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

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An original 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 one to control and magnify intra- as well as interdot few-exciton effects. Such a 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. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1399003]

In recent years semiconductor nanostructures have attracted enormous attention.[1] In particular, the ability of building *zero-dimensional* systems, such as semiconductor quantum dots (QDs),[2] has led to a technological revolution: QD applications in fact range from laser emitters[3] to charge-storage devices,[4] and from fluorescent biological markers[5] to quantum information processing devices.[6,7] With QDs, the flexibility in controlling carrier densities reaches its extreme: it is possible to inject into a QD even just a single electron[8] or a single exciton.[9,10] The QDs discrete energy spectrum, on the other hand, results in rich optical response and in 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 important technological applications, like *all-optical* quantum gates[7] and ultrafast optical switches.[11] As we shall see, the proposed field-induced effects may be easily understood and quantitatively estimated using the simple analytical model described below.[12]

Let us 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."[10] In particular, then, if we consider two excitons with opposite spins in the lower energy state, the associated biexcitonic shift[13] is equal to zero. Let us 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* at will 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 QDs 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 the formation of *tunable bonds* between QDs, i.e., *artificial macromolecules*.

The external field can be even used to *turn off* Coulomb interactions. Let us consider for simplicity a single exciton in a QD: What happens if we keep increasing the external electric field? Like in 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 at which the electron-hole charge separation is so large that the two particles can be described as noninteracting.

In order to test the viability of the proposed scheme, we have performed a realistic calculation[14] 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 illustrates the dipole–dipole coupling energy, i.e., the biexcitonic shift,[13] 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

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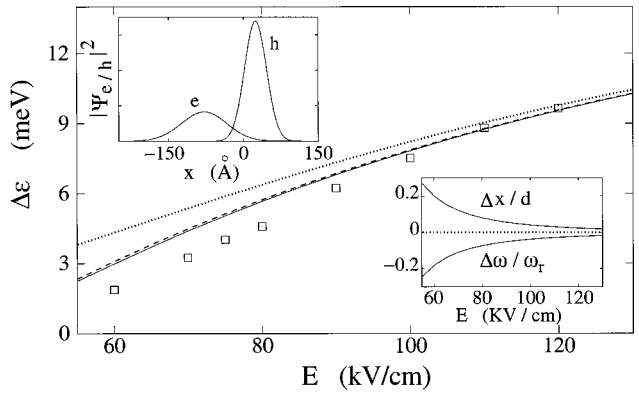

FIG. 1. Biexcitonic shift $\Delta\epsilon$ vs external field E for a system characterized by the parameters $m_e=0.067m_0$, $m_h=0.34m_0$ (m_0 is the metallic electron mass), $\hbar\omega_e=30$ meV and $\hbar\omega_h=24$ meV. The squares indicate the results of the fully three-dimensional calculation, 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 $\Delta x/d$ with respect to the external field.

with the typical resolution of current ultrafast spectroscopy.[1] We stress that, in order to minimize the loss of oscillator strength due to field-induced charge separation, 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 end, 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 $\vec{E}$ the in-plane electric field, the single-exciton Hamiltonian will then be

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

where the $\pm$ sign and the subscripts e and h refer, respectively, to the electron and hole. Here, $\vec{d}_i=e\vec{E}/m_i\omega_i^2$ 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 λ_z^2. It is then possible to separate the Hamiltonian, Eq. (1), as $\mathbf{H}=\mathbf{H}_\parallel(\{\vec{r}_i\})+\mathbf{H}_\perp(z_e)+\mathbf{H}_\perp(z_h)$ where $\mathbf{H}_\perp(z_i)=p_{z_i}^2/2m_i+V_i(z_i)$ 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 $\vec{R}=[m_e(\vec{r}_e+\vec{d}_e)+m_h(\vec{r}_h-\vec{d}_h)]/M$ $(M=m_e+m_h)$ and $\vec{r}=\vec{r}_h-\vec{r}_e$, the in-plane Hamiltonian $\mathbf{H}_\parallel(\{\vec{r}_i\})$ becomes

