Differences between photoluminescence spectra of type-I and type-II quantum dots

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Abstract. Semiconductor quantum dots which trap simultaneously electrons and holes are called quantum dots of type-I. Contrary to these structures, empty dots of type-II attract only one type of charged carriers and repel the other. Particularities of confining potential are unaccessible by any direct measurements, thus recognition of quantum dot type by indirect method is highly desired. Our proposal is to distinguish between the two types of quantum dots via a comparison of photoluminescence spectra of these structures, which differ in both cases qualitatively.

1. Introduction

Contrary to quantum dots (QDs) of type-I which trap simultaneously electrons and holes, empty dots of type-II attract charge carriers of one type and repel the other \cite{1}. Fig. 1 illustrates this classification in terms of band layout - QDs of type-I have contravariant band layout (Fig. 1a) while QDs of type-II have covariant band layout (Fig. 1b).

This differentiation is especially interesting for self-assembled QDs which are formed as a result of strain between two lattices with significantly mismatched lattice constants (Stransky-Krastanow method) \cite{2}. Depending on the type of used materials on the one hand we have self-assembled quantum dots which attract holes and repel electrons (e.g. GaSb/GaAs \cite{3} or Ge/Si \cite{4, 5}), and on the other hand we deal with self-assembled QDs which attract electrons and repel holes (InP/InGaP \cite{6, 7, 8} or InP/GaAs \cite{9}). Moreover, it has been verified that for certain self-assembled QDs (e.g. QDs on GaAs substrate) strain forces can considerably alter local band edges (creating a ring-shaped potential well around the dot for heavy-hole confinement) and thus convert a quantum dot type \cite{9} from I to II.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure1}
\caption{Band layout for QD of type-I (a); band layout for QD of type-II (b), cb - conduction band, vb - valence band. The strain on the edges of self-assembled QD of type-I, eg. GaAs/InAs, can change its type (a) $\rightarrow$ (b) (a particular case of strong modification concerns heavy-hole band).}
\end{figure}
Particularities of confining potential are unaccessible by any direct measurements, thus recognition of the type of a dot by indirect method is highly desired. Our proposal is to distinguish between two types of quantum dots via a comparison of photoluminescence (PL) spectra of these structures, which differ in both cases qualitatively.

Although quantum dots of type-II attract only one type of carrier and repel the other, the electron-hole pair (exciton) can still be captured due to the Coulomb attraction of opposite charges. Moreover, in the presence of moderate magnetic field at least one type of carrier can be confined in the double-well structure (formed by the interplay of the Coulomb force and the repulsive quantum dot potential). This results in a characteristic multi-peak structure of PL spectrum of type-II dots, highly sensitive to magnetic field magnitude and size of the dot. Such a behavior gives us a possibility to distinguish between type-II and - much less complex - type-I dots by comparison of these qualitative PL features.

2. Model

We use a Gaussian-shape well as a more realistic model of confining QD potential comparing to commonly used parabolic approximation. This approach conserves the parabolic curvature in the vicinity of potential’s center and simultaneously allows for an escape of highly excited carriers.

We restrict our considerations to the quasi-two-dimensional quantum dot, which can be modelled by a 2D Gaussian \[ V(r) = \mp V_0 \exp\left(-r^2/L^2\right) \] where the upper sign corresponds to an electron and the lower to a hole (or oppositely). \( L \) is the dot radius, and \( V_0 \) is the depth of the confining potential which for simplicity is considered equal for both electron and hole. We also assume a constant curvature \( \omega_0 \) of the potential (for Gaussian confinement given by \( \omega_0^2 = 2V_0/m^*L^2 \)), which leads to the condition \( V_0/L^2 = \text{const.} \)

It is then convenient to introduce a scale parameter \( \alpha = \hbar \omega_0/V_0 = 2\lambda_0^2/L^2 \) (where \( \lambda_0^2 = \hbar/m^*\omega_0 \)), which allows us to control the dot diameter and connected confining potential depth in a way that the curvature remains constant. The physical meaning of parameter \( \alpha \) is illustrated in Fig. 2a.

Additionally, we consider a magnetic field aligned across the quantum dot plane, i.e. along the \( z \) axis, which within the symmetrical gauge leads to the formula for the bare lateral potential

\[
V_i(r_i) = \mp V_0 \exp\left(-\frac{r_i^2}{L^2}\right) + \frac{1}{8} m_i^* \omega_{ci}^2 r_i^2,
\]

where the upper sign corresponds to the electron (\( i = e \)) and the lower sign to the hole (\( i = h \)), \( m_e^* \) and \( m_h^* \) are the effective masses of the electron and the hole, and \( \omega_{ci} = eB/m_i^*c \) are the cyclotronic frequencies.

Figure 2. A diagram explaining the meaning of parameter \( \alpha = \hbar \omega_0/V_0 \), controlling the QD size at constant potential curvature (a). Hartree potential for the hole (b) and the electron (c) in magnetic field \( B = 4T \) with respect to the dot dimension governed by size parameter \( \alpha \) in type II QD.
The energies and wave functions of the QD are obtained by solving the Hartree equations in the effective mass approximation. The wave function of the whole exciton is assumed as a product of single-particle wave functions (Hartree approximation) \( \Psi(\mathbf{r}_e, \mathbf{r}_h) = \Phi_e(\mathbf{r}_e) \Phi_h(\mathbf{r}_h) \), and is found via self-consistent solution of the single-particle Schrödinger equation for the electron and the hole in QD, with the confinement given by the effective Hartree potentials of the following form

\[
U_e(\mathbf{r}_e) = V_e(\mathbf{r}_e) - \frac{e^2}{\epsilon} \int d\mathbf{r}_h \frac{|\Phi(\mathbf{r}_h)|^2}{|\mathbf{r}_e - \mathbf{r}_h|},
\]

for the electron. The expression for the hole is obtained from (2) by interchange of the subscripts: \( e \leftrightarrow h \). The details of this calculations can be found in [10] (for low level of excitation) and in [12] (for high level of excitation).

