Universität
Münster

Controlling the optical excitation of semiconductor nanostructures by vortex beams

Tilmann Kuhn
Institute of Solid State Theory
University of Münster

Characterization and control of quantum materials with optical vortex beams

living.knowledge

SPICE Workshop, Ingelheim, June 10th – 12th, 2025

many thanks to:

Guillermo F. Quinteiro Rosen (Universidad Nacional del Nordeste, Corrientes, Argentina)

Pablo Tamborenea (Universidad de Buenos Aires, Argentina)

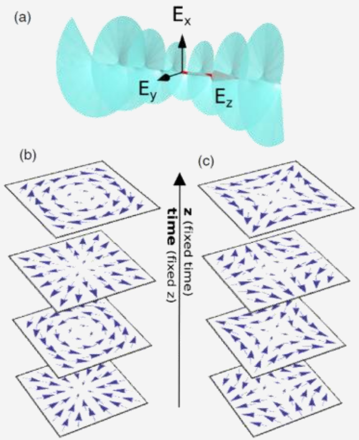
Doris Reiter (TU Dortmund, Germany)

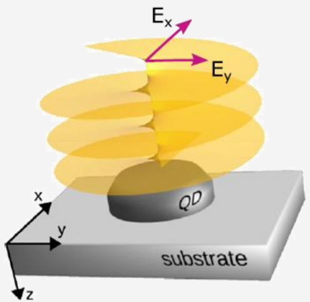
Matthias Holtkemper (University of Münster, Germany)

Universität Münster

Overview

- vortex beams
  - paraxial and propagation-invariant beams
  - construction of vortex beams
  - properties of vortex beams
- excitation of quantum dots by vortex beams
  - basic properties of quantum dots
  - selection rules for excitation by vortex beams
  - reconstruction of exciton states
  - dark exciton preparation using structured light
- conclusions





Universität Münster

Overview

- vortex beams
  - paraxial and propagation-invariant beams
  - construction of vortex beams
  - properties of vortex beams
- excitation of quantum dots by vortex beams
  - basic properties of quantum dots
  - selection rules for excitation by vortex beams
  - reconstruction of exciton states
  - dark exciton preparation using structured light
- conclusions

Propagation-invariant and paraxial beams

- light propagation in free space

  from Maxwell's equations: wave equation for electric (and magnetic) field

  $$\nabla^2 \mathbf{E}(\mathbf{r},t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E}(\mathbf{r},t) = 0 \qquad \text{with} \quad c^2 = (\mu_0 \epsilon_0)^{-1}$$

- for harmonic (single frequency) beam with $k = \omega/c$: Helmholtz equation

  $$\nabla^2 \mathbf{E}(\mathbf{r},t) + k^2 \mathbf{E}(\mathbf{r},t) = 0$$

- assumption: **propagation-invariant ("non-diffracting") beam** propagating in z-direction

  $$\mathbf{E}(\mathbf{r},t) = \tilde{\mathbf{E}}(\mathbf{r}_\perp) e^{i(q_z z - \omega t)} + \text{c.c.}$$

- leads to

  $$\nabla_\perp^2 \tilde{\mathbf{E}}(\mathbf{r}_\perp) + (k^2 - q_z^2)\tilde{\mathbf{E}}(\mathbf{r}_\perp) = 0$$

- factorization in cartesian coordinates (⟶ plane waves), polar coordinates (⟶ Bessel beams), …
- from Maxwell's equation $\nabla \cdot \mathbf{E} = 0$:

  transverse components $\tilde{\mathbf{E}}_\perp(\mathbf{r}_\perp)$ can be independently chosen among solutions of Helmholtz equation

  longitudinal component determined by $\tilde{E}_z(\mathbf{r}_\perp) = \frac{i}{q_z} \nabla_\perp \cdot \tilde{\mathbf{E}}_\perp(\mathbf{r}_\perp)$

Universität Münster

Propagation-invariant and paraxial beams

- except for special cases: beam with radial structure (e.g., finite beam waist w_0) experiences diffraction
- Fraunhofer diffraction on a slit with width w_0 :

  $\rightarrow$ width of central maximum grows according to $w/w_0 = 2\lambda z/w_0^2 = 4\pi z/kw_0^2$
- characteristic length scale for widening of beam: diffraction length $l = kw_0^2$
- divergence of beam described by **paraxial parameter** $f = \dfrac{w_0}{l} = \dfrac{1}{w_0 k}$
- ansatz for **paraxial beam** propagating in z-direction

$$\mathbf{E}(\mathbf{r},t) = \tilde{\mathbf{E}}(\mathbf{r})e^{i(kz-\omega t)} + \text{c.c.}$$

- leads to paraxial Helmholtz equation

$$\nabla_\perp^2 \tilde{\mathbf{E}}_\perp(\mathbf{r}) + 2i\,k\,\partial_z \tilde{\mathbf{E}}_\perp(\mathbf{r}) = -\partial_z^2 \tilde{\mathbf{E}}_\perp(\mathbf{r}) \approx 0$$

r.h.s is of order f^2

- longitudinal component from Maxwell's equation $\boldsymbol{\nabla}\cdot\mathbf{E} = 0$:

$$\tilde{E}_z(\mathbf{r}) = i f w_0 \boldsymbol{\nabla}_\perp \cdot \tilde{\mathbf{E}}_\perp(\mathbf{r})$$

of order f

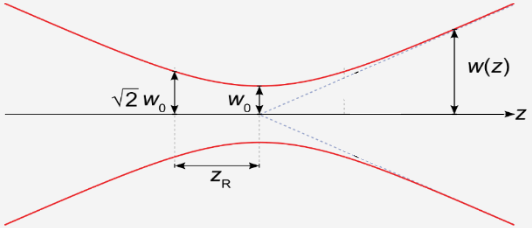
Laguerre-Gauss and Bessel beams

- solution of paraxial Helmholtz equation: **Laguerre-Gauss beams**

$$\tilde{\mathbf{E}}(\mathbf{r}) = E_0 \mathbf{e}_{\sigma} \sqrt{\frac{2p!}{\pi(p+|\ell|)!}} \frac{1}{w(z)} \left(\frac{r\sqrt{2}}{w(z)}\right)^{|\ell|} e^{i\ell\varphi} e^{i\psi(z)} L_p^{|\ell|}\left(\frac{2r^2}{w^2(z)}\right) \exp\left[-\frac{r^2}{w^2(z)}\right] \exp\left[-ik\frac{r^2}{2R(z)}\right]$$

with $w(z) = w_0\sqrt{1+(z/z_R)^2}$, $z_R = (1/2)kw_0^2$, $R(z) = z[1+(z_R/z)^2]$, $\psi(z) = -(|\ell|+2p+1)\arctan(z/z_R)$

and $L_p^{|\ell|}$: generalized Laguerre polynomial



- solution of full Helmholtz equation (transverse components also of paraxial equation): **Bessel beams**

$$\tilde{\mathbf{E}}(\mathbf{r}) = iE_0 J_\ell(q_r r) e^{i\ell\varphi} \mathbf{e}_\sigma + \sigma \frac{q_r}{q_z} \frac{E_0}{\sqrt{2}} J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi} \mathbf{e}_z$$

with $J_\ell(q_r r)$: Bessel function of first kind and order ℓ

Universität Münster

Potentials and gauges

- derivation of electric and magnetic fields from potentials

$$\mathbf{E}(\mathbf{r},t) = -\partial_t \mathbf{A}(\mathbf{r},t) - \nabla\Phi(\mathbf{r},t)$$

$$\mathbf{B}(\mathbf{r},t) = \nabla \times \mathbf{A}(\mathbf{r},t)$$

- equations of motion for $\mathbf{A}$ and Φ decouple in
  - Coulomb gauge $\nabla \cdot \mathbf{A}(\mathbf{r},t) = 0$
  - Lorenz gauge $\nabla \cdot \mathbf{A}(\mathbf{r},t) + \frac{1}{c^2}\frac{\partial\Phi(\mathbf{r},t)}{\partial t} = 0$
- in Coulomb gauge far from sources: $\Phi = 0$ $\rightarrow$ special case of Lorenz gauge
- discussion of propagation-invariant and paraxial beams holds also for $\mathbf{A}$
- in strictly paraxial limit: $\mathbf{A}$ is transverse, satisfies
  - Helmholtz equation $\nabla_\perp^2 \tilde{\mathbf{A}}_\perp(\mathbf{r}_\perp) + \left(k^2 - q_z^2\right)\tilde{\mathbf{A}}_\perp(\mathbf{r}_\perp) = 0$ (propagation-invariant beam)
  - paraxial Helmholtz equation $\nabla_\perp^2 \tilde{\mathbf{A}}_\perp(\mathbf{r}) + 2ik\partial_z \tilde{\mathbf{A}}_\perp(\mathbf{r}) = 0$ (paraxial beam)
- construction of A_z and Φ from Lorenz gauge condition

