OPTICAL SPECTRA LINE WIDTH IN QUANTUM DOTS

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The single quantum dots (QDs) are similar to individual atoms or molecules. Despite the relative simplicity of such systems their behavior has not yet been satisfactorily understood.

The electronic system of quantum dots relaxes quickly the electronic energy, the spectral lines of the optical transitions show a finite width, and in certain cases they display a special shape of the spectral density homogeneous line profile[1]. While the rapidity of the electronic energy relaxation has been interpreted in the frames of at least two mechanisms, the one considering the purely electron-electron interaction, and the other using only the electron-phonon interaction, the question of the optical line shape of the individual quantum dot electronic system has so far been discussed perhaps only with help of the electron-phonon interaction.

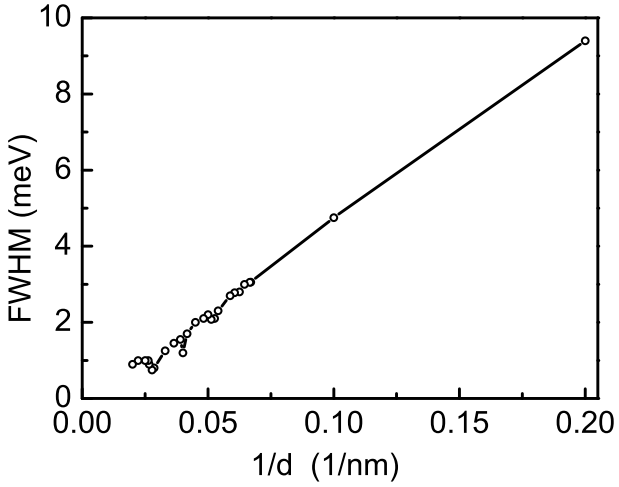

Fig. 1 The full width at half maximum (FWHM) of the main peak of the lower energy (n=0) electronic spectral density of the InAs quantum dot, at lattice temperature of 10 K, is plotted against the inverse of the quantum dot lateral size *d*. At every value of the quantum dot lateral size at first the stationary distribution of the electron density among the two energy levels is computed. Then the FWHM of the spectral density main feature is determined.

In the present contribution we discuss the theory of the full width at the half maximum (FWHM) of the main peaks of the optical transitions in the electronic subsystem of the individual quantum dots, basing on the electronic interaction with the longitudinal optical phonons. The theoretical conclusions are then compared to experiments.

From the theoretical point of view, we can view the optical transition of the photon emission in quantum dots as an annihilation of an electron in the conduction band states with a hole in the valence band states. In this process we neglect the motion of holes and approximate their infuence by a static charge added to the potential well of the electron[1]. Assuming that the spectral density of such a heavy hole can be expressed as a delta function, then the spectral line shape of the luminescence transition can then be expressed approximately as the spectral density of the electron in the conduction band states of quantum dot.

The quantum dot will be assumed to have a cubic shape with infinitly deep electronic potential. We shall take into account only the electronic ground state (n = 0) and one of the triply degenerate excited states (n = 1). The electronic spin will be neglected.

The phonon-based theory of the electronic optical transitions in quantum dots is considered in the self-consistent Born approximation (SCBA) for the electronic self-energy. This approximation of a multiphonon nature is applied both to the formulation of the electronic spectral density and to the theory of the electronic kinetics in the quantum dots. The Reader can see the details in the ref. 1 and the references cited in. To be specific, let us also remark that we ignore the so called ‚overheating' effect[1] here. The electronic spectral density in a quantum dot was considered theoretically in an earlier reference[4], under the assumption that the electron in an isolated quantum dot is influenced mainly by its interaction with the longitudinal optical phonons. The electronic spectral density is given by the imaginary part of the nonequilibrium Green's function. The Green's function is determined by the electronic self-energy, which is found by solving the self-consistent equation for the electronic self-energy for the two energy levels of the present model, assuming the electron-LO-phonon coupling[1].

We expect that in the experiment, the optical transitions are due to the annihilation of the hole in the valence band state with the electron in the lowest energy state (n=0) of the two conduction band states. The electronic spectral density of the n=0 is calculated numerically in the dependence on the lattice temperature and on the latteral size d of the quantum dot. The material parameters for the determination of the electron effective mass and the static and high-frequency

dielectric constants are taken those of InAs or CdSe bulk materials.

