Dark Exciton Preparation in a Quantum Dot by a Longitudinal Light Field Tuned to Higher Exciton States

M. Holtkemper,1 G. F. Quinteiro,2 D. E. Reiter,1 and T. Kuhn1

1Institut für Festkörpertheorie, Universität Münster, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany
2IMIT and Departamento de Física FacENA, Universidad Nacional del Nordeste, Corrientes, Argentina

(Dated: March 9, 2020)

Several important proposals to use semiconductor quantum dots in quantum information technology rely on the control of the dark exciton ground states, such as dark exciton based qubits with a µs life time. In this paper, we present an efficient way to occupy the dark exciton ground state by a single short laser pulse. The scheme is based on an optical excitation with a longitudinal field component featured by, e.g., radially polarized beams or certain Laguerre-Gauss or Bessel beams. Utilizing this component, we show within a configuration interaction approach that high-energy exciton states composed of light-hole excitons and higher dark heavy-hole excitons can be addressed. When the higher exciton relaxes, a dark exciton in its ground state is created.

With their discrete energy states, semiconductor quantum dots (QDs) are designated to play an important role in solid-state quantum information technology [1], for example as sources for single or entangled photons [2–6] or as realization of qubits [7–9]. For qubits, it is especially promising to use the dark excitons as information storage, since they exhibit extraordinary long life and coherence times [10–14]. Furthermore, dark excitons are important intermediate states in some state preparation protocols, e.g. for biexciton [15] or impurity spin preparation [16].

The long lifetime of dark excitons is mainly due to the vanishing or very weak dipole coupling to the light field which, in turn, makes their optical generation challenging. Several workarounds have been developed to overcome this difficulty. By using a non-resonant cw excitation and a subsequent random optical charging of the QD excited biexcitons with various spin configurations are formed. After emission of a photon and subsequent phonon-induced hole relaxation certain spin configurations lead to the formation of a dark exciton [12]. Alternatively, a weak mixing between bright and dark exciton can be induced via an external magnetic field [17, 18] or is the result of a broken $C_{2z}$-symmetry of the QD [19]. This makes a direct optical excitation of the dark exciton possible [20], however with an oscillator strength which is some orders of magnitude smaller than the absorption into the bright exciton. A (desired) increase of the optical excitation by a stronger coupling between dark and bright excitons is inevitably associated with a further (undesired) distortion of the spin character of the dark exciton resulting, e.g., in a decrease of the lifetime.

In this paper, we propose a scheme to circumvent this connection of absorption strength and spin distortion by moving the necessary spin coupling to higher excited states. There the spin coupling can be strong essentially without affecting the pure spin character of the ground states. In detail, our scheme utilizes suitable complex light fields with a strong longitudinal field component. Such field components can excite otherwise optically forbidden light-hole (LH) excitons. Valence band mixing couples those LH excitons preferentially to higher heavy-hole (HH) excitons involving a dark spin configuration, building strongly mixed higher exciton states. Due to their dark HH exciton contribution, these states will then relax by phonon emission into the dark HH exciton ground state. Our proposal complements studies on the great benefits of the use of higher QD exciton states [21–25] or complex light fields beyond Gaussian beams [26, 27] for QD based quantum information technology.

A key role for our excitation process is a component of the light field polarized in the growth direction of the QD sample. At normal incidence along the growth direction this corresponds to a component in propagation direction. While such a longitudinal field component is absent in plane-wave-like fields and is very small in beams which are well described by the paraxial approximation, it can become dominant close to the beam axis in certain strongly non-paraxial or in tightly focused beams, such as certain Laguerre-Gaussian, Bessel or radially polarized beams [28–31]. This dominance can be strong enough to safely neglect all but the longitudinal field components in the region close to the beam center. Such longitudinal components have been successfully used to probe single molecules [29] or to trap metallic Rayleigh particles [30].

To provide a clear and well-defined theoretical description, here we consider Bessel beams, which constitute an exact and complete set of solutions to Maxwell’s equations. They are propagation-invariant (non-diffracting) beams. Although strictly speaking Bessel beams have an infinite lateral extension because of their weak decay in radial direction, approximations of such beams have been realized in various experiments [32–35]. Furthermore, it can be shown that the tight focusing of cylindrically symmetric vector beams typically also results in Bessel function-like field distributions in the transverse and longitudinal components [28].

