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Statistics of energy levels and photoluminescence in arrays of quantum dots

E. V. Tsiper,* A. L. Efros
Department of Physics, University of Utah, Salt Lake City, Utah 84112

A model is proposed which allows to explain quantitatively the behavior of PL and PLE spectra of large arrays of isolated quantum dots (QDs). Recent experiment[1] shows existence of a large Stokes-like shift between PL and PLE. The spectra consist of broadened peaks with the broadening of the order of the distance between them. The PL peaks show a peculiar interplay when the excitation energy is varied. An interplay is absent in the PLE spectra.

The presented theory explains all these features based on a picture of statistical distribution of energy levels in quantum dots. It is assumed that the luminescence occurs only from the lowest energy state of an excited QD due to the fast relaxation. We suggest that QDs which are pumped into this lowest energy state do not contribute to the spectra due to the extremely small linewidth associated with a single QD. The corresponding response is masked with the high intensity of the exciting light. The first peak observed is due to the QDs that absorb light into the next excited state. The shift is therefore equal to the average distance between the two lowest energy states of QDs in the array.

It is shown that the first two peaks observed are the result of the lifted degeneracy of the excited state of QD. An assumption is made that the fluctuation of the position of this level and its splitting have the same origin. This explains the same scale of the broadening of peaks and of the distance between them and allows to describe the whole lineshape with a universal curve.

The intensity measured in both PLE and selectively-excited PL can be expressed as a single function of excitation and detection energies. This function is in turn determined by the distribution function of *pairs* of energy levels: $I(E_{ex}, E_{det}) \propto \sum_{m=1}^{\infty} |M_m|^2 P(E_{det}{=}E_0, E_{ex}{=}E_m)$, where E_m are the energy levels, and M_m are the matrix elements of the transitions. The smooth broadening of the specftra occurs due to the large number of single-QD contributions.

Different QDs have energy levels shifted randomly due to a number of factors, including compositional disorder, rough boundary, strain etc. Most of these factors can be accounted for by introducing a random potential $V(\mathbf{r})$ with a proper correlation function. For example, composition disorder is equivalent to the white-noise random potential. The potential V also splits degenerate energy levels if the degeneracy exists. Quite important is that the random shifts and splittings of different energy levels in the same QD are *correlated*, since they are determined by the same realization of the random potential V. At the same time, realizations of V in different QDs are independent.

The distribution function $P(E_0, E_m)$, which determines the shape of both PLE and selectively-excited PL spectra near $E_{det} = E_0$, $E_{ex} = E_m$, is calculated for the

case of axially-symmetric QDs. It is assumed that matrix elements of the potential V are normally distributed (though, correlated) random values. This assumption holds for the case of compositional disorder as well as for some type of distortions of the shape of the QDs. The result [2] is plotted in Fig. 1 (a,b).

The spectrum depends crucially on the *correlation coefficient* ρ, $\rho \leq 1$, which determines the correlation between E_0 and E_m. This coefficient is $\rho = 0.795$ for compositional disorder for two lowest levels of cylindrical QD. For the rough boundary $\rho = 1$. If both factors are present, ρ should take some intermediate value. The value $\rho = 0.94$ in Fig. 1(a) gives the best fit to the experimental data of Ref. [1].

The prediction can be made about an enhanced sensitivity of the spectra to the magnetic field. In an axially-symmetric QD, the magnetic field splits each energy level except the lowest one into two sublevels $\pm m\hbar\omega/2$ with opposite projections $\pm m$ of the angular momentum (ω being the cyclotron frequency). However, only those QDs are affected, for which the magnetic-field induced splitting is larger than the splitting Δ in zero field. Such dots are more likely to contribute to the center part of the broadened peak. Therefore, the central part of the peak is sensitive to a relatively small magnetic field. The calculated shape of the PLE spectrum in magnetic field is presented in Fig. 1 (c,d).

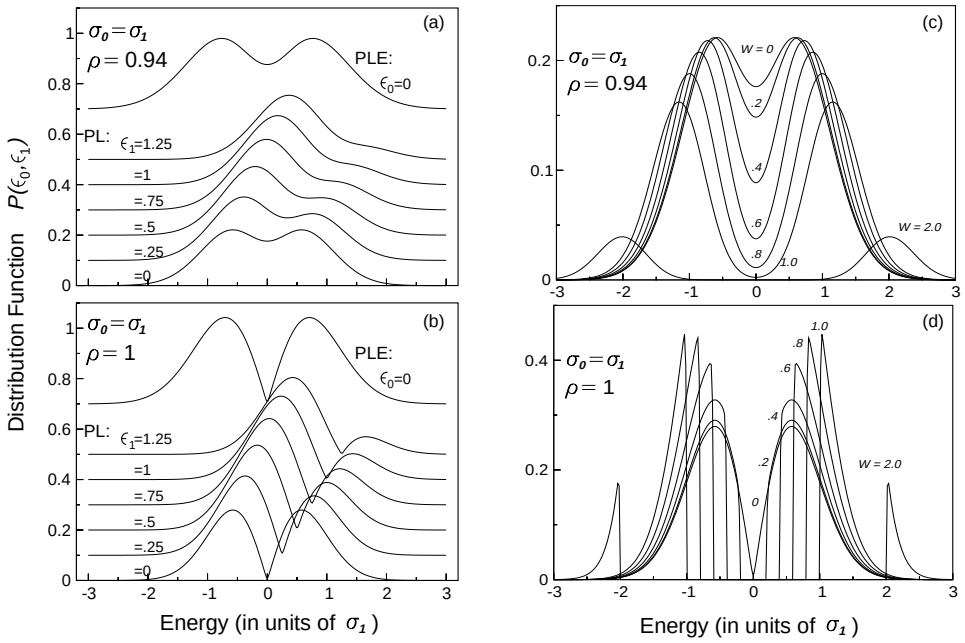

Fig. 1 (a,b) PL and PLE spectra without magnetic field. The widths $\sigma_{0,1}$ of the distributions of the first two excited states of QD are taken to be equal to each other. ϵ_1 is the deviation, in units of σ_1, of the excitation energy from the maximum of the PLE peak. (c,d) The PLE lineshape as a function of magnetic field. Parameter w= $m\hbar\omega/\sigma_0$.

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*e-mail: rr@physics.utah.edu.

[1] S. Fafard, D. Leonard, J. L. Merz, and P. M. Petroff, Appl. Phys. Lett. **65**, 1388, 1994.

[2] E. V. Tsiper, accepted to Phys. Rev. B, preprint No. cond-mat/9508149