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

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

In the limit $(\omega_e^2-\omega_h^2)/(\omega_e^2+\omega_h^2)\ll1$, the two coordinates are only weakly coupled, and the Schrödinger equation associated with the CM coordinate $\vec{R}$ is exactly solvable. In the general case we will concentrate on the ground state, although generalization to higher states is straightforward. We can approximate the ground state of $\mathbf{H}_\parallel$ as $\Psi(\vec{r},\vec{R})=\Psi_x(x)[1/(\lambda_r^2\pi)^{1/4}]e^{-(y^2/2\lambda_r^2)}[1/(\lambda_R^2\pi)^{1/2}]e^{-(R^2/2\lambda_R^2)}$, where x and y denote, respectively, the components of $\vec{r}$ parallel and perpendicular to the field $\vec{E}$, $\lambda_r=\sqrt{\hbar/\mu\omega_r}$ and $\lambda_R=\sqrt{\hbar/M\omega_R}$. By averaging $\mathbf{H}_\parallel$ over $\Psi(\vec{r},\vec{R})$, we obtain the effective Hamiltonian $\mathbf{H}_{\rm eff}=\frac{1}{2}\hbar\omega_r+\hbar\omega_R+p_x^2/2\mu+V_{\rm eff}(x)$, characterized by the effective potential

$$V_{\rm eff}(x)=\frac{1}{2}\mu\omega_r^2(x-d)^2+V_C\left(\frac{x^2+\lambda_z^2}{2\lambda_r^2}\right), \tag{3}$$

with $V_C(u)=-(e^2/\epsilon\sqrt{\pi}\lambda_r)e^uK_0(u)$, K_0 being the zero-order Bessel function.

If we are interested in the low-energy states, we can approximate $V_{\rm eff}$ around its minimum V_0 with a parabolic potential, i.e., $V_{\rm eff}(x)\approx V_0+\frac{1}{2}\mu\tilde{\omega}^2(x-x_0)^2$. Within such an approximation scheme, the eigenvalues and eigenfunctions of $\mathbf{H}_{\rm eff}$ 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 the electron and hole can be considered 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 Δx is obtained:

$$\frac{\Delta x}{d}=-\frac{\lambda_r}{a^*}\frac{\exp(\xi)}{\sqrt{\pi}}\frac{\Delta K}{1-\frac{\lambda_r}{a^*}\frac{\exp(\xi)}{\sqrt{\pi}}\left[\frac{d^2}{\lambda_r^2}A(\Delta K,K_1)+\Delta K\right]}, \tag{4}$$

with $\xi=(d^2+\lambda_z^2)/2\lambda_r^2$, K_1 the first-order Bessel function, $\Delta K=K_0(\xi)-K_1(\xi)$, $A(\Delta K,K_1)=2\Delta K+K_1(\xi)/\xi$ and $a^*=\hbar^2\epsilon/\mu e^2$ the reduced Bohr radius. Notice that the prefactor λ_r/a^* is a measure of the system confinement. In a similar way, setting $\tilde{\omega}=\omega_r+\Delta\omega$ in $\mu\tilde{\omega}=d^2V_{\rm eff}/dx^2|_{x_0}$, we can calculate the effect of Coulomb attraction on the potential shape. The result is cumbersome and not particularly enlighting; here we will report only the important limit $\lambda_r^2/d^2\ll1$ (high fields), in which $\Delta\omega/\omega_r=-(\Delta x/d)\propto-(\lambda_r/a^*)\times(\lambda_r^3/d^3)$. The condition $\Delta x/d\lesssim20\%$ quantitatively defines the "intermediate and strong" electric field regime. It is easy to show that, in the regime of interest, the correction of 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 wave functions sitting in different dots. The latter, in turn, are taken as products of the in-plane wave functions times the ground-state wave functions along the z direction. The desired biexcitonic shift $\Delta\epsilon$ is then obtained by averaging the corresponding two-exciton Hamiltonian over such a factorized ground state. In 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: Fig. 1 shows the difference between the exact results (squares), the approximate results (solid curve) and the results obtained neglecting the Coulomb correlation completely (dotted line). The dashed curve shows the approximate results obtained by setting $\Delta\omega/\omega_r=0$: as anticipated, this correction is generally negligible. The lower inset presents the behavior of $\Delta x/d$ and $\Delta\omega/\omega_r$ with respect to the external field $\vec{E}$. We stress that the proposed model allows 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 long computational time.[7]

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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