Fine structure of excited excitonic states in quantum disks

M. M. Glazov, E. L. Ivchenko, R. v. Baltz and E. G. Tsitsishvili

A.F. Ioffe Physico-Technical Institute RAS, 194021 St.-Petersburg, Russia
Universität Karlsruhe, 76128 Karlsruhe, Germany

We report on a theoretical study of the fine structure of excited excitonic levels in semiconductor quantum disks. A particular attention is paid to the effect of electron-hole long-range exchange interaction. We demonstrate that, even in the axisymmetric quantum disks, the exciton P-shell is split into three sublevels. The analytical results are obtained in the limiting cases of strong and weak confinement. A possibility of exciton spin relaxation due to the resonant LO-phonon-assisted coupling between the P and S shells is discussed.

I. INTRODUCTION

In the envelope-function approximation, the excitonic states in semiconductors and semiconductor quantum dots can be classified by referring to the orbital shape of the exciton envelope function (S-, P-, D-like) and the character of the electron and hole Bloch functions (bright and dark states). For example, the state $|P_z y\rangle$ means a bright $P_z$-shell exciton with the electron-hole dipole moment directed along the $y$ axis. Theoretically, the fine structure of excited states of zero-dimensional (0D) excitons has been studied for excitons localized, respectively, by rectangular islands in a quantum-well structure, quantum disks with a Gaussian lateral potential, and lens-shaped quantum dot. However, up to now the splitting of excited-exciton levels has been analyzed for a fixed orbital shell, e.g., the $P_z$ shell, which is valid in the case of strongly anisotropic confinement so that the orbital splitting of the $P$-like shells $P_x$ and $P_y$ exceeds by far the exchange-interaction energy. Here we show that, for axially symmetric or square quantum dots, one has, in addition, to take into account the exchange-interaction-induced mixing between the excitonic states $|P_z y\rangle$ and $|P_{xy}\rangle$. Moreover, for the first time we focus on the interplay between the exchange interaction and anisotropic shape of the dot and develop an analytic theory in the particular case where the orbital and exchange splittings are comparable.

II. ELECTRON-HOLE EXCHANGE INTERACTION IN QUANTUM DISKS

The two-particle excitonic wave function can be written as a linear combination of products $\Psi_{s,j}(r_e, r_h)|s, j\rangle$, where $|s, j\rangle$ is a product of the electron and hole Bloch functions, $s$ and $j$ are the electron and hole spin indices, $\Psi(r_e, r_h)$ is the envelope function, and $r_{e,h}$ is the electron (hole) 3D radius-vector. In the following, for the sake of simplicity, we concentrate on heavy-hole excitons with $j = \pm 3/2$ and the long-range mechanism of electron-hole exchange interaction which allows to discuss only the bright excitonic states $|s, j\rangle$ with $s + j = \pm 1$ or their linear combinations $|\alpha\rangle$ with the dipole moment $\alpha = x, y$. The short-range mechanism may be taken into consideration similarly to [1] if one includes an admixture of light-hole states into the heavy-hole exciton wave function.

We consider a quantum disk formed by a 2D harmonic potential $V(r_e, r_h) = A_e \rho_e^2 + A_h \rho_h^2$ in a quantum well grown along the $z$-axis. Here the 2D vector $\rho_{e,h}$ determines the in-plane position of an electron or a hole, and $A_{e,h}$ are positive constants. Assuming that the confinement along the growth direction is stronger than both the quantum-disk and Coulomb potentials the envelope for the electron-hole pair wavefunction can be written as

$$\Psi(r_e, r_h) = \psi(r_e, \rho_e) \varphi_e(z_e) \varphi_h(z_h),$$

where $\varphi_e, h(z_e, h)$ are the respective $z$-envelopes of electron and hole, and $\psi(r_e, \rho_e)$ is an in-plane wavefunction of exciton calculated with allowance for both Coulomb interaction and quantum disk potential. The form of the function $\psi(r_e, \rho_e)$ depends on the relationship between the potential radius and 2D-exciton Bohr radius, $a_B$. In the weak confinement regime, i.e. in a quantum disk with the diameter exceeding $a_B$, the two-particle envelope probe function $\psi(r_e, \rho_e)$ is a product $F(R)f(\rho)$ of two functions describing, respectively, the in-plane motion of the exciton center of mass $R = (X, Y)$ and the relative electron-hole motion, $\rho = r_e - r_h$. On the other hand, in small quantum disks with strong confinement where single-particle lateral confinement dominates over the Coulomb interaction, the pair envelope can also be presented as a product of two functions, $\psi_e(r_e)\psi_h(r_h)$, but here they describe the independent in-plane localization of an electron and a hole.

For the exciton envelope functions presented in the form (1), the matrix element $H_n^e_{n'}^h$ of long-range exchange interaction taken between the exciton states $n'$ and $n$ is written as follows

$$\frac{1}{2\pi\epsilon\infty} \left(\frac{e\hbar|p_0|}{m_0 E_g}\right)^2 \int dR \frac{K_{\alpha}^e K_{\alpha'}^h}{K} \tilde{\psi}_{n'}^e(K) \tilde{\psi}_{n}^h(K),$$

where the exciton-state index $n$ includes the dipole moment $\alpha$, $\epsilon_{\infty}$ is the high-frequency dielectric constant, $m_0$ is the free electron mass, $E_g$ is the band gap, $p_0$ is the interband matrix element of the momentum operator, and we introduced the 2D Fourier-transform

$$\tilde{\psi}(K) = \int dRe^{-iKR}\psi(R, R).$$
of the function $\psi(\rho_e, \rho_h)$ at the coinciding coordinates, $\rho_e = \rho_h \equiv R$.

