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Theory of the optical absorption of light carrying orbital angular momentum by semiconductors

G. F. QUINTEIRO[(a)] and P. I. TAMBORENEA

Departamento de Física "J. J. Giambiagi", Universidad de Buenos Aires - Ciudad Universitaria, Pabellón I, C1428EGA, Ciudad de Buenos Aires, Argentina

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Abstract – We develop a free-carrier theory of the optical absorption of light carrying orbital angular momentum (twisted light) by bulk semiconductors. We obtain the optical transition matrix elements for Bessel-mode twisted light and use them to calculate the wave function of photo-excited electrons to first-order in the vector potential of the laser. The associated net electric currents of first and second-order on the field are obtained. It is shown that the magnetic field produced at the center of the beam for the $\ell = 1$ mode is of the order of a millitesla, and could therefore be detected experimentally using, for example, the technique of time-resolved Faraday rotation.

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It is well known from classical electromagnetism that light can carry spin and orbital angular momentum. While the former was detected for the first time in the 1930s [1], the latter became widely available for experimental study only recently after the work of Allen *et al.* [2] In a seminal paper, those authors showed that light carrying an integer amount of *orbital* angular momentum ($\hbar l$, with l an integer) may be generated in the laboratory using conventional laser beams. Since then, research on the subject of light carrying orbital angular momentum (OAM), or twisted light (TL) [3–5] has spanned a large number of areas, namely, generation of beams [4], interaction with mesoscopic particles (optical tweezers) [6–8], entanglement with spins for potential applications in quantum information processing [9], interaction with atoms and molecules [10,11], cavity-QED [12], and interaction with Bose-Einstein condensates [13,14]. Nevertheless, the interaction with solid-state systems, although potentially important for technological applications, has not been explored so far. In this letter, we present the first theoretical predictions about the interaction of TL with bulk semiconductors. We consider band-to-band transitions, *i.e.* optical transitions with light frequencies above the bandgap, so that free carriers rather than excitons are produced. We show that there is a transfer of OAM between the light and the photoexcited electrons so that a net electric current initially confined to the beam area appears. The magnetic field induced by these photocurrents is estimated.

A beam of TL presents an azimuthal phase dependence —helical wavefront— responsible for the OAM, and a radial dependence of the *Laguerre-Gaussian* (LG) or *Bessel mode* type. We will focus on the Bessel modes, but our results are applicable to the LG-mode with slight changes. The vector potential in the Coulomb gauge with cylindrical coordinates $\{r_\parallel, \phi, z\}$ is [15]

$$\mathbb{A}_{\mathbf{q}l\pm}(\mathbf{r},t) = A_0 e^{i(q_z z-\omega t)}\Big[\boldsymbol{\epsilon}_\pm J_l(q_\parallel r_\parallel)e^{il\phi} \mp i\hat{z}\frac{q_\parallel}{q_z}J_{l\pm1}(q_\parallel r_\parallel)e^{i(l\pm1)\phi}\Big] + \text{c.c.}, \qquad (1)$$

with polarization vectors $\boldsymbol{\epsilon}_\pm = \hat{x} \pm i\hat{y}$, Bessel functions J_l, and parameters $q_\parallel \ll q_z$.

Semiconductors are solids which at zero temperature have the highest occupied (valence) and lowest empty (conduction) energy bands separated by a gap E_g. In this respect, they are closer to insulators than to metals. However, in typical semiconductors $E_g \simeq 1\,\text{eV}$, making possible the transitions between the valence and conduction bands by optical excitation. In a crystalline semiconductor, electrons in the valence and conduction bands, denoted by $\lambda = \{v, c\}$, occupy the Bloch states $\varphi_{\lambda\mathbf{k}}(\mathbf{r}) = L^{-3/2}e^{i\mathbf{k}\cdot\mathbf{r}}u_{\lambda\mathbf{k}}(\mathbf{r})$, with $u_{\lambda\mathbf{k}}(\mathbf{r})$ a cell-periodic function (lattice constant a), $\hbar\mathbf{k}$ the crystal momentum, and L the

(a) E-mail: gquinteiro@df.uba.ar

linear size of the semiconductor. For concreteness, here we consider transitions from the heavy-hole valence bands. The orbital angular momentum equal to 1 of the p-type orbital states plus the electron spin add up to a total angular momentum of 3/2 with a z-projection of $\pm 3/2$ for these bands. These states are denoted $|3/2, \pm 3/2\rangle$. The conduction band states, being s-type (zero orbital angular momentum), are $|1/2, \pm 1/2\rangle$. We can consider a simplified two-band model which grasps the main features of a semiconductor [16], and can be considered a good model of a real bulk system under the following two conditions: i) an applied strain splits the heavy-hole and light-hole valence bands; ii) only circularly polarized light is considered, producing optical transitions of only one electron spin type between valence and conduction bands. For example, circularly polarized light with photon spin equal to -1 induces transition between the states $|3/2, 3/2\rangle$ and $|1/2, 1/2\rangle$.

