In-plane radiative recombination channel of a dark exciton in self-assembled quantum dots

T. Smoleński,1 T. Kazimierczuk,1 M. Goryca,1 T. Jakubczyk,1 L. Klopotowski,2 L. Cywiński,2 P. Wojnar,2 A. Golnik,1 and P. Kossacki1

1Institute of Experimental Physics, Faculty of Physics, University of Warsaw, ul. Hożą 69, 00-681 Warsaw, Poland
2Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/64, 02-688 Warsaw, Poland

We demonstrate evidence for a radiative recombination channel of dark excitons in self-assembled quantum dots. This channel is due to a light hole admixture in the excitonic ground state. Its presence was experimentally confirmed by a direct observation of the dark exciton photoluminescence from a cleaved edge of the sample. The polarization resolved measurements revealed that a photon created from the dark exciton recombination is emitted only in the direction perpendicular to the growth axis. Strong correlation between the dark exciton lifetime and the in-plane hole g-factor enabled us to show that the radiative recombination is a dominant decay channel of the dark excitons in CdTe/ZnTe quantum dots.

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Self-assembled semiconductor quantum dots (QDs) are recognized as a medium for storage and manipulation of quantum information. Important applications, involving emission of single photons or entangled photon pairs, rely heavily on properties of confined excitonic complexes. In particular, the central point of many schemes is a neutral exciton consisting of a single electron and a single hole. However, the properties of such a complex depend on the relative orientation of the angular momenta of confined carriers. An electron and a hole with antiparallel angular momenta form an optically active bright exciton \( (X_b) \) with total angular momentum projection on the QD growth axis \( J_z = \pm 1 \), while parallel orientation of angular momenta leads to the formation of a dark exciton \( (X_d) \) with \( J_z = \pm 2 \). Due to the lack of dipole allowed recombination channel, the lifetime of the latter may extend above microseconds. The presence of the dark excitons is often neglected, however due to their persistent nature, dark excitons can have detrimental effect on the properties of QD devices. On the other hand, recent findings show that the dark exciton can be also used as a qubit, which turns its long lifetime into an advantage. The important role of the dark excitons and their lifetime is therefore of principal interest for quantum information processing.

Studies of dark excitons are hindered by their absence in the photoluminescence (PL) spectrum. Dark excitons become visible after introducing a mixing between bright and dark configurations, either by an in-plane magnetic field or by the exchange interaction with magnetic dopant. The lifetime of the dark exciton is widely believed to be determined by a spin-flip process turning a dark exciton into a bright one. The main argument for such a mechanism is a biexponential decay of the bright exciton, which was observed in both III-V and II-VI QDs. However, the spin-flip cannot be considered the sole decay mechanism of the dark exciton. It is especially evident in the analysis of the low temperature behavior of this process. In particular, turning the dark exciton into a bright one requires absorption of energy from the phonon bath equal to (isotropic) electron-hole exchange constant, and therefore predicted lifetime of the dark exciton can, in principle, be infinitely prolonged by lowering the temperature. A presence of another decay mechanism was already suggested in Ref. [15], where the authors concluded that the lifetime of the dark exciton is limited by an unknown “non-radiative decay channel”, rather than by the spin-flip process.

In this Communication, we demonstrate that the dark exciton can recombine radiatively, emitting linearly polarized light in a direction perpendicular to the growth axis. This fact, together with our observation of a strong quantitative correlation between the \( X_d \) radiative lifetime and the in-plane hole g-factor, proves that this recombination channel is due to an admixture of the light hole component in the hole ground state. We also provide an evidence that the radiative recombination can be dominant decay channel of the dark exciton in CdTe/ZnTe QDs. However, our findings should apply also to other QDs exhibiting heavy-light hole mixing.

Our experiments were performed on samples containing self-assembled CdTe/ZnTe QDs grown by the amorphous tellurium desorption method. The measurements were carried out in a micro-photoluminescence setup described in Refs. [17] and [18]. Sample was placed inside either a continuous-flow or a magneto-optical bath cryostat at temperature 1.5-10K. During the time-resolved measurements the QDs were excited nonresonantly by a frequency doubled pulses from a Ti:sapphire laser. The laser repetition rate was effectively reduced by an avalanche photodiode (APD) with sub-nanosecond temporal reso-
lution.

