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Physica E 35 (2006) 229–233
PHYSICA E
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Optical near-field mapping of bright and dark quantum dot states ☆

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Available online 13 October 2006

Abstract

We theoretically investigate scanning near-field optical microscopy (SNOM) of semiconductor quantum dots. A general theoretical framework is developed that accounts for photo excitation and relaxation in complex dielectric environments. We find that in the near-field regime bright and dark excitonic states become mixed, opening new channels for the coupling to the electromagnetic field.

PACS: 73.21.La; 78.67.−n; 71.35.−y

Keywords: Quantum dots; Scanning near-field optical microscopy (SNOM); Open system

1. Introduction

Optics is a unique tool that allows a remote imaging of objects. For nanosystems, with dimensions L ranging from a few to several tens of nanometers, problems arise because of the micrometer wavelength λ of light that limits the resolution of conventional optics to $\lambda/2 \gg L$. Let us look more closely at the reasons for this limitation. We consider a quasi-two-dimensional nanostructure located in the xy-plane that radiates at frequency ω. The electric component of the field at position z away from the nanostructure will be given by some Fourier transform [1]

$$\boldsymbol{E}(\boldsymbol{r},t) = \sum_{\sigma,k_x,k_y} \boldsymbol{E}_\sigma(k_x,k_y)\,\exp\,\mathrm{i}(k_x x + k_y y + k_z z - \omega t), \quad (1)$$

with σ the light polarization. Maxwell's equations tells us that for light propagating away from the nanostructure the relation

$$k_z = \sqrt{k^2 - k_x^2 - k_y^2}, \quad k^2 > k_x^2 + k_y^2 \quad (2)$$

must be fulfilled, where $k = \omega c^{-1}$ is the light wavevector. Because of this inequality a propagating light wave can never carry a spatial resolution greater than

$$\Delta \approx \frac{2\pi}{k_{\max}} = \frac{2\pi c}{\omega} = \lambda, \quad (3)$$

which is reminiscent of the Abbe limit. However, something is missing. For larger values of the transverse wave vector we get

$$k_z = \mathrm{i}\sqrt{k_x^2 + k_y^2 - k^2}, \quad k^2 < k_x^2 + k_y^2. \quad (4)$$

These evanescent waves, which provide information about the fine spatial details of the nanostructure, decay exponentially with z. The measurement of such waves is the objective of scanning near-field optical microscopy (SNOM) [2,3]. Conveniently, this is achieved by exciting the nanostructure through the tip of an optical fiber (illumination mode) or collecting luminescence of the nanostructure through the tip (collection mode). Near-field spectroscopy has been successfully used for the measurement of single and coupled semiconductor nanostructures [4–6], molecules [7,8], or metallic nanostructures [9,10].

If the spatial near-field resolution falls below the extension of confined quantum systems, it becomes possible to directly map out the spatial probability distribution of the wavefunction. This was recently achieved by Matsuda et al. [11] for quantum dots where the quantum confinement is induced by local monolayer fluctuations in the thickness of a semiconductor quantum

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doi:10.1016/j.physe.2006.08.012

well. With $\Psi_x(\boldsymbol{r}_e, \boldsymbol{r}_h)$ the electron–hole wavefunction of the quantum dot, the matrix elements for the optical near-field transitions are of the form [12–14]

$$\int d\boldsymbol{r}\, \boldsymbol{E}_\omega(\boldsymbol{r}_{tip} - \boldsymbol{r})\Psi_x(\boldsymbol{r}, \boldsymbol{r}), \tag{5}$$

with $\boldsymbol{E}_\omega(\boldsymbol{r}_{tip} - \boldsymbol{r})$ the electromagnetic distribution around the near-field tip. In contrast to the far-field, where the matrix elements governing the light-matter coupling are determined by the quantum states alone, in the near-field the pertinent matrix elements (5) become a convolution of the quantum states with the electro-magnetic field profile of the SNOM tip. This allows to break in the near-field the usual optical selection rules, and to excite dark states whose excitation is forbidden by symmetry in the far-field.

In this paper we theoretically investigate the spectral response of a semiconductor quantum dot excited in the optical near-field. We find that in order to observe dark states directly by means of optical spectroscopy it not only suffices to excite these states, e.g. by means of symmetry breaking in the near-field, but additionally new decay channels have to be opened that allow coupling to propagating photon modes.

2. Theory

A proper theoretical treatment of optical near-field microscopy of nanostructures should account on the same footing for the carrier states, the excitation of the near-field probe, the optical decay of the carrier states in presence of a possibly modified dielectric environment, and for other environment couplings. All this constitutes a formidable theoretical challenge. Let us start with the carrier states. We consider quantum confined states induced by monolayer fluctuations in the thickness of a GaAs/AlGaAs semiconductor quantum well, Fig. 1(a) [11,14]. Since the confinement length (several tens of nm) is much larger than both the lattice constant and the excitonic Bohr radius, we adopt the usual envelope function and rigid-exciton (rigid-biexciton) approximations, the latter assuming that the correlated electron–hole wavefunctions factorize into a center-of-mass and a relative part given by that of the quantum well [16]. The resulting two-dimensional Schrödinger equation for the exciton and biexciton center-of-mass wavefunctions are then solved numerically [14,15]. We describe the light-matter coupling within the usual dipole and rotating wave approximations [15]

$$H_{op}(t) = \int d\boldsymbol{r}\, (\boldsymbol{P}(\boldsymbol{r})\boldsymbol{E}^-(\boldsymbol{r}, t) + \boldsymbol{P}^\dagger(\boldsymbol{r})\boldsymbol{E}^+(\boldsymbol{r}, t)). \tag{6}$$