3. Low level of excitation

At low level of excitation, when no more than single exciton is localized in the dot, for type-I QD only one peak is present in PL spectrum, while for QD of type-II we deal with a doublet of peaks (in the absence of the magnetic field). The reason for appearance of two peaks in the latter case is the double-well shape of the effective confining potentials for the hole and the electron simultaneously located in the dot. As we can see in Fig. 2b and Fig. 2c, for almost all values of the size parameter \( \alpha \), the Hartree potentials for the electron and the hole both exhibit a double-well structure. Such a shape of the potential together with the same rotational symmetry of the two lowest lying states of the electron as the hole leads to the appearance of metastable states (against dipole optical transitions) producing four theoretically possible peaks in PL spectrum of type-II quantum dot (some of these states can however be bound only in the presence of a magnetic field and only for certain values of parameter \( \alpha \)).

The physical reason for a double-well structure in type-II quantum dots can be explained as a rearrangement of bare lateral potential due to e-h attraction. In comparison, in the case of type-I QDs e-h Coulomb interaction only deepens both potentials and does not change them qualitatively. They still have the single-well structure for both carriers resulting in a single peak in the PL spectrum.

Inclusion of a perpendicularly oriented magnetic field enhances the electron and the hole localization. In our calculations this manifests itself in the appearance of an additional quadratic term in the bare lateral potential (1), however in classical terms this enhancement of localization takes place due to the well-known cyclotronic effect (the electron and the hole rotate in the opposite directions in the plane perpendicular to the magnetic field axis). For type-I QDs the cyclotronic effect narrows both \( e \) and \( h \) potentials, which results in a small blue-shift of PL features, but for QDs of type-II the influence of the magnetic field is much more sophisticated: the attractive bare potential for the electron is enhanced while the repulsive potential for the hole is diminished (or conversely, for the opposite band layout). The Hartree calculus for type-II QD indicates, that we have generally three distinct types of magnetic-field evolution of the PL spectrum: (a) for large dots (small \( \alpha \)) there are two peaks for all field magnitudes below a critical value, at which the second peak disappears, (b) for medium-sized dots there are two similar peaks for the field-free case, the third one emerges at moderate field magnitudes (\( B \leq 2 − 4T \)) and then disappears for medium fields (\( \sim 4T \)). Finally, for higher fields (\( \sim 6 − 8T \)) also the second peak disappears, (c) for small dots (\( \alpha > 0.7 \)) there is only one peak for all values of the magnetic field. This complicated behavior corresponds to the fact, that some of the possible four e-h states (corresponding to both \( e \) and \( h \) double-well effective potentials) are bound only for certain combinations of dot sizes and magnetic field magnitudes. The maximum number of bound states of a single exciton - three - predicted within our model at any magnetic field and dot size, agrees with the experiment of Bayer et al. [13]. Another convincing experimental confirmation of our theoretical predictions can be found in [14] (compare Fig. 1 and Fig. 4).
4. High level of excitation

For higher levels of excitation, when we deal with more than one exciton per dot, in addition to the simple exciton one has to consider possible formation of exciton-like multi-particle complexes e-e-h and e-h-h, which are called trions. For simplicity we consider only an e-e-h trion. Although similar, the Hartree analysis of this case [12] differs from the one for simple e-h exciton. In particular, various possibilities of mutual spin alignment of the electron pair (multiple singlet and triplet states) inside the e-e-h trion play a meaningful role. As a consequence, for sufficiently large type-II QDs ($\alpha < 0.5$), three peaks in PL spectrum can occur already in zero magnetic field, which further split into four when the magnetic field is applied. The reason for this is that without magnetic field both electrons can either occupy their ground states, both occupy the excited metastable states, or one occupies the ground state and the other the excited metastable state. In the first two cases the electron pair spins have opposite directions and only a singlet-type wave function for them is possible, whereas in the third case the applied magnetic field can split the symmetric-singlet state and the antisymmetric-triplet state. For small type-II QDs ($\alpha > 0.5$) only one state for the electron-hole pair is possible (singlet) and only one peak in PL spectrum is expected. All these theoretical predictions find a convincing experimental confirmation for known QD structures (compare Fig. 1 and Fig. 4 in [14] and Fig. 1 in [15]).

In the case of type-I quantum dots several levels in the electron and the hole potential wells can be occupied at high activation level. This can result in additional PL peaks due to the e-h annihilation from these higher excitonic states, however their mutual positioning should not be sensitive to the dot dimension and the magnetic field (except for the red shift with growing QD dimension), as opposed to the characteristic behavior exhibited by type-II QDs.

5. Conclusion

The presented analysis indicates that type-I and type-II QDs can effectively be distinguished with high fidelity via only optical observation of photoluminescence features. The key distinction between these two types of QDs lies in characteristic PL sensitivity to the dot dimension and the external magnetic field.

For low level of excitation [10], a presence of a dublet of PL peaks (strongly sensitive to the dot dimension) is predicted for almost all sizes of type-II QDs (except for very small dots), while for type-I QDs only single peak is present. Moreover, for medium-sized type-II QDs in the presence of medium magnetic field third peak can appear in the spectrum, while for higher magnetic fields only single peak remains for all QD sizes.

At high level of excitation [11], in type-II quantum dots three PL peaks appear at zero magnetic field, further splitting into four in magnetic field presence, while for type-I QDs the additional peaks would correspond to e-h annihilation from higher excitonic states, being insensitive to variation of the dot dimension and magnetic field.

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