Microscopic quantum theory of spatially resolved photoluminescence in semiconductor quantum structures

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We present a microscopic analysis of spatially resolved photoluminescence and photoluminescence excitation spectroscopy in semiconductor quantum structures. Such theoretical and numerical framework provides a general basis for the description of spectroscopic imaging in which the excitation and detection energies and spatial positions can all independently be scanned. The numerical results clarify the impact of the near-field optical setup on the obtained images and resolutions. [DOI: 10.1063/1.1711184]

In recent years, measurements based on spatially resolved photoluminescence (PL) provided direct information on the spatial and energy distribution of light emitting nanometric centers of semiconductor quantum structures, thus opening a rich area of physics involving spatially resolved quantum systems in a complex solid state environment.1–6 In particular, near-field optical microscopy and spectroscopy can detect the optical characteristics of individual quantum dots (QDs) among, e.g., a high-density ensemble of naturally formed QDs,7 whereas usual far-field methods provide only ensemble-averaged properties. Detailed simulations of Zimermann, Runge, and Savona8,9 have clarified many aspects of the intrigued nonequilibrium dynamics giving rise to photoluminescence spectra in these quantum structures. However, theoretical simulations of near-field imaging spectroscopy of semiconductor quantum structures focus on calculations of local absorption.10–13 In contrast, as a matter of fact, almost all experimental images are obtained from PL measurements. Here we present a microscopic theory of spatially resolved photoluminescence in quantum structures that includes both light quantization (essential to describe radiative recombination) and phonon scattering. The theory also includes the description of spatially confined excitation (illumination mode) and/or detection (collection mode). It is worth noting that we are faced with a strictly nonequilibrium problem. Nonequilibrium here arising from both radiative recombination (preventing full thermalization) and from the local nature of the excitation source (in the illumination-mode setup). We are also faced with the problem of the differences between illumination (I) and collection (C) scanning near-field optical microscopy (SNOM) instruments. The equivalence between these two working modes has been established on the basis of the reciprocity theorem for electromagnetic fields.14 However, this theorem holds for linear and passive media. While semiconductor structures, at low excitation densities, show a linear behavior, phonon scattering and radiative recombination prevent them from being passive.

The positive frequency components of the operator de-
A spatially correlated process. We used 

tions, both modeled as a zero mean, Gauss distributed and 

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I 

initial condition. Following this procedure we obtain 

signal mode 

contains the overlap of the exciton wave functions with the 

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R 

s 

oscillator strength: 

rate for radiative recombination proportional to the exciton 

E I 

the spatial overlap between the illuminating field 

resonant with the input light: 

G 

the exciton wave functions corresponding to exciton levels 

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b 

r 

a 

S 

C 

o 

\( \beta \rightarrow \alpha \) depend on the lattice deformation potentials and the overlap between the exciton states. 

2 \( \Gamma \) = \( r \alpha + \sum_{\beta} \gamma_{\beta \rightarrow \alpha} \) is the total out-scattering rate, \( r \alpha \) being the rate for radiative recombination proportional to the exciton oscillator strength: 

\( r \alpha = r_0 \left| \int d^2 \mathbf{R} \psi_\alpha(\mathbf{R}) \right|^2 \) (we assume that the tip-sample interaction does not alter the radiative decay rates). In the above equation the generation term depends on the spatial overlap between the illuminating field \( E_I(\mathbf{r}) \) and the exciton wave functions corresponding to exciton levels resonant with the input light: \( G_\alpha(\mathbf{R}) = r_0 | \psi_\alpha(\mathbf{R}) |^2 L_o(\omega) \) with 

\( \pi L_o(\omega) = \Gamma^2 (\omega - \omega_\alpha)^2 + \Gamma^2 \) and \( \psi_\alpha(\mathbf{R}) = \int d^2 \mathbf{R} \tilde{E}_\alpha(\mathbf{R}) \psi_\alpha(\mathbf{R}) \) where \( \tilde{E}_\alpha(\mathbf{R}) = \int E_I(\mathbf{r}) f(z) dz \). In the subsequent numerical calculations concerning the illumination mode we will assume an input light field with a given Gaussian profile centered around the tip position \( \mathbf{R}_I \): 

\( \tilde{E}_\alpha(\mathbf{R}) = \tilde{E}_\alpha^0 (\mathbf{R} - \mathbf{R}_I) \). In this case the generation term becomes function of the beam position and shape (spatial resolution).