Potentials and gauges

- choice: $\Phi^{(1)} = 0$ (3 component A, 3CA)

$$\mathbf{A}_\perp^{(1)}(\mathbf{r},t) = \mathbf{A}_\perp(\mathbf{r},t),$$
$$\partial_z A_z^{(1)}(\mathbf{r},t) = -\nabla_\perp \cdot \mathbf{A}_\perp(\mathbf{r},t),$$
$$\Phi^{(1)}(\mathbf{r},t) = 0.$$

- choice: $A_z^{(0)} = 0$ (2 component A, 2CA)

$$\mathbf{A}_\perp^{(0)}(\mathbf{r},t) = \mathbf{A}_\perp(\mathbf{r},t),$$
$$A_z^{(0)}(\mathbf{r},t) = 0,$$
$$\Phi^{(0)}(\mathbf{r},t) = -i\frac{c^2}{\omega}\nabla_\perp \cdot \mathbf{A}_\perp(\mathbf{r},t).$$

- generalization, weighting with parameter γ

$$\mathbf{A}_\perp^{(\gamma)}(\mathbf{r},t) = \mathbf{A}_\perp(\mathbf{r},t),$$
$$\partial_z A_z^{(\gamma)}(\mathbf{r},t) = -\gamma\nabla_\perp \cdot \mathbf{A}_\perp(\mathbf{r},t),$$
$$\Phi^{(\gamma)}(\mathbf{r},t) = -i(1-\gamma)\frac{c^2}{\omega}\nabla_\perp \cdot \mathbf{A}_\perp(\mathbf{r},t).$$

- all potentials satisfy Lorenz gauge condition and wave equation $\rightarrow$ electromag. fields satisfy Maxwell's equations
- A_z and Φ of first order in paraxial parameter $f = (w_0 k)^{-1}$ or $f = q_r/q_z$
- potentials for different γ are not related by a gauge transformation: lead to different electromagnetic fields

Application to Bessel beams

- ansatz: transverse vector potential for vortex beam

$$\mathbf{A}_\perp(\mathbf{r},t) = A_0 J_\ell(q_r r) e^{i\ell\varphi} e^{i(q_z z - \omega t)} \mathbf{e}_\sigma$$ with in-plane polarization vector $\mathbf{e}_\sigma = (\mathbf{e}_x + i\sigma \mathbf{e}_y)/\sqrt{2}$, $\sigma = \pm 1$

  - vector potential with well-defined orbital (OAM) and spin (SAM) angular momentum $\hbar\ell$ and $\hbar\sigma$, respectively
  - radial shape: Bessel function of first kind and order ℓ, satisfies $J_{-\ell}(q_r r) = (-1)^\ell J_\ell(q_r r)$
  - q_r^{-1} characterizes radial length scale (beam waist), close to beam center $J_\ell(q_r r) \propto (q_r r)^{|\ell|}$
  - structure of beam unchanged for $(\sigma,\ell) \Leftrightarrow (-\sigma,-\ell)$, in the following $\ell \geq 0$

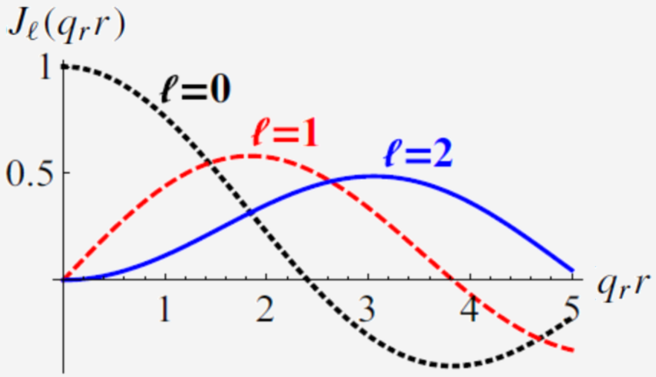
- construction of missing components: divergence of the transverse potential

$$\nabla_\perp \cdot \mathbf{A}_\perp(\mathbf{r},t) = -\sigma \frac{A_0}{\sqrt{2}} q_r J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi} e^{i(q_z z - \omega t)}$$

- potentials satisfying Lorenz gauge (with $\overline{\gamma} = 1 - \gamma$)

$$\tilde{\mathbf{A}}^{(\gamma)}(\mathbf{r}) = A_0 J_\ell(q_r r) e^{i\ell\varphi} \mathbf{e}_\sigma - i\gamma\sigma \frac{q_r}{q_z} \frac{A_0}{\sqrt{2}} J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi} \mathbf{e}_z$$

$$\tilde{\Phi}^{(\gamma)}(\mathbf{r}) = i\overline{\gamma} \frac{c^2}{\omega} \sigma \frac{A_0}{\sqrt{2}} q_r J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi}$$



Bessel beams: electromagnetic fields

- field components from $\mathbf{E}(\mathbf{r},t) = -\partial_t \mathbf{A}(\mathbf{r},t) - \nabla\Phi(\mathbf{r},t)$, $\mathbf{B}(\mathbf{r},t) = \nabla \times \mathbf{A}(\mathbf{r},t)$
- with $\mathbf{E}(\mathbf{r},t) = \tilde{\mathbf{E}}(\mathbf{r}) e^{i(q_z z - \omega t)}$ and $\mathbf{B}(\mathbf{r},t) = \tilde{\mathbf{B}}(\mathbf{r}) e^{i(q_z z - \omega t)}$, $E_0 = \omega A_0$, $B_0 = q_z A_0$
- for $\gamma = 1$ (3CA)

$$\tilde{\mathbf{E}}^{(1)}(\mathbf{r}) = i E_0 J_\ell(q_r r) e^{i\ell\varphi} \mathbf{e}_\sigma + \sigma \frac{q_r}{q_z} \frac{E_0}{\sqrt{2}} J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi} \mathbf{e}_z$$

$$\tilde{\mathbf{B}}^{(1)}(\mathbf{r}) = \sigma B_0 J_\ell(q_r r) e^{i\ell\varphi} \mathbf{e}_\sigma + \sigma \frac{B_0}{2} \left(\frac{q_r}{q_z}\right)^2 [J_{\ell+\sigma+1}(q_r r) e^{i(\ell+\sigma+1)\varphi} \mathbf{e}_- + J_{\ell+\sigma-1}(q_r r) e^{i(\ell+\sigma-1)\varphi} \mathbf{e}_+]$$

$$- i \frac{B_0}{\sqrt{2}} \frac{q_r}{q_z} J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi} \mathbf{e}_z$$

with $\mathbf{e}_\pm = \mathbf{e}_{\sigma=\pm 1}$

- properties
  - longitudinal components of first order in paraxial parameter $f = (q_r/q_z)$
  - transverse component of $\mathbf{E}$ circularly polarized
  - transverse component of $\mathbf{B}$ elliptically polarized, correction of order f^2
  - all non-paraxial corrections exhibit mixing of orbital and spin angular momentum
    longitudinal fields $\ell + \sigma$, transverse fields $\ell + \sigma \pm 1$

Bessel beams: electromagnetic fields

- for $\gamma = 0$ (2CA)

$$\tilde{\mathbf{E}}^{(0)}(\mathbf{r}) = iE_0 J_\ell(q_r r)e^{i\ell\varphi}\mathbf{e}_\sigma + i\sigma\frac{E_0}{2}\left(\frac{cq_r}{\omega}\right)^2 [J_{\ell+\sigma+1}(q_r r)e^{i(\ell+\sigma+1)\varphi}\mathbf{e}_- - J_{\ell+\sigma-1}(q_r r)e^{i(\ell+\sigma-1)\varphi}\mathbf{e}_+]$$
$$+\sigma c^2\frac{q_z q_r}{\omega^2}\frac{E_0}{\sqrt{2}}J_{\ell+\sigma}(q_r r)e^{i(\ell+\sigma)\varphi}\mathbf{e}_z$$

$$\tilde{\mathbf{B}}^{(0)}(\mathbf{r}) = \sigma B_0 J_\ell(q_r r)e^{i\ell\varphi}\mathbf{e}_\sigma - i\frac{B_0}{\sqrt{2}}\frac{q_r}{q_z}J_{\ell+\sigma}(q_r r)e^{i(\ell+\sigma)\varphi}\mathbf{e}_z$$

