Optical control of exciton and spin states in a quantum dot by excitation with twisted light

T Kuhn1, D E Reiter1,2 and G F Quinteiro3
1 Institut für Festkörpertheorie, Universität Münster, 48149 Münster, Germany
2 Imperial College London, South Kensington Campus, London SW7 2AZ, UK
3 Departamento de Física and IFIBA, Universidad de Buenos Aires, Buenos Aires, Argentina
E-mail: Tilmann.Kuhn@uni-muenster.de

Abstract. We discuss how the excitation of a quantum dot can benefit from the special properties of twisted light beams. First we address the question of the appropriate theoretical description of the light-matter coupling in this system, i.e., of the most appropriate gauge. Then we discuss the optical transitions induced by different types of twisted light beams showing that both the envelope and the spin state of the exciton can be well controlled.

1. Introduction
Highly inhomogeneous light beams carrying a phase singularity at the beam center – also called vortex beams or twisted light (TL) – attract much attention for their interesting effects in different areas of physics [1]. Theoretical studies in solid-state physics have predicted, for instance, that TL can induce electric currents in quantum rings [2] and new electronic transitions (forbidden for plane waves) in quantum dots (QDs) [3, 4]. In this contribution we will discuss how the excitation of a QD by different types of TL beams can be used to control both the envelope and the spin parts of the generated excitons.

2. Gauge-invariant formulation of the twisted-light–matter interaction
We study the excitation of a QD by a TL Bessel beam with the vector potential given in cylindrical coordinates $(r, \varphi, z)$ by

$$A^{(\ell,\sigma)}(r, t) = \frac{1}{2} A_0 \left[ J_{\ell}(q_r r) e_r + i \sigma J_{\ell}(q_r r) e_\varphi - i \sigma q_z J_{\ell+\sigma}(q_r r) e_z \right] e^{i(q_z-s-\omega t)+i(\ell+\sigma)\varphi} + \text{c.c.} \tag{1}$$

Here, $A_0$ is the amplitude, $\omega$ the frequency, $q_r$ and $q_z$ the wave vectors in longitudinal and radial direction, respectively, and $e_r, e_\varphi, e_z$ are unit vectors. $J_\ell$ is a Bessel function with the integer $\ell$ characterizing the topological charge of the beam, $\sigma = \pm 1$ describes the handedness of the circular polarization (CP), and c.c. denotes the complex conjugate. Equation (1) is an exact solution of the vectorial Helmholtz equation and it satisfies the Coulomb and radiation gauge, i.e., $\text{div} A(r, t) = 0$ and $U(r, t) = 0$, $U$ being the scalar potential. For beams with large beam waist, i.e., $(q_r/q_z) \ll 1$, the $z$-component can often be neglected, which corresponds to the paraxial approximation. Here we will consider tightly focused beams, in which $q_r \approx q_z$, and therefore keep the full vector potential. For simplicity, we will refer to $\ell$ as the orbital angular momentum (OAM), although this strictly holds only in the paraxial limit.
The general form of the light-matter interaction is given in terms of a Hamiltonian \( \sim \mathbf{p} \cdot \mathbf{A}(\mathbf{r}, t) \) with the momentum operator \( \mathbf{p} \). This form has the drawback that it depends on the gauge. Although the results are of course gauge independent as long as exact calculations are being performed, this does not hold anymore if approximations are made. The light-matter coupling in QDs is typically treated in terms of the dipole coupling by the Hamiltonian \(-\mathbf{d} \cdot \mathbf{E}(\mathbf{r}_0, t)\), where \( \mathbf{d} \) is the dipole operator and \( \mathbf{E}(\mathbf{r}_0, t) \) is the electric field at the position of the QD. Being described in terms of the electric field, it is obviously gauge invariant. However, if the QD is placed close to the center of a TL beam – which is necessary to benefit from the peculiarities of this beam – the field is strongly inhomogeneous over the QD and it even often vanishes directly at the center, such that this form cannot be used. We have recently shown [5] that a gauge (the TL gauge) exists where the coupling can still be written in terms of the electric field, however, in a form different from the dipole coupling. For a TL beam with given \( \ell \) and \( \sigma \) the coupling Hamiltonian in the TL gauge then reads

\[
H_{\text{TL-matter}} = \frac{1}{|\ell| + 1} e r E_r^{(\ell, \sigma)}(\mathbf{r}, t) + \frac{1}{|\ell + \sigma| + 1} e z E_z^{(\ell, \sigma)}(\mathbf{r}, t)
\]  

