Field-induced Coulomb coupling in semiconductor macroatoms:
Application to “single-electron” quantum devices

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Abstract

A novel approach for the control of exciton-exciton Coulomb coupling in semiconductor macroatoms/molecules is proposed. We show that by applying properly tailored external fields, we can induce —or significantly reinforce— excitonic dipoles, which in turn allows to control and magnify intra- as well as inter-dot few-exciton effects. Such dipole-dipole interaction mechanism will be accounted for within a simple analytical model, which is found to be in good agreement with fully three-dimensional calculations. The proposed approach may play an important role for the design and realization of fully-optical quantum gates as well as ultrafast optical switches.
In recent years semiconductor nanostructures have gathered enormous attention. In particular, the ability of building zero-dimensional systems, such as semiconductor quantum dots (QD’s), led to a technological revolution: QD’s applications in fact range from laser emitters to charge-storage devices, from fluorescent biological markers to quantum information processing devices. With QD’s, the flexibility in controlling carrier densities reaches its extreme: it is possible to inject in a QD even just a single electron or a single exciton. Their discrete energy spectrum, on the other side, results in a rich optical response and in a weak interaction of the quantized carrier system with environmental degrees of freedom (phonons, plasmons, etc). At the same time, their reduced spatial extension —up to few nanometers— leads to a significant increase of Coulomb interactions among carriers. This, together with the granular nature of charge injection/photogeneration previously mentioned, leads to pronounced few-particle effects.

In this Letter we shall show how an external electric field can be used as a simple, effective way to tailor few-exciton interactions in single as well as coupled QD structures. This fine-tuning possibility may allow for important technological applications, like all-optical quantum gates and ultrafast optical switches. As we shall see, the proposed field-induced effects may be easily understood and quantitatively estimated using the simple analytical model described below.

Let’s consider first a single QD structure. In the usual situation, the electron and hole charge distributions corresponding to the same excitonic state are spatially superimposed. Assuming that the typical lengths associated to the confining potential for electron and hole are the same, it is possible to show that there is no net Coulomb interaction among excitons belonging to the same “shell”. In particular, then, if we consider two excitons with opposite spins in the lower energy state, the associated biexcitonic shift is equal to zero. Let’s apply now a constant electric field. This will pull apart charges of opposite sign, creating an electrical dipole for each exciton. It follows that the net Coulomb interaction among excitons is now different from zero even in the same shell. In this way an external electric field, removing part of the system symmetries, allows us to turn on and to tune exciton-exciton interactions.

A similar argument can be applied to the interaction between excitons in a coupled QD structure, i.e., to a semiconductor macromolecule. Even if charge distributions for electrons and holes in the same shell are somewhat different, the interaction between excitons sitting in different and far enough QD’s will be negligible. If we now apply a constant in-plane electric field (see below), we create again electron-hole dipoles inside each QD. Therefore, the polarized excitons will now interact with a strength that is roughly proportional to the square of the field-induced excitonic dipole. Once more, the presence of the field is found to turn on exciton-exciton interactions, thus allowing for the formation of tunable bonds between QD’s, i.e., artificial macromolecules.

The external field can be even used to turn off Coulomb interactions. Let’s consider for simplicity a single exciton in a QD: what happens if we keep increasing the external electric field? After the regime of strong electron-hole Coulomb interaction just described, the electric field will start to dominate, and the Coulomb correlation becomes a mere perturbation, up to the point in which the electron-hole charge separation is so large that the two particles can be described as non-interacting.

In order to test the viability of the proposed scheme, we have performed a realistic
calculation\(^2\) of field-induced exciton-exciton interaction in a GaAs-based coupled QD structure. The upper inset of Fig. 1 shows the electron and hole particle distributions corresponding to the excitonic ground state along the field direction: we can clearly recognize the field-induced electron-hole charge separation. Figure 1 reports the dipole-dipole coupling energy, i.e., the biexcitonic shift,\(^3\) as a function of the in-plane field \(E\). Here, our exact calculation (squares) is compared to the result of the model (solid curve) described below. As we can see, we obtain energy shifts of the order of a few meV, fully compatible with the typical resolution of current ultrafast spectroscopy.\(^4\) We underline that, in order to minimize the loss of oscillator strength due to the field-induced charge separation, a careful optimization of the system parameters is needed.

