Magnetic-optical transitions induced by twisted light in quantum dots

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Abstract. It has been theoretically predicted that light carrying orbital angular momentum, or twisted light, can be tuned to have a strong magnetic-field component at optical frequencies. We here consider the interaction of these peculiar fields with a semiconductor quantum dot and show that the magnetic interaction results in new types of optical transitions. In particular, a single pulse of such twisted light can drive light-hole-to-conduction band transitions that are cumbersome to produce using conventional Gaussian beams or even twisted light with dominant electric fields.

Introduction
Twisted light (TL) is light having a helical wave front. Such light exhibits several interesting features: Due to the azimuthal phase dependence a phase singularity occurs at the beam axis, leading to the name optical vortex. Furthermore, in addition to spin angular momentum (SAM) associated with the handedness of circular polarization, such light fields carry orbital angular momentum (OAM). Depending on the combination of SAM and OAM, TL beams may have strong field components along the propagation direction or strong magnetic field close to the beam axis. The research in TL spans nowadays several areas of fundamental and applied physics [1] including its potential for quantum communication [2].

Of particular interest is the interaction of a TL beam with matter, which opens up the possibility to address unusual transitions [3,4] and to excite typically dark modes in plasmonics [5]. In this paper we focus on the interaction of TL with a semiconductor quantum dot (QD). QDs are discussed for many applications in optoelectronics and spintronics [6] which, however, requires a precise control of the optically excited states. Here we will show that the interaction of a QD with TL having a strong magnetic component may be used for the excitation of specific light-hole (LH) excitons, which cannot easily be addressed by conventional Gaussian beams or even twisted light with dominant electric fields.

Twisted light with a strong magnetic component
TL can be characterized by the handedness of its circular polarization $\sigma$ (SAM) and its OAM quantum number $\ell$. We recently identified two distinct classes of TL [3,4], that we named parallel and antiparallel class reflecting the relative orientation of SAM and OAM. Figure 1 depicts the electric field profiles for a beam with $\ell = 2$ and $\sigma = \pm 1$ showing the pronounced difference in the topology of the field lines.
Starting from the vector potential for a Bessel beam with given values of $\ell$ and $\sigma$ propagating in $z$-direction, which is an exact solution of the vectorial Helmholtz equation, the behavior of the electric and magnetic field in a region close to the beam axis has been derived in \[3\]. Remarkably, it has been found that especially the antiparallel class is characterized by unconventional features which become particularly strong in the case of tightly focused beams: (i) The electric field close to the beam axis is dominated by its longitudinal component; (ii) for $|\ell| \geq 2$ the beam in that region is dominated by the magnetic field. The virtue of employing the longitudinal electric field component of antiparallel beams with $|\ell| = 1$ for the excitation of LH excitons has already been discussed in \[3\]. Here we complement this work by showing that the use of the dominant magnetic field present in an antiparallel beam with $|\ell| = 2$ again opens up new excitation pathways which are usually unaccessible.

Close to the singularity the field amplitudes read (see Table 1 in \[3\])

$$
\tilde{E}_x(r_\perp) = i \frac{E_0}{8} (q_r r)^2 e^{i2\varphi}, \\
\tilde{E}_y(r_\perp) = \frac{E_0}{8} (q_r r)^2 e^{i2\varphi}, \\
\tilde{E}_z(r_\perp) = -i \frac{E_0}{2} q_r (q_r r)^2 e^{i\varphi}, \\
\tilde{B}_x(r_\perp) = -i \frac{B_0}{2} q_r (q_r r)^2 e^{i\varphi}, \\
\tilde{B}_y(r_\perp) = -i \frac{B_0}{2} q_r (q_r r)^2 e^{i\varphi}.
$$

Here, $q_r$ and $q_z$ are the transverse and longitudinal wave vector, where $q_r$ is inversely proportional to the beam waist, $r_\perp$ is the in-plane coordinate with $|r_\perp| = r$ and azimuthal angle $\varphi$. We will consider the excitation by tightly focused TL, i.e., beams with the paraxial parameter $q_r/q_z \approx 1$. $E_0$ and $B_0 = E_0/c$ are the respective amplitudes.

