of Antwerp (VIS), and the European Commission GROWTH program NANOMAT project, Contract No. G5RD-CT-2001-00545.

*Electronic mail: karen.janssens@ua.ac.be
†Electronic mail: bart.partoens@ua.ac.be
‡Electronic mail: francois.peeters@ua.ac.be

[1] For a review, see, e.g., D. Bimberg, M. Grundmann, and N. N Ledentsov, in *Quantum Dot Heterostructures* (Wiley, New York, 1999).
[2] S. Schmitt-Rink, D.A.B. Miller, and D.S. Chemla, Phys. Rev. B **35**, 8113 (1987).
[3] E. Biolatti, R.C. Iotti, P. Zanardi, and F. Rossi, Phys. Rev. Lett. **85**, 5647 (2000).
[4] 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).
[5] A. Patanè, A. Levin, A. Polimeni, F. Schindler, P.C. Main, L. Eaves, and M. Henini, Appl. Phys. Lett. **77**, 2979 (2000).
[6] J.A. Barker and E.P. O'Reilly, Phys. Rev. B **61**, 13 840 (2000).
[7] W. Sheng and J.-P. Leburton, Phys. Rev. B **63**, 161301 (2001).
[8] A. Vasanelli, R. Ferreira, H. Sakaki, and G. Bastard, Solid State Commun. **118**, 459 (2001).
[9] W. Sheng and J.-P. Leburton, Phys. Rev. Lett. **88**, 167401 (2002).
[10] K.L. Janssens, B. Partoens, and F.M. Peeters, Phys. Rev. B **64**, 155324 (2001).
[11] M. Califano and P. Harrison, Phys. Rev. B **61**, 10 959 (2000).
[12] J.A. Maddi, T.P. Dinneen, and H. Gould, Phys. Rev. A **60**, 3882 (1999).
[13] 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).
[14] C. Pryor, M.-E. Pistol, and L. Samuelson, Phys. Rev. B **56**, 10 404 (1997).
[15] K.L. Janssens, B. Partoens, and F.M. Peeters, cond-mat/0201026 (unpublished).