Assuming a beam propagating along the $z$-direction
reads the complex conjugate, the electric field of a Bessel beam reads

\[ E_x(r) = i\sigma \tilde{E}_y(r) = \frac{E_0}{\sqrt{2}} J_\ell(q_r r)e^{i\ell\varphi} \]

\[ E_z(r) = \frac{E_0 q_r}{\sqrt{2} q_z} J_{\ell+\sigma}(q_r r)e^{i(\ell+\sigma)\varphi} \]

with the Bessel function of first kind and \( \ell \)-th order \( J_\ell \), the electric field amplitude \( E_0 \), the frequency \( \omega \) and the wave vector components in propagation and radial directions, \( q_z \) and \( q_r \), respectively. The latter quantities are related by \( q_z^2 + q_r^2 = (n\omega/c)^2 \), \( n \) being the index of refraction of the medium. We consider a CdSe QD with \( n = 2.8 \) [36] and realistic highly non-paraxial (tightly focused) beams with \( q_r/q_z = 1 \) [37, 38]. A Bessel beam is characterized by the indices \( \sigma = \pm 1 \) and \( \ell = 0, \pm 1, \ldots \). Since for all Bessel functions \( J_\ell \) with \( n > 0 \) the intensity of the beam profile has a minimum around the beam axis (thus at the position of the QD), there are only two types of beams with a high intensity at the QD: First, beams with \( \ell = 0 \) and \( \sigma = \pm 1 \), which results in a predominantly transverse field component. Selection rules and spectra are similar to those of typical Gaussian (or plane wave like) beams. Second, beams with \( \ell = \pm 1 \) and \( \sigma = \mp 1 \), which results in a predominantly longitudinal field component. We note, that one can also superpose the two Bessel beam modes \( \ell = \pm 1 \) and \( \sigma = \mp 1 \) to a radially or azimuthally polarized beam, where the radially polarized beam also features the pronounced longitudinal field component [28–30] while the azimuthally polarized beam remains purely transverse [28]. In the following we will consider the absorption of the two cases of a transverse and a longitudinal field component.

We are interested in the optical excitation of a self-assembled QD, which has a disk-like shape of a few nm in size. The confinement of electron and hole states is modeled by a three-dimensional, anisotropic harmonic oscillator potential with confinement lengths \( L_x, L_y \) and \( L_z \). The electronic states are described within an envelope function formalism. Therein, the wave function is separated into a Bloch part and an envelope part. For the Bloch states, we take into account the HH valence band with a total angular momentum projection (or pseudo spin) of \( J_h = \pm 3/2 \), the LH valence band with \( J_h = \pm 1/2 \) and the conduction band for electrons with \( J_e = \pm 1/2 \). According to the confinement, the envelope states are given by harmonic oscillator basis functions with their lowest-lying states just increasing in the in-plane quantum numbers. As usual, we group the states into \( s, p, d, \ldots \)-shells. We will use small letters for HHs and capital letters for LHs. Excitons as well as their exciting transitions are indicated by the valence band to conduction band state involved (e.g. \( d \rightarrow s \) is for the excitation of a HH from the \( d \)-shell to an electron in the \( s \)-shell). The Bloch part of the wave function determines the spin selection rules. For transverse electric fields only electron-hole pairs with a total angular momentum projection of \( \pm 1 \) can be excited, i.e., HH\( \pm 1 \) excitons with \( J_h = \pm 3/2 \) and \( J_e = \mp 1/2 \) and LH\( \pm 1 \) excitons with \( J_h = \pm 1/2 \) and \( J_e = \pm 1/2 \). Transitions into the “dark” HH\( \pm 2 \) excitons with \( J_h = \pm 3/2 \) and \( J_e = \pm 1/2 \) and LH\( \pm 0 \) excitons with \( J_h = \pm 1/2 \) and \( J_e = \mp 1/2 \) are forbidden [12]. For longitudinal field components, new spin selection rules apply and the light-hole excitons LH\( \pm 0 \) [27] can be excited, while HH\( \pm 1 \), HH\( \pm 2 \) and LH\( \pm 1 \) are spin forbidden. The transitions between different shells are governed by the overlap integral of the envelope functions. Since the confinement lengths for conduction and valence band states are taken to be the same, envelope functions from different shells are orthogonal and only transitions between the envelope states from the same shell are possible. Exemplary transitions for an excitation with the transverse and longitudinal field are sketched in gray and green in Fig. 1, respectively.

