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Exciton–exciton interaction engineering in coupled GaN quantum dots

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We present a fully three-dimensional study of the multiexciton optical response of vertically coupled GaN-based quantum dots via a direct-diagonalization approach. The proposed analysis is crucial in understanding the fundamental properties of few-particle/exciton interactions and, more important, may play an essential role in the design/optimization of semiconductor-based quantum information processing schemes. In particular, we focus on interdot exciton–exciton coupling, the key ingredient in recently proposed all-optical quantum processors. Our analysis demonstrates that there is a large window of realistic parameters for which both biexcitonic shift and oscillator strength are compatible with such implementation schemes.
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Semiconductor quantum dots (QDs) are systems of paramount interest in nanoscience and nanotechnology.[1] They are the natural evolution of band-gap/wave-function engineering in semiconductors; however, in contrast to quantum wells and wires, they exhibit a discrete—i.e., atomic-like—energy spectrum and, more important, their optical response is dominated by few-particle/exciton effects. Moreover, several QD-based technological applications have been proposed, such as lasers,[2] charge-storage devices,[3] fluorescent biological markers,[4] and all-optical quantum information processors.[5]

So far, self-organized QDs have been successfully fabricated using a wide range of semiconductor materials; they include III–V QD structures based on GaAs as well as on GaN compounds. GaAs-based QDs have been well characterized and their electronic structures have been widely studied,[6,7] whereas the characterization of GaN-based systems is still somewhat fragmentary and their electronic properties have been studied only recently.[8] GaAs- and GaN-based nanostructures exhibit very different properties: GaN systems have a wider band gap (3.5 eV) compared to GaAs-based ones (1.5 eV). Moreover, whereas GaAs and most of the other III–V compounds have a cubic (zincblende) structure, GaN (as well as other nitrides) has a hexagonal (wurzite) structure, which leads to strong built-in piezoelectric fields (of the order of MV/cm). As a consequence of such built-in fields, in these nanostructures excitonic transitions are redshifted, and the corresponding interband emission is fractions of eV below the bulk GaN band gap.

In this letter, we shall provide a detailed investigation of the interplay between single-particle carrier confinement and two-body Coulomb interactions in coupled GaN-based QDs. In particular, we shall analyze exciton–exciton dipole coupling versus oscillator strength: we demonstrate that it is possible to tailor and control such nontrivial Coulomb interactions by varying the QD geometry (e.g., base and height), since this in turn modifies the wave functions of electrons and holes confined into the QDs as well as intrinsic electric fields; at the same time, our investigation shows that the oscillator strength of the ground-state exciton decreases superexponentially with increasing QD height.

The relevance of our analysis is twofold: (i) we address a distinguished few-particle phenomenon typical of nitride QDs, i.e., the presence of an intrinsic exciton–exciton dipole coupling induced by built-in polarization fields; (ii) we provide detailed information on the set of parameters needed for the experimental realization of the quantum information processing strategy proposed in Ref. 9, in which a large biexcitonic shift is necessary for energy selective addressing of the different few-particle excitations with fs/ps laser pulses.

More specifically, in our analysis we consider the range of GaN/AlN QDs presented in Ref. 10: the dot height will vary from 2 to 4 nm and the QD-based diameter from 10 to 17 nm, assuming a linear dependence between these two parameters in agreement with experimental and theoretical findings.[8]

As already underlined, the peculiarity of wurzite GaN heterostructures is the strong built-in electric field, which is the sum of the spontaneous polarization and the piezoelectric field. Spontaneous polarization charge accumulates at the GaN/AlN interfaces as a consequence of a slight distortion of GaN and AlN unit cells, compared to those of an ideal hexagonal crystal. Piezoelectric fields are caused by uniform strain along the (0001) direction. Contrary to GaN/AlGaN quantum wells—where the spontaneous-polarization contribution is dominant—[11] in QDs the strain-induced piezoelectric field and the spontaneous-polarization potential are of similar magnitude and sign, both oriented along the growth direction. The strength of the intrinsic field along such a direction is almost the same inside and outside the dot, but it

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is opposite in sign. The built-in electric field in GaN QDs and AlN barriers is calculated according to[11]

$$F_d = \frac{L_{\mathrm{br}}(P_{\mathrm{tot}}^{\mathrm{br}} - P_{\mathrm{tot}}^{d})}{\epsilon_0(L_d \epsilon_{\mathrm{br}} + L_{\mathrm{br}} \epsilon_d)}, \tag{1}$$

where $\epsilon_{\mathrm{br},(d)}$ is the relative dielectric constant of the barrier (of the quantum dot), $P_{\mathrm{tot}}^{\mathrm{br},(d)}$ is the total polarization of the barrier (of the quantum dot), and $L_{\mathrm{br},(d)}$ is the width of the barrier (the height of the dot). The value of the field in the barrier F_{br} is obtained by exchanging the indices br and d. Equation (1) is derived for an alternating sequence of quantum wells and barriers, but it is a good approximation also in the case of an array of similar QDs in the growth (z) direction. The lateral shape of the QD is simply approximated by a bidimensional parabolic potential, which mimics the strong in-plane carrier confinement caused by the built-in electric field and preserves the spherical symmetry of the ground state.[8] Our approach is supported by the agreement with the experimental findings in Ref. 8. The polarization is the sum of the spontaneous polarization charge that accumulates at GaN/AlN interfaces and the piezoelectric one. All the parameters are taken from Ref. 11 (adapted for the case $x=1$ for Al percentage in the barrier).