In this study we investigate the presence of a radiative emission of the dark exciton without external magnetic field. Such an emission is not observed in PL measurements performed in a typical experimental setup with optical axis parallel to the QD growth axis. However, the dark exciton PL can be revealed by using an in-plane setup geometry (i.e., the optical axis perpendicular to the growth axis). Figure 1(a) presents PL spectra of the neutral exciton measured in such a geometry under nonresonant, continuous wave excitation. Under strong excitation power (short dot repopulation time) the bright exciton PL dominates the spectrum, as $X_d$ lifetime is usually much shorter compared to the dark exciton one. However, in the low excitation power regime, the dot repopulation time is long enough to allow undisturbed recombinations of the $X_d$, the dark exciton transition line is clearly visible (its identification is based on the spectral position, which matches relative energy of the dark exciton determined in the magneto-photoluminescence experiments). This result indicates the presence of radiative recombination channel of the dark exciton confined inside a QD. Until now, such an effect was demonstrated only in [111]-grown pyramidal QDs. We emphasize that dark exciton PL could be discovered only using the in-plane geometry. It is related to the fact that $X_d$ emission line is almost fully linearly polarized along the QD growth axis (Fig. 1(b)).

In order to understand the origin of the observed radiative $X_d$ decay mechanism, one has to take into account the valence band mixing, which is usually present in both III-V and II-VI QDs. We consider the heavy ($|± 3/2⟩$) and light ($|± 1/2⟩$) hole states, which are mixed due to the presence of a QD shape or strain anisotropy. In the leading order, $|± 3/2⟩$ states are only mixed with $|± 1/2⟩$ (which yields the lowest-energy hole states in a QD to be given by $|φ_{±h}⟩ = |± 3/2⟩ + e± |± 1/2⟩$ (where $e$ represents the strength of the valence band mixing, and $θ$ its direction). The ground states of the dark exciton are now of the forms $|φ_{h+}⟩ = |e⟩|φ_{h+}⟩$, $|φ_{h−}⟩ = |e⟩|φ_{h−}⟩$, where $|e⟩$, $|φ_{e}⟩$, $|φ_{h}⟩$, $|φ_{h±}⟩$ correspond to the same data points. However, in the low excitation power regime the bright exciton PL dominates the spectrum, as $X_d$ lifetime is usually much shorter compared to the dark exciton lifetime. The 90° and 270° directions are along the QD growth axis. Symbols for angles $ϕ$ and $φ + 180°$ correspond to the same data points.
The lifetime of the dark exciton for this QD yields the fitted curve described by Eq. (3). The zero magnetic field of the in-plane magnetic field. The solid line represents the without polarization resolution. Solid lines represent the fits of the dark exciton for different magnetic fields, measured without polarization resolution. Solid lines represent the fits of the dark exciton for different magnetic fields, measured without polarization resolution. The zero magnetic field is related to X_d with larger admixture of the bright excitons, while the slow decay time originates from PL of X_d with lower bright exciton admixture. This explanation stays in an agreement with more abrupt reduction of the fast decay time with increasing magnetic field. In the following analysis we focus on the dark exciton with stronger field-induced mixing. Figure 2(c) shows the inverse lifetime of this dark exciton as a function of the magnetic field.

We use the data in Fig. 2(c) to extrapolate the X_d lifetime to \( B = 0 \) T and thus obtain the zero-field dark exciton lifetime. To do so, we use a simple model of the neutral exciton in the in-plane magnetic field given by the following Hamiltonian:

\[
\mathcal{H} = -2\delta_0 \hat{S}^e_z \hat{S}^h_z + \frac{\delta_1}{2} (\hat{S}^e_z \hat{S}^h_z + \hat{S}^e_z \hat{S}^h_z) \\
+ g_e \mu_B \mathbf{B} \cdot \hat{S}_e + g_h \mu_B \mathbf{B} \cdot \hat{S}_h,
\]