We shall now show that the field-induced effects previously introduced can be described in terms of a simple analytical model. To this purpose, the QD carrier confinement along the growth \((z)\) direction can be modeled as a narrow harmonic potential (or as a square box) \(V(z)\); the confinement in the QD \((\vec{r})\) plane is described as a two-dimensional (2D) parabolic potential. By denoting with \(E\) the in-plane electric field, the single-exciton Hamiltonian will then be

\[
\mathbf{H} = \sum_{i=e,h} \left[ \frac{\mathbf{p}_i^2}{2m_i} + \frac{1}{2} m_i \omega_i^2 |\vec{r}_i| + \epsilon_i + V_i(z_i) \right] - \frac{\epsilon^2}{\epsilon \sqrt{|\vec{r}_e - \vec{r}_h|^2 + |z_e - z_h|^2}},
\]

where the ± sign and the subscripts \(e\) and \(h\) refer, respectively, to electron and hole. Here, \(\vec{d}_i = \frac{eE}{m_i \omega_i^2}\) is the single-particle charge displacement induced by the field. We want to show that, under suitable conditions, Eq. (1) can be analytically solved and all the important quantities can be easily estimated with a good degree of accuracy.

Due to the strong single-particle confinement along the \(z\) direction, we will approximate \(|z_e - z_h|^2\) with its average value \(\lambda_z^2\). It is then possible to separate the Hamiltonian (1) as \(\mathbf{H} = \mathbf{H}_\parallel(\{\vec{r}_i^\parallel\}) + \mathbf{H}_\perp (z_i) + \mathbf{H}_\perp (z_h)\) where \(\mathbf{H}_\perp (z_i) = \frac{\mathbf{p}_i^2}{2m_i} + V_i(z_i)\) is the single-particle Hamiltonian along the growth direction —exactly solvable for the case of a parabolic potential as well as of an infinite-height square well. By further defining the center of mass (CM) and relative coordinates \(\vec{R} = \frac{m_e \vec{r}_e + m_h \vec{r}_h}{M} (M = m_e + m_h)\) and \(\vec{r} = \vec{r}_h - \vec{r}_e\), the in-plane Hamiltonian \(\mathbf{H}_\parallel(\{\vec{r}_i^\parallel\})\) becomes

\[
\mathbf{H}_\parallel(\vec{R}, \vec{r}) = \frac{P^2}{2M} + \frac{1}{2} M \omega_R^2 R^2 + \frac{p}{2 \mu} + \frac{1}{2} \mu \omega_e^2 |\vec{d} - \vec{r}|^2 + \mu (\omega_e^2 - \omega_h^2) \vec{R} \cdot (\vec{d} - \vec{r}) - \frac{\epsilon^2}{\epsilon \sqrt{r^2 + \lambda_z^2}},
\]

where \(\mu = \frac{m_e m_h}{M}\) is the reduced mass, \(\omega_R^2 = \frac{\omega_e^2 + \omega_h^2}{2} (1 + \Delta)\), \(\omega_e^2 = \frac{\omega_e^2 + \omega_h^2}{2} (1 - \Delta)\), \(\Delta = \frac{m_e - m_h}{m_e + m_h} \frac{\omega_e^2 - \omega_h^2}{\omega_e^2 + \omega_h^2}\) and \(\vec{d} = \vec{d}_e + \vec{d}_h\) denotes the total electron-hole in-plane displacement.

