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(Article begins on next page)

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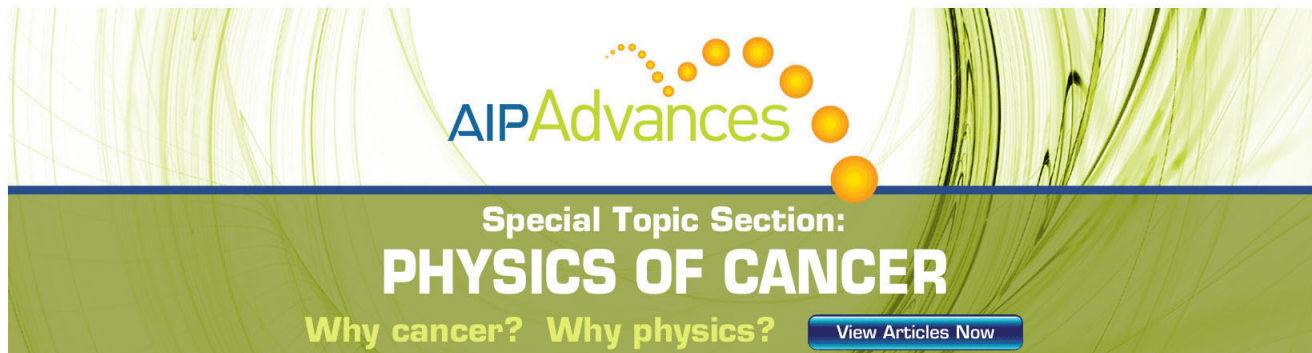
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All-optical single-electron read-out devices based on GaN quantum dots

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We study few-particle interactions in GaN-coupled quantum dots and discuss how the built-in field characteristic of these structures strongly reinforce dipole–dipole and dipole–monopole interactions. We introduce a semi-analytical model that allows for a rapid and easy estimate of the magnitude of few-particle interactions and whose predictions are closer than 10% to “exact” results. We apply our study to the design of an all-optical read-out device that exploits long-range dipole–monopole interactions and may be also used to monitor the charge status of a quantum dot system.

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Quantum dots (QDs) are quasi-zero-dimensional systems that have focused the attention of the scientific community as a playground for testing few-particle fundamental physics and for their increasing technological applications.^{1–5} In particular, semiconductor self-assembled QDs have received much attention recently due to the symmetry of their confinement potential (which paves the road to an easier engineering of their electronic structure) and to the possibility of optically-driving their excitations; this in turn allows for ultrafast state manipulation and coherent control of the few-particle dynamics, a basic request for quantum computing devices.^{4,5} In this panorama, GaN self-assembled QDs occupy a distinguished place since, while their characterization is still incomplete, interesting properties such as their long spin decoherence time⁶ or the presence of a built-in electric field—as strong as few MeV/cm (Ref. 7)—have been already demonstrated. Such a field, of polarization and piezoelectric origin, is antiparallel to the growth direction inside the QDs, inverting its sign outside.⁷ Its importance in relation to single-electron devices relies on the possibility of creating strong bonds between neighboring QDs that can be switched on/off optically. Under its action in fact, electrons and holes are driven in opposite directions, so as to create electrical dipoles that interact with energies of the order of few meV;⁸ this effect is, for example, the key ingredient for a recently proposed quantum computation/information processing device.⁸

In this letter, we shall (i) provide a very accurate semi-analytical method to estimate few-particle interactions between stacked QDs without resorting to heavy numerical calculations and (ii) propose an all-optical read-out device based on such interactions.