We now proceed to develop an analytical description of the interband transitions in a two-band model of a bulk semiconductor induced by twisted light. Our analysis tackles the coherent optical excitation to conduction-band states, *i.e.* we consider the case $\hbar\omega > E_g$. In this regime the creation of excitons is negligible and one only has to take into account the free carriers transferred by optical excitation from the valence to the conduction band [16]. The lowest-order contribution to the light-matter interaction, using the minimal-coupling Hamiltonian, is $H_I = -(Q/m)\mathbf{p}\cdot\mathbf{A}_{\mathbf{q}l\sigma}(\mathbf{r})$ with $\mathbf{A}_{\mathbf{q}l\sigma}(\mathbf{r})$ the transverse or xy part of $\mathbb{A}_{\mathbf{q}l\pm}(\mathbf{r},t)$ (eq. (1)), and $\{\mathbf{p}, m, Q\}$ the momentum operator, mass, and charge of the particle involved. The complete semiclassical Hamiltonian —operators for electrons and classical variables for the light field— consists then of two terms, the bare electron energy and the interaction H_I

$$H = \sum_{\lambda\mathbf{k}} E_{\lambda\mathbf{k}} a^\dagger_{\lambda\mathbf{k}} a_{\lambda\mathbf{k}} + \sum_{\lambda'\mathbf{k}'\lambda\mathbf{k}} \frac{-Q}{m} \langle\lambda'\mathbf{k}'|\mathbf{p}\cdot\mathbf{A}_{\mathbf{q}l\sigma}(\mathbf{r})|\lambda\mathbf{k}\rangle a^\dagger_{\lambda'\mathbf{k}'} a_{\lambda\mathbf{k}},$$

with $a^\dagger_{\lambda'\mathbf{k}'}/a_{\lambda\mathbf{k}}$ the creation/annihilation operators for electrons in a Bloch state of band $\lambda = v$, c and quasi-momentum $\hbar\mathbf{k}$, and $E_{\lambda\mathbf{k}}$ its bare energy. The action of $\mathbf{p} = -i\hbar\boldsymbol{\nabla}$ onto the wave function $\varphi_{\lambda\mathbf{k}}(\mathbf{r}) = \langle\mathbf{r}|\lambda\mathbf{k}\rangle$ yields two terms: $\mathbf{p}\varphi_{\lambda\mathbf{k}}(\mathbf{r}) = \hbar\mathbf{k}\varphi_{\lambda\mathbf{k}}(\mathbf{r}) - i\hbar L^{-3/2} e^{i\mathbf{k}\cdot\mathbf{r}} \nabla u_{\lambda\mathbf{k}}(\mathbf{r})$. For interband transitions —our interest— the first term is negligible; then

$$H_I|_{\lambda'\mathbf{k}'\lambda\mathbf{k}} = \frac{i\hbar}{L^3}\frac{Q}{m}\int_{L^3} \mathrm{d}\mathbf{r} e^{-i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{r}} \times u^*_{\lambda'\mathbf{k}'}(\mathbf{r})[\mathbf{A}_{\mathbf{q}l\sigma}(\mathbf{r})\cdot\nabla]u_{\lambda\mathbf{k}}(\mathbf{r}).$$

This expression can be simplified by using a standard procedure in solid-state physics. The integration over the complete system is replaced by an integration over a unit cell and a sum over all cells

$$H_I|_{\lambda'\mathbf{k}'\lambda\mathbf{k}} = -\frac{Q}{m}\frac{1}{N}\sum_c e^{-i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_c} \mathbf{A}_{\mathbf{q}l\sigma}(\mathbf{R}_c)\cdot \frac{1}{a^3}\int_{a^3} \mathrm{d}\mathbf{r} u^*_{\lambda'\mathbf{k}'}(\mathbf{r})[-i\hbar\nabla]u_{\lambda\mathbf{k}