Using a simple model of uncoupled electron-hole pairs inside the QD, we can calculate the corresponding absorption spectrum, which is shown in Fig. 2(a) for the two types of exciting beams. Here we have considered a QD with a size of \( (L_x \times L_y \times L_z) = (5.4 \times 5.4 \times 2.0) \) nm\(^3\) and material parameters for CdSe as in Ref. [24]. The small lines at the bottom mark the positions of all states, which are in general multiply degenerate. We find that the corresponding spectra indeed match our expectations. More specifically, for the transverse field (bottom line) we see the \( s \rightarrow s \) and \( p \rightarrow p \) HH\( \pm 1 \) transitions and the \( S \rightarrow s \) LH\( \pm 1 \) transition. For the longitudinal field (top line) only the \( S \rightarrow s \) LH\( \pm 0 \) transition is excited in the given energetic range, however, the strength is similar to the case of the transverse field excitation.

Valence band mixing and correlation effects induced
by Coulomb interaction become important especially for higher exciton states [24]. To get a more realistic description of a QD, we extend the envelope function approximation by including the direct Coulomb interaction, the short range exchange Coulomb interaction as well as valence band mixing via the Luttinger Hamiltonian within a configuration interaction approach to obtain the exciton eigenstates of the QD. The interactions lead to a strong mixing of the electron-hole pair states. Details of the model can be found in Ref. [24].

The corresponding spectra are shown in Fig. 2(b) for the two types of excitation discussed above. Below the spectra again all existing states are indicated. Many degeneracies seen in the uncoupled model, associated typically with different spin configurations, are now lifted. For example, at the ground state transition (s → s) a splitting into two levels appears, the lower one reflecting the two dark HH±2 and the upper one the two bright HH±1 excitons. Both doublets can be further split by breaking the cylindrical symmetry of the QD confinement. The lowest transition involving light-holes, S → s splits into three levels, a doubly degenerate LH±1 transition as well as two energetically separated states containing the LH±0 excitons, from which one is accessible by the longitudinal field.

Let us now turn to the spectra obtained by taking Coulomb interaction and valence band mixing into account. The first thing to notice is that all the transitions from the reduced model still prevail in the full model, however at different energies due to the Coulomb shifts. Also, the relative intensity of the lines changes. Note that for reasons of clarity we use the same labels for the transitions. However, due to the mixing there are in general contributions from other electron-hole pair states. In addition, due to the state mixing several new absorption lines show up. In the considered energetic range these are d → s and D → s transitions.

To better understand the observed spectra, in Table I we compare the dark groundstate exciton and the ones excited by the transverse and longitudinal fields (see Fig. 2(b)). In addition, we provide the ratio R of dark (HH±2) to bright (HH±1) contributions and B is the relative brightness of the transitions.

![FIG. 2. Optical spectra of a QD in (a) the reduced QD model and (b) the full QD model including Coulomb interaction and valence band mixing. In each part the bottom line shows the spectrum for the transverse field and the upper line for the longitudinal field. The small lines at the bottom mark the positions of all available states, which are in general multiply degenerate.](image)

| Coupling to light | Transition | Spin contributions | $R$ | $B$ |
|------------------|------------|--------------------|-----|-----|
| Long. | s → s | 0.99 : 0.00: 0.01: 0.01 | 0.00 | 0.01 |
| Long. | S → s | 0.12 : 0.23 : 0.00 : 0.65 | 0.5 | 39% |
| Long. | d → s | 0.02 : 0.79 : 0.01 : 0.18 | 0.0 | 7% |
| Long. | d → s | 0.05 : 0.63 : 0.02 : 0.31 | 0.1 | 18% |
| Long. | D → s | 0.23 : 0.25 : 0.01 : 0.51 | 0.9 | 9% |
| Long. | S → s | 0.39 : 0.10 : 0.51 : 0.00 | 3.8 | 47% |
| Long. | d → s | 0.48 : 0.11 : 0.40 : 0.02 | 4.5 | 63% |
| Long. | D → s | 0.24 : 0.22 : 0.54 : 0.01 | 1.1 | 33% |

TABLE I. Different spin contributions of the dark exciton and the ones excited by the transverse and longitudinal fields for a QD with size $5.4 \times 5.4 \times 2$ nm$^3$. $R$ is the ratio of dark (HH±2) to bright (HH±1) contributions and $B$ is the relative brightness of the transitions.
cussed above, the valence band mixing causes a coupling between HH±2 and LH±0, leading to two mixed eigenstates. The LH±0 spin components of these eigenstates makes them accessible by the longitudinal light field.