- properties
  - longitudinal components and OAM – SAM mixing as before
  - transverse component of $\mathbf{B}$ circularly polarized
  - transverse component of $\mathbf{E}$ elliptically polarized

Bessel beams: electromagnetic fields

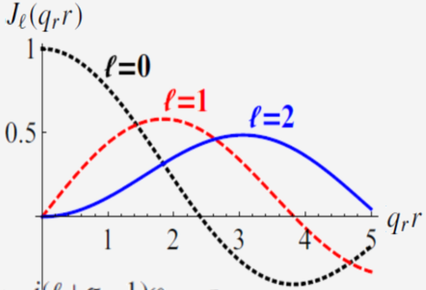
- for other values of γ: both $\mathbf{E}$ and $\mathbf{B}$ elliptically polarized
- special case for $\gamma_s = \frac{1}{1+\frac{\omega}{cq_z}} = \frac{1}{1+\sqrt{1+\left(\frac{q_r}{q_z}\right)^2}}$

$$\tilde{\mathbf{E}}^{(\gamma_s)}(\mathbf{r}) = i\frac{E_0}{2}\Big[\Big(1-\sigma\frac{cq_z}{\omega}\Big)J_{\ell+\sigma+1}(q_r r)e^{i(\ell+\sigma+1)\varphi}\mathbf{e}_- + \Big(1+\sigma\frac{cq_z}{\omega}\Big)J_{\ell+\sigma-1}(q_r r)e^{i(\ell+\sigma-1)\varphi}\mathbf{e}_+ - i\sigma\sqrt{2}\frac{cq_r}{\omega}J_{\ell+\sigma}(q_r r)e^{i(\ell+\sigma)\varphi}\mathbf{e}_z\Big]$$

$$\tilde{\mathbf{B}}^{(\gamma_s)}(\mathbf{r}) = -i\sigma\frac{B_0}{cE_0}\frac{\omega}{q_z}\tilde{\mathbf{E}}^{(\gamma_s)}(\mathbf{r})$$

- properties
  - $\mathbf{E}$ and $\mathbf{B}$ fields completely symmetric
  - $E/(cB) = 1$ as for plane waves
  - fields in agreement with superposition of plane waves with given circular polarization

Bessel beams: parallel and antiparallel class



- 3CA fields:

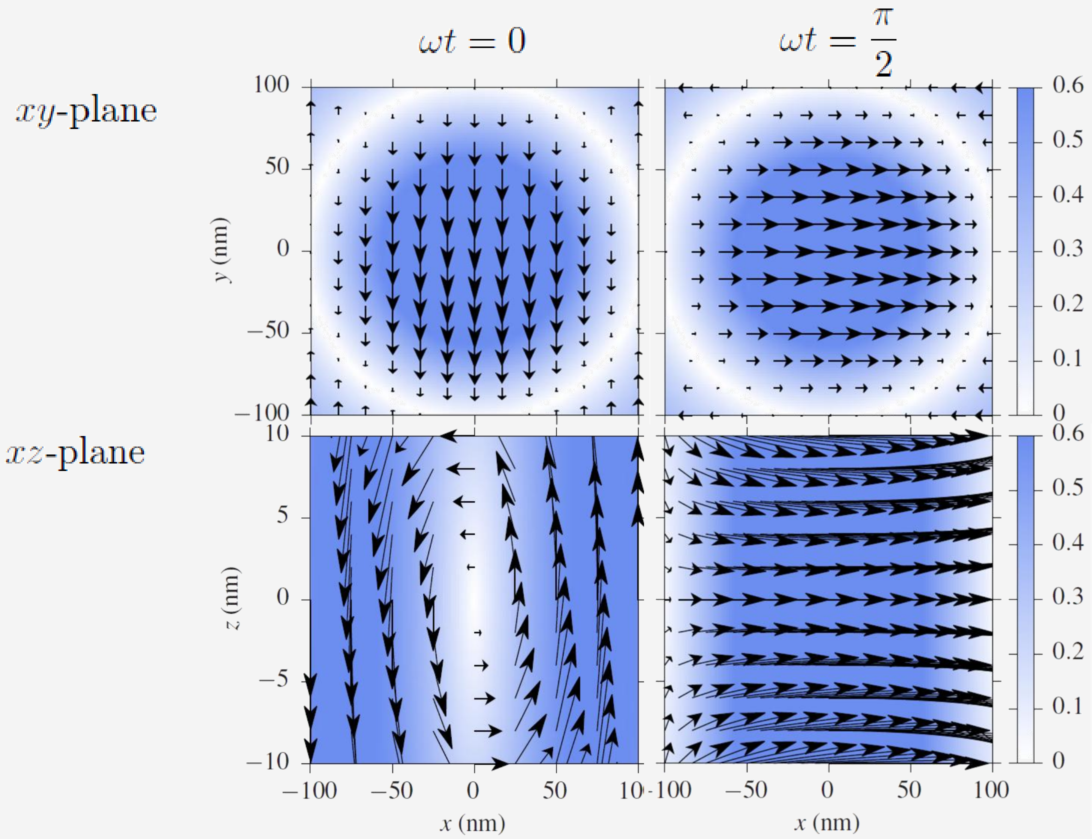
$$\tilde{\mathbf{E}}^{(1)}(\mathbf{r}) = iE_0 J_\ell(q_r r) e^{i\ell\varphi}\mathbf{e}_\sigma + \sigma \frac{q_r}{q_z}\frac{E_0}{\sqrt{2}} J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi}\mathbf{e}_z$$

$$\tilde{\mathbf{B}}^{(1)}(\mathbf{r}) = \sigma B_0 J_\ell(q_r r) e^{i\ell\varphi}\mathbf{e}_\sigma + \sigma \frac{B_0}{2}\left(\frac{q_r}{q_z}\right)^2 [J_{\ell+\sigma+1}(q_r r) e^{i(\ell+\sigma+1)\varphi}\mathbf{e}_- + J_{\ell+\sigma-1}(q_r r) e^{i(\ell+\sigma-1)\varphi}\mathbf{e}_+]$$

$$- i\frac{B_0}{\sqrt{2}}\frac{q_r}{q_z} J_{\ell+\sigma}(q_r r) e^{i(\ell+\sigma)\varphi}\mathbf{e}_z$$