Once the exciton densities have been derived, the frequency integrated PL can be readily obtained. It results in 

\( \mathcal{I}(\mathbf{R}, \omega) = r_0 \sum_\alpha | \psi_\alpha(\mathbf{R}) |^2 L_o(\omega) \) where \( \psi_\alpha^C \), analogously to \( \psi_\alpha^I \), contains the overlap of the exciton wave functions with the signal mode \( \tilde{E}_\alpha(\mathbf{R}) \) delivered by the tip (collection mode). According to the quantum regression theorem, 

\( \langle \hat{S}^z(\tau) \hat{S}^z(\tau) \rangle \) has the same dynamics of \( \langle \hat{S}^z(\tau) \hat{S}^z(0) \rangle \) (proportional to the exciton operator), but with \( \langle \hat{S}^z(\tau) \hat{S}^z(0) \rangle \) as initial condition. Following this procedure we obtain 

\( \mathcal{I}(\mathbf{R}, \omega) = r_0 \sum_\alpha | \psi_\alpha^C(\mathbf{R}) |^2 L_o(\omega) N_\alpha \).

We apply the above developed theoretical scheme to analyze the spatially resolved light emission of a QD system arising from interface fluctuations of GaAs QWs. The effective disordered potential felt by 1s excitons used in our simulations is obtained summing up two different contributions, both modeled as a zero mean, Gauss distributed and spatially correlated process. We used \( \xi = 16 \) nm (the correlation length), \( v_0 = 1.5 \) meV (the width of the energy distribution); and \( \xi = 8 \) nm, \( v_0 = 0.5 \) meV, respectively.

Figure 1(a) displays the specific realization of the effective disordered potential used for all the calculations. Figures 1(b)–1(d) show energy-integrated PL images obtained after uniform illumination of the sample at energy \( \omega_I = 1 \) meV (the zero of energy is fixed at the energy of the 1s exciton in absence of disorder) and collecting locally the emitted light [C mode with spatial resolution full width at half maximum (FWHM) = 47 nm]. It is worth noting that the energy-integrated excitonic local density of states does not depend on position. So the observed structures are a direct consequence of the increasing ratio between radiative and nonradia-
Also, the I-v when sample involving length scales significantly larger than incoherent energy transfer between different regions of the sample is excited at quite a high energy and the far-field and local absorption spectra. Figure 3 shows good selectivity for PL measurements (C-tuned, C- modes give the same results when the C modes almost coincide). On the contrary, the I mode is selective only for photoluminescence excitation (PLE) measurements (ωC fixed, ωI tuned), being of poor selectivity for PLE. This picture can be better pointed out by looking at the sections of these gray-scale plots shown in Fig. 3. Panel 3(a) displays for reference the far-field and local absorption spectra. Figure 3(b) shows that the C and I-C modes give the same results when the sample is excited at quite a high energy and ωC is tuned. Also, the I and I-C modes almost coincide [see Fig. 3(c)] when ωC is fixed at a low energy absorption peak. However, Fig. 3(d) shows that in other circumstances the three working modes produce different results. It is also interesting to notice the differences in the peaks height between PLE and local absorption spectra. The strong nonequivalence between different SNOM kinds, here observed, is consequence (and at the same time a direct proof) of the nonlocal nature of the incoherent energy transfer between different regions of the sample involving length scales significantly larger than SNOM spatial resolutions (even in presence of well defined dots). Further evidence is given by the strong differences between the calculated local absorption and emission spectra in some working mode. Finally, we note the thermal activation of light up-conversion due to phonon-assisted exciton scattering clearly visible in the lower panels of Fig. 2.

In summary we have presented a microscopic quantum theory of spatially resolved photoluminescence in quantum wells with interface fluctuations. The theory establishes a clear functionality of the different SNOM kinds when applied to the analysis of semiconductor quantum structures. Moreover, this formulation permits one to model PL excitation spectroscopy in which the excitation and detection energies and spatial positions can all independently be scanned. The numerical results here presented constitute an intriguing example of the impact of sample temperature, exciton localization, and microscope setup in the formation of subwavelength-resolution images.