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Quantum mechanical repulsion of exciton levels in a disordered quantum well evidenced by near-field spectroscopy

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Abstract

Near-field photoluminescence spectra of a single quantum well dominated by emission from localized excitons are subjected to a statistical analysis of the two-energy autocorrelation function. This analysis is compared to a theoretical model of the exciton center-of-mass motion in a two-dimensional spatially correlated disordered potential. The spatial and energetic parameters of the disorder potential underlying the excitonic motion are extracted and clear signatures of quantum-mechanical energy level repulsion are revealed. © 2002 Elsevier Science B.V. All rights reserved.

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Localized states play a key role for the optical and transport properties of disordered quantum systems, e.g., thin quantum wells. Here, exciton localization on a nanometer scale due to unavoidable growth irregularities, i.e. interface roughness and alloy fluctuations, gives rise to pronounced inhomogeneous broadening of far-field optical spectra. In experiments with high spatial and spectral resolution, the smooth, inhomogeneously broadened photoluminescence (PL) spectra break up into many narrow emission spikes from single localized excitons whose individual spectral widths are often determined by experimental resolution or by the natural line width [1–4]. So far, it has been difficult to infer precise information about the underlying disorder potential or the nature of the excitonic wave functions from such spectra.

In this paper, we demonstrate both experimentally and theoretically that this information is contained in the energetic correlations of single exciton spectra. A statistical analysis of the two-energy autocorrelation function of near-field PL spectra of a single quantum well reveals the correlation length of the underlying disorder potential and a clear signature of the quantum-mechanical nature of the disordered system [5].

The sharp luminescence lines characterizing spatially resolved spectra are a direct consequence of exciton localization within a disorder-induced potential. In the center-of-mass approximation [6], the potential $V(R)$ felt by the exciton results from a convolution of

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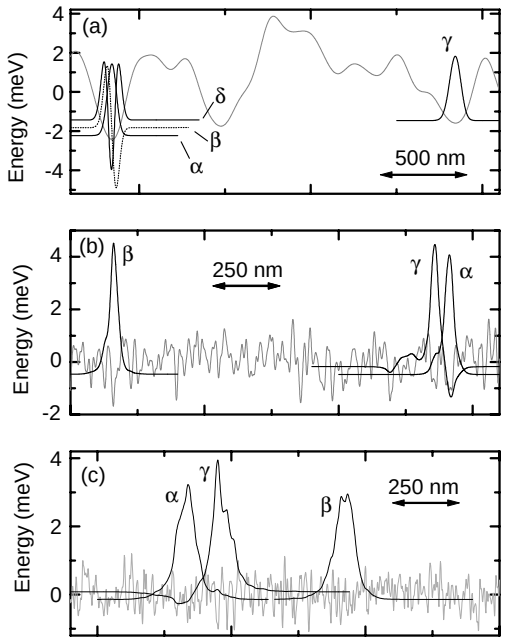

Fig. 1. Schematic illustration of the level repulsion effect for three different disordered potential configurations characterized by $\sigma/E_c = 36, 1, 0.01$, respectively. Spatially overlapping localized wave functions are energetically nondegenerate.

the microscopic confinement energy profile with the exciton 1s-wave function. Within this picture, the energetic position of the localized emissions is governed by the solution of the stationary Schrödinger equation for the excitonic eigenstates within the effective disorder potential $V(R)$. Following Ref. [7], in our model the disorder potential $V(R)$ is characterized by the average potential amplitude σ, and the spatial correlation length ξ [7]. It is known [7] that all the average properties resulting from the solution of the Schrödinger equation depend on the dimensionless parameter σ/E_c, where $E_c = \hbar^2/(2M\xi^2)$ with M the exciton mass.

A general feature of all quantum systems is the so-called level repulsion, i.e. if the wave functions of two eigenstates are spatially overlapping, their eigenenergies cannot be degenerate but split into two levels of different energy. In the following, we illustrate this quantum effect for the case of excitonic wave functions in one-dimensional model potentials that are characteristic for the effective disorder potential in typical semiconductor nanostructures. In Fig. 1, exciton wave functions are sketched for three characteristic sets of parameters of the disorder. (a) The disorder potential is characterized by a correlation length, $\xi = 50$ nm, much larger than the exciton Bohr radius, i.e. the distance between two adjacent potential minima is much larger than 10 nm. In this case, for $\sigma/E_c = 36$, excitons are mostly localized in single potential minima. The three wave functions α, β, and δ can be regarded as the ground state and the first two excited states of an exciton localized in a single potential minimum and their energies are well separated. The wave function γ, localized in a different distant local minimum, has zero spatial overlap with the former three wave functions, therefore there is no restriction on its energy position (in this example γ turns out to be nearly degenerate with δ). If the correlation length is of the order of the exciton Bohr radius [$\xi = 13$ nm, $\sigma/E_c = 1$, Fig. 1(b)], there is sufficient overlap between wave functions in *neighboring* minima, α and γ, and their eigenenergies are separated by quantum-mechanical level repulsion. On the contrary, wave functions α and β are spatially separated and may thus be nearly degenerate. In the case of weak localization the correlation length is smaller than the exciton Bohr radius [ξ=2 nm, σ/E_c=0.012, Fig. 1(c)]. In this limit, wave function localization is strongly linked to multiple random scattering processes of excitonic wave functions [8] and the wave functions are delocalized over several minima of the disorder potential. Nevertheless, level repulsion works in this case as in the previous ones and spatially overlapping wave functions are not allowed to be degenerate.