The above theoretical scheme has been applied to realistic state-of-the-art GaN QDs. The difference between the well width of two neighboring QDs is assumed to be 8% to allow energy-selective generation of ground-state excitons in neighboring QDs. The barrier width is such to prevent single-particle tunneling and to allow, at the same time, significant dipole–dipole Coulomb coupling: the giant internal field, in fact, strongly modifies the conduction and valence bands along the growth directions and causes the separation of electrons and holes, driving the first one towards the QD top and the latter towards its bottom. This corresponds to the creation of intrinsic dipoles. If we consider two stacked dots occupied by one exciton each, the resulting charge distribution can be seen as two dipoles aligned along the growth direction. This is evident in Fig. 1, where we plot the electron and hole single-particle distributions corresponding to the lowest biexcitonic state (with parallel-spin excitons) in our GaN-based semiconductor "macromolecule." The creation of stacked dipoles results in a negative exciton–exciton coupling (or biexcitonic shift).

The theoretical approach employed to study the optical response of our GaN nanostructure is a generalization (to nitride materials) of the fully three-dimensional exact-diagonalization scheme proposed in Ref. 7. More specifically, we consider electrons (e) and holes (h) confined within stacked QDs, as depicted in Fig. 1. As usual, the confinement potential is modeled as parabolic in the $x-y$ plane and as a square-well potential modified by the built-in electric field along the growth (z) direction. The many-exciton optical spectra, i.e., the absorption probability corresponding to the generic $N \rightarrow N'$ transition, is evaluated as described in Ref. 7. In particular, here we focus on the excitonic ($0 \rightarrow 1$) and biexcitonic ($1 \rightarrow 2$) optical spectra in the presence of the built-in electric field. For all the structures considered, the two lowest optical transitions correspond to the formation of direct ground-state excitons in dots a and b, respectively. The biexcitonic ($1 \rightarrow 2$) optical spectrum describes the creation

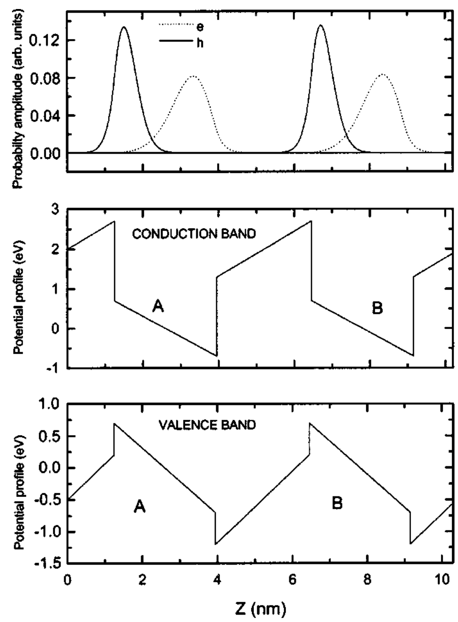

FIG. 1. Electron and hole particle distribution, conduction- and valence-band structure along the growth direction for two coupled GaN dots of, respectively, 2.5 and 2.7 nm of height, separated by a 2.5 nm AlN barrier. In the upper panel the dotted line corresponds to the hole, and the solid one to the electron spatial distribution of the biexcitonic ground state.

of a second electron–hole pair in the presence of a previously generated exciton. Here, we shall consider parallel-spin configurations only.

Let us focus on the biexcitonic shift corresponding to the energy difference between the ground-state biexcitonic transition (given a ground-state exciton in dot a) and the ground-state excitonic transition of dot b. This quantity—a "measure" of the ground-state exciton–exciton coupling—plays a crucial role in all-optical quantum processors, being the key ingredient for conditional-gating schemes.[5,9]

Figure 2 shows how the biexcitonic shift increases with the height of the dot. The barrier width is kept fixed and equal to 2.5 nm. In curve (A) both the height and diameter D of the dots are varied according to the relation $D=3.5L_d+3$ nm,[10] while in (B) only the height of the dot is changed. We notice that, for realistic parameters, it is possible to achieve biexcitonic shifts up to 9.1 meV.

A few comments are now in order: (i) When a barrier of only 2.5 nm separates two stacked GaAs-based QDs, the excitonic wave function is molecular like,[12] forming bonding and antibonding states spread over the whole macromolecule for both electron and hole. This effect is maximum for dots of the same size but persists even when their dimensions are slightly different. In GaN QDs, instead, over the range of parameters used, the lowest states preserve their atomic-like shape since both electron and hole effective masses and valence/conduction-band discontinuities are much higher than in GaAs, therefore, decreasing the atomic-like wave-function overlap responsible for the molecular bonding. (ii) The excitonic dipole length is roughly proportional to the height of the dot because of the strong built-in electric field; therefore, it is crucial to evaluate the dependence of the exciton–exciton interaction on the height of the QDs. (iii) The spreading of the wave function affects the biexcitonic shift, as one can notice by comparing curves A and B in Fig.