Let us consider first a single exciton confined in a GaN QD. We shall work in the usual envelope function approximation and focus on the system ground state, although our method can be easily extended to the other low-energy level states. The Hamiltonian of the excitonic system is

$$H = \sum_{\alpha=e,h} [H_{z\alpha}^0 + m_{\alpha}\omega_{\alpha}^2(x_{\alpha}^2 + y_{\alpha}^2)/2] - e^2/\epsilon|\mathbf{r}_e - \mathbf{r}_h|$$

with

$$H_{z(e/h)}^0 = (p_{z(e/h)}^2/2m_{e/h}) + V_z(z_{e/h}) \pm eE(z)z_{e/h}.$$

The terms in square brackets correspond to the Hamiltonian of a particle confined by a parabolic potential in the in-plane directions and by the strongly confining potentials V_z in the growth direction (usually modeled as a square potential), $E(z)$ is the built-in electric field, e is the absolute value of the electron charge, and ϵ is the dielectric constant of the medium. In this letter, Greek letters will indicate the indexes e and h corresponding, respectively, to electrons and holes.

The ground state $\psi_{e/h}(z_{e/h})$ of the one-dimensional Hamiltonian $H_{z(e/h)}^0$, can be easily calculated by exact diagonalization. We can then resort to the separable effective Hamiltonian $\tilde{H} = H_{ze}^0 + H_{zh}^0 + H_I$, where

$$H_I(\mathbf{R}, \mathbf{r}) = \left[\frac{p_R^2}{2M} + \frac{1}{2} M \omega_R^2 R^2 \right] + \left[\frac{p_r^2}{2\mu} + \frac{1}{2} \mu \omega_r^2 r^2 - \frac{e^2}{\epsilon \sqrt{r^2 + \langle (z_e - z_h)^2 \rangle}} \right] + \mu(\omega_e^2 - \omega_h^2) \mathbf{R} \cdot \mathbf{r}. \quad (1)$$

Here $\mathbf{R} = [m_e(x_e, y_e) + m_h(x_h, y_h)]/M$ and $\mathbf{r} = (x_e - x_h, y_e - y_h)$ are the in-plane center of mass and relative coordinates, respectively $M = m_e + m_h$, $\mu = m_e m_h / M$, $\omega_R^2 = (m_e \omega_e^2 + m_h \omega_h^2)/M$, $\omega_r^2 = (m_h \omega_e^2 + m_e \omega_h^2)/M$, and $\langle (z_e - z_h)^2 \rangle \equiv \langle \psi_e(z_e) \psi_h(z_h) | (z_e - z_h)^2 | \psi_e(z_e) \psi_h(z_h) \rangle$. The original problem has now been reduced to solving the Schrödinger equation for $H_I(x, y)$. By approximating the ground state solution of H_I with the factorized form $\psi_R(\mathbf{R}) \psi_{\text{rel}}(\mathbf{r})$, where $\psi_R(\mathbf{R}) = \sqrt{M \omega_R / \hbar \pi} \exp(-M \omega_R R^2 / 2\hbar)$, we get

$$\tilde{H}_{\text{rel}}(r) = \langle \psi_R | H_I | \psi_R \rangle = \hbar \omega_R + \frac{p_r^2}{2\mu} + V(r), \quad (2)$$

with

$$V(r) = \frac{1}{2} \mu \omega_r^2 r^2 - e^2 / \epsilon \sqrt{r^2 + \langle (z_e - z_h)^2 \rangle}.$$

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For calculating the properties of low-energy states, $V(r)$ can be approximated around its minimum as $V(r) \approx V_0 + \mu \tilde{\omega}_r^2 r^2/2$, where

$$\mu \tilde{\omega}_r^2 = \mu \omega_r^2 + \frac{e^2}{\epsilon \langle (z_e - z_h)^2 \rangle^{3/2}}. \quad (3)$$

We underline that the expression (3) includes corrections due to the Coulomb interaction between electron and hole. As we will see later, such corrections strongly improve the precision of the approximation.⁹ The eigenvalue problem related to Eq. (2) is now exactly solvable, and its ground state is given by $\psi_{\text{rel}}(r) = \sqrt{\mu \tilde{\omega}_r / \hbar \pi} \exp(-\mu \tilde{\omega}_r r^2 / 2\hbar)$. The approximated form for the total excitonic wave function is then

$$\psi_{xc} \approx \psi_e(z_e) \psi_h(z_h) \psi_R(R) \psi_{\text{rel}}(r). \quad (4)$$

Next, we will discuss the biexciton system. If the barrier between two stacked quantum dots QD0 and QD1 is such that particle tunneling is negligible, or if the mismatch between relevant single particle levels in QD0 and QD1 is sufficiently large due to the built-in electric field and to size differences between the two dots, each direct low-energy exciton in the macromolecule QD0+QD1 will be strictly confined to a single dot, so that when considering a biexciton formed by one exciton in QD0 and the second in QD1, we can safely approximate its wave function as the product $\psi_{\text{bi}} \approx \psi_{xc0} \psi_{xc1}$, where 0, 1 indicates QD0 and QD1, respectively.