After excitation, higher excitons tend to relax into lower lying exciton states; a process which dominantly occurs via the emission of phonons typically on a timescale of a few hundred fs [23]. Spin flip processes via the coupling to nuclear spins [39] occur on a ns timescale and are therefore negligible. Since the emission of phonons does not change spins, the spin state of the initial and final eigenstate of a relaxation need to be similar (the overlap needs to be large). Since the HH ground states (s → s transitions) have very pure spin states of either HH±2 or HH±1 type (cf. Table I), the relative contribution R between HH±2 and HH±1 within an excited eigenstate can be used to estimate the relative occupation between the HH±2 and the HH±1 ground states after relaxation. For an efficient mechanism to occupy the dark exciton ground state, the considered eigenstate should have a high value of R, but also a high brightness B, which is indeed the case for the S → s and d → s transitions excited by the longitudinal field.

To check whether the large values of R and B are specific for the chosen QD geometry, we performed parameter studies as in Ref. [24] on a variety of QD geometries. The obtained values of R and B are given for several realistic QD geometries in Tab. II. For each geometry, we considered two eigenstates: The one mainly consisting of S → s LH±0 and the brightest eigenstate mainly consisting of dark HH±2 spin contributions. While all scenarios provide suitable values for R and B, some geometries provide quite appealing combinations such as R = 110 and B = 20%. One should also note that the strength of valence band mixing can be adjusted by strain-tuning [40].

The proposed scheme is related to findings in Ref. [41], where a small coupling between the dark HH±2 ground state and LH±0 was found to enable an optical recombination of the dark exciton ground state by emitting a photon in in-plane direction (polarized in z-direction). This recombination was identified as the main relaxation process of the dark ground state. However, the coupling is small, because S → s LH±0 and s → s HH±2 are strongly separated in energy. Considering higher excited HH-shells, like the d-shell, these couplings become strong and enable the proposed efficient scheme to excite the dark HH ground state.

In conclusion, we propose an efficient and viable scheme to initialize the dark exciton using a single excitation at normal incidence. Due to long lifetimes, such dark excitons are useful for information storage in quantum technologies. Our scheme utilizes light beams with a pronounced longitudinal component tuned in resonance to certain higher exciton states. These states are characterized by a strong valence band mixing between optically active LH±0 excitons and higher dark HH±2 excitons, which enables a large coupling to the light field and, at the same time, an ultrafast relaxation path into the dark exciton ground state. We showed in a parameter study using a configuration interaction approach, that our findings are not specific to a certain QD geometry but should...
appear rather generically.

Acknowledgment: G. F. Q. thanks the ONRG for financial support through NICOP grant N62909-18-1-2090

[1] P. Michler, Quantum dots for quantum information technologies, Vol. 237 (Springer, 2017).

[2] O. Gazzano and G. S. Solomon, “Toward optical quantum information processing with quantum dots coupled to microstructures,” JOSA B 33, C160 (2016).

[3] P. Senellart, G. Solomon, and A. White, “High-performance semiconductor quantum-dot single-photon sources,” Nat. nanotechnol. 12, 1026 (2017).

[4] D. Huber, M. Reindl, J. Aberl, A. Rastelli, and R. Trotta, “Semiconductor quantum dots as an ideal source of polarization-entangled photon pairs on-demand: A review,” J. Opt. 20, 073002 (2018).

[5] S. L. Portalupi, M. Jetter, and P. Michler, “InAs quantum dots grown on metamorphic buffers as non-classical light sources at telecom C-band: A review,” Semicond. Sci. Technol. 34, 053001 (2019).