In the study of the line width behavior we pay attention to the zero-phonon feature of the electronic spectral density of the lower-energy state n=0. The resulting plot of FWHM against $1/d$ is shown in Fig. 1. The open circles mark the computed points, while the continuous lines serve as a guide for the eye. Let us remind that the electronic spectral density depends on the distribution of the electron among the two energy levels. In the present model this electronic distribution need to be calculated separately and is given by the condition that the electronic relaxation rate between the two energy levels, due to the electron-phonon interaction, is zero[5].

Comparing experimental characteristics of the optical spectra[2,3] with the theory we see an interesting agreement between the experiment and the theory based on the SCBA approximation. In particular, in Fig. 1 the calculated FWHM dependence on $1/d$ is shown as nearly linear, which feature is in agreement with experimental measurements[3] in CdSe nanocrystals. The slight modulation, to which we paid attention especially in the region of about $(1/d) \in (0.02, 0.07)$, occurs in InAs approximately near certain resonance situations. These situations occur at such $1/d$, at which the electronic energy level separation ΔE equals an integer (m) multiple of the optical phonon energy E_{LO}, in other words, at $\Delta E = mE_{LO}$. We do not pay attention in the present work to the individual resonance features in the Fig. 1, posponing a detailed consideration of the shape of the resonance features to a later work. One of the reasons may be that the technological control of changing the quantum dot or nanocrystal size may be difficult. An experimental observation of this modulation of the mostly straight line would nevertheless give a considerable support to the importance of the electron-LO-phonon interaction in QDs.

The same calculations as in InAs were performed for the case of the CdSe quantum dots, Fig. 2. On average, the presented calculated data show an approximately straight line going through the origin of coordinates after the linear extrapolation. This character is in agreement with the measurement[2]. The theoretical results show a rather large modulation of the dependence of FWHM on the inverse lateral size, probably due to the much larger polaron constant of the CdSe material. While in InAs quantum dots the magnitude of FWHM parameter appears to have values about comparable to the the FWHM values in experiments[3], in CdSe the values of FWHM are about one order larger than shown by measurements[2]. From this reason one may consider an improvement of the approximations used in the present theory of the line width. It is interesting that the experimental results obtained by measuring the line width in CdSe nanocrystallites[6] give FWHM values rather close by order of magnitude to our theoretical estimate.

The data shown in Fig. 2 have not been possible to be extended to include $1/d$ larger than about 0.14 of 1/nm. The reason is that beyond this limit of the values of $1/d$ the individual peaks of the multiphonon spectral structure of the lower energy state (n=0) coalesce in the calculated spectrum of CdSe quantum dots.

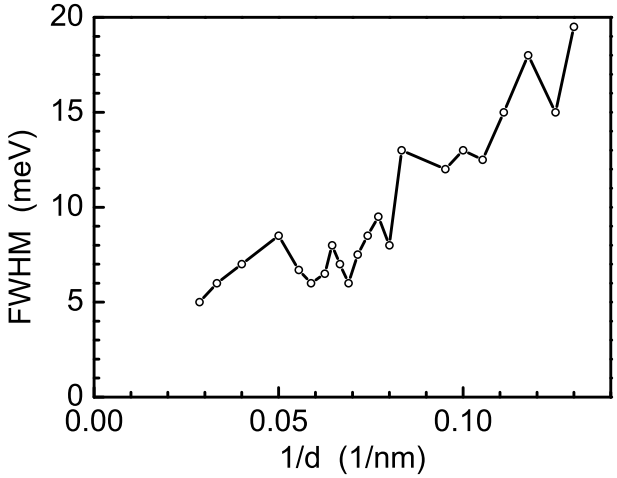

Fig. 2 The plot of FWHM for CdSe quantum dot. See caption of Fig. 1 for other data.

In conclusion, to the best knowledge of the authors the resonance modulations measurements of the dependence of FWHM on the inverse lateral size of the dot probably have not been reported. The reason might have been that the experiments did not include the data corresponding to the resonance features for small integers m, for which the resonances may be the most pronounced. Besides the measurements like those on CdSe quantum dots[2,6], and on InAs quantum dots[3], more detailed measurement would be welcome in order to verify the resonance features calculated here.

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