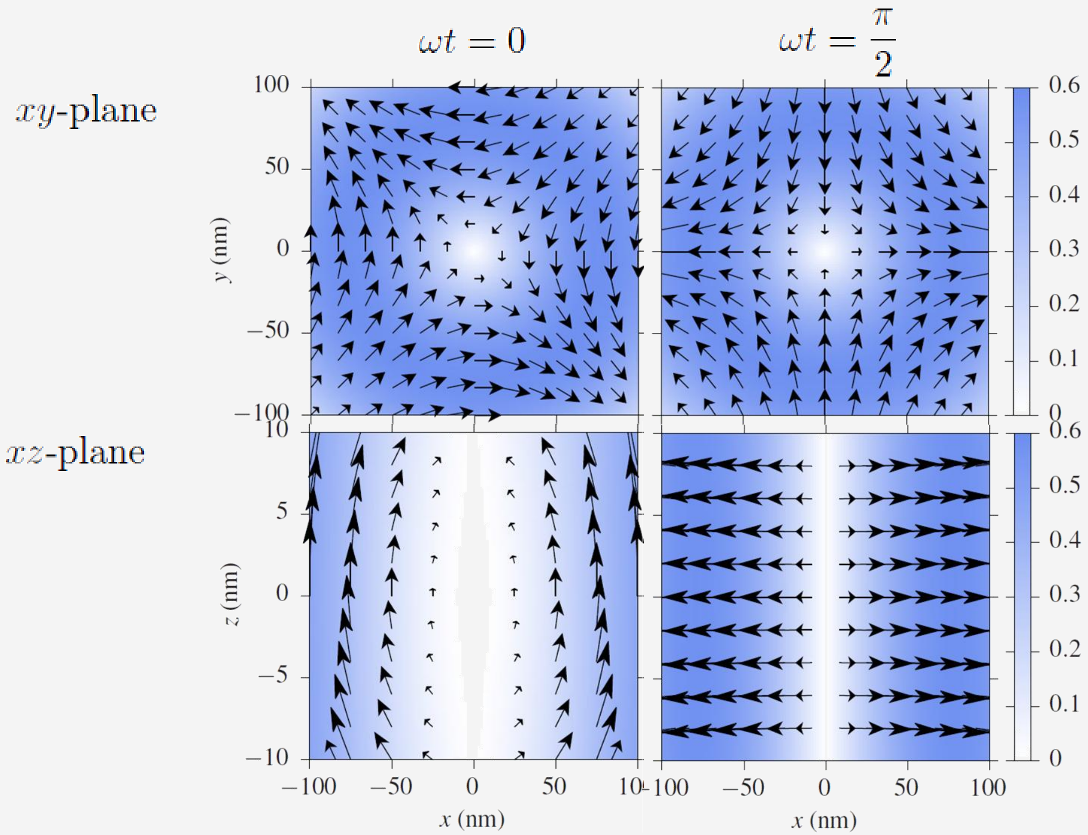
- beams can be separated into two classes:
  - parallel class: $\mathrm{sign}(\sigma) = \mathrm{sign}(\ell) \;\rightarrow\; |\ell+\sigma| > |\ell|$
  - antiparallel class: $\mathrm{sign}(\sigma) \neq \mathrm{sign}(\ell) \;\rightarrow\; |\ell+\sigma| < |\ell|$
- in parallel class non-paraxial corrections in higher order than $|\ell| \rightarrow$ close to beam center <u>less important</u>
- in antiparallel class non-paraxial corrections in lower order than $|\ell| \rightarrow$ close to beam center <u>more important</u>
- particularly interesting cases:
  - $\ell = 1\,,\ \sigma = -1$: non-vanishing longitudinal electric and magnetic field at the beam center
  - $\ell = 2\,,\ \sigma = -1$: non-vanishing transverse magnetic field at beam center

Electric field profiles for $\ell = 0, \sigma = 1$



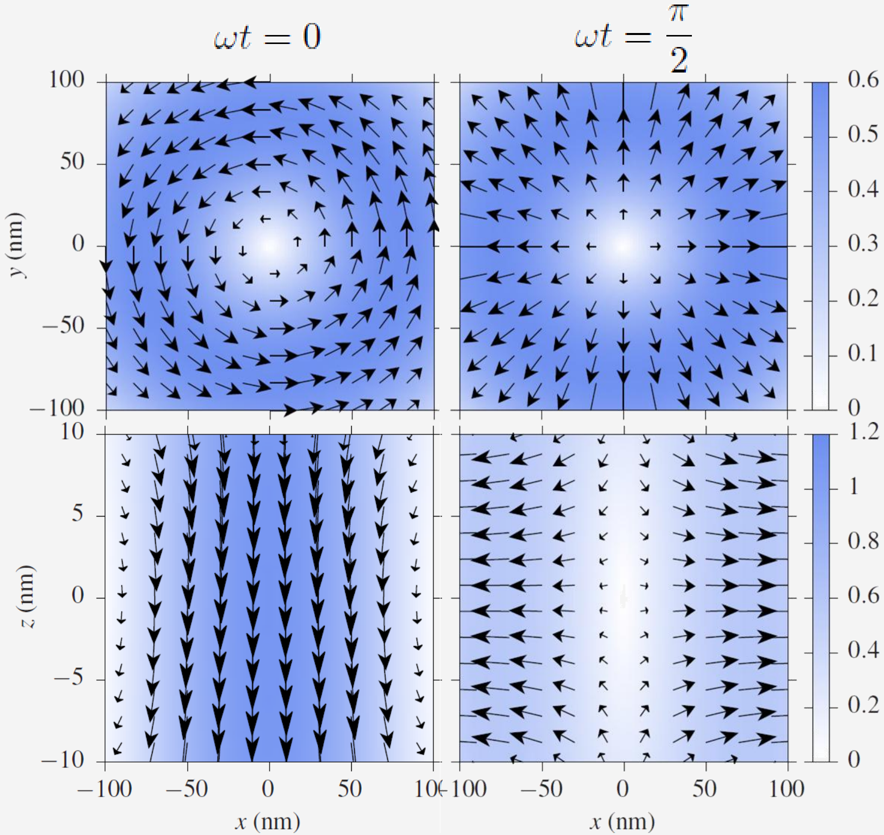
- in-plane component dominates in region around beam center
- z-component vanishes at beam center
- behaves similar to plane wave

Electric field profiles for $\ell = 1, \sigma = 1$



- beam from parallel class
- in-plane component dominates around beam center
- singularity has saddle-point like structure at any time
- z-component vanishes at beam center

Electric field profiles for $\ell = 1, \sigma = -1$



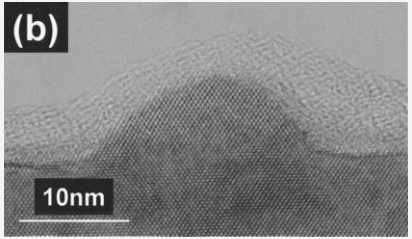
- beam from antiparallel class
- in-plane component alternately azimuthally and radially polarized
- singularity cycles through center, source, center, sink, …
- z-component dominates close to beam center

Overview

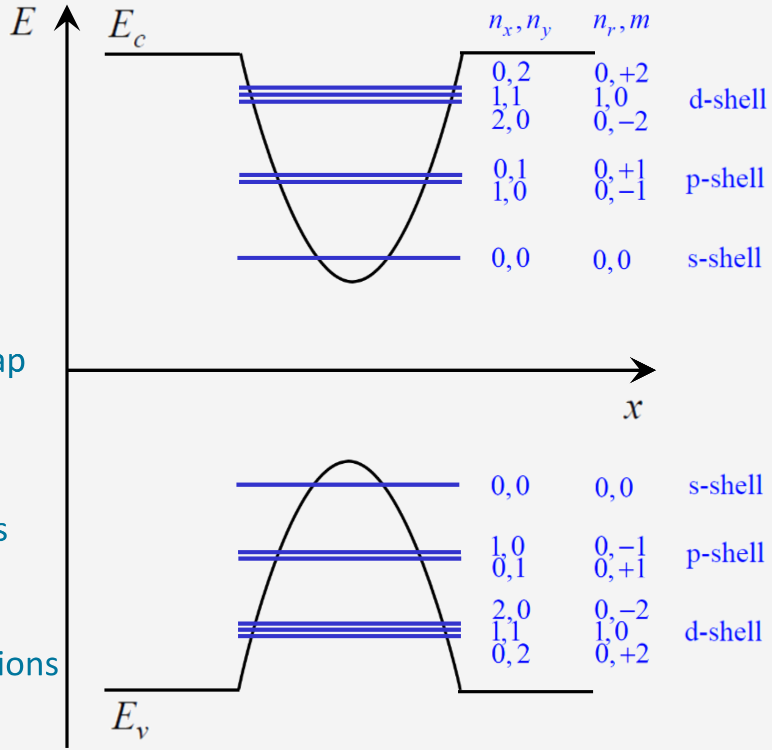
- vortex beams
  - paraxial and propagation-invariant beams
  - construction of vortex beams
  - properties of vortex beams
- excitation of quantum dots by vortex beams
  - basic properties of quantum dots
  - selection rules for excitation by vortex beams
  - reconstruction of exciton states
  - dark exciton preparation using structured light
- conclusions

Quantum dots: confinement and envelope level structure

- self-assembled quantum dots, grown by Stranski-Krastanov growth



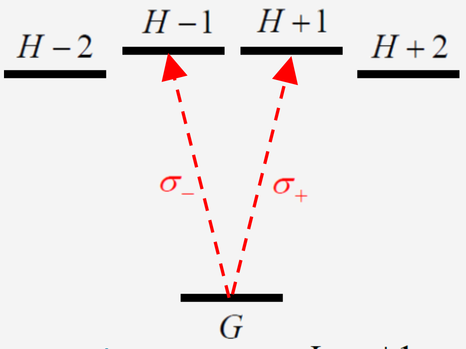
P.W. Fry et al.,
Phys. Rev. B 62, 16784 (2000)

