

Insensitivity of spin dynamics to the orbital angular momentum transferred from twisted light to extended semiconductors

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We study the spin dynamics of carriers due to the Rashba interaction in semiconductor quantum disks and wells after excitation with light with orbital angular momentum. We find that although twisted light transfers orbital angular momentum to the excited carriers and the Rashba interaction conserves their total angular momentum, the resulting electronic spin dynamics is essentially the same for excitation with light with orbital angular momentum $l = +|l|$ and $l = -|l|$. The differences between cases with different values of $|l|$ are due to the excitation of states with slightly different energies and not to the different angular momenta per se, and vanish for samples with large radii where a k -space quasi-continuum limit can be established. These findings apply not only to the Rashba interaction but also to all other envelope-function approximation spin-orbit Hamiltonians like the Dresselhaus coupling.

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Light with orbital angular momentum (OAM), referred to as twisted light, is a relatively new field of research which has become increasingly popular [1–17] since Allen *et al.* showed how twisted light beams can be easily generated from conventional laser beams [18]. Recently, the theoretical foundation of the optical excitation of solids and nanostructures with twisted light has been established [19–27], and experimental studies with twisted light on semiconductors have been carried out [28, 29].

One motivation for such studies is the prospect of using the large amounts of angular momentum that twisted light can carry in order to control the spin dynamics of electrons, thus adding a flexible tool to the active field of spin control [30–39]. In this context two different mechanisms need to be distinguished. First, angular momentum as well as energy selection rules can lead to selective optical excitation of carriers with a preferred spin direction. This mechanism enables fast spin-selective preparation of states during the photoexcitation process and has recently been studied for strongly confined systems such as quantum dots [27] and quantum rings [23]. Secondly, the spin-orbit interaction—like the Rashba [40] and Dresselhaus [41] couplings in semiconductor structures—is expected to couple the OAM of carriers transferred from the twisted light [19, 22] to their spin degree of freedom. This would provide a slower carrier spin control which would be dynamical and would remain active after the twisted light pulse.

In this Letter, we study the spin dynamics of carriers in semiconductor quantum disks and wells excited with twisted light taking into account the Rashba spin-orbit interaction. Our central finding is that, rather unexpectedly, the spin dynamics of the photo-excited electrons differs only slightly after excitation with light with and without OAM in the limit of large quantum disks, becoming insensitive to the OAM content of the twisted

light beam for extended quantum wells. This result is consistent with the outcome of recent experiments which did not show traces of the OAM transferred from twisted light to bulk GaAs in spin-resolved photoemission measurements [29].

Analytically, we find that the Rashba interaction, while conserving the total angular momentum of the electrons, has matrix elements which are independent of the OAM quantum number in the (k -space) quasi-continuum limit. As a consequence, the induced spin dynamics is almost identical, in particular, for twisted light with components of the OAM in the growth direction $l = +|l|$ and $l = -|l|$. This finding can be generalized to all possible effective spin-orbit interactions stemming from a lattice-periodic potential in the envelope-function approximation, e. g. the Dresselhaus coupling. Our results suggest that for the search for materials supporting twisted-light based spin control it is most promising to concentrate on small quantum structures and other discrete systems.

The discussion of the optical excitation of electrons with twisted light is especially clear when a basis of cylindrical states is chosen [22]. The wave functions of these basis states are expressed in cylindrical coordinates $\{r, \phi, z\}$ as

$$\psi_{bm\nu}(r, \phi, z) = \mathcal{N}_{m\nu} J_m(k_{m\nu}r) e^{im\phi} \Phi_b(z), \quad (1)$$

where J_m is the m -th Bessel function, $\Phi_b(z)$ is the z -envelope of the b subband (the band index includes the spin quantum number) and $\mathcal{N}_{m\nu} = [\sqrt{\pi}R J_{m+1}(k_{m\nu}R)]^{-1}$ is the normalization factor. For a circular quantum disk with radius R , width L and growth direction z , the boundary conditions $\psi_{bm\nu}(R, \phi, z) = 0$ are satisfied, if $k_{m\nu} = u_{m,\nu}/R$ where $u_{m,\nu}$ is the ν -th zero of the m -th Bessel function. Note that $\psi_{bm\nu}$ is an eigenstate of the z -component of the envelope OAM operator with eigenvalue $\hbar m$ and $k_{m\nu}$ determines the kinetic