[6] S. Rodt, S. Reitzenstein, and T. Heindel, “Deterministically fabricated solid-state quantum-light sources,” Journal of Physics: Condensed Matter 32, 153003 (2020).

[7] E. Biollati, R. C. Iotti, P. Zanardi, and F. Rossi, “Quantum information processing with semiconductor macroatoms,” Phys. Rev. Lett. 85, 5647 (2000).

[8] A. J. Ramsay, “A review of the coherent optical control of the exciton and spin states of semiconductor quantum dots,” Semicond. Sci. Technol. 25, 103001 (2010).

[9] R. J. Warburton, “Single spins in self-assembled quantum dots.” Nature Mat. 12, 483 (2013).

[10] J. McFarlane, P. A. Dalgaro, B. D. Gerardot, R. H. Hadfield, R. J. Warburton, K. Karrai, A. Badolato, and P. M. Petroff, “Gigahertz bandwidth electrical control over a dark exciton-based memory bit in a single quantum dot.” Appl. Phys. Lett. 94, 093113 (2009).

[11] J. Johansen, B. Julsgaard, S. Stobbe, J. M. Hvam, and P. Lodahl, “Probing long-lived dark excitons in self-assembled quantum dots,” Phys. Rev. B 81, 081304 (2010).

[12] E. Poem, Y. Kodriano, C. Tradonsky, N. H. Lindner, B. D. Gerardot, P. M. Petroff, and D. Gershoni, “Accessing the dark exciton with light,” Nature Phys. 6, 993 (2010).

[13] Y. H. Huo, V. Krápek, O. G. Schmidt, and A. Rastelli, “Spontaneous brightening of dark excitons in GaAs/AlGaAs quantum dots near a cleaved facet,” Phys. Rev. B 95, 165304 (2017).

[14] T. Heindel, A. Thoma, I. Schwartz, E. R. Schmidgall, L. Gantz, D. Cogan, M. Strauß, P. Schnauber, M. Gschrey, J.-H. Schulze, S. Rodt, D. Gershoni, and S. Reitzenstein, “Accessing the dark exciton spin in deterministic quantum-dot microlenses,” APL Photonics 2, 121303 (2017).

[15] S. Lübker and D. E. Reiter, “A review on optical excitation of semiconductor quantum dots under the influence of phonons,” Semicond. Sci. Technol. 34, 063002 (2019).

[16] D. E. Reiter, T. Kuhn, and V. M. Axt, “All-optical spin manipulation of a single manganese atom in a quantum dot,” Phys. Rev. Lett. 102, 177403 (2009).

[17] S. Lüker, T. Kuhn, and D. E. Reiter, “Direct optical state preparation of the dark exciton in a quantum dot,” Phys. Rev. B 92, 201305 (2015).

[18] S. Germanis, P. Atkinson, R. Hostein, C. Gourdon, V. Voliotis, A. Lemaître, M. Bernard, F. Margaillan, S. Majrab, and B. Eble, “Dark-bright exciton coupling in asymmetric quantum dots,” Phys. Rev. B.

[19] M. Zielinski, Y. Don, and D. Gershoni, “Atomic theory of dark excitons in self-assembled quantum dots of reduced symmetry,” Phys. Rev. B 91, 085403 (2015).

[20] I. Schwartz, E. R. Schmidgall, L. Gantz, D. Cogan, E. Bordo, Y. Don, M. Zielinski, and D. Gershoni, “Deterministic writing and control of the dark exciton spin using single short optical pulses,” Phys. Rev. X 5, 011009 (2015).

[21] J. Huneke, I. D’Amico, P. Machnikowski, T. Thomay, R. Bratschitsch, A. Leitenstorfer, and T. Kuhn, “Role of Coulomb correlations for femtosecond pump-probe signals obtained from a single quantum dot,” Phys. Rev. B 84, 115320 (2011).

[22] T. Suzuki, R. Singh, G. Moody, M. Aßmann, M. Bayer, A. Ludwig, A. D. Wieck, and S. T. Cundiff, “Dephasing of InAs quantum dot p-shell excitons studied using two-dimensional coherent spectroscopy,” Phys. Rev. B 98, 195304 (2018).