III. P-ORBITAL EXCITONS IN AXIALLY-SYMMETRIC DISKS

We start with an axially-symmetric quantum disk and calculate the fine structure of the $P$-orbital exciton level and then proceed to a slightly anisotropic disk. The straightforward calculation shows that the long-range exchange Hamiltonian has the following non-zero matrix elements

$$\langle P_x | H^{\text{long}} | P_x \rangle = \langle P_y | H^{\text{long}} | P_y \rangle = \lambda \rightleftharpoons \langle P_x | H^{\text{long}} | P_y \rangle = \eta \rightleftharpoons \langle P_y | H^{\text{long}} | P_x \rangle = \mu \rightleftharpoons$$

where $\lambda = 3\eta = 3\mu$. According to Eq. (3) and in agreement with the angular-momentum considerations, the $P$-shell of the bright exciton in an axisymmetric quantum disk is split into three sublevels, see Fig. 1. The outermost sublevels labelled $0^\ell$, $0^L$ are nondegenerate and characterized by a zero total angular-momentum $z$-component. The central doubly-degenerate sublevel corresponds to the angular-momentum component $\pm 2$. The intersublevel energy spacing, $\Delta$, equals to $2\eta = 2\mu$.

The splitting $\Delta$ depends on the character of exciton confinement in a disk. Let us assume $A_{e,h} = \hbar^2 / 2m_{e,h}a^4$, where $a$ is the disk effective radius, $m_{e,h}$ are the electron and hole effective masses. It follows then that in the strong confinement regime, $a \ll a_B$, one has

$$\Delta = \frac{3\sqrt{2\pi}}{16a^2\hbar\omega_0} \left( \frac{\hbar|p_0|}{m_aE_g} \right)^2. \quad (4)$$

In the opposite limiting case $a \gg a_B$ where the exciton is quantized as a whole we obtain

$$\Delta = \frac{3\sqrt{\pi}}{2a_{\text{exc}}\hbar\omega_0} \left( \frac{\hbar|p_0|}{m_aE_g} \right)^2 \quad (5)$$

with $a_{\text{exc}} = a(\mu/M)^{1/4}$ being the radius of exciton in-plane confinement, $M = m_e + m_h$ and $\mu = m_em_h/M$.

IV. P-SHELL EXCITONS IN ANISOTROPIC QUANTUM DISKS

We introduce a slightly elliptical lateral potential replacing the disk radius $a$ by the effective radii $a_x = a_d$, $a_y = a + d$ along the $x$ and $y$ axes, respectively, and assuming $d \ll a$. At zero $d$ the exciton $P$-states are partially split by the exchange interaction. The anisotropy of a quantum disk results in a full removal of the degeneracy and formation of four sublevels. It is convenient to introduce, as a parameter describing the anisotropy, a half of the $P_x-P_y$ splitting, $E_{\text{anis}}$, calculated neglecting the exchange interaction. Its value depends on the model of exciton quantization. If an electron and a hole are quantized independently ($a \ll a_B$) then, for the $|P_e, S_h \rangle$ excited state, we have

$$E_{\text{anis}} = \frac{d}{a} \frac{2\hbar}{m_a a^2} \quad (6)$$

and, for the $|S_e, P_h \rangle$ state, $E_{\text{anis}}$ differs from Eq. (6) by the replacement $m_e \rightarrow m_h$. Here $|S_e, P_h \rangle$ means an exciton formed by a $S_e$-shell electron and a $P_h$-shell hole. If exciton is quantized as a whole then in Eq. (6) one should replace $m_e$ by the exciton translational mass $M = m_e + m_h$ and $a$ by $a_{\text{exc}} = a(\mu/M)^{1/4}$.

Figure 1 shows splitting of the $P$-shell bright-exciton level as a function the ratio $\xi = E_{\text{anis}}/\Delta$. For small values of $\xi$ the splitting of the doublet $\pm 2\Delta$ is proportional to $\xi^2$. In the limit of strong anisotropy, $E_{\text{anis}} \gg \Delta$, the $P$-shell exciton states form two doublets, $|P_e, \alpha \rangle$ and $|P_h, \alpha \rangle$ ($\alpha = x, y$), separated by $2E_{\text{anis}}$ and each split by $\Delta$.

V. RESONANT P-S EXCITONIC POLARONS

Finally, we briefly discuss the $P-S$ excitonic polaron which is formed as a result of the resonant coupling of the $P$- and $S$-like levels by a longitudinal optical (LO) phonon. We use an approach developed in Ref. to calculate the LO-assisted resonant coupling between $S$- and $P$-shell electron states confined in a quantum dot. For the Fröhlich interaction, the constant of exciton-phonon $P-S$ coupling is given by

$$\gamma = \sqrt{\frac{2\pi e^2 \hbar \Omega}{V x^2} \sum_q \left| T(q) \right|^2}. \quad (7)$$
Here $\Omega$ is the LO-phonon frequency, $V$ is the 3D normalization volume, $\kappa^{-1} = \kappa_{\infty}^{-1} - \kappa_{0}^{-1}$, and

$$ I(q) = \int dr_e dr_h \psi_p(r_e, r_h) \psi_s(r_e, r_h) (e^{iqr_e} - e^{iqr_h}). $$

Depolarization of the linearly-polarized $P$-$S$ excitonic polaron excited via the $P$-shell is determined by the ratio of $\gamma$ and the exchange splitting $\Delta$. If $\Delta \gg |\gamma|$ the initial linear polarization is being rapidly lost and the exciton photoluminescence is practically depolarized. On the contrary, if $\Delta \ll |\gamma|$ then the polarization relaxes on a much longer time scale and can be well preserved.

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