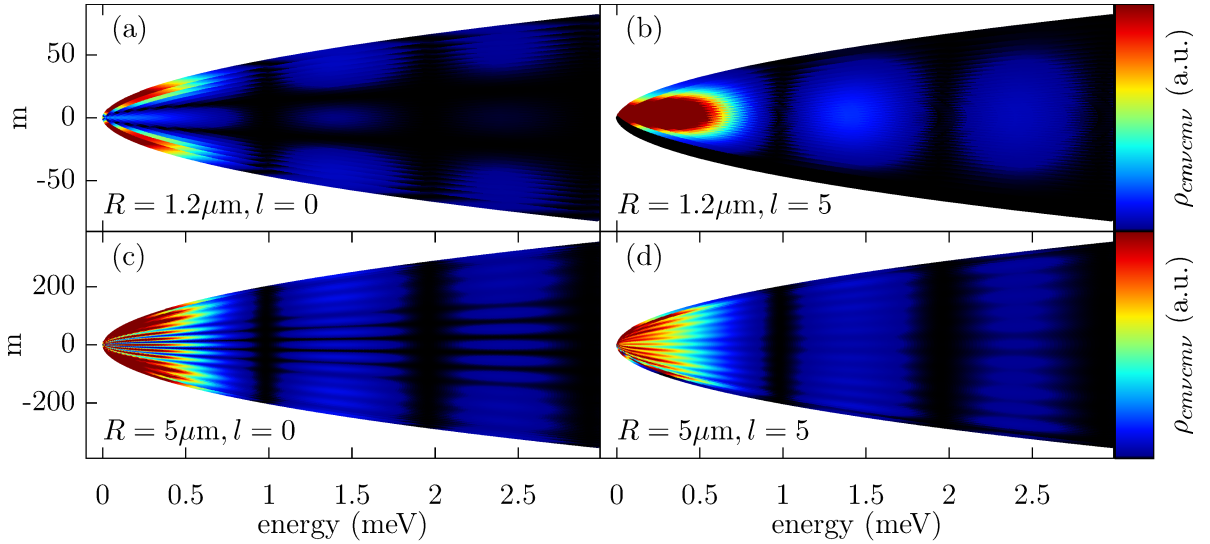


FIG. 1. Diagonal elements of the density matrix $\rho_{cm\nu cm\nu}$ after an excitation of a cylindrical quantum disk with radius R with a box-shaped pulse of length $t = 5$ ps and orbital angular momentum l of the light. (a) and (b) show the occupations of a $R = 1.2 \mu\text{m}$ quantum disk with $l = 0$ and $l = 5$, respectively. (c) and (d) display the occupations for a larger $R = 5 \mu\text{m}$ disk with $l = 0$ and $l = 5$. For a better comparison, the matrix elements are plotted against the energies $\epsilon_{cm\nu}$ instead of the indices ν and the absolute values of the occupations are rescaled.

energy of the state $\psi_{bm\nu}$, since in a parabolic band b with effective mass m_b^* the energy of the state is given by $\epsilon_{bm\nu} = \hbar^2 k_{m\nu}^2 / (2m_b^*)$. Note also how the precise location of the energy eigenvalues is given by the zeros of the Bessel functions; this detailed information will be “smeared out” in the limit $R \rightarrow \infty$ as the allowed values of k become a quasi-continuum. For the sake of simplicity, we restrict our discussion to a case where only spin degenerate conduction ($b = c = \{-1/2, 1/2\}$) and heavy-hole ($b = v = \{-3/2, 3/2\}$) bands are considered.

The matrix elements of the twisted-light-matter interaction Hamiltonian H_I (in the dipole approximation for only the z -component) in the cylindrical basis states was derived in Ref. 22:

$$\langle cm'\nu' | H_I | vm\nu \rangle = \xi_{cv\nu'\nu m'} e^{-i\omega t} \delta_{m, m'-l}, \quad (2a)$$

$$\xi_{cv\nu'\nu m'} = -\frac{e}{m_e} A_0 \epsilon_\sigma \cdot \mathbf{p}_{cv} \langle \Phi_c | \delta_{k_z, q_z} | \Phi_v \rangle \mathcal{N}_{m'\nu'} \mathcal{N}_{m\nu} \times \int_0^R dr r J_l(q_\parallel r) J_{m'}(k_{m'\nu'} r) J_{m'-l}(k_{m\nu} r), \quad (2b)$$

where e and m_e are the electron charge and (bare) mass, A_0 is the field strength, ω is the light frequency, q_\parallel the in-plane and q_z the growth-direction part of the light wave vector, k_z is the electron wave vector in the growth direction, l the OAM of the light, ϵ_σ is the light polarization vector and \mathbf{p}_{cv} is the dipole matrix element between heavy-hole and conduction band states. Note that \mathbf{p}_{cv} contains spin selection rules. Let us consider the case where due to excitation with circularly polarized light only spin-up electrons are excited.