Here, $\boldsymbol{P}(\boldsymbol{r}) = \boldsymbol{\mu}\Psi_h(\boldsymbol{r})\Psi_e(\boldsymbol{r})$ is the interband polarization operator accounting for the destruction of an electron–hole pair at position $\boldsymbol{r}$, $\boldsymbol{\mu}$ is the dipole moment of the bulk semiconductor, and $\boldsymbol{E}^\pm$ are the electric field components propagating with positive or negative frequency components. For simplicity, we shall not indicate explicitly the spin and polarization degrees of freedom. Eq. (6) holds for both classical and quantum light fields, where in the former case $\boldsymbol{E}^\pm$ is a c-number and in the latter case an operator. The optical matrix elements for the vacuum-to-exciton and exciton-to-biexciton transitions $\boldsymbol{P}_{0x}(\boldsymbol{r})$ and $\boldsymbol{P}_{xb}(\boldsymbol{r})$, respectively, can be computed according to [14,15]

$$\boldsymbol{P}_{0x}(\boldsymbol{r}) = \boldsymbol{\mu}\Psi_x(\boldsymbol{r}, \boldsymbol{r}),$$
$$\boldsymbol{P}_{xb}(\boldsymbol{r}) = \boldsymbol{\mu}\int d\boldsymbol{r}_e\, d\boldsymbol{r}_h\, \Psi_x^*(\boldsymbol{r}_e, \boldsymbol{r}_h)\Psi_b(\boldsymbol{r}, \boldsymbol{r}, \boldsymbol{r}_e, \boldsymbol{r}_h), \tag{7}$$

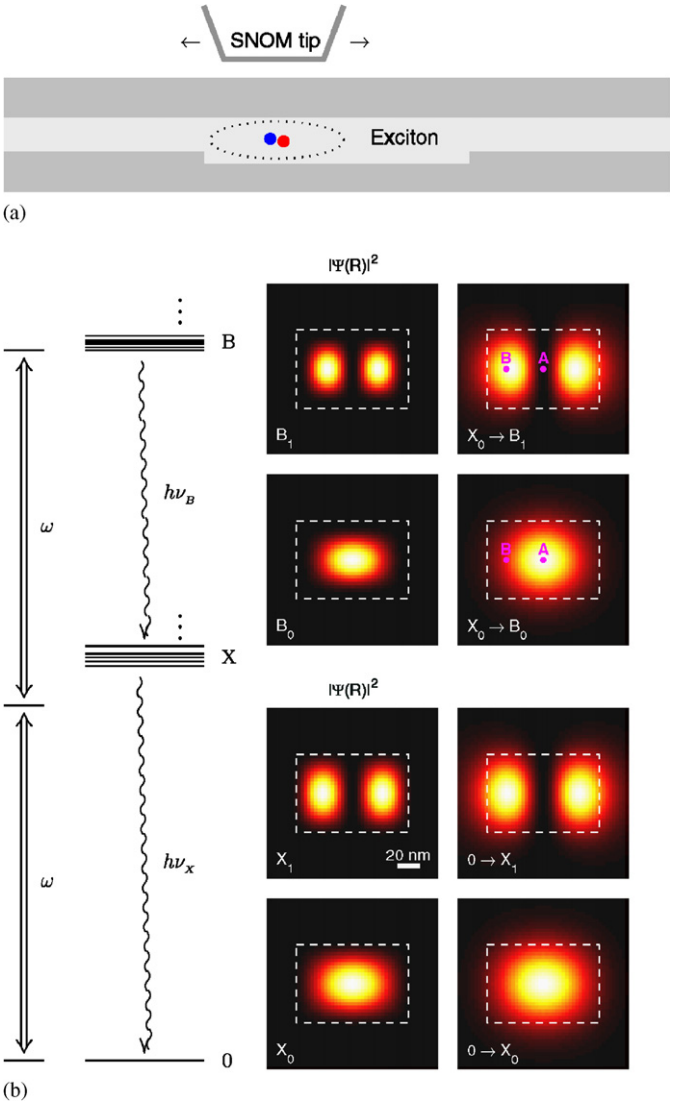

Fig. 1. (a) Schematic sketch of the confinement potential for excitons and biexcitons, which is induced by local monolayer fluctuations in the thickness of a semiconductor quantum well, and the probing SNOM tip. (b) Photoexcitation scenario assumed in our calculations. A biexciton state is excited by SNOM within a two-photon process. Because of the biexciton binding $\delta E_b \sim 4$ meV the luminescence of the biexciton-to-exciton transition is red-, and the exciton-to-groundstate transition blue-shifted with respect to ω. The insets report the square modulus of the center-of-mass part of the exciton and biexciton wave functions [14,15] (left columns) and the spatial near-field maps (right columns) for a terrace of dimension 100×70 nm (dashed line) and for a Bethe–Bouwkamp near-field probe with a full-width of half maximum of approximately 25 nm. In the optical far-field only X_0 and B_0 are allowed, whereas X_1 and B_1 are forbidden because of symmetry.