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Exciton relaxation and quantum mechanical level repulsion

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Abstract. Theoretical results for spatially resolved exciton spectra of quantum wells with disorder are discussed. As a consequence of quantum mechanical level repulsion, the autocorrelation of the sharp peaks corresponding to individual quantum states yields information on their spatial extent. Level repulsion is present not only in the density of states but as well in absorption and even luminescence. More generally, the question under what conditions level repulsion can be observed in disordered systems with localized states is addressed.

Introduction

Recent years have seen a strong interest in spatially resolved spectra from both microphotoluminescence (μPL) and near-field scanning optical microscopy (NSOM). The inhomogeneously broadened macroscopic luminescence is resolved into often hundreds of narrow lines with widths of a a few tens of μeV, compatible with the radiative broadening [ ]. We show that a statistical analysis of their autocorrelation allows to "measure the size of the wave functions".

First, we discuss in Sec. 1 the phenomenon of level repulsion. In order to study luminescence, we discuss in Sec. 2 the relaxation kinetics of excitons. An illustrative example is given in Sec. 3.

1 Level repulsion in quantum mechanical systems

It was understood early in the development of quantum mechanics that eigenenergies repel each other. Textbook examples include the avoided crossing of two levels as function of an external parameter and random matrix theory [ ]. Eugene Wigner and Freeman Dyson proposed the latter as statistical description for spectra of highly excited nuclei. For the model of large Hamilton matrices with randomly chosen matrix elements, they could show that the probability to find a small level spacing is reduced by a factor

$$C_\rho(E) = \left\langle \sum_{m,n} \delta(E - \epsilon_m + \epsilon_n) \right\rangle \sim \left| \frac{E}{\Delta_\rho} \right|^\beta \tag{1}$$

with the average level distance Δ_ρ and an integer exponent $\beta = 1, 2, 4$ depending on the time-reversal properties of the system. For exciton spectra in the absence of magnetic fields, we have real orthogonal matrices (Gaussian Orthogonal Ensemble, GOE) and consequently $\beta = 1$.

If, in contrast to the random matrix model where each state interacts with every other state, the system is composed of M independent compartments not interacting with each other, the level repulsion effect is reduced by a factor $1/M$

$$C_\rho(E) \sim \left(1 - \frac{1}{M}\right) + \frac{1}{M} \left| \frac{E}{\Delta_\rho} \right|^\beta \quad . \tag{2}$$

1.1 Localization and level repulsion

The absence or presence of level repulsion has been proven recently as a very valuable tool in the analysis of numerical simulations for the Anderson localization transition [3]. Thus it might be questionable to look for level repulsion in spatially resolved spectra of two dimensional excitons at all.

We will now argue, that at least three different scenarios allow to see level repulsion in localized systems, in spite of the statement of Eq. (2).

(i) Spatially resolved excitation or detection: μPL or NSOM experiments excite excitons only in a region A_{det} which is not much larger than the area A_{loc} of the excitons center-of-mass wavefunction. Eq. (2) becomes

$$C_\rho(E) \sim \left(1 - \frac{A_{\mathrm{loc}}}{A_{\mathrm{det}}}\right) + \frac{A_{\mathrm{loc}}}{A_{\mathrm{det}}} \left|\frac{E}{\Delta_\rho}\right|^\beta \tag{3}$$

which allows to estimate A_{loc}.

(ii) Matrixelement-weighted correlations: Many physical processes are determined not by correlations of energy levels, i.e., by the probability to find a given energy spacing, but by the combination of energy spacing and large transition matrix elements. Combinations such as $\langle \sum_{m,n} |M_{mn}|^2 \delta(E - \epsilon_m + \epsilon_n) \rangle$ can show level repulsion even in disordered systems, because near-degeneracies between spatially well separated regions do not contribute due to vanishing matrix elements [4].

(iii) Expectation values whose uncorrelated part vanish: If the dominant contribution vanishes, the $1/M$ term can survive. Generally, susceptibilities have this form. This has been used by Mott in his classical estimate for the susceptibility of the Anderson insulator. Recently, it has been pointed out that the long-time Rayleigh-scattering signal is of this type. The average electric fields vanish and only the correlated part survives [5].

According to *(i)*, level repulsion should be present in a spatially resolved density of states (DOS). Actual experiments almost exclusively measure luminescence, which differs from DOS in two aspects. First, only optically active states with a large zero-momentum content contribute. Second, luminescence spectra reflect the relaxation kinetics. Low-lying states carry a large weight in luminescence. Both aspects can be expected to decrease the amount of level repulsion: Optical active states are rare and thus one would expect it to be improbable that two states close in energy are both optically active. This effect should already be present in spatially resolved absorption spectra, if they could be measured. Regarding relaxation, one could at low temperature imagine a scenario of only local minima being populated and one might expect local minima to be well separated and thus non-interacting. These two arguments show that a detailed analysis for specific samples and temperatures is needed which includes both optical matrix elements and relaxation kinetics.