Let us consider the biexcitonic shift $\Delta\epsilon$, i.e., the energy shift in the transition related to the creation of a second exciton in the presence of a first one. This quantity is essential for performing, for example, conditional operations in quantum computational devices;^{4,5,8,10} it is then crucial to have a quick way to estimate $\Delta\epsilon$ in order to define the correct range for the structure parameters. For the case we are analyzing, where no significant excitonic tunneling is present between different QDs and only the Hartree term plays a relevant role, a good approximation for $\Delta\epsilon$ will be the Coulomb interaction average $\Delta\epsilon = \sum_{\alpha, \beta = e, h} \Delta\epsilon_{\alpha 0, \beta 1}$, with

$$\Delta\epsilon_{\alpha 0, \beta 1} = s_{\alpha, \beta} \langle \psi_{xc, 0} \psi_{xc, 1} | \frac{e^2}{\epsilon |\mathbf{r}_{\alpha 0} - \mathbf{r}_{\beta 1}|} | \psi_{xc, 0} \psi_{xc, 1} \rangle \quad (5)$$

with $s_{\alpha, \beta} = -1$ if $\alpha \neq \beta$; $s_{\alpha, \beta} = 1$, otherwise. $\Delta\epsilon_{\alpha 0, \beta 1}$ represents the Coulomb interaction between particle α in QD0 and particle β in QD1 in the presence of the other particles composing the biexciton (i.e., partially including correlation effects). $\Delta\epsilon_{\alpha 0, \beta 1}$ is an integral over the coordinates of all the four particles considered; in general, in fact, due to Coulomb interaction, it is not possible to *exactly* factorize $\psi_{xc, i}$ into single particle components. Let us now consider a ground state biexciton and approximate $\psi_{xc, i}$ with Eq. (4). We stress that, being the factorization done over the collective coordinates internal to the single QD, such expression *includes* to a certain extent the Coulomb interaction between electron and hole in the same QD. It is now possible to integrate analytically over most of the variables. If we additionally approximate the electron and hole single particle wave functions along the z direction as

$$\psi_{\alpha, i}(z_{\alpha, i}) \approx \exp[-(z_{\alpha, i} - \langle z_{\alpha, i} \rangle)^2 / 2\lambda_{\alpha, i}^2] / (\sqrt{\pi} \lambda_{\alpha, i})^{1/2}$$

where

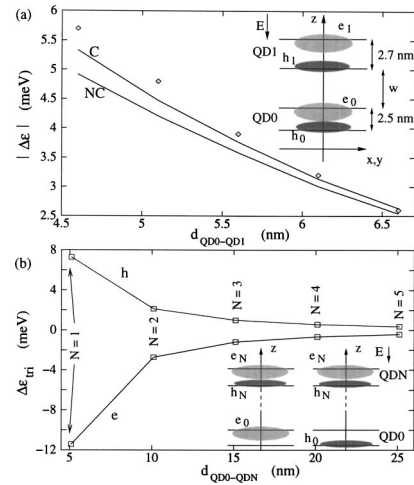


FIG. 1. (a) Biexcitonic shift $\Delta\epsilon$ vs distance between nearest-neighbor dots QD0 and QD1. The barrier width w is varied between 2–4 nm. $\Delta\epsilon$ is obtained using Eq. (6) (solid line). Diamonds correspond to the “exact” results in Ref. 9. Curve C (NC) includes (does not include) Coulomb corrections. Inset: Schematic view of the macromolecule QD0+QD1. The position along the growth direction occupied by each dot is marked by couples of parallel lines. The electronic (light gray, e_i) and hole (dark gray, h_i) clouds are sketched as well. (b) $\Delta\epsilon_{\text{int}}$ vs distance between QD0 and QDN. The barrier width is fixed at $w = 2.5$ nm. The coordination number N is indicated for each point. Curve e (h) corresponds to an electron (hole) in QD0. Inset: same as for inset of (a), but for the stacked-QD array QD0...QDN, when QD0 is occupied by an electron (left) or by a hole (right).

