Local optical absorption by confined excitons in single and coupled quantum dots

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Abstract We study theoretically the local absorption spectra of single and double semiconductor quantum dots, where the three-dimensional confinement leads to an enhancement of Coulomb effects. We show that because of such Coulomb correlations the intensity of certain optical peaks as a function of the resolution can exhibit an unexpected non-monotonic behavior for spatial resolutions comparable with the excitonic Bohr radius.

1 Introduction

Recently, much attention has been devoted to semiconductor quantum dots (QDs) [1–3], where the three-dimensional confinement gives rise to a discrete atomic-like density of states. In turn, the coupling to the solid-state environment is suppressed and Coulomb correlations are strongly enhanced. For these reasons, QDs display a rich number of interesting and new physical phenomena. However, despite the continuing progress, all the available fabrication approaches still suffer from the effects of inhomogeneity and dispersion in dot size, which leads to large linewidths when optical experiments are performed on large QD ensembles. A major advancement in the field has come from different types of local optical experiments, that allow the investigation of individual QDs thus avoiding inhomogeneous broadening [3]. Among local spectroscopies, near-field optical approaches [4,5] are especially interesting as they bring the spatial resolution well below the diffraction limit of light.

As the resolution increases, local optical techniques allow direct access to the space and energy distribution of quantum states within semiconductor nanostructures. Theoretically, the interaction between a given near-field probe and the carrier states inside the dots is not at all obvious: In analogy with ultrafast time-resolved spectroscopies, that have revealed the importance of phase coherence in the quantum-mechanical time evolution of photoexcited carriers, it may be expected that spatial interference of quantum states plays a dominant role when variations of the electromagnetic (EM) field occur on an ultra-short length scale.

2 Theory

Let us consider the situation where a nanostructure (single dot or coupled dots) is investigated by a local near-field probe (with $\mathcal{E}_\omega(\mathbf{r})$ the EM field profile at photon energy $\hbar\omega$). For photon energies around the effective semiconductor band gap, this external light field creates electron-hole pairs which propagate in presence of the dot confinement potential and of their mutual Coulomb interactions. Within linear response the optical near-field properties are thus governed by the electron-hole (exciton) energies E_λ and wavefunctions $\Psi^\lambda(\mathbf{r}_e,\mathbf{r}_h)$, with quantum number λ labelling the different exciton states. It turns out to be convenient to consider for the EM field distribution a given profile ξ centered around the beam position $\mathbf{R}$, i.e., $\mathcal{E}_\omega(\mathbf{r}) = \mathcal{E}_\omega\xi(\mathbf{r}-\mathbf{R})$. Then, the local spectrum for a given tip position $\mathbf{R}$ can be expressed as [6]:

$$\alpha_\xi(\mathbf{R},\omega) \propto \sum_\lambda \left| \int d\mathbf{r}\, \Psi^\lambda(\mathbf{r},\mathbf{r})\xi(\mathbf{r}-\mathbf{R}) \right|^2 \mathcal{D}_\gamma(E_\lambda - \hbar\omega),$$

with $\mathcal{D}_\gamma$ a broadened delta function where the phenomenological damping constant γ accounts for environment couplings of excitons (e.g., phonons).

Within our computational approach we obtain: the single-particle energies $\epsilon^{e,h}$ and wavefunctions $\phi^{e,h}(\mathbf{r})$ for electrons and holes, respectively, by solving the single-particle Schrödinger equation for arbitrary confinement potentials within the enevelope-function and effective-mass approximations (see Ref. [7] for details); E_λ and Ψ^λ by expanding the electron-hole Hamiltonian $\mathcal{H}$ in the single-particle bases $\phi^{e,h}$ and through direct diagonalization of $\mathcal{H}$ (note that within this scheme Coulomb interactions are accounted for in a first-principles manner).

3 Results

Figure 1 reports the calculated local absorption spectra $\alpha_\xi(X,\hbar\omega)$ for a single QD with a prototypical dot confinement, which is parabolic in the (x,y)-plane and box-like along the z-axis.[1] For ξ we use a Gaussian with $\xi(x,y,z) \propto \exp -(x^2+y^2)/2\sigma^2$ (because of the narrow well width of the dot confinement the z-dependence of the EM near-field profile is neglected). The values of σ reported on the right-hand side of Fig. 1 correspond to the regimes of: the far-field limit (σ=50 nm); a resolution comparable to the extension of the quantum states

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[1] In our cylindrical QD the confinement energies due to the in-plane parabolic potential are 20 meV for electrons, and 3.5 meV for holes; with this choice, electron and hole wavefunctions have the same lateral extension. The quantum-well confinement along z is such that the intersubband splittings are much larger than $\omega_o^{(e,h)}$. Material parameters for GaAs are used.

