Photoluminescence spectra and level repulsion in quantum wires

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Luminescence spectra of V-groove GaAs/AlGaAs quantum wires are investigated by spatially resolved photoluminescence (PL) spectroscopy using a low temperature scanning near-field optical microscope (SNOM). The statistical analysis of these spectra in terms of the autocorrelation function reveals the presence of level repulsion. We find that the comparison between the experimental autocorrelation curve and the theoretical one obtained from the simulation of PL spectra, assuming for each exciton state a Lorentzian lineshape, is unsatisfactory. This is due to the presence of a broad background in the measured spectra, which is absent in the simulated ones. We propose an improvement of the theoretical model in order to include this feature and propose as explanation for the origin of this broad background the coupling between a single exciton state and the phonon thermal bath. The addition of this last element in our model is very important to establish a better quantitative agreement between the simulated and measured autocorrelation functions.

1 Introduction

Many experimental and theoretical studies have been devoted over the last two decades to the understanding of the qualitative and quantitative consequences of interface roughness in semiconductor nanostructures on the electronic states and their optical and transport properties [1]. One of the most important consequences is that interface roughness leads to fluctuations of the confining electric potential and creates localized states, thus opening the way to the study of fundamental properties of disorder in semiconductor nanostructures. One general prediction of the quantum theory of disorder is the appearance of correlations in the statistics of energy-level distances for pairs of localized states with finite spatial overlap [2]. This correlation manifests itself in different ways depending on the shape of the disorder potential. It can go from the simple discretization of quantum levels in a single potential well to the more subtle correlations existing between wave functions weakly localized in a random potential with very short spatial correlation length. It is now widely accepted to group these different scenarios under the common designation of level repulsion [3].

Recently this effect has been evidenced on excitonic levels in disordered quantum wells [4, 5] and quantum wires (QWRs) [6] by measuring near-field PL spectra. In these papers the comparison between experiment and theory was made assuming the equivalence between measured PL spectra and computed absorption spectra. We have shown in a former publication [7], that the measured near-field spectra in QWR...
samples present signatures of a nonthermalized exciton population. We argued that a correct treatment of exciton relaxation processes mediated by phonons in a disordered potential would be necessary to model correctly the measured PL spectra and to perform a direct comparison between experiment and theory.

In this paper we present experimental evidence for level repulsion in QWRs and we develop a model allowing to perform this comparison. First, we show that the assumption of a Lorentzian lineshape for the exciton emission lines in our near-field PL spectra simulations leads only to a partial agreement with the experimental curve and that this is due to the presence of a broad background in the measured spectra, which is absent in the simulated PL. We propose as origin of this background the coupling of a single exciton state to the acoustic phonon thermal bath, which in the following we will call the diagonal coupling between exciton and phonons. This phenomenon has already been experimentally observed and theoretically described in the context of strongly confined quantum dots [8, 9, 10]. Including this element in our model the agreement between experiment and theory is improved.

2 Experiment

SNOM PL spectra are recorded at low temperature \(T = 8\) K in collection mode with 250 nm spatial resolution. The QWR-sample is excited with the 2.41 eV line of the \(\text{Ar}^+\) laser over a 50 \(\mu\)m diameter spot. The excitation power is 5 \(\mu\)W. The emitted PL is collected by a pulled and uncoated optical fiber tip and dispersed by a double monochromator (25 \(\mu\)eV spectral resolution). The detection is performed with a cooled CCD camera. The near-field spectra are collected along the wires at regular intervals of 350 nm. We took a number of spectra around 350. The average of the normalized spectra shows a smooth and broad emission peak with \(\text{FWHM} = 8\) meV, which corresponds to the far field emission spectrum (left panel in Fig. 1). In contrast, an individual near-field spectrum features several intense and sharp emission lines (right panel in Fig. 1). These well-resolved peaks are due to the emission of excitons localized within the collecting near-field spot by the quantum wire inhomogeneities. Note the presence of a broad background, extending below these narrow emission peaks.

It is expected, in a single near-field PL spectrum, that an important part of the exciton states are spatially correlated, and that their wave functions overlap, which must lead to quantum mechanical level repulsion. We applied the same statistical analysis to our data as in former publications [4, 5, 6] and analyzed the near-field spectra in terms of the autocorrelation function \(\Delta C(\Delta E)\). This function is related to the probability

![Fig. 1: Sum of the measured near-field spectra (left panel) and typical near-field spectra at fixed spatial positions along the QWR (right panel).](image1)

![Fig. 2: Experimental (left panel) and simulated (right panel) autocorrelation function at \(T = 8\) K. Level repulsion is evident from the shoulder in the experimental curve around 2 meV. The theoretical autocorrelation function instead features a strong peak at about the same energy.](image2)
to find two exciton emission lines separated energetically by $\Delta E$. The signature of level repulsion is a dip in $\Delta C$ for $\Delta E \to 0$.