$$\langle z_{\alpha, i} \rangle = \langle \psi_{zi}(z_{\alpha, i}) | \hat{z} | \psi_{zi}(z_{\alpha, i}) \rangle$$

and

$$\lambda_{\alpha, i}^2 \equiv 2 \langle \psi_{zi}(z_{\alpha, i}) | \hat{z}^2 | \psi_{zi}(z_{\alpha, i}) \rangle$$

we obtain

$$\begin{aligned} |\Delta\epsilon_{\alpha 0, \beta 1}| &= \frac{e^2}{\epsilon} \sqrt{\frac{\tilde{D}_{\alpha 0, \beta 1}}{\lambda_{\alpha 0}^2 + \lambda_{\beta 1}^2}} \int_{-\infty}^{\infty} dz \exp\left[-\frac{z^2}{\lambda_{\alpha 0}^2 + \lambda_{\beta 1}^2}\right] \\ &\quad \exp[(z + \Delta z_{\alpha 0, \beta 1})^2 \tilde{D}_{\alpha 0, \beta 1}] \\ &\quad \times \{1 - \phi[\sqrt{(z + \Delta z_{\alpha 0, \beta 1})^2 \tilde{D}_{\alpha 0, \beta 1}}]\}, \quad (6) \end{aligned}$$

where $\phi(x) = (2/\sqrt{\pi}) \int_0^x \exp(-t^2) dt$ is the error function, $\Delta z_{\alpha 0, \beta 1} \equiv \langle z_{\alpha 0} \rangle - \langle z_{\beta 1} \rangle$, and $\tilde{D}_{\alpha 0, \beta 1} = D_{\alpha 0} D_{\beta 1} / (D_{\alpha 0} + D_{\beta 1})$. Here $D_{e/h, i} = [(\mu/\hbar) \{B_{e/h} - [(\omega_R - \tilde{\omega}_r)^2 / B_{h/e}]\}]_i$, with $B_{e/h, i} = [\tilde{\omega}_r + \omega_R (m_{e/h} / m_{h/e})]_i$ and $i = 0, 1$ indicating to which QD the involved parameters belong. With the use of Eq. (4), the twelve-dimensional integral in Eq. (5) has been reduced to the one-dimensional integral in Eq. (6).

Figure 1(a) shows biexcitonic shift values associated to the system QD0+QD1 in the inset, when the barrier width w is varied between 2 and 4 nm and QD heights are respectively 2.5 and 2.7 nm. $\Delta\epsilon$ is obtained using Eq. (6) (solid line), and compared to the “exact” results in Ref. 11 calculated by direct diagonalization of the fully interacting biexcitonic Hamiltonian. As can be seen in Fig. 1(a), Eq. (6) captures most of the information: its estimates are in fact at most 7% from the exact values. The curve labeled C includes Coulomb correlation effects in the excitonic wave functions, while the NC curve does not. We stress that, in this formulation, *including Coulomb interaction, does not imply more complex calculations*, since it is done by the simple substitution $\omega_r \rightarrow \tilde{\omega}_r$ [see Eq. (3)]. The precision of the results is highly affected by such corrections.

Similarly to the biexcitonic shift, Coulomb interaction modifies the transition energy in the absorption spectrum corresponding to the creation of an exciton in a certain dot in the presence of an electron (hole) in another dot.¹² In this case, though, the (dipole–monopole) interaction decreases as $\sim d/w^2$ (d the dipole length, roughly equal to the GaN QD height), that is, much slower than the dipole–dipole interaction $\sim d^2/w^3$ characterizing the biexcitonic shift. This implies that, it should be possible to detect such a shift [and consequently the electron (hole) presence], even if the two dots are not neighbors.