In Ref. 22 equations of motion were presented for the density matrix under the influence of twisted light switched on with constant amplitude at $t = 0$. In the low-excitation limit, i.e. initially empty conduction and filled valence bands excited with a moderate light field so that the occupations can be well approximated by an expansion up to second order in the field strength, we find from Eqs. (16) and (17) of Ref. 22:

$$\rho_{cm\nu c'm'\nu'}(t) = \sum_{m_1\nu_1\nu} \delta_{\frac{3}{2}, \nu} \xi_{cv\nu\nu_1 m} \xi_{c'\nu\nu_1 m'}^* \left[\frac{1 - e^{-i(\epsilon_{cm\nu} - \epsilon_{c'm'\nu'})t/\hbar}}{\epsilon_{cm\nu} - \epsilon_{c'm'\nu'}} \left(\frac{1}{\epsilon_{c'm'\nu'} - \epsilon_{vm_1\nu_1} - \hbar\omega} - \frac{1}{\epsilon_{cm\nu} - \epsilon_{vm_1\nu_1} - \hbar\omega} \right) + \frac{1}{(\epsilon_{cm\nu} - \epsilon_{vm_1\nu_1} - \hbar\omega)(\epsilon_{c'm'\nu'} - \epsilon_{vm_1\nu_1} - \hbar\omega)} \left(e^{i(\epsilon_{c'm'\nu'} - \epsilon_{vm_1\nu_1} - \hbar\omega)t/\hbar} + e^{-i(\epsilon_{cm\nu} - \epsilon_{vm_1\nu_1} - \hbar\omega)t/\hbar} - 2 \right) \right] \delta_{mm'} \delta_{c\frac{1}{2}} \delta_{c'\frac{1}{2}}. \quad (3)$$

Thus, the optical excitation yields only diagonal ele-

ments of $\rho_{cm\nu c'm'\nu'}$ with respect to m . Also, we find

from Eqs. (2) that for every electron with OAM m there occurs an unoccupied state with OAM $m - l$ in the valence band, i.e. a hole with OAM $l - m$. From this we can conclude that the total envelope OAM l^{tot} induced in valence and conduction band together is $l^{tot} = lN_e\hbar$ where N_e is the number of excited electrons or, equivalently, holes. The distribution of the total orbital momentum into conduction and valence band contributions depends on the details of the band structure, the pulse duration and the laser frequency. For example, for the effective masses $m_c^* = 0.067m_e$ and $m_{hh}^* = 0.45m_e$ for conduction and heavy-hole bands, respectively, an excitation with a circularly polarized twisted-light pulse with OAM $l = 5$, pulse duration $t = 5$ ps, and central frequency resonant to the band gap leads to a total OAM in the conduction band of $l_c^{tot} = 2.866N_e\hbar$ for $R = 1.2\mu\text{m}$, $l_c^{tot} = 3.145N_e\hbar$ for $R = 5\mu\text{m}$, and $l_c^{tot} = 2.914N_e\hbar$ for $R = 10\mu\text{m}$.

Figure 1 shows the diagonal elements of the density matrix $\rho_{cm\nu cm\nu}$ for the excitation conditions described above. The oscillatory structure of the occupations along the energy axis can be attributed to the finite pulse duration via the energy-time uncertainty relation. Along the m axis, there are also oscillations in the occupations. Since their frequency depends strongly on the radius R of the sample and they get smeared out for large values of R , we attribute these oscillations to finite size effects. Note that in Fig. 1(b), where the occupation for light with $l = 5$ is plotted, the states with the 5 lowest values of m for every energy shell are empty (seen more clearly at low energies), since there are no valence band states which satisfy the condition $m' = m - l$ of the matrix element in Eq. (2a). Figures 1(c) and (d) show that for the larger $R = 5\mu\text{m}$ quantum disk, the difference between the occupations after $l = 0$ and $l = 5$ excitations diminishes visibly.