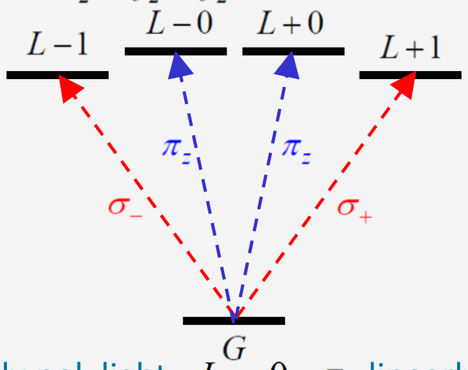


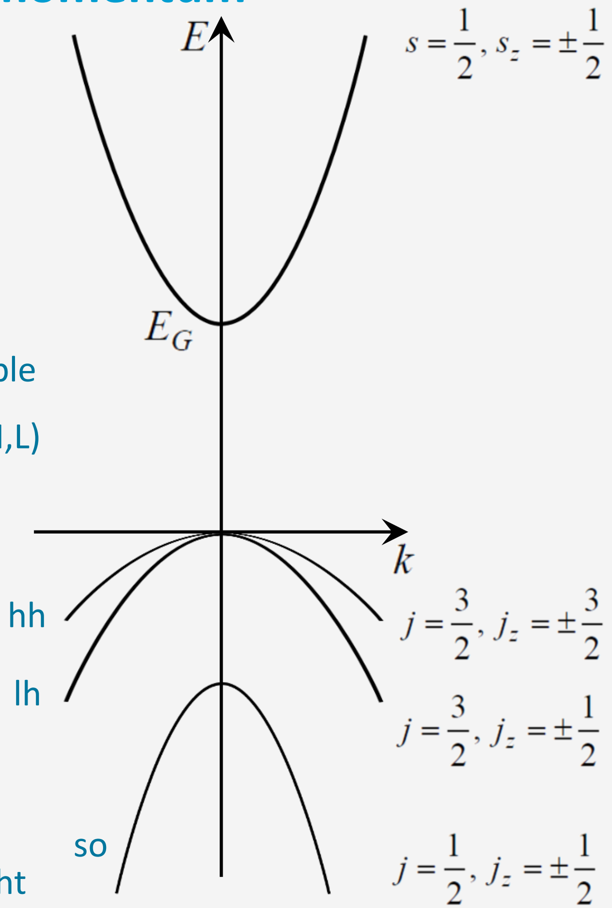
- material with smaller band gap embedded in material with larger gap e.g., InAs in GaAs, GaAs in AlGaAs
- thickness variation leads to approximately harmonic lateral confinement potential for envelope functions of electrons and holes
- two-dimensional harmonic oscillator, for cylindrical symmetry:
  - factorization in cartesian coordinates: Hermite-Gauss wave functions
    $$E = \hbar\omega_{\mathrm{conf}}\left(n_x + n_y + 1\right)$$
  - factorization in polar coordinates: Laguerre-Gauss wave functions
    $$E = \hbar\omega_{\mathrm{conf}}\left(2n_r + |m| + 1\right)$$ m : envelope angular momentum

Quantum dots: spin and intrinsic angular momentum

- full wave functions: product of envelope, Bloch and spin functions
- typical band structure of bulk III-V semiconductors
- conduction band: originates from s-orbitals $l = 0,\ j = s = \frac{1}{2}$
- valence bands: originate from p-orbitals $l = 1,\ s = \frac{1}{2},\ j = \frac{1}{2}, \frac{3}{2}$
- with spin-orbit interaction:
  heavy hole (hh), light hole (lh) and split-off (so) band; so band typically negligible
- excitation of electron-hole pairs: excitons; characterization by valence band (H,L) and angular momentum of electron+hole $J_z = j_z^e + j_z^h$







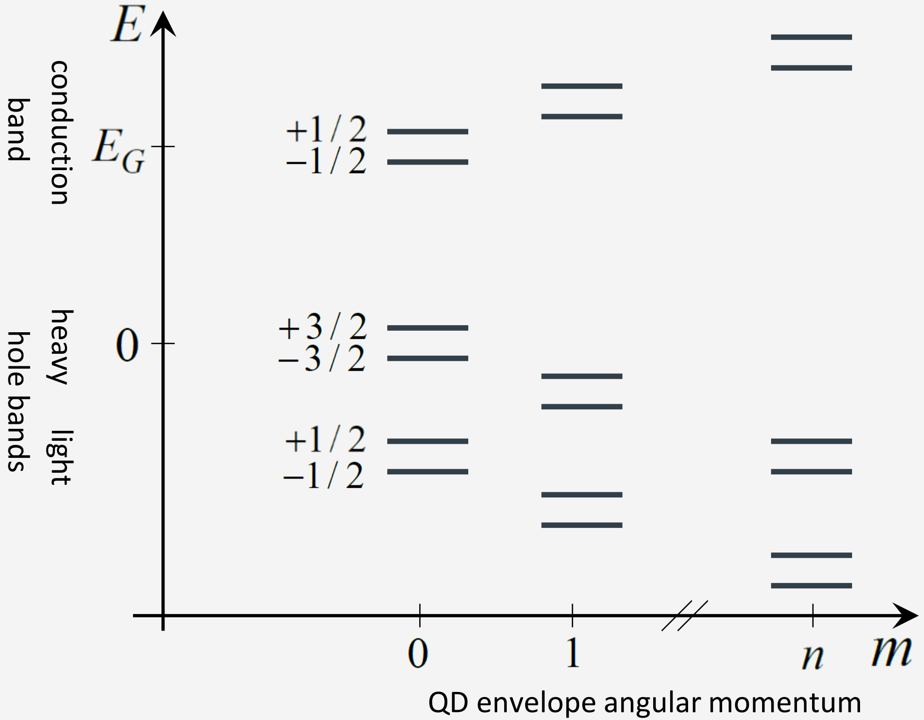
- optical excitation: $J_z = \pm 1$: $\sigma_\pm$ circularly pol. light; $J_z = 0$: π_z linearly pol. light

Quantum dots: complete level structure (idealized)

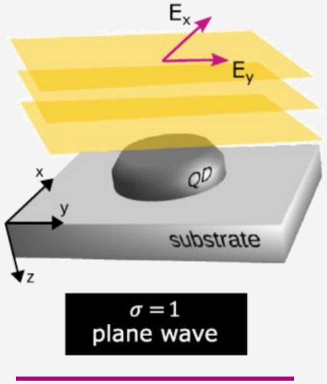
characterization of optical transitions (excitons) by:

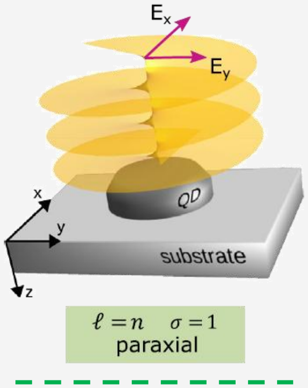
- intrinsic (band+spin) angular momentum in valence and conduction bands
- orbital angular momentum m of envelope function
- radial quantum number n_r of envelope function (typically conserved, since light beam diameter much larger than QD size)

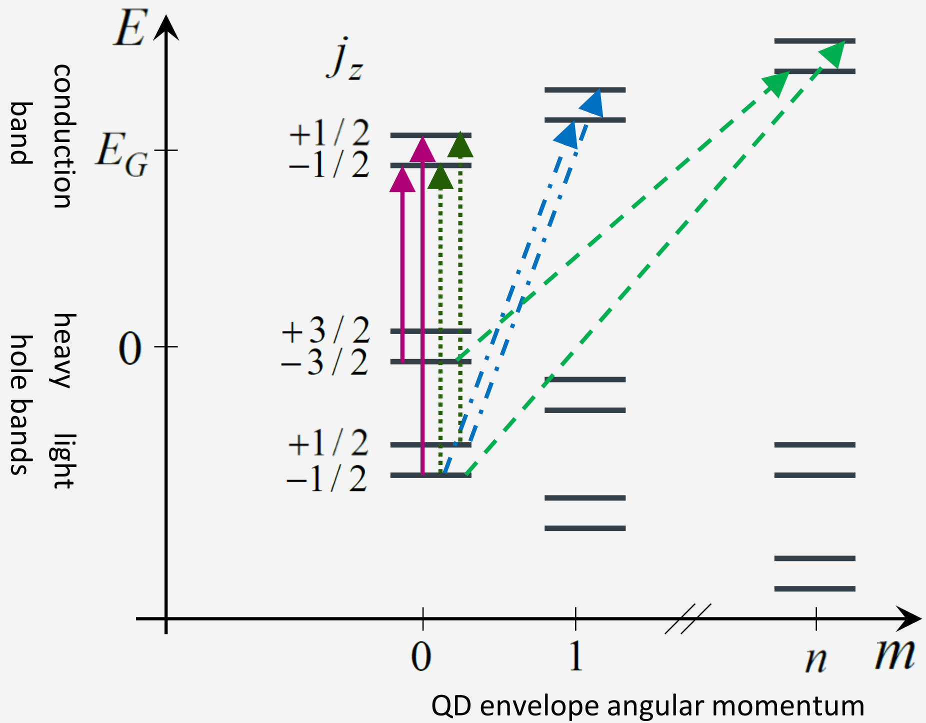
- here: intrinsic angular momentum in electron picture; for holes in valence band opposite signs