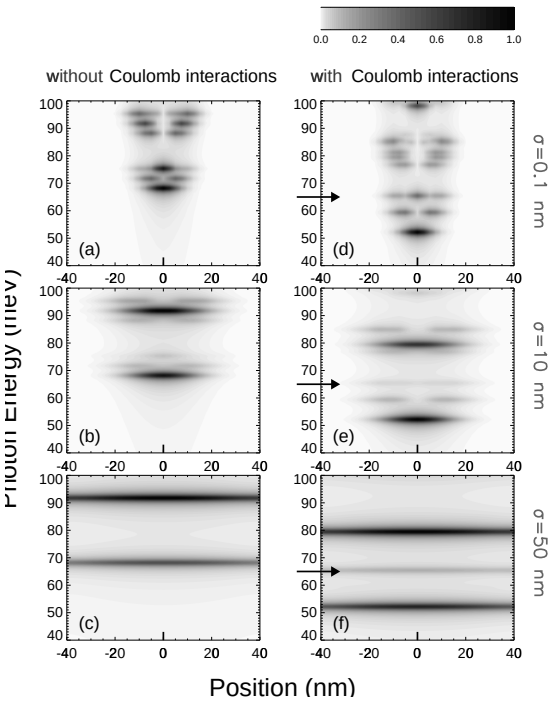

Fig. 1 Local absorption spectra $\alpha_\xi(X, \hbar\omega)$ for a single QD with (Figs. 1(d–f)) and without (Figs. 1(a–c)) Coulomb interactions and for different values of σ; here, the tip is swept along the x-direction (with $Y = 0$), i.e., passing through the center of the QD. Photon energy $\hbar\omega$ is measured with respect to the bandgap.

(σ=10 nm; exciton Bohr radius of GaAs $a_o \approx$12 nm); a (rather unphysical) regime of an extremely narrow probe pulse (σ=0.1 nm) which provides a direct measure of the exciton wavefunctions. A comparison of Figs. 1(a–c) with Figs. 1(d–f) reveals that Coulomb interaction induces several pronounced effects: An almost rigid redshift of the spectra (due to the attractive electron-hole Coulomb interactions); a transfer of oscillator strength from transitions at higher energies to those at lower energies; the appearance of new features in the optical spectra (see, e.g., the features indicated by arrows).

Varying σ, these features show an interesting non-monotonic behavior (in contrast to the other transitions which, with increasing σ, either remain strong or gradually disappear): They are quite strong at σ=0.1 nm, almost disappear at σ=10 nm, and become visible again in the far-field limit. Figure 2 shows the relative weight of the three excitons with $E_\lambda \sim 65$ meV to the local spectra (averaged over all tip positions $\mathbf{R}$), I_σ^λ: While the contribution of excitons (b) (with p-type symmetry) disappear monotonically, the contribution of exciton (a) (with s-type symetry) displays a non-monotonic behavior. As discussed in detail in Ref. [7] those contributions to exciton state (a) which are responsible for the oscillator strength in the optical far field are due to Coulomb coupling (this can also be inferred from Fig. 1(c) where no corresponding peak is visible).

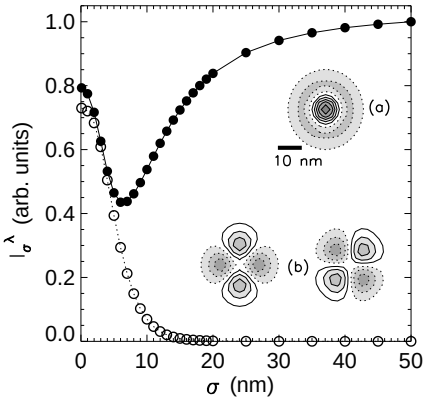

Fig. 2 The relative contribution I_σ^λ as a function of σ, of the excitons which are responsible for the non-monotonic behavior of the feature at 65 meV in Fig. 1 (arrows). Inset: Contour plot of the wavefunctions of these excitons, $\Psi^\lambda(\mathbf{r}, \mathbf{r})$ (solid and dashed lines correspond to positive and negative values, respectively.): Full (open) circles correspond to exciton (a) (excitons (b)).

Finally, we shortly comment on the optical near-field spectra of coupled dots. Quite generally, reducing the distance d between two quantum dots (artificial atoms) carriers can tunnel between the two dots and start to form an *artificial molecule*. Consequently, while for large d carrier states are degenerate, reducing d results in a lifting of degeneracy: E.g., the groundstates approximately split into a (lower-energy) symmetric and (higher-energy) antisymmetric superposition of the two states. As shown in Ref. [7], in the optical far-field only the symmetric superposition couples to the light field—in contrast to the optical near-field spectra which clearly show the formation of "binding" and "anti-binding" states. We also find for coupled dots features with non-monotonic behavior as a function of spatial resolution. Thus, such non-monotonic behavior provides a striking *fingerprint* of Coulomb correlations in the optical near-field spectra and appears quite generally in zero-dimensional nanostructures where carrier states are confined on a length scale comparable to the Bohr radius.

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