When approaching the beam center, i.e., for $r \to 0$, we find that the electric field $E$ and the longitudinal magnetic field $B_z$ vanish while the transverse magnetic field $B_\perp$ remains finite. Therefore, the transverse magnetic field dominates for interaction with particles or nanostructures, which are much smaller than the beam waist and are placed at or close to $r = 0$, which is in fact a surprising effect at optical frequencies.

Recently, we have developed the theory of light-nanostructure interaction for the electric and magnetic components of general antiparallel TL beams \[3,4\]. It turned out that the interaction can be written in a form similar to the well-known dipole approximation for electric and magnetic fields \[4\]. In particular, for TL with $\ell = 2$ and $\sigma = -1$, in which the magnetic field dominates, the Hamiltonian can be written as \[4\]

$$
H_I = -\frac{q}{2m} \mathbf{B}_\perp(t) \cdot (\mathbf{r} \times \mathbf{p}) = -i \frac{qB_0}{m} (r_+ p_z - z p_+) e^{-i\omega t} + \text{c.c.}
$$

(1)

with $q$ and $m$ being the electron charge and mass, respectively, $\omega$ is the frequency of the light, $r_\pm = x \pm iy$, $p_\pm = p_x \pm ip_y$, and c.c. denoting the complex conjugate.

**Figure 1.** Electric field patterns for the antiparallel (left) and parallel (right) class for an orbital angular momentum of $\ell = 2$. 
Quantum dot light-hole states

We consider a semiconductor QD, where the conduction band states are $s$-type ($|s\rangle$) and can be classified by the electron spin given by the quantum number $s_z = \pm 1/2$. For the valence band states ($|p_x\rangle, |p_y\rangle, |p_z\rangle$), due to their $p$-type character, one finds states with total orbital angular momenta $j = 3/2$ and $j = 1/2$, where the $j = 3/2$ states are split into heavy holes with $j_z = \pm 3/2$ and LHs with $j_z = \pm 1/2$. Here, we focus in the LH states, such that the Bloch function for electrons and holes read:

\[
|s_z = +1/2\rangle = |s\rangle |\uparrow\rangle \quad |j_z = +1/2\rangle = \frac{1}{\sqrt{6}} \left( |p_x\rangle - i |p_y\rangle \right) |\uparrow\rangle + 2 |p_z\rangle |\downarrow\rangle \\
|s_z = -1/2\rangle = |s\rangle |\downarrow\rangle \quad |j_z = -1/2\rangle = -\frac{1}{\sqrt{6}} \left( |p_x\rangle + i |p_y\rangle \right) |\downarrow\rangle - 2 |p_z\rangle |\uparrow\rangle
\]

where the arrow indicates the electron spin. Note that the sign of $j_z$ refers to the angular momentum of the holes which is opposite to that of valence band electrons.

To describe the full wave function of electrons and holes in the QD, we further apply the envelope function approximation. Within this scheme the Bloch functions are multiplied by the respective envelope functions for electrons and holes,

\[
F_{n,m}^{e/h}(r) = R_{n,m}^{e/h}(r) e^{im\varphi} Z^{e/h}(z) ,
\]

which have been further separated into the radial part $R_{n,m}^{e/h}(r)$, an envelope angular momentum part $e^{im\varphi}$, and a part $Z^{e/h}(z)$ in $z$-direction. Assuming a flat QD, we restrict ourselves to the ground state in $z$-direction assuming wave functions $Z^{e/h}$ with well-defined parity.

The interaction of twisted light with light holes

To calculate the matrix elements between LH valence and conduction band states we follow the standard procedure in the envelope-function formalism: We split the integration into a sum over all lattice vectors $\mathbf{R}$ and an integral over the unit cell indicated by the coordinate $\mathbf{r}'$. Finally, we replace the sum over lattice vectors by an integral over the whole system.