Let us consider the response of an array of slightly differently stacked GaN dots, whose height is ~ 2.5 nm¹³ and which are separated by barriers 2.5 nm wide. In the hypothesis that the dot QD0 contains an electron (hole), we can calculate, by using Eq. (6) and by not including Coulomb interaction in the wave functions related to QD0, the energy shift connected to the creation of an exciton in QDN, where $N=1,2,3,\dots$ is the coordination number with respect to QD0. The system is sketched in the inset of Fig. 1(b). The calculated shift $\Delta\varepsilon_{\text{tri}}$ is plotted in Fig. 1(b) as a function of the distance between the centers of QD0 and QDN. The coordination number of the latter dot is indicated as well. The curve labeled by e(h) corresponds to the presence of an electron (hole) in QD0. For $N=1$, $|\Delta\varepsilon_{\text{tri}}| \sim 10$ meV, but even considering N as high as 5, the energy shift is still of the order of ~ 0.5 meV, that is, able to be resolved by laser pulses as short as 2–3 ps. The asymmetry between the e and the h curves reflects the corresponding asymmetry between electron and hole wave functions; the sign of $\Delta\varepsilon_{\text{tri}}$ is related to the sign of the particle in QD0.

Starting from these simple observations, we can think of a noninvasive optical read-out device for a memory which has been written as the presence (logic state 1) or the absence (logic state 0) of a charge in each QD:³ by using a laser probe centered at the chosen QDN excitonic transition energy, the logic state 1 (0) will correspond to the absence (presence) of the corresponding excitonic peak in the absorption spectrum. This scheme could be also used to measure the qubit state in quantum computing devices,¹⁴ and could, in general, be a valid alternative to the reading done, for example, by point contacts, since it avoids charge fluctuations due to the presence of currents in the system.¹⁵ In addition, for far enough distances between the “written” QD0 and the “reading” QDN, the interaction becomes proportional to the total charge inside QD0, so in principle a measure of $\Delta\varepsilon_{\text{tri}}$ could be used to count the electrons (holes) that have been injected in QD0. The sign of $\Delta\varepsilon_{\text{tri}}$ would be related to the sign of the net charge present in QD0. The desired QDN excitonic transition can be generated by energy-selective schemes⁵ or near-field techniques. A plus of the proposed device is that, by using a long-distance interaction, the presence of the exciton in the reading dot would not significantly perturb the system in the written one.

A similar scheme can, of course, be implemented in different semiconductor QDs (as, for example, GaAs QDs), with the caveat of using an external in-plane electric field to reinforce dipole–monopole interactions.¹⁶ The main advantage of using GaN QDs is the strong *built-in* electric field which on the one hand simplifies the setup, and on the other

hand will never ionize the trapped particles. Moreover, in GaN QDs, Coulomb interaction is maximized, since (i) this system naturally *aligns* the charges along the growth direction, and (ii) due to the strength of the built-in field, the wave function spreading in the growth direction is reduced. It is important to stress that, for the device to work, it is sufficient to have a static electric field in the *reading* QD so, for example, a GaN QD layer could be grown on a different substrate. We finally underline that the parameters used in our calculations are in the reach of present experimental techniques.

In summary, we have proposed an all-optical read-out device based on long-range exciton–single-charge interactions. The latter were calculated using our semi-analytical model, which allows for a quick and very precise estimate of few-particle interactions in stacked GaN quantum dots. The device exploits the built-in electric field characteristic of GaN-based heterostructures, but may be adapted to nanostructures based on different semiconductor compounds.

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¹⁵As for point contacts, though, since both reading schemes are based on Coulomb interaction, the measure outcome could be influenced by the presence of spurious charges in the system.

¹⁶For the effect of an in-plane electric field on excitonic wave functions in GaAs QD see Ref. 5. Recently, significant built-in electric fields have been reported also in InGaAs self-assembled QD (R. Rinaldi, private communication).