(2)

where \( \hat{S}^e_z \) and \( \hat{S}^h_z \) are the electron spin operators, \( \hat{S}_e \) and \( \hat{S}_h \) are the 1/2 spin operators in the two-dimensional subspace of heavy hole states. The first two terms represent the isotropic and anisotropic parts of the electron-hole exchange interaction. The remaining terms represent the Zeeman energies of the electron and the hole, with their in-plane g-factors equal to \( g_e \) and \( g_h \), respectively. The hole Zeeman term includes the \( \hat{r}_{2\theta} \) tensor, which is the rotation matrix through \( 2\theta \), where \( \theta \) is an angle between direction of the axis of exchange interaction anisotropy and the direction related to the valence band mixing. All the parameters in the Hamiltonian were directly extracted from the polarization resolved PL measurements in different magnetic fields for each studied QD. In particular, exchange energies were identified as the splitting between the dark and bright states (\( \delta_0 \)), and the splitting between two bright configurations (\( \delta_1 \)), obtained at zero magnetic field. Here we neglect \( \delta_2 \) splitting of the dark excitons, which is \( \sim \mu_e^2 \). The carrier g-factors were extracted from the energy positions of four-fold split trion emission lines in magnetic field. Futhermore, the angle \( \theta \) was obtained from the polarization resolved X_d and trion PL measurements. By diagonalization of the Hamiltonian for a given magnetic field, one obtains four eigenstates. Two lower energy states \( |\psi_i(B)\rangle \) with \( i = 1, 2 \) correspond to mostly dark states. Their overlap with zero-field bright states \( |\pm 1\rangle \) is given by \( f_i(B) = |\langle 1|\psi_i(B)\rangle|^2 + |\langle -1|\psi_i(B)\rangle|^2 \). This overlap is proportional to the oscillator strength of \( |\psi_i(B)\rangle \) radiative recombination induced by an in-plane magnetic field. If there were no other decay mechanisms, the inverse lifetime of dark excitons would be proportional to \( f_i(B) \) implying that at \( B = 0 \) T the X_d lifetime would be.

![Figure 2: (color online) (a) PL spectra of a single QD for different values of the in-plane magnetic field. (b) PL decay curves of the dark exciton for different magnetic fields, measured without polarization resolution. Solid lines represent the fits of the dark excitons with almost equal emission energies, but different efficiencies of mixing with bright excitons. (c) Inverse lifetime of the shorter lived dark exciton as a function of the in-plane magnetic field. The solid line represents the fitted curve described by Eq. (3). The zero magnetic field lifetime of the dark exciton for this QD yields \( \tau_0 = 32 \pm 3 \) ns.](image-url)
where $\tau_0$ is its zero-field lifetime, and $\gamma$ is a constant related to the radiative lifetime of the bright exciton at $B = 0T$. Our calculations of Eq. 3 perfectly reproduce the dependence of $X_d$ lifetime on the magnetic field, as it is shown in Fig. 2(c). In order to draw robust conclusions, similar measurements were performed on several randomly selected QDs. The obtained zero-field dark exciton lifetime $\tau_0$ varied from about 30ns to over 20µs. Figure 3 presents the square root of $1/\tau_0$ as a function of $g_h$ for each studied QD. Within the experimental uncertainties, we observe a direct correlation between the dark exciton zero-field decay rate and the in-plane hole $g$-factor (i.e., the magnitude of the valence band mixing). This clearly implies that the radiative recombination is the only important decay channel of the dark exciton at $B = 0T$ in the case of CdTe/ZnTe QDs. In particular, such a result shows that the typically invoked spin-flip process turning the dark exciton into the bright exciton in the QDs. In particular, the coupling between the dark exciton and the in-plane radiation demonstrated in our work may be used for a direct optical control of the dark exciton qubit with lifetime much longer than a qubit based on the bright exciton in a QD. Therefore neither fast nor slow component of the bright exciton decay can be attributed to the feeding by the spin-flip effect.

We identify the faster $X_b$ decay time with the radiative lifetime of the bright exciton. The slower decay, which corresponds to about 5% of the total line intensity, is tentatively attributed to the recapturing of the carriers by the QD. Qualitative discrepancy between the experimental $X_b$ decay and the one calculated in the spin-flip model (dashed line in Fig. 4) clearly shows that the spin-flip process is ineffective in the case of CdTe/ZnTe QDs.

All presented results are a clear evidence for the radiative recombination channel of the dark exciton. This radiative recombination can dominate over other decay mechanisms of the dark exciton. We believe that our findings will stimulate the studies on the role of the dark exciton in the QDs. In particular, the coupling between the dark exciton and the in-plane radiation demonstrated in our work may be used for a direct optical control of the dark exciton qubit with lifetime much longer than a qubit based on the bright exciton in a QD.

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∗ Electronic address: Tomasz.Smokowski@fuw.edu.pl

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