Following this procedure, in the Hamiltonian (1) the products of coordinates and momenta separate into terms such as $(R_z' + r_z' + R_z + r_z)(P_z + p_z)$. Keeping in mind the orthogonality and parity of the Bloch functions under study, one finds that all matrix elements involving only intracell coordinates $\mathbf{r}'$ or only envelope coordinates $\mathbf{R}$ vanish. Furthermore, the integral over the envelope functions including either $Z$ or $P_z$ vanish, i.e., \( \int dZ Z^{e}(Z) Z^{h}(Z) = \int dZ Z^{e}(Z) P_z Z^{h}(Z) = 0 \), due to the same parity of the ground state functions $Z^{e/h}(z)$. The only remaining matrix element then reads

\[
\langle n', m'; s_z | H_I | n, m; j_z \rangle = -i \frac{qB_0}{m} \int d^3 R F_{n',m'}^{e*}(\mathbf{R}) \left[ R_z + M_{p_z} - P_z M_z \right] F_{n,m}^{h}(\mathbf{R}) \delta_{s_z, -j_z} ,
\]

where we have introduced the microscopic matrix elements

\[
M_z = \langle s_z = \pm 1/2 | z' \rangle \quad M_{p_z} = \langle s_z = \pm 1/2 | p_z' \rangle
\]

In Eq. (3), we find that due to the optical selection rules only electron-hole pairs having opposite angular momenta of the Bloch state are excited, because only the matrix element $M_z$ and $M_{p_z}$ survive. In other words, only excitons with a total angular momentum of zero (i.e. $J_z = s_z + j_z = 0$) are excited. This is very different compared to the excitation with plane waves. Plane waves have an angular momentum of $\pm 1$ and thereby excite excitons with a total angular
momentum $J_z = \pm 1$. To excite LH excitons in a QD with $J_z = 0$, one would have to apply an excitation from the side of the QD, which typically requires cleaving the sample.

We can now evaluate the macroscopic integrals over $R$ for the two different parts. For the first term, we rewrite $R_+ = R e^{i\Phi}$. For the second term we write the momentum operator in cylindrical coordinates $P_+ = -i\hbar e^{i\Phi} \left[ \partial_R + (i/R) \partial_{\Phi} \right]$. Thus, both terms contribute a term $\sim e^{i\Phi}$ to the angle integral. This shows, that by exciting the QD with a magnetic TL beam having $\ell = 2$ and $\sigma = -1$, the envelope state changes by $\Delta m = 1$ while the total band+spin angular momentum of the exciton is zero. Such a transition is forbidden for excitation with a plane wave due to dipole selection rules, and it is also not present in the case of excitation by a TL beam with dominant electric field in $z$-direction (i.e., an antiparallel TL beam with $|\ell| = 1$).

Figure 2. Optical transition between light-hole and conduction band state in a QD induced by single pulses of twisted light having $\ell = 2$ and $\sigma = -1$. For comparison also transition due to beams with $\ell = 0$ and $\ell = 1$ are included. Note that for clarity reasons holes states with $m = \pm 1$ have been removed.

Figure 2 combines our new results for a TL beam with $\ell = 2$ and $\sigma = -1$ with previous results reported in Ref. [7] for beams with $\ell = 0$ and $\ell = 1$. Note that for reasons of clarity the Figure shows only the lowest hole shell with $m = 0$. The diagonal transitions with $\Delta m = 0$ and $\Delta m = -1$ induced correspondingly by TL beams with $(\sigma = -1, \ell = 0)$ and $(\sigma = -1, \ell = -1)$ are not depicted. We thus find, that by using TL beams with $|\ell| \leq 2$ all possible exciton states in the lowest two shells with $m = 0$ and $m = \pm 1$ can be excited.

Conclusions

We have demonstrated that the optical-magnetic interaction in case of excitation with a TL beam with OAM $\ell = 2$ and SAM $\sigma = -1$ generates LH exciton states with no band+spin angular momentum ($J_z = 0$) but with envelope angular momentum $m = 1$, and thus produces states that are not accessible with plane waves or even TL with dominant electric interaction. Combining these findings with previous results for beams with other values of OAM and SAM, we find that TL beams are beneficial to obtain a full control of exciton states in a QD.

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