The strength of level repulsion depends on the properties of the disordered potential felt by the exciton. A quantitative estimate of the level repulsion behavior as function of the only independent parameter of the problem, σ/E_c, is based on the two-energy autocorrelation of individual excitonic spectra [7]:

$$R_c(\Delta E) = R(\Delta E) - R_0(\Delta E) \propto \left\langle \int \mathrm{d}E' D_n(E') D_n(E' - \Delta E) \right\rangle - \int \mathrm{d}E' \langle D_n(E') \rangle \langle D_n(E' - \Delta E) \rangle. \quad (1)$$

Here, $D_n(E)$ is the optical density of a single spot n. In our work $D_n(E)$ is represented by the measured PL local spectra [5] and $\langle \cdots \rangle$ denotes the average over many measured spots.

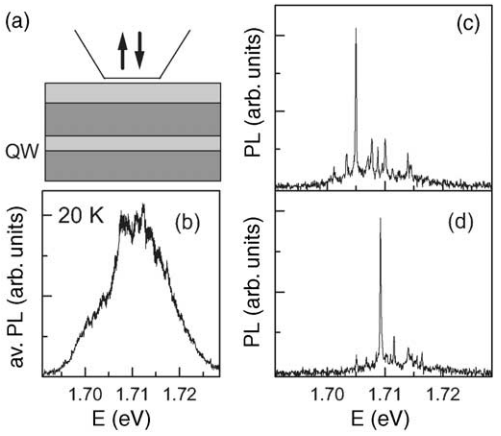

Fig. 2. (a) Schematic experimental setup. The quantum well sample is excited and the PL emitted by the sample is collected through the near-field fiber probe; (b) averaged (far-field) PL spectrum ($T = 20$ K); (c, d) representative near-field PL spectra recorded at two different spatial positions ($T = 20$ K).

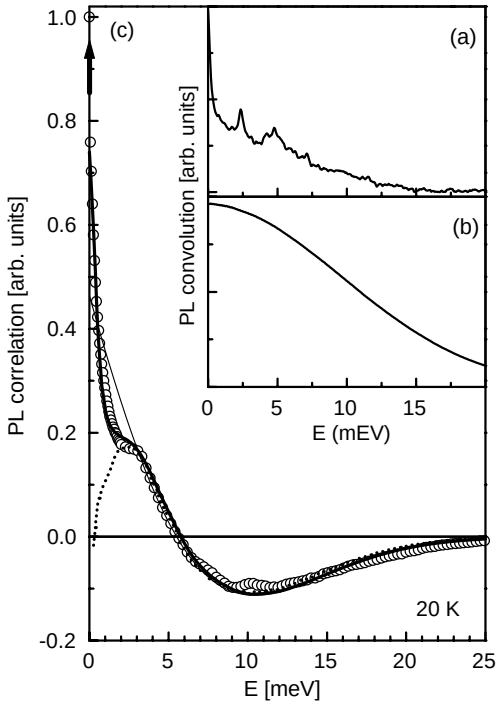

Fig. 3. (a) Two-energy autocorrelation function $R_c(\Delta E)$ of a set of 400 near-field PL spectra (circles) at $T = 20$ K compared to the quantum-mechanical theoretical simulation (solid line). The thin line shows the classical limit. The dotted line represents the raw numerical simulation without homogeneous line broadening—the δ-like self-correlation peak is denoted by the arrow at $\Delta E = 0$; (b, c) autoconvolution $R(\Delta E)$ of a single near-field spectrum and of the far-field spectrum, respectively.

The results in Fig. 1 demonstrate that in order to observe the level repulsion effect, one has to investigate the disordered system with a high spatial resolution of the optical experiment. With decreasing spatial resolution, the contribution from spatially non-overlapping and energetically degenerate states strongly increases, and the features of level repulsion are expected to be obscured. This is why we employ near-field scanning optical microscopy (NSOM) [9] providing a combined spatial and spectral resolution of 150 nm and 100 μV to measure spatially resolved photoluminescence (PL) spectra. In the investigated buried nanostructures evanescent fields components are expected to be of minor relevance and the image contrast is similar to that in far-field microscopy, yet with considerably improved spatial resolution. The excitation energy and excitation power coupled into the fiber probe had values of 1.96 eV and 800 nW, respectively. As a model system, we investigated a 3 nm thick GaAs/AlGaAs quantum well grown on a GaAs (3 1 1)A substrate [10] at a sample temperature of 20 K.