(2)

with \( E^{(\ell, \sigma)} = -\frac{\partial \mathbf{A}^{(\ell, \sigma)}}{\partial t} \) and \( e \) being the elementary charge. In [5] we have shown that this is always a good approximation for beams in which \( \text{sign}(\ell) = \text{sign}(\sigma) \), which we call the parallel class. In the anti-parallel class \( \text{sign}(\ell) \neq \text{sign}(\sigma) \) in general the magnetic terms cannot be neglected. This different behavior can be traced back to the electric field profiles, which are shown in Fig. 1 for the example of TL beams with \( \ell = 1 \) and \( \sigma = \pm 1 \) traveling along the \( z \)-axis. In both cases the in-plane field is described by the function \( J_1 \) and therefore has a vortex at the beam center. The in-plane field of the parallel class [Fig. 1(c)] can be well approximated by a gradient of a scalar potential, which is obviously not true for the field profile in the anti-parallel case [Fig. 1(a)]. However, while in the parallel class the electric field close to the beam center is dominated by the in-plane part (the \( z \)-component involves the function \( J_0 \)), in the anti-parallel class the \( z \)-component (involving the function \( J_0 \)) dominates [Fig. 1(b)]. It is essentially homogeneous in that region and therefore can also be written as a gradient field. Hence, if there is a coupling to the \( z \)-component, this usually dominates and the TL gauge is valid also here.

3. Twisted-light–induced transitions in quantum dots

We will analyze the transitions that can be induced by exciting a QD, which is placed close to the beam axis, by different types of TL beams. We assume that the lateral size of the QD is smaller than \( q_-^{-1} \) and the height is smaller than \( q_z^{-1} \), such that the lowest orders in the expansions in
the VB Bloch functions with total angular momentum $(H\ell)$ states with angular momentum band (CB) and valence band (CB). Typically the CB Bloch functions are of a product of an envelope function for the case of excitation by the in-plane electric field of a TL beam (a) with $\ell = 0$ and (b) with $\ell = \pm 1$. The labels denote the initial (VB) and final (CB) shell of the transition.

$(q_x r)$ and $(q_z z)$ are sufficient. The electron and hole wave functions in a QD can be written as a product of an envelope function $\phi^{e,h}(r)$ and the lattice-periodic Bloch functions of conduction band (CB) and valence band (CB). Typically the CB Bloch functions are of $s$-type with total angular momentum $J^c = \frac{3}{2}$ and $J^c_s = \pm \frac{1}{2}$, i.e., $|1/2, +1/2\rangle = |s \uparrow\rangle$ and $|1/2, -1/2\rangle = |s \downarrow\rangle$, while the VB Bloch functions with total angular momentum $J^h = \frac{3}{2}$ can be separated into heavy hole (HH) states with $J^h_z = \pm 3/2$ and light hole (LH) states with $J^h_z = \pm 1/2$ given by

$$
\begin{align*}
|3/2, +3/2\rangle &= 2^{-1/2} (|p_x + ip_y \rangle \uparrow), \quad |3/2, +1/2\rangle = 6^{-1/2} (|p_x + ip_y \rangle \downarrow - 2|p_z \rangle \uparrow), \\
|3/2, -3/2\rangle &= 2^{-1/2} (|p_x - ip_y \rangle \downarrow), \quad |3/2, -1/2\rangle = 6^{-1/2} (-|p_x - ip_y \rangle \uparrow - 2|p_z \rangle \downarrow). 
\end{align*}
$$

For the calculation of the TL-induced transition rates – and thus of the absorption spectrum – the matrix elements of the interaction Hamiltonian (2) between VB and CB states are required. Since the electric field varies slowly over an elementary cell and the Bloch functions of CB and VB have opposite parity, the transition matrix elements can be separated into a product of the bulk dipole matrix element $d_{ev}$ and envelope integrals $G^{eh}$ with

$$
d_{ev} = \langle s|x|p_x \rangle = \langle s|y|p_y \rangle = \langle s|z|p_z \rangle, \quad G^{eh} = \int \phi^{e*}(r)E(r)\phi^{h}(r)d^3r. \tag{4}
$$

We model the QD by harmonic confinement potentials for electrons and holes, where we assume that the confinement in $z$-direction (growth direction) is much stronger than in $x$- and $y$-direction. The energy levels of electrons and holes are then grouped into shells denoted by $s, p, d, f \ldots$ We assume a slight in-plane asymmetry, such that the states in higher shells are split, and characterize the transition by the initial (VB) and final (CB) shell.