Now, we focus on the spin dynamics after the optical excitation. We study the effects of spin-orbit coupling mechanisms, considering for concreteness the Rashba Hamiltonian [40], H_R , which is usually the dominant mechanism in quasi-two-dimensional systems. In order to better work with the cylindrical states given in Eq. (1), we switch from the usual cartesian-coordinate expression of H_R to its expression in polar coordinates:

$$H_R = \hbar\alpha_R(k_y\sigma_x - k_x\sigma_y) = \hbar\alpha_R(s^+\partial^- - s^-\partial^+), \quad (4a)$$

$$\partial^\pm := \frac{\partial}{\partial x} \pm i\frac{\partial}{\partial y} = e^{\pm i\phi} \left(\frac{\partial}{\partial r} \pm \frac{i}{r} \frac{\partial}{\partial \phi} \right), \quad (4b)$$

where α_R is the Rashba coefficient, σ_i and s^\pm are the Pauli matrices and spin raising and lowering operators, respectively. With the relation

$$\begin{aligned} & \int_0^R dr r J_m(pr) J_m(qr) = \\ & = R \frac{p J_m(qR) J'_m(pR) - q J_m(pR) J'_m(qR)}{q^2 - p^2}, \end{aligned} \quad (5)$$

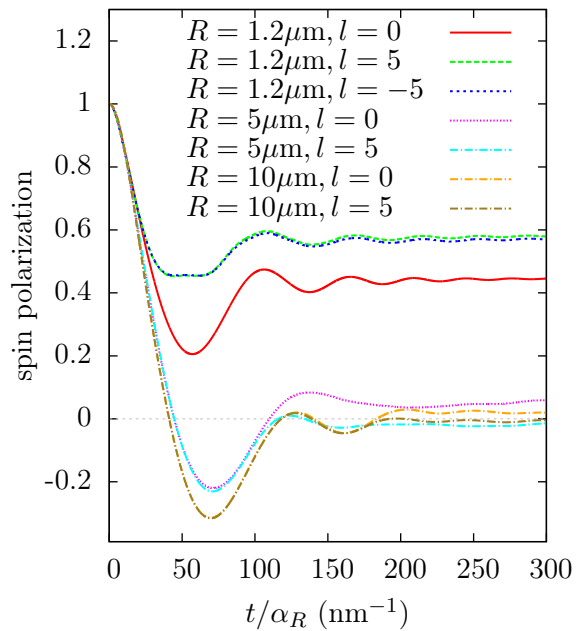


FIG. 2. Spin dynamics after excitation with twisted light with orbital angular momentum $l = \{-5, 0, 5\}$ and quantum-disk radius $R = 1.2\mu\text{m}$ and with $l = \{0, 5\}$ and $R = \{5\mu\text{m}, 10\mu\text{m}\}$.

it is straightforward to calculate the matrix elements of H_R with respect to the cylindrical states:

$$\langle c'l'm'\nu' | \partial^\pm | cm\nu \rangle = \delta_{m', m \pm 1} \frac{2}{R} \frac{k_{m\nu} k_{m'\nu'}}{k_{m\nu}^2 - k_{m'\nu'}^2}, \quad (6a)$$

$$\begin{aligned} \langle c'l'm'\nu' | H_R | cm\nu \rangle &= \frac{\hbar\alpha_R k_{m\nu} k_{m'\nu'}}{R(k_{m\nu}^2 - k_{m'\nu'}^2)} \times \\ &\times (s_{c'l}^+ \delta_{m', m-1} - s_{c'l}^- \delta_{m', m+1}). \end{aligned} \quad (6b)$$

It can be seen from the form of the Rashba Hamiltonian in cylindrical coordinates that an electron with spin-up (down) and envelope OAM m flips to a state with spin-down (up) and OAM $m + 1$ ($m - 1$). In this sense, ∂^\pm can be regarded as raising and lowering operators in m . If H_R is applied a second time, the electronic state is transferred back to the initial spin and OAM state, while a change in ν is possible. Note that the sum $J = m + s$ of the envelope OAM m and the spin s is conserved by the Rashba Hamiltonian.

Having derived the matrix elements of H_R in cylindrical coordinates, it is straightforward to calculate numerically the time evolution of the density matrix, where the initial conditions correspond to the final occupations generated by optical excitation with light with OAM l , illustrated in Fig. 1. The resulting dynamics for the total conduction-band spin is shown in Fig. 2. We show results for three different values of the disk radius, $R = \{1.2, 5, 10\}\mu\text{m}$. As in the case of optical excitation with light with zero OAM, the Rashba interaction leads to a dephasing of the initial electron spins. Since

for small disks only a finite number of states contributes noticeably to the dynamics, oscillations are found which do not cancel completely so that for long times the total spin reaches a non-zero value. Note that the curves for excitation with $l = 5$ and $l = -5$ almost coincide (shown for $R = 1.2 \mu\text{m}$). This unexpected result shows clearly the insensitivity of the spin dynamics, in the presence of the Rashba spin-orbit interaction, to the content of OAM transferred from twisted light to the electron gas. For the same quantum disk, an excitation with $l = 0$ produces spin dynamics different from the $l = \pm 5$ excitation, but this difference decreases for larger radii. This tendency can be seen by comparing the excitation with $l = 5$ and $l = 0$ for the three different values of R used in Fig. 2.