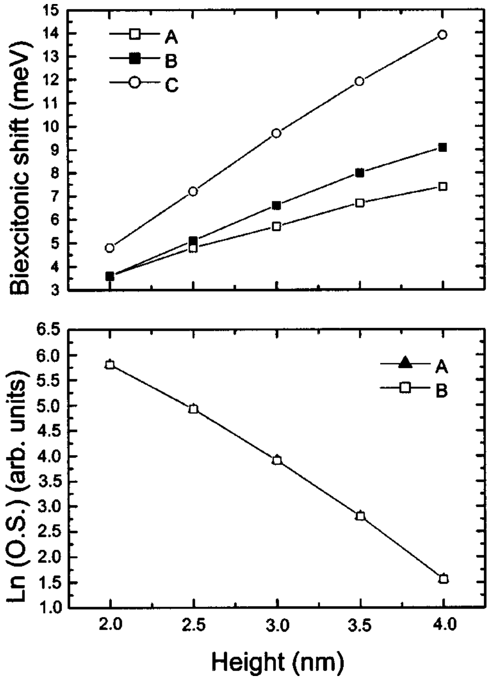

FIG. 2. Biexcitonic shift (upper panel) and oscillator strength (lower panel) of the ground-state transition in dot *b* for two coupled GaN dots separated by a barrier of 2.5 nm vs QD height. In curve (B) only the height of the dots is changed ($D=10$ nm), while in curve (A) D is varied proportionally to the height from 10 to 17 nm. Curve (C) shows the biexcitonic shift in the point-like charge approximation. The parameters used are: effective masses $m_e=0.2m_0$ and $m_h=m_0$; in-plane parabolic confinement energy $\hbar\omega_e=74$ meV and $\hbar\omega_h=33$ meV for the (B) curve and $\hbar\omega_e=74-290$ meV and $\hbar\omega_h=33-130$ meV for the (A) curve.

2. The biexcitonic shift is larger (up to 20% for the parameters considered here) when the wave function is more localized, since the system is closer to the idealized "point-like" particle case [see curve (C) in Fig. 2]. (iv) Our results demonstrate that there exists a wide range of parameters for which the biexcitonic shift is at least a few meV. This is a central prerequisite for realizing energy-selective addressing with subpicosecond laser pulses, as requested, for example, by all-optical quantum information processing schemes.[5,9]

Our analysis shows that the best strategy to achieve a large biexcitonic shift is to grow "high" and "small diameter" dots. The drawback is that the oscillator strength (OS) of the ground-state transition strongly decreases with the height of the dot, since it is proportional to the overlap of electron and hole wave functions. A small value of the OS enhances the well-known difficulties of single-dot signal detection. The lower panel in Fig. 2 shows that the OS corresponding to the excitonic ground state of dot *b* decreases superexponentially with the height of the dot. As shown by the fact that curves (A) and (B) practically coincide, the height of the dot is the only parameter relevant for the OS value. Indeed, the wave-function spreading due to the width of the dot does not influence the electron–hole overlap.

In the range of height values considered in Fig. 2, the OS varies over three orders of magnitude, so care must be taken in a future quantum information processing experiment in order to optimize at the same time biexcitonic shift and OS. Our analysis suggests that a reasonable way to do so is to maximize the product between the biexcitonic shift and the logarithm of the oscillator strength. Such a quantity is plotted in Fig. 3 and it is the largest for a QD height of 2.5–3 nm. The curve presents a well-defined maximum corresponding to a quantum-dot height of 2.8 nm (parabolic fit).

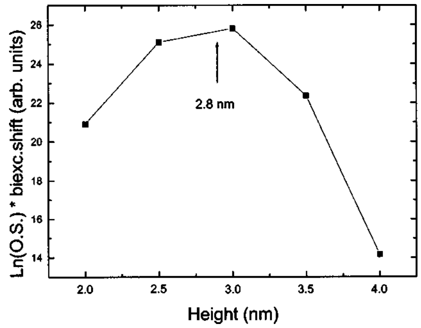

FIG. 3. Figure of merit (biexcitonic shift times logarithm of oscillator strength) vs QD height. Arrow indicates the maximum obtained by a parabolic fit.

In conclusion, we have performed a detailed investigation of exciton–exciton interaction as well as of its effect on the multiexciton optical response in state-of-the-art GaN-based nanostructures. We have shown how it is possible to engineer the interdot biexcitonic shift by varying the height and width of the dots. Our analysis provides precious indications for the realization of GaN-based quantum information processing clarifying, in particular, the crucial interplay between biexcitonic shift and oscillator strength.

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