[23] C. Hinz, P. Gunsheinerner, C. Traum, M. Holtkemper, B. Bauer, J. Haase, S. Mahapatra, A. Frey, K. Brunner, D. E. Reiter, T. Kuhn, D. V. Seletskiy, and A. Leitenstorfer, “Charge and spin control of ultrafast electron and hole dynamics in single CdSe/ZnSe quantum dots,” Phys. Rev. B 97, 045302 (2018).

[24] M. Holtkemper, D. E. Reiter, and T. Kuhn, “Influence of the quantum dot geometry on p-shell transitions in differently charged quantum dots,” Phys. Rev. B 97, 075308 (2018).

[25] C. Qian, X. Xie, J. Yang, K. Peng, S. Wu, F. Song, S. Sun, J. Dang, Y. Yu, M. J. Steer, et al., “Enhanced strong interaction between nanocavities and p-shell excitons beyond the dipole approximation,” Phys. Rev. Lett. 122, 087401 (2019).

[26] G. F. Quintheieiro, D. E. Reiter, and T. Kuhn, “Formulaion of the twisted-light–matter interaction at the phase singularity: The twisted-light gauge,” Phys. Rev. A 91, 033808 (2015).

[27] G. F. Quinteiro and T. Kuhn, “Light-hole transitions in quantum dots: Realizing full control by highly focused optical-vortex beams,” Phys. Rev. B 90, 115401 (2014).

[28] K. S. Youngworth and T. G. Brown, “Focusing of high numerical aperture cylindrical-vector beams,” Optics Express 7, 77 (2000).

[29] L. Novotny, M. R. Beversluis, K. S. Youngworth, and T. G. Brown, “Longitudinal field modes probed by single molecules,” Phys. Rev. Lett. 86, 5251 (2001).

[30] Q. Zhan, “Trapping metallic Rayleigh particles with radial polarization,” Optics Express 12, 3377 (2004).

[31] G. F. Quinteiro, F. Schmidt-Kaler, and C. T. Schmiegelow, “Twisted-light–ion interaction: The role of longitudinal fields,” Phys. Rev. Lett. 119, 253203 (2017).

[32] J. Durnin, J. J. Miceli, and J. H. Eberly, “Diffraction-free beams,” Phys. Rev. Lett. 58, 1499 (1987).

[33] M. Woerdeman, C. Alpmann, M. Esseling, and C. Dzenz, “Advanced optical trapping by complex beam shaping,” Laser Photon. Rev. 7, 839 (2013).

[34] M. Ettorre, S. C. Pavone, M. Casaletti, and M. Al-
bani, “Experimental validation of Bessel beam generation using an inward Hankel aperture distribution,” IEEE Transactions on Antennas and Propagation 63, 2539 (2015).

[35] G. Milione, A. Dudley, T. A. Nguyen, O. Chakraborty, E. Karimi, A. Forbes, and R. A. Alfano, “Measuring the self-healing of the spatially inhomogeneous states of polarization of vector Bessel beams,” J. Opt. 17, 035617 (2015).

[36] S. Ninomiya and S. Adachi, “Optical properties of cubic and hexagonal CdSe,” J. Appl. Phys. 78, 4681 (1995).

[37] W. Chen and Q. Zhan, “Realization of an evanescent Bessel beam via surface plasmon interference excited by a radially polarized beam,” Opt. Lett. 34, 722 (2009).

[38] H. Huang, Q. Li, J. Fu, J. Wu, F. Lin, and X. Wu, “Efficient subwavelength focusing of light with a long focal depth,” Nanoscale 7, 16504 (2015).

[39] I. A. Merkulov, A. L. Efros, and M. Rosen, “Electron spin relaxation by nuclei in semiconductor quantum dots,” Phys. Rev. B 65, 205309 (2002).

[40] Y. H. Huo, B. J. Witek, S. Kumar, J. R. Cardenas, J. X. Zhang, N. Akopian, R. Singh, E. Zallo, R. Grifone, D. Kriegner, et al., “A light-hole exciton in a quantum dot,” Nature Phys. 10, 46 (2014).

[41] T. Smoleniski, T. Kazimierczuk, M. Goryca, T. Jakubczyk, P. Wojnar, A. Golnik, P. Kossacki, et al., “In-plane radiative recombination channel of a dark exciton in self-assembled quantum dots,” Phys. Rev. B 86, 241305 (2012).