In the limit \(\frac{\omega_e^2 - \omega_h^2}{\omega_e^2 + \omega_h^2} \ll 1\), the two coordinates are only weakly coupled, and the Schrödinger equation associated to the CM coordinate \(\vec{R}\) is exactly solvable. In the general case we will concentrate on the ground state, though the generalization to higher
states is straightforward. We can approximate the ground state of $H_{\parallel}$ as $\Psi(\vec{r}, \vec{R}) = \Psi_x(x) 1/(\lambda^2 \pi)^{1/2} e^{-\frac{x^2}{2\lambda^2_x}} 1/(\lambda^2 \pi)^{1/2} e^{-\frac{y^2}{2\lambda^2_y} e^{-\frac{\vec{r}^2}{2\lambda^2_R}}}$, where $x$ and $y$ denote, respectively, the components of $\vec{r}$ parallel and perpendicular to the field $\vec{E}$, $\lambda_r = \sqrt{\frac{\hbar}{\mu \omega_r}}$ and $\lambda_R = \sqrt{\frac{\hbar}{\mu \omega_R}}$. By averaging $H_{\parallel}$ over $\Psi(\vec{r}, \vec{R})$, we obtain the effective Hamiltonian $H_{\text{eff}} = \frac{1}{2} \hbar \omega_r + \hbar \omega_R + \frac{\vec{r}^2}{2a^2} + V_{\text{eff}}(x)$, characterized by the effective potential

$$V_{\text{eff}}(x) = \frac{1}{2} \mu \omega^2_r (x - d)^2 + V_C \left( \frac{x^2 + \lambda_x^2}{2 \lambda^2_r} \right),$$

with $V_C(u) = -\frac{e^2}{\sqrt{\pi \lambda} e^{au}} K_0(u)$, $K_0$ being the zero-order Bessel function.

If we are interested in the low-energy states, we can approximate $V_{\text{eff}}$ around its minimum $V_0$ with a parabolic potential, i.e., $V_{\text{eff}}(x) \approx V_0 + \frac{1}{2} \mu \omega^2(x - x_0)^2$. Within such approximation scheme, the eigenvalues and eigenfunctions of $H_{\text{eff}}$ can be evaluated analytically. As already pointed out, if the external field is strong enough (and we will quantitatively define “enough” later), the Coulomb attraction between electron and hole can be considered as a perturbation. In this regime its main effect is to reduce the electron-hole displacement from $d$ to $x_0$. For intermediate and strong fields, we can then write the effective displacement as $x_0 = d - \Delta x$, with $\Delta x \ll d$. In this regime the following analytical expression for $\Delta x$ is obtained:

$$\frac{\Delta x}{d} = -\frac{\lambda_r \exp(\xi)}{a^*} \sqrt{\pi} \left[ 1 - \frac{\lambda_r \exp(\xi)}{a^*} \frac{\Delta K}{d^2} A(\Delta K, K_1) + \Delta K \right],$$

with $\xi = (d^2 + \lambda^2_x)/2\lambda^2_r$, $K_1$ the first-order Bessel function, $\Delta K = K_0(\xi) - K_1(\xi)$, $A(\Delta K, K_1) = 2\Delta K + \frac{\xi}{\lambda_x^2} A(\Delta K, K_1)$, and $a^* = \hbar^2/\mu \omega_x$, the reduced Bohr radius. Notice that the prefactor $\frac{\lambda_r}{a^*} \exp(\xi)$ is a measure of the system confinement. In a similar way, setting $\tilde{\omega} = \omega_r + \Delta \omega$ in $\mu \tilde{\omega} = d^2 V_{\text{eff}}/dx^2 |_{x_0}$, we can calculate the effect of the Coulomb attraction on the potential shape. The result is cumbersome and not particularly highlighting; here we will report only the important limit $\frac{\lambda_x^2}{a^*} \ll 1$ (high fields), in which $\frac{\Delta \omega}{\omega_r} = -\frac{\Delta \omega}{d} \propto -\frac{\lambda_x^2}{a^*} \lambda_x^3$. The condition $\Delta x/d \lesssim 20\%$ quantitatively defines the “intermediate and strong” electric field regime. It is easy to show that, in the regime of interest, the correction on the wave function due to $\Delta \omega/\omega_r$ is negligible with respect to the correction given by the shift $\Delta x/d$.