Using our model, we solved the Boltzmann rate equation for the exciton population $N_\alpha$, as described in [7], and calculated the near-field PL spectra, assigning to each exciton state $\psi_\alpha$ (with energy $\epsilon_\alpha$) a Lorentzian lineshape:

$$I(z, \omega) \propto \sum_\alpha N_\alpha \frac{\Gamma_\alpha^{\text{rad}}(\Gamma_\alpha^{\text{phon}} + \Gamma_\alpha^{\text{rad}})}{(\omega - \epsilon_\alpha/\hbar)^2 + (\Gamma_\alpha^{\text{phon}} + \Gamma_\alpha^{\text{rad}})^2} \cdot f(z).$$ (1)

$\Gamma_\alpha^{\text{rad}}$ and $\Gamma_\alpha^{\text{phon}}$ are respectively the radiative recombination rates and the phonon scattering rates to other exciton states. $f(z)$ is a spatial filter function resulting from the convolution of a Gaussian function and the square of the wave functions which selects the luminescence of states localized in a segment of about 250 nm around the coordinate $z$ of the wire. We performed then the same statistical analysis on these simulated spectra, as on the measured ones.

The experimental autocorrelation function features a strong peak at $\Delta E = 0$ (left panel of Fig. 2). This peak is due to the self-convolution of each emission line and to the self-convolution of the noise spikes in the spectra. Some evidence of the expected dip, when $\Delta E$ is close to zero, can be seen as a shoulder at about 2 meV, but is largely hidden by the self-convolution of the single emission lines. However, the emission lines are narrow enough and the effect of level repulsion should appear. This shoulder is the signature of quantum mechanical level repulsion of excitons in QWRs. The theoretical curve features instead a much more pronounced peak at about 2 meV and the level repulsion dip is clearly visible. Thus the comparison of the experimental and theoretical autocorrelation curves is not very satisfactory (right panel of Fig. 2).

3 Absorption lineshape If the only contribution to the lineshape of an exciton state were the sum of the radiative recombination and phonon scattering rates its form would be Lorentzian. In particular the contribution of the diagonal terms in the phonon scattering rates matrix as calculated in [7] is strictly zero. However this is a result obtained in first order perturbation theory. It is possible to treat the diagonal problem of the coupling of a single exciton state to the phonon thermal bath exactly and it has been shown theoretically [8, 9] and experimentally [10] that it introduces important changes in the absorption spectrum. This diagonal contribution of the exciton-phonon interaction does not involve transitions between different exciton states (i.e. relaxation processes), it is a pure dephasing mechanism. The theoretical frame is that of the independent bose model [12] and the main change in the absorption spectrum is the appearance of a broad background below the sharp zero-phonon peak.

We consider the coupling of one exciton state $\psi_\alpha$ (with energy $\epsilon_\alpha$) with the acoustic phonon thermal bath through the deformation potential matrix element $M_{\alpha \beta}^q$. The expression for the polarization induced by a $\delta$-like laser pulse is [8]:

$$P_\alpha(t) = -i\theta(t)\exp\left[\frac{-i\epsilon_\alpha t}{\hbar}\right] + R_\alpha(t) - R_\alpha(0) - t\dot{R}(0) - (\Gamma_\alpha^{\text{phon}} + \Gamma_\alpha^{\text{rad}})t]$$

$$R_\alpha(t) = \sum_q (M_{\alpha \beta}^q)^2 \left[ \frac{N_q}{\omega_q^2} e^{i\omega_q t} + \frac{N_q + 1}{\omega_q^2} e^{-i\omega_q t} \right],$$ (2)

where $\omega_q$ is the acoustic phonon frequency. The absorption spectrum is obtained by Fourier transforming the polarization and taking the square modulus. Now the formula for the simulated near-field PL spectra reads:

$$I(z, \omega) \propto \sum_\alpha N_\alpha \left| \int dt P_\alpha(t) e^{i\omega t} \right|^2 \cdot f(z).$$ (3)
As shown in Fig. 3 the change obtained in a single near-field spectrum is not very strong. However a slight increase of the PL intensity between the narrow emission lines is evident. On the contrary the difference in the autocorrelation function is remarkable (see Fig. 4). Its shape has completely changed and in particular the dip in the autocorrelation function has completely disappeared. This leads to a much better agreement between experimental and simulated curve, even if small differences still remain visible.

4 Conclusions We present evidence for level repulsion in QWRs using near-field spectroscopy and statistical analysis. The comparison with a simple model for exciton photoluminescence based on Lorentzian lineshapes leads only to a partial agreement between measured and simulated autocorrelation curves. The diagonal coupling of a single exciton states to the phonon bath and the subsequent appearance of a broad background reveals an important effect in the autocorrelation function and is essential in order to obtain a good agreement between measurements and theoretical model.

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