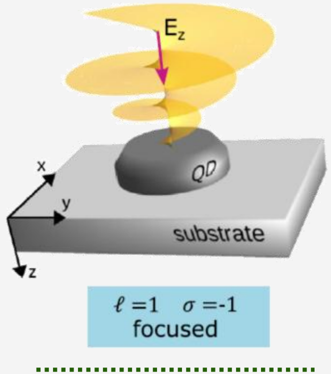


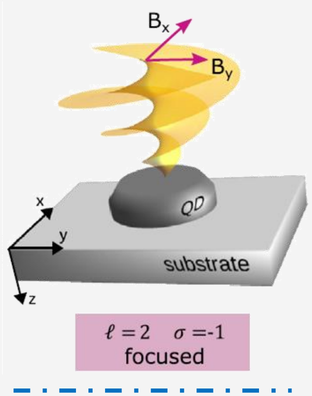
Optical transitions in a cylindrically symmetric QD







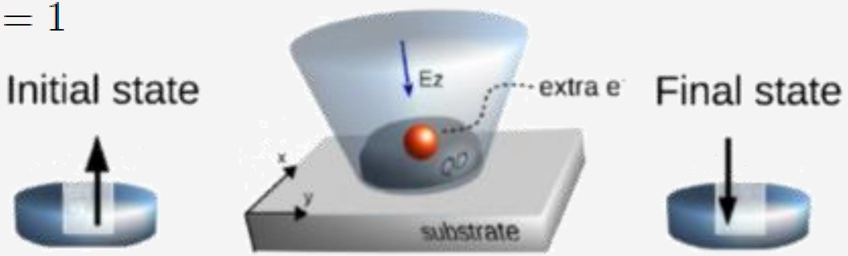


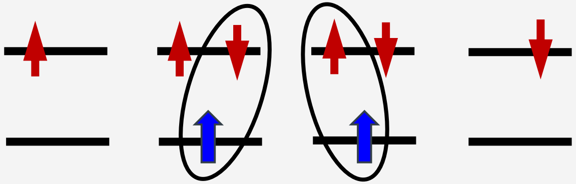


Control of the spin of an electron in a negatively charged QD

E. Pazy et al., Europhys. Lett. 62, 175 (2003)

- proposal by E. Pazy et al. (2003) for spin-based quantum computation (without structured light)
- goal: flip of the spin of an additional electron in the QD
- initial state: electron with $s_z = +\frac{1}{2}$
- creation of lh exciton with $J_z = 0$ by beam with $\ell = 1, \sigma = -1$
- removal of lh exciton with $J_z = +1$ by beam with $\ell = 0, \sigma = 1$
- final state: electron with $s_z = -\frac{1}{2}$
- without structured light: excitation from the side necessary
- requires cleaving of sample,
  difficult in particular for arrays of QDs





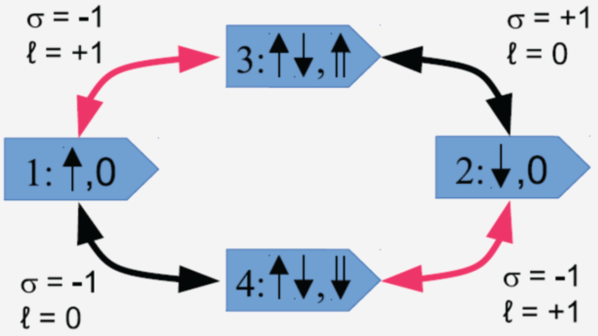
G.F. Quinteiro, T. Kuhn, Phys. Rev. B 90, 115401 (2014)

Universität Münster

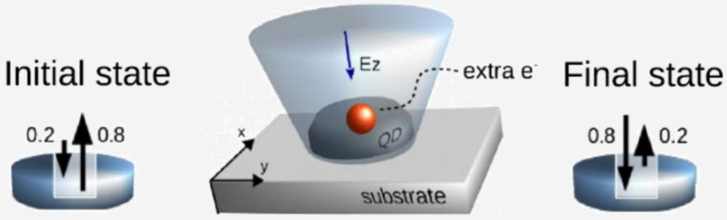
Control of the spin of an electron in a negatively charged QD

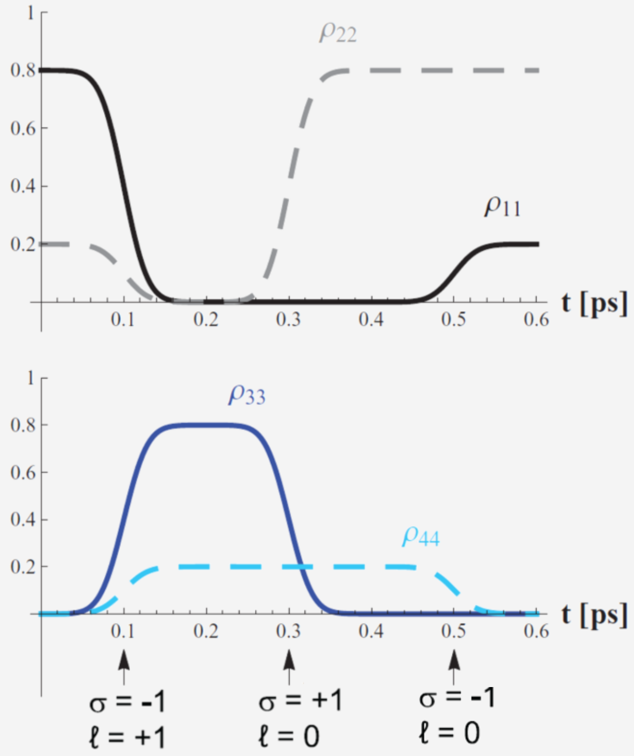
- extension to initial superposition states:

  three-pulse excitation



- inversion of initial state



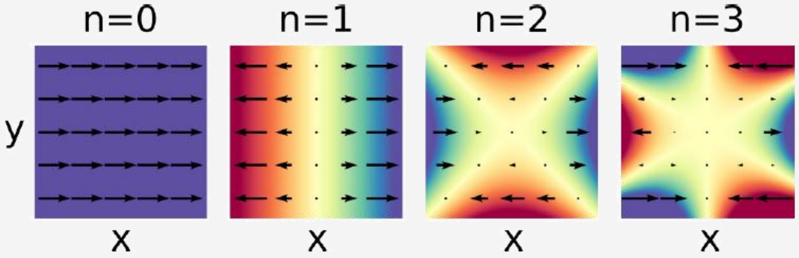


G.F. Quinteiro, T. Kuhn,
Phys. Rev. B 90, 115401 (2014)

Reconstruction of exciton wave functions by structured light

- reconstruction based on selection rules for envelope functions
- here: real wave functions, more realistic in case of small deviations from circular symmetry
- relevant property of light beam: nodal structure near beam center
- linearly polarized Bessel beam, approximation $J_n(q_r r) \sim (q_r r)^n$ (since $q_r^{-1} \gg L_{x/y}$)

$$\tilde{\boldsymbol{A}}_{n,\theta,\alpha}(\boldsymbol{r}) = A_0 \left(\frac{r}{R_{\text{QD}}} \right)^n \cos(n\varphi - \theta)\, \boldsymbol{e}_\alpha$$

