Comment to article "A light–hole exciton in a quantum dot" by Y.H.Huo et al, Nature Physics 10, 46 (2014).

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Abstract

The exciton ground state in strained quantum dots similar to those fabricated in article specified in the title is shortly discussed within a relevant model Hamiltonian. Some characteristics of the light–hole exciton ground state reached in a dot under the tensile biaxial strain appear to be sensitive to the strain anisotropy breaking a purity of this state. It refers in particular to a degree of the in–plane polarization of the emission and the fine structure of the ground state.

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In recent paper "A light–hole exciton in a quantum dot" by Y.H. Huo et al.[1] is reported about a creation of the light–hole exciton ground state by applying biaxial tensile strain to an initially unstrained quantum dot. This conclusion is based, in particular, on the observation of the \( z \)-polarized line in emission spectra of strained dots - an obvious sign that the ground state is "mainly light–hole" exciton. A degree of the in–plane polarization anisotropy of the emission contains a definite information about a "purity" of this state. The last item is discussed by authors in Supplementary Section II.6 on a basis of theoretical results from Ref.[2]. While we acknowledge the calculation way of the linear polarization degree used in Ref.[2], an application of the particular results received there (formulae (1) and (7)) to the scenario taking place in Ref.[1] appears to be questionable. In this comment we simply would like to give some additions and specifications.

It is a question about the states of the heavy–hole (HH) and light–hole (LH) exciton with total angular momentum projection \( |J_z| = 1 \). For the in–plane symmetrical dots, this (two–fold degenerate) bright state is circularly polarized and therefore contributes to unpolarized emission. Any perturbation providing a coupling between the states with \( J_z \) of different signs leads generally to a formation of elliptically polarized states. Considering the HH–LH coupling of this type, the low–energy exciton states have a form (for simplicity, we take the same envelope function for the heavy and light hole)

\[
\Psi_{L}^{(\pm)} \approx \left[ w_{hh} \left( \pm \frac{1}{2} ; \pm \frac{3}{2} \right) + w_{lh} \left( \pm \frac{1}{2} ; \mp \frac{1}{2} \right) \right].
\] (1)

The (square) amplitude \( |w_{hh(lh)}| \)^2 = \( 0.5 \left[ 1 \pm \Delta_{th}^{(0)} \left( \sqrt{(\Delta_{th}^{(0)})^2 + 4\rho^2} \right)^{-1} \right] \) (with the mixing amplitude \( \rho \) and the LH–HH splitting \( \Delta_{th}^{(0)} > 0 \)) determines the exciton character \( P_{hh(lh)} = |w_{hh(lh)}|^2 \), that is the probability for the exciton to be HH(LH) exciton. Evidently, the above ground state is mainly of the heavy–hole type in a weak coupling limit, where \( \rho \ll \Delta_{th}^{(0)} \), while both the HH and LH exciton characters are of the same order at \( \rho \gg \Delta_{th}^{(0)} \). Having in mind a contribution of the bright exciton to a recombination, a degree of the linear polarization of the emission is expressed generally as

\[
C = \frac{2 \sqrt{3P_{hh}P_{lh}}}{3P_{hh} + P_{lh}}.
\] (2)

Here it is considered that the recombination probability is three times larger for the HH than LH exciton, see e.g.[3]. From Eq.(2) follows that the linear polarization is equal to zero in absence of the HH–LH mixing, as expected, whereas it becomes close to unity in a strong
coupling limit where $\rho \gg \Delta^{(0)}_{lh}$. The above scenario is familiar for conventional quantum dots showing the anisotropic effects of an intrinsic nature and keeping the exciton ground state of the heavy–hole type, see e.g. Ref.[4] and Ref.[2] as well.

Simulating the experimental conditions realized in Ref.[1], the excitonic states with $|J_z| = 1$ in a strained dot can be described by a model $(4 \times 4)$ Hamiltonian having a block–diagonal matrix form. Both matrixes, one in the $\{|-\frac{1}{2}; +\frac{3}{2}\}, \{|-\frac{1}{2}; -\frac{1}{2}\}\}$ basis and another in the $\{|\frac{1}{2}; -\frac{3}{2}\}, \{|\frac{1}{2}; +\frac{1}{2}\}\}$ basis, are identical and given by

$$
\begin{pmatrix}
0 & \gamma \Delta_d \\
\gamma^* \Delta_d & \Delta^{(0)}_{lh} - \Delta_d
\end{pmatrix}.
$$

(3)

Here the energy position of the HH exciton is set to zero and the strain–induced splitting $\Delta_d$, positive (negative) for the tension (compression), is determined by the relative deformation $(\varepsilon_\parallel - \varepsilon_\perp)$. The adjustable parameter $\gamma$ measures the HH–LH coupling due to the strain anisotropy, which is expected to be rather weak. The low–energy states of the bright exciton have still the above structure Eq.(1), now with the (absolute value) amplitudes

$$
|w_{lh(hh)}| = \frac{1}{\sqrt{2}} \left[ 1 \pm \frac{1 - \delta_d}{\sqrt{(1 - \delta_d)^2 + 4|\gamma|^2\delta_d^2}} \right]^\frac{1}{2},
$$

(4)

where $\delta_d = \Delta_d/\Delta^{(0)}_{lh}$. In Fig.1a we plot the exciton characters $P_{hh}$ and $P_{lh}$ calculated from Eq.(1) in a dependence on dimensionless variable $\delta_d$ at the anisotropic parameter $|\gamma| = 0.1$. It is seen that while the HH exciton fully dominates in the ground state for a compressive strain, the exciton character can be shifted to dominantly LH at a tensile strain. A step–like switching happens at $\delta_d = 1$, where the strain effect compensates the confinement–induced splitting $\Delta^{(0)}_{lh}$, and at an increase in tension only twice the LH exciton character almost completely prevails. This result is close to that reported for the valence band ground state in a specific quantum dot calculated in an empirical pseudopotential based approach in Ref.[1].