Let us start by considering a beam with $\ell = 0$ and $\sigma = \pm 1$. In such a beam the in-plane components of the field dominate. Its radial dependence is given by $J_0(q_x r)$ and is therefore essentially homogeneous over the QD. Hence the transitions induced by this beam are the same as for plane waves. Both in the case of transitions from the HH and the LH band excitons with $J_z = J^c_z + J^h_z = \pm 1$ are excited. A qualitative absorption spectrum is plotted in Fig. 2(a). We find strong peaks when the VB and CB shells agree and weaker peaks when the the VB and CB shell index differs by two (in general by an even number). The presence of these peaks is due to a slight difference between electron and hole wave functions of the same shell. Similar spectra have been obtained with more refined exciton models and are also seen in experiments [6].

As the first genuine TL beam we consider a beam with $\ell = \pm 1$ and $\sigma = \ell$, i.e., from the parallel class. Also here the in-plane components dominate; however, now their radial dependence is...
given by $J_1(qr)$, which in the region of the dot can be approximated by $(qr)^r$, and the angular dependence is $e^{\pm i\varphi}$. At the beam center there is a vortex with vanishing electric and magnetic field. The electric field has odd parity, such that only transitions between shells differing by one (in general by an odd number) can be excited. Such a spectrum is qualitatively shown in Fig. 2(b). Since only in-plane dipole matrix elements are involved, the spectrum is again qualitatively the same for HH and LH excitons. Thus we find that with this kind of TL beams excitons which are usually dark due the symmetry of their envelope functions can be generated.

Finally we consider a beam with again $\ell = \pm 1$ but now $\sigma = -\ell$, i.e., a beam belonging to the anti-parallel class. In such a beam the radial dependence of the in-plane components is $\sim J_1(qr)$, while that of the z-component is $\sim J_0(qr)$. Therefore the latter one is much larger and, as soon as the dipole matrix element has a non-vanishing z-component, this will give rise to the dominant transition. While for HH excitons there are no such transitions, LH excitons with angular momentum $J_z = 0$ can be excited [see Eq. (3)]. Since the integrals over the envelope functions are the same as in the case of the beam with $\ell = 0$, the spectrum will again look similar to Fig. 2(a). We thus find that this kind of TL beam can be used to generate excitons with the same envelope structure as in the case of plane waves, but with different spin configuration.

4. Discussion and conclusions

We have shown that TL beams, due to their more complex spatial structure compared to plane waves, provide additional flexibility which can be used to excite specific transitions in QDs: (i) beams with dominant in-plane components and $\ell \neq \pm 1$ can excite transitions from the $p$- to the $s$-shell and vice versa; (ii) beams with dominant $z$-component can excite LH excitons with $J_z = 0$. Let us comment in some more detail on this latter aspect.

For different applications in quantum information processing using the switching either of the electron spin in a charged QD [7] or of the spin of a magnetic impurity in a QD [8] LH excitons have been shown to be useful or even necessary for the desired operation. This is due to the possibility to optically address excitons with $J_z = 0$. With plane waves this requires cleaving the sample and exciting from the side, in addition to an excitation from the top, which is still necessary to address excitons with $J_z = \pm 1$. TL therefore provides the big advantage of addressing all transitions with co-propagating beams. QD structures, in which the lowest exciton transition is of LH type, have indeed recently been fabricated [9] using a suitable strain engineering. Therefore the combination of these structures with TL excitation opens up new ways for spin control phenomena in QD systems.

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References

[1] Andrews D L 2008 Structured light and its applications: An introduction to phase-structured beams and nanoscale optical forces (New York: Academic Press)
[2] Quinteiro G F and Berakdar J 2009 Optics Express 17 20465
[3] Quinteiro G F and Tamborenea P I 2009 Phys. Rev. B 79 155450
[4] Quinteiro G F and Kuhn T 2014 Phys. Rev. B 90 115401
[5] Quinteiro G F, Reiter D E and Kuhn T 2015 Phys. Rev. A 91 033808
[6] Huneke J et al. 2011 Phys. Rev. B 84 115320
[7] Pazy E et al. 2003 Europhys. Lett. 62 175
[8] Reiter D E, Kuhn T and Axt V M 2011 Phys. Rev. B 83 155322
[9] Huo Y H et al. 2014 Nat. Phys. 10 46