To understand this finding, it is useful to analyze the case of an infinitely extended quantum well, obtained letting $R \rightarrow \infty$. In this limit, the discrete $k_{m\nu}$ become continuous and the eigenstates can be written as

$$\psi_{bkm}(r, \phi, z) := \sqrt{\frac{k}{2\pi}} J_m(kr) e^{im\phi} \Phi_b(z), \quad (7)$$

with the orthogonality relation $\langle bkm|b'k'm' \rangle = \delta_{bb'} \delta_{mm'} \delta(k - k')$. Using that Bessel functions satisfy

$$\int_0^\infty dr r J_m(kr) J_m(k'r) = \frac{1}{k} \delta(k - k'), \quad (8)$$

the corresponding matrix elements become

$$\langle c'k'm'|\partial^\pm|ckm \rangle = \mp k \delta_{m', m \pm 1} \delta(k - k') \quad (9a)$$

$$\langle c'k'm'|H_R|ckm \rangle = \hbar \alpha_R k \delta(k - k') \times \\ \times (s_{c'e}^+ \delta_{m', m-1} + s_{c'e}^- \delta_{m', m+1}). \quad (9b)$$

Thus, in the quasi-continuum limit, the prefactor of the Rashba-interaction depends only on the energy of the state via k but not on m . The spin dynamics is therefore a precession of the electron spin with a k -dependent frequency which, for a given k , is the same for all different values of m . Since the effect of the excitation with twisted light was mainly that states with different m are excited, it is now easy to see why, for extended systems, the spin dynamics due to the Rashba Hamiltonian is almost the same for excitations with light with and without OAM.

It is noteworthy that this statement is also true for all effective Hamiltonians with a microscopic origin in the lattice-periodic crystal potential such as the Dresselhaus [41] spin-orbit coupling when treated in the envelope-function approximation: For a lattice-periodic potential, the solutions of the corresponding Schrödinger equation are given by the Bloch theorem as $\psi(\mathbf{r}) \propto e^{i\mathbf{k}\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r})$ with lattice-periodic Bloch function $u_{n\mathbf{k}}(\mathbf{r})$, band index n and wave-vector \mathbf{k} . The envelope-function approximation consists of integrating over the plane-wave part of the wave function yielding an effective Hamiltonian [42] H_{eff} for the $u_{n\mathbf{k}}$ which is diagonal in \mathbf{k} and the matrix

elements can be written as a power series in \mathbf{k} . The resulting effective Hamiltonian can be rewritten by decomposing k_x and k_y in terms of ∂^+ and ∂^- , as done in Eqs. (4). Thus, the dependence of the matrix elements of H_{eff} in cylindrical states on m is of the same character as for the Rashba Hamiltonian and vanishes in the quasi-continuum limit. For systems with finite size, however, a weak dependence on m can be found due to the m -dependence of the possible k -values in the prefactor of the Rashba Hamiltonian in Eq. (6b). This means that, e.g. for small quantum disks, where the energy separation between the discrete cylindrical states becomes important, the OAM of the exciting light can influence the spin dynamics significantly.

In conclusion, we have shown that although the OAM of twisted light can be transferred into the envelope OAM of electrons, the usual solid state spin-orbit interactions, such as Rashba and Dresselhaus interactions, do not couple the envelope OAM of the carriers to their spin degree of freedom in such a way that a significant difference in the spin dynamics after excitation with light with and without OAM is found in large extended systems. This finding can explain that in recent experiments [29] no influence of the light's OAM on the spin polarization was found. However, for cylindrical quantum disks with small radii, the discreteness of the states plays an important role so that the spin dynamics indeed depends on the OAM of the light. Nevertheless, also for small systems, the spin dynamics after excitation with OAM $l = |l|$ and $l = -|l|$ are very similar, in contrast to optical excitation with opposite circular polarization, where the spin dynamics acquires a different sign. Thus, twisted-light based spin control is fundamentally different from traditional spin orientation and combining these control schemes should therefore open new ways for spin manipulation. Our findings have direct implications for the search for systems where optical excitation with twisted light can be used to manipulate the spin dynamics: A continuum of states has to be avoided. Such systems can be confined structures like quantum dots [27] and rings [21, 23], discrete states due to multi-particle effects, e.g. excitons or localized states due to impurities and surface effects.

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