Based on the analytical model proposed so far, we have investigated the biexcitonic shift previously discussed (see Fig. 1) more specifically, we have approximated the biexcitonic ground state as the product of two excitonic wavefunctions sitting in different dots. The latter, in turn, are taken as products of the in-plane wavefunctions times the ground-state wavefunctions along the $z$ direction. The desired biexcitonic shift $\Delta \epsilon$ is then obtained averaging the corresponding two-exciton Hamiltonian over such factorized ground state. Within this approximation, $\Delta \epsilon$ can be reduced to an easy-to-calculate sum of, at most, two-dimensional integrals. In the corresponding validity region the estimate provided by the model is accurate: Figure 1 shows the difference between the exact results (squares), the approximate results (solid curve) and the results obtained neglecting Coulomb correlation completely (dotted line). The dashed curve shows the approximate results obtained setting $\Delta \omega/\omega_r = 0$: as anticipated, this correction is generally negligible. The lower inset presents
the behavior of $\Delta x/d$ and $\Delta \omega/\omega_r = 0$ with respect to the external field $\vec{E}$. We stress that the proposed model allows for a quick scan of the whole parameter space, useful especially when it is complex to determine the correct operative region and the exact numerical calculation requires a long computational time.

In conclusion, we have discussed how an external electric field can be used to turn on and off exciton-exciton interactions in a QD system and we have provided a simple analytical model to calculate its main properties. The proposed strategy can play an important role in the design and optimization of semiconductor-based quantum devices, like ultrafast optical switches, single-electron devices, and quantum-information processors.

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REFERENCES

1 See, e.g., J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanosstructures* (Springer, Berlin, 1996).

2 See, e.g., L. Jacak, P. Hawrylak, A. Wojs, *Quantum Dots* (Springer, Berlin 1998).

3 H. Saito et al., Appl. Phys. Lett. 78, 267 (2001); O. Shchekin et al., Appl. Phys. Lett. 77, 466 (2000).

4 G. Yusa, H. Sakaki, Appl. Phys. Lett. 70, 345 (1997); J. Finley et al., Appl. Phys. Lett. 73, 2618 (1998); T. Lundström et al., Science 286, 2312 (1999).

5 M. Jr Bruchez et al., Science 281, 2013 (1998).

6 M. Sherwin et al., Phys. Rev.A 60,3508 (1999); T. Tanamoto, Phys. Rev.A 61, 22305 (2000); F. Troiani et al., Phys. Rev.B 62, 2263 (2000).

7 E. Biolatti et al., Phys. Rev. Lett. 85, 5647 (2000).

8 S. Tarucha et al., Phys. Rev. Lett. 77, 3613 (1996); L. Landin et al., Science 280, 262 (1998); U. Banin et al., Nature 400, 542 (1999).

9 R. Rinaldi et al., Phys. Rev.B 62, 31592 (2000).

10 P. Hawrylak et al., Phys. Rev. Lett. 85, 389 (2000); M. Bayer et al., Nature 405, 923 (2000).

11 C. Coriasso, D. Campi, and A. Stano, Opt. Lett. 23, 183 (1998)

12 Similar models have been proposed for different situations in Ref. 2

13 The biexcitonic shift is defined as the difference between excitonic and biexcitonic optical transition energies.

14 Our fully three-dimensional calculation of the biexcitonic ground state is based on the direct-diagonalization approach proposed in Ref. 2.
FIG. 1. Biexcitonic shift $\Delta \epsilon$ vs external field $E$ for a system characterized by the parameters $m_e = 0.067m_0$, $m_h = 0.34m_0$ ($m_0$ the metallic electron mass), $\hbar \omega_e = 30$ meV and $\hbar \omega_h = 24$ meV. The squares indicate the results of the fully three dimensional calculation, the solid line represents the results of the proposed model, the dotted line is the result obtained when Coulomb correlations are completely neglected and the dashed line corresponds to setting $\Delta \omega = 0$ in the model. The upper inset shows the electron and hole particle density corresponding to the excitonic ground state along the field direction; the lower inset presents the behavior of the two key quantities $\Delta \omega/\omega_r$ and $\Delta x/d$ in respect to the external field.