In Fig. 2, results from the PL experiments are summarized. The near-field PL spectra recorded with high spatial and spectral resolution are dominated by a set of spectrally sharp and intense emission lines. In contrast, the spatially averaged PL spectrum is characterized by a 15 meV broad structureless emission band (Fig. 2(b)).

Figs. 3(a) and (b) show the autoconvolution function $R(\Delta E)$ for a single near-field spectrum [Fig. 2(d)] and for the normalized far-field spectrum [Fig. 2(b)], respectively. The main characteristics of the single near-field spectrum autoconvolution are (i) the narrow spike around $\Delta E = 0$, due to the self-convolution of the individual emission spikes, (ii) different correlation resonances at higher energy values, reflecting a typical energy separation of 2–3 meV between PL spikes characteristic for this individual spectrum, and (iii) a large background, that monotonically decreases. The first two features, and in particular the correlation resonances, are absent in the autoconvolution function of the far-field spectrum. In fact, this spectrum is dominated by states which do not overlap and

therefore do not show correlation. In order to decide if, in the near-field autoconvolution curve, the absence of correlation resonances for $\Delta E < 1$–2 meV can be related to level repulsion, an average over more than 400 individual data sets is performed. In Fig. 3(c) the experimental curve $R_c(\Delta E)$ (open circles), obtained by applying [Eq. (1)], represents the near-field spectrum autoconvolution average from which the uncorrelated part due to the far-field autoconvolution has been subtracted. The peaks at nonzero energies which are found in the individual correlation functions [Fig. 3(a)], are averaged out. However, the self-correlation peak and the quantum mechanical level repulsion, which manifests itself in a clear shoulder at $\Delta E =$ 1–2 meV, are clearly recognizable. This curve is compared to that calculated within a quantum theoretical treatment of the exciton center-of-mass motion in a two-dimensional spatially correlated disordered potential [6]. The pronounced level repulsion dip, that is predicted in the theoretical simulation (dotted line) is weakened into a shoulder, since the theoretical δ-like self-correlation peak is broadened by the homogeneous linewidth of the individual exciton spikes. The agreement between the experimental correlation and the theoretical simulation, Lorentz-convoluted in order to account for the finite PL line width (solid line), is excellent and strikingly different from the classical limit (thin line) which neglects level repulsion due to wavefunction overlap. Treating excitons as classical particles means to neglect the kinetic term in the center-of-mass Schrödinger equation [6]. Consequently all the energies of the disorder-induced potential are solutions of the eigenvalue problem. Hence, $R_c(\Delta E)$ in the classical limit is solely given by the energy correlation of the potential itself. For values that are larger than the typical level repulsion correlation energies, the quantum mechanical contribution to the autocorrelation vanishes and the data points follow the classical limit.

By comparing autocorrelations for different values of σ and ξ to the experimental data, we find that σ can be extracted with high accuracy by the large energy part of the classical autocorrelation. The potential correlation length ξ that is difficult to derive with other experimental methods, strongly influences the energy position and amplitude of the level repulsion dip. This allows to determine for the parameters of the investigated quantum well disorder potential the following values, $\sigma = (5.3 \pm 0.2)$ meV and $\xi = (17 + 8, -3)$ nm.

In conclusion, by combining experimental near-field spectra and theoretical simulation based on a center-of-mass model we provide the first clear evidence for level repulsion in a disordered quantum well and open the way to a systematic characterization of disordered semiconductor systems by means of correlation techniques providing new insight into the growth-related features of semiconductor nanostructures.

References

[1] K. Brunner, et al., Phys. Rev. Lett. 73 (1994) 1138.
[2] H.F. Hess, et al., Science 264 (1994) 1740.
[3] D. Gammon, et al., Phys. Rev. Lett. 76 (1996) 3005.
[4] F. Intonti, et al., Phys. Rev. B 63 (2001) 075313.
[5] F. Intonti, et al., Phys. Rev. Lett. 87 (2001) 076801.
[6] R. Zimmermann, F. Große, E. Runge, Pure and Appl. Chem. 69 (1997) 1179.
[7] V. Savona, R. Zimmermann, Phys. Rev. B 60 (1999) 4928.
[8] P.W. Anderson, Phys. Rev. 109 (1958) 14982.
[9] G. Behme, et al., Rev. Sci. Instrum. 68 (1997) 3458.
[10] R. Nötzel, et al., Jpn. J. Appl. Phys. 35 (1996) L297.