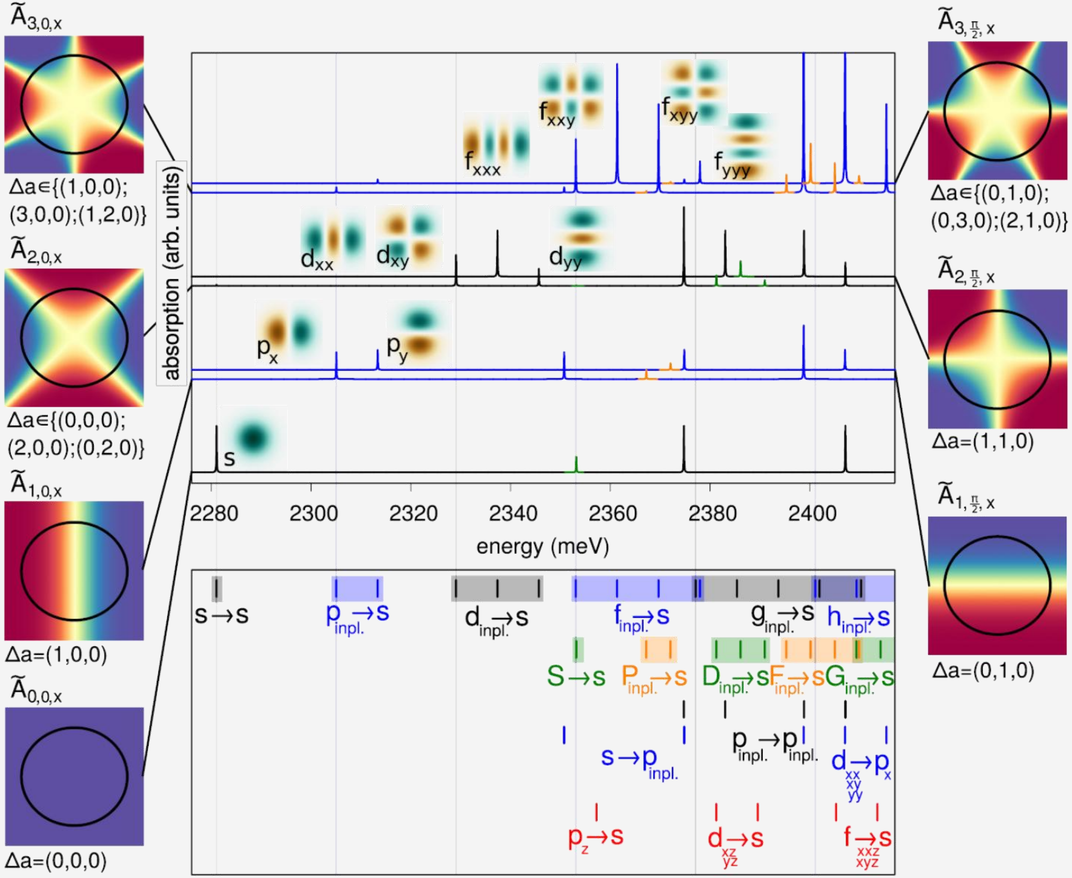


M. Holtkemper, G.F. Quinteiro, D.E. Reiter, T. Kuhn, Phys. Rev. B 102, 165315 (2020)

- model: slightly asymmetric QD, $5.8 \times 5.0 \times 2.0\ \text{nm}^3$
- conduction band, hh and lh bands, Coulomb interactions
- full configuration interaction (CI) calculation

Universität Münster

Reconstruction of exciton wave functions by structured light

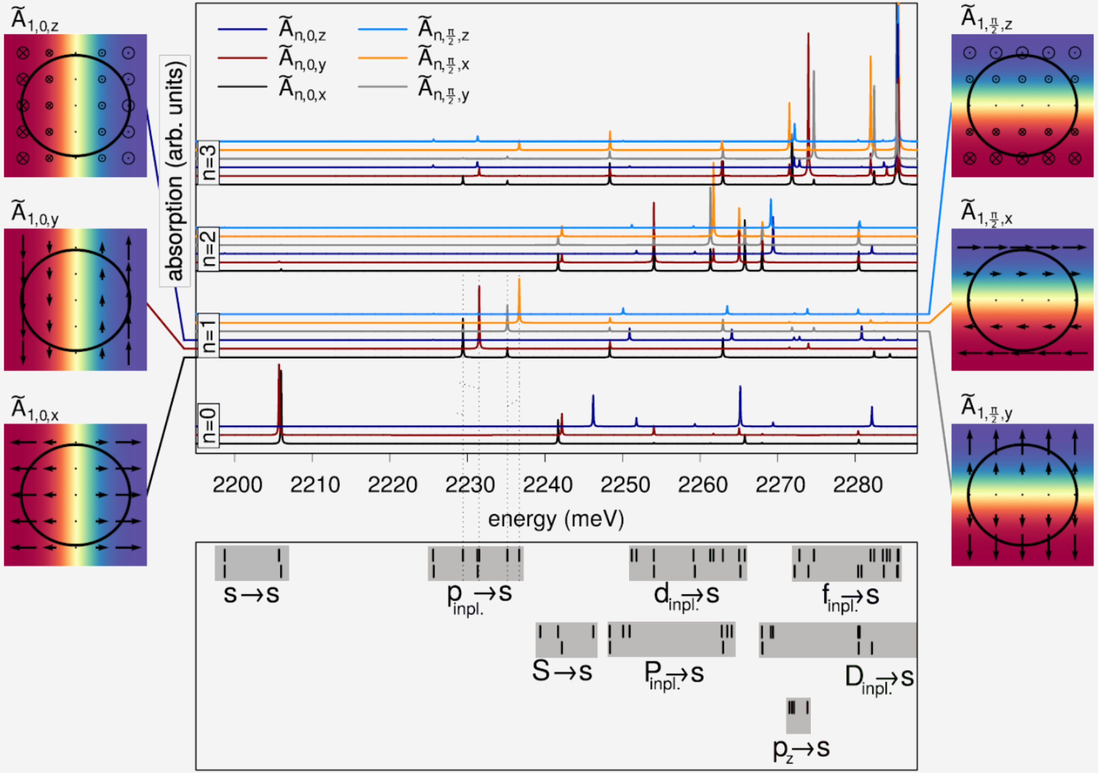


- simplified model, without Coulomb interaction and valence band mixing
- all possible excitonic states
- measurement of absorption e.g. by PLE
- absorption spectrum for $n=0$ (plane wave)
- absorption spectra for $n=1$
- absorption spectra for $n=2$
- absorption spectra for $n=3$

M. Holtkemper, G.F. Quinteiro, D.E. Reiter, T. Kuhn, Phys. Rev. B 102, 165315 (2020)

Universität Münster

Reconstruction of exciton wave functions by structured light



- complete model, including Coulomb inter-action and valence band mixing
- all possible excitonic states
- additional splittings due to exchange interaction → bright-dark splittings
- systematic study for field profiles
- absorption spectra for $n = 0$
- absorption spectra for $n = 1$
- absorption spectra for $n = 2$
- absorption spectra for $n = 3$

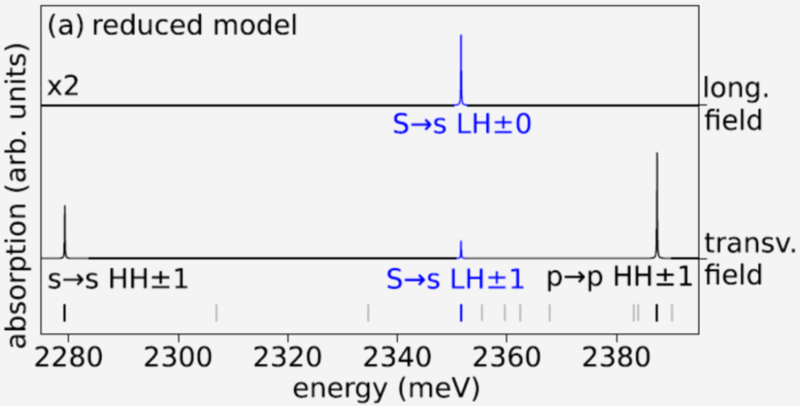
M. Holtkemper, G.F. Quinteiro, D.E. Reiter, T. Kuhn, Phys. Rev. B 102, 165315 (2020)