Similarly to the bright exciton, the (anisotropic) strain couples the dark states of the HH exciton with $J_z = |2|$ to the states of the LH exciton with $J_z = 0$. Now, however, the exchange interaction must be considered explicitly. The corresponding Hamiltonian represents four–by–four matrix in the $\{|\frac{1}{2}; +\frac{3}{2}\}, (2^{-0.5})[-i| + \frac{1}{2}; -\frac{1}{2}) + | -\frac{1}{2}; +\frac{1}{2}\}, (2^{-0.5})[+i| + \frac{1}{2}; -\frac{1}{2}) + | -\frac{1}{2}; +\frac{1}{2}\} basis. Solving the system makes possible to receive an information on the fine structure splitting of the exciton ground state. For a dot with the
FIG. 1: Evolution of the exciton characters of the ground state with in–plane biaxial strain at $|\gamma| = 0.1$ (a). Energy splitting between the optically allowed excitonic states at $|\gamma| = 0.1$ (b).

"mainly LH” exciton ground state ($\delta_d > 1$), the energy splitting between the optically active excitons is approximated by

$$E_{B_z} - E_{B_{x,y}} \simeq \Delta_{st} \left[ 1 - \left( \frac{2\gamma \delta_d}{\delta_d - 1} \right)^2 \right].$$  \hspace{1cm} (5)

Here the energy $E_{B_z}$ and $E_{B_{x,y}}$ refers to the $z$–polarized exciton and the exciton doublet polarized in the growth plane of a dot, respectively, and the exchange energy is $\Delta_{st}$. To illustrate, in Fig. 1b is shown the (relative) energy splitting Eq.(5) as a function of dimensionless parameter $\delta_d$ at $|\gamma| = 0.1$ for a dot with the LH exciton character $P_{lh} \gtrsim 0.88$ (corresponding to $\delta_d \gtrsim 1.3$). It is seen that the energy distance between the high–energy exciton $B_z$ and the low–energy doublet $B_{x,y}$ grows with an increase of a strain and limits to $\Delta_{st}$ at $\delta_d \gg 1$, as expected for the "pure” LH exciton \cite{3}. These results are similar to experimental findings and theoretical calculations from Ref. \cite{1}.

For the in–plane polarized doublet in a strained dot, in Fig. 2 we plot a degree of the linear polarization given by Eq.(2) as a function of the LH exciton character $P_{lh}$. Since a contribution of the HH exciton to recombination is (three times) larger than a contribution of the LH exciton, the calculated curve is not symmetric with respect to $P_{lh} \leftrightarrow 1 - P_{lh} = P_{hh}$ replacement and the polarization rate shows a sharp falling at $P_{lh} \rightarrow 1$. A speed with which the ground state becomes the almost pure LH exciton depends on a degree of the HH–LH coupling. Indeed, according to Eq.(1), for the tensile strain the LH exciton character limits to $P_{lh} \simeq 1 - \left( \varrho / \Delta_{lh}^{(-)} \right)^2$ at $\delta_d > 1$, where $\varrho = |\gamma| E_d$ and $\Delta_{lh}^{(-)} = E_d - \Delta_{lh}^{(0)} > 0$. Obviously,
FIG. 2: Degree of the linear polarization as a function of the LH exciton character.

the HH exciton character in this case is $P_{hh} \approx \left( \varrho / \Delta_{lh}^{(-)} \right)^2$.

In this regard, an application of results from Ref. [2] to (at least) the tensile strained dots is, evidently, not correct. Remember that for the ground state in a quantum dot, the exciton characters are presented in Ref. [2] by $P_{lh} = \beta^2$ and $P_{hh} = 1 - \beta^2$ with $\beta = \varrho_s / \sqrt{\Delta_{HL}^2 + \varrho_s^2}$ (formula (1), where $\Delta_{HL}$ and $\varrho_s$ denotes the valence band splitting and the coupling amplitude, respectively). These results, written down in a weak mixing limit, are able to describe in general the effects of a weak intrinsic anisotropy and/or an external compressive strain. For tensile strained dots, however, formal equating of the "mixing parameter" $\beta$ to unity (to obtain for the polarization rate the desired result $C = 0$) means a very strong coupling limit and is not adequate to the real physical scenario described above.

On the contrary, to minimize the optical anisotropy effect in a strained quantum dot with a dominant light–hole exciton ground state, the applied (tensile) strain is required to be highly isotropic in the growth plane of a dot. Indeed, even if the exciton ground state has about 95% LH character a degree of the linear polarization is calculated from Eq. (2) to be $C \approx 0.6$. Note that isotropic strain is also desirable to avoid the strain–induced source of the spin relaxation within the exciton ground state limiting the generation of single photons from a dot [3].

Strictly speaking, even in a fully isotropic case the HH and LH exciton states, those with the same momentum $J_z = 1(-1)$, are coupled by the short–range exchange interaction
(evidently, the heavy– and light–hole valence band states experience any such mixing). This kind of a coupling provides a smooth switching from the HH to LH exciton in the (tensile) strained dot as before, but preserves the circular polarization of the exciton ground state. It is possible that such a scenario is relevant for the strained quantum dot reported in Ref. [1], in which a negligible degree of the linear polarization $C = 0.01$ was measured (inset 3 in Fig. S17). In any case this extremely small polarization rate points to a very weak LH–HH coupling and the almost pure LH exciton ground state.

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