2 Relaxation kinetics

We discuss relaxation in terms of a kinetic equation for the occupation N_α of the disorder eigenstates ψ_α [6]:

$$\partial_t N_\alpha = g_\alpha + \sum_\beta \gamma_{\alpha \leftarrow \beta} N_\beta - \left(r_\alpha + \sum_\beta \gamma_{\beta \leftarrow \alpha}\right) N_\alpha \tag{4}$$

with generation rates g_α, radiative recombination r_α, and acoustic phonon scattering rates $\gamma_{\beta \leftarrow \alpha}$ [7]. The generation rate is determined by the specific experimental set-up. All other rates are calculated from numerically determined disorder eigenfunctions.

We focus here on deformation potential scattering involving acoustic phonons ($E = \hbar\omega_q = \hbar uq$), which is thought to dominate the exciton relaxation at least in the III-V materials at low, but not extremely low temperatures where piezoelectric scattering becomes relevant. The factorization approximation for the wave function,

$$\Psi(\vec{r}_e\,\vec{r}_h) = \psi_\alpha(\vec{\rho}\,)\,\varphi_{1s}(\vec{r}\,)\,u_e(z_e)\,u_h(z_h)\ , \tag{5}$$

has been shown to be applicable for typical III-V quantum wells [6].

Apart from thermal phonon occupation, the main ingredient of the scattering rate $\gamma_{\beta\leftarrow\alpha}$ is the center of mass matrix element $\int \psi_\alpha(\vec{\rho}\,)\,e^{i\vec{Q}\cdot\vec{\rho}}\,\psi_\beta(\vec{\rho}\,)$. Similarly, the radiative rate can be written in the factorization approximation as

$$r_\alpha = \frac{4\,e^2\,p_{cv}^2\,n\,E_X}{3\,\hbar^2\,c^3}\ \varphi_{1s}(0)^2\ \langle u_e|u_h\rangle^2\ M_\alpha^2 \tag{6}$$

with the momentum matrix element $p_{cv} = \langle s|\hat{p}|p_z\rangle$, index of refraction n, and exciton energy E_X. A reduction by scattering into dark exciton states is not included here. The confinement wave function overlap $\langle u_e|u_h\rangle$ typically is close to unity for the dominant transition. For strongly localized excitons (neglect of the finite photon momentum and thus polariton effects [6]) the center of mass part of (6) reduces to [6]

$$M_\alpha = \int d^2\vec{\rho}\ \psi_\alpha(\vec{\rho}\,)\ . \tag{7}$$

3 Results

An illustrative example for the solution of Eq. (4) and the resulting autocorrelation is given in Fig. 1. A rather small detection area of 120 nm × 120 nm corresponding to a good NSOM resolution is used. Averaging of the order of 250 spectra should be enough to give a well defined level repulsion dip at small energy difference. Required is however a very good energy resolution of the spectrometer. An almost complete level repulsion such as seen

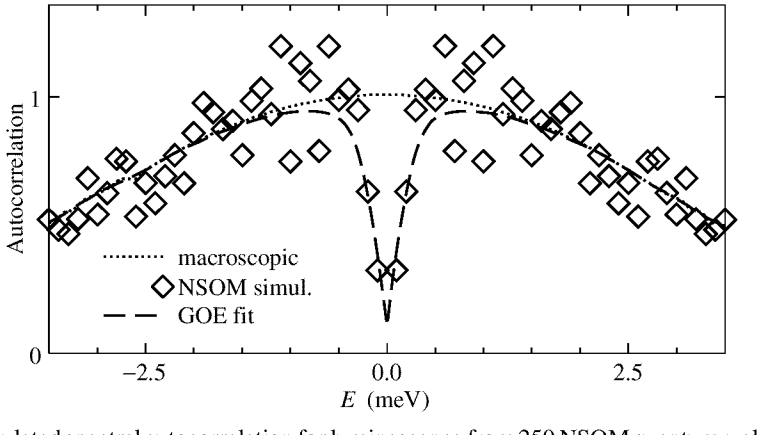

Fig. 1. Simulated spectral autocorrelation for luminescence from 250 NSOM quantum well spectra, each corresponding to an area 120×120 nm. Parameters of a 5 nm wide GaAs/$Al_{.3}Ga_{.7}As$ quantum well with disorder strength $\langle v^2\rangle^{1/2} = 8$ meV are used. GOE-fit (dashed) to the simulation data (diamonds) and corresponding fit to the autocorrelation of the sum of all spectra (macroscopic spectrum) (from Ref. [11].

in Fig. 1 implies, c.f. Eq. (2,3), that almost all wave functions within the detection area interact with each other and A_{loc} is only slightly smaller than A_{det}. This is confirmed by direct analysis of the numerically determined wave functions.

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