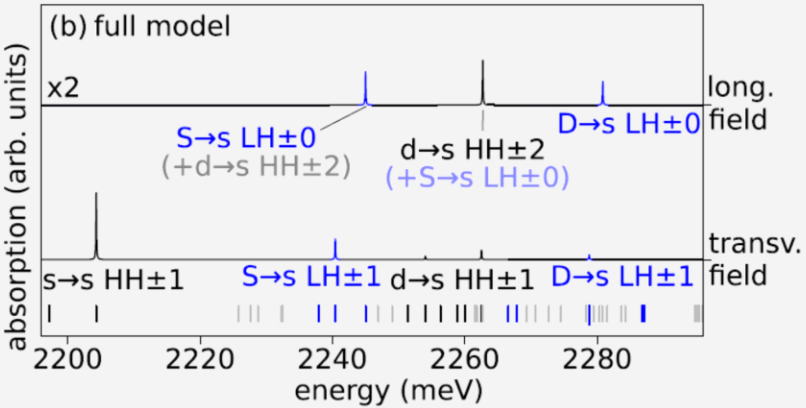
Dark exciton preparation by excitation with a longitudinal light field

- energetically lowest to excitons: HH±2, intrinsic angular momentum (spin) ±2
- cannot emit a photon, cannot relax to lower energy state: optically dark, long lifetime
- interesting for quantum applications: quantum memory
- due to missing light coupling difficult to excite

- possible schemes:
  - use in-plane magnetic field
    (S. Lüker, T. Kuhn, D.E. Reiter, Phys. Rev. B 92, 201305(R) (2015))
    disadvantage: magnetic field induced coupling also reduces lifetime
  - use excitation to higher states and subsequent relaxation
    take advantage of spin selection rules during relaxation
- here: excitation into states with strong band mixing
- making use of properties of vortex light

Dark exciton preparation by excitation with a longitudinal light field





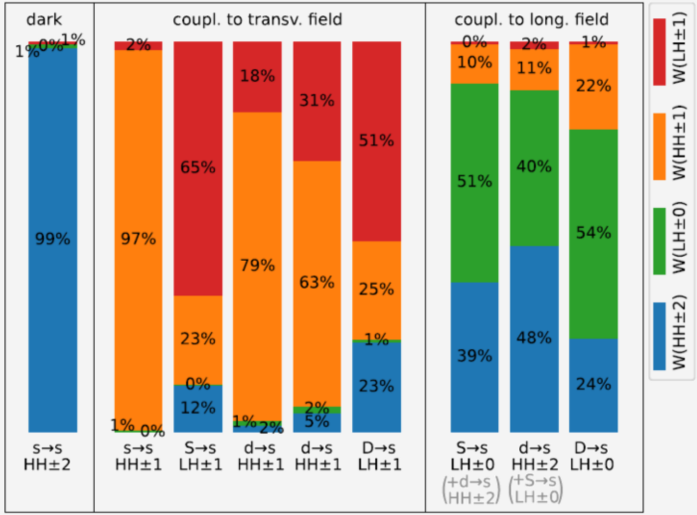
- absorption spectrum of reduced model, no Coulomb interaction and valence band mixing
- transverse field: excitation of excitons with spin ±1
- longitudinal field: excitation of excitons with spin 0

- absorption spectrum of complete model, with Coulomb interaction and valence band mixing
- additional lines due to band and spin mixing
- particularly pronounced mixing between HH±2 and LH±0

M. Holtkemper, G.F. Quinteiro, D.E. Reiter, T. Kuhn, Phys. Rev. Res. 3, 013024 (2021)

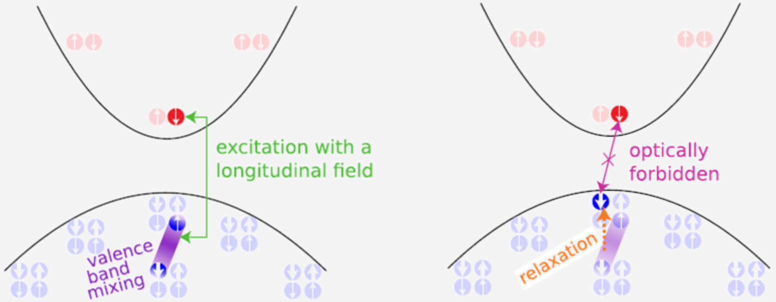
Dark exciton preparation by excitation with a longitudinal light field

- spin mixing of different exciton states:



- lowest two excitons (dark and bright) very pure spin states
- higher excitons typically strongly mixed
- dominant mixing either HH±1 - LH±1 or HH±2 - LH±0

- idea: excitation with longitudinal light field creates LH±0 excitons
- due to mixing contribution of higher HH±2 excitons

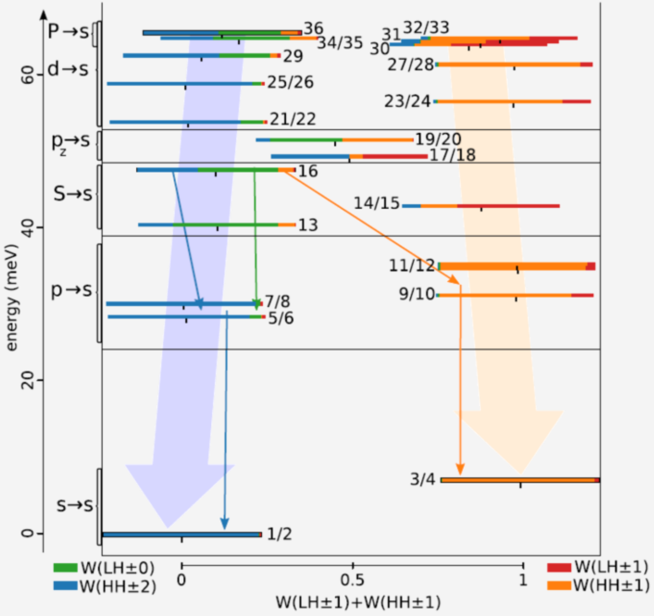


- spin-conserving relaxation into lowest HH±2 exciton

M. Holtkemper, G.F. Quinteiro, D.E. Reiter, T. Kuhn,
Phys. Rev. Res. 3, 013024 (2021)

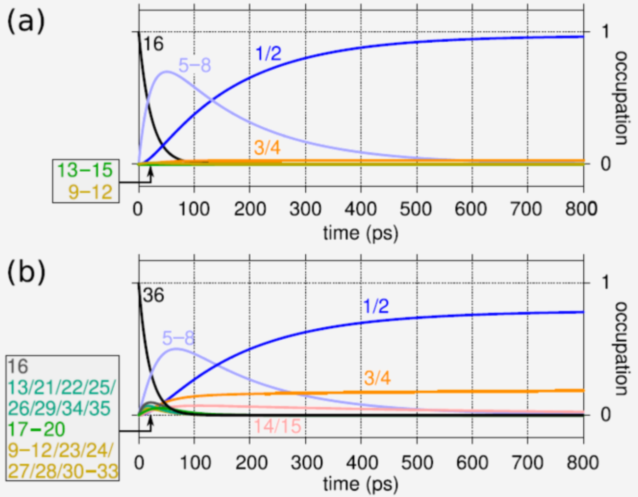
Dark exciton preparation by excitation with a longitudinal light field

- classification of exciton states according to their spin 1 contribution



- most states fall into one of the two classes
- a few states outside of this classification

- simulation of exciton relaxation due to coupling to acoustic and optical phonons
- results for two different initial states



- efficient population of dark exciton ground state

M. Holtkemper, G.F. Quinteiro, D.E. Reiter, T. Kuhn, Phys. Rev. Res. 3, 013024 (2021)

Conclusions

- vortex beams
  - paraxial and propagation-invariant beams: *Laguerre-Gauss and Bessel beams*
  - construction of vortex beams: *different assumptions on gauge leads to different beams*
  - properties of vortex beams: *parallel and antiparallel class*
- excitation of quantum dots by vortex beams
  - basic properties of quantum dots: *spin, band and envelope angular momentum*
  - selection rules for excitation by vortex beams: *specific transitions addressed by specific beams*
  - reconstruction of exciton states: *absorption spectra for different beams reflect structure of wave function*
  - dark exciton preparation using structured light: *use spin mixing of higher states and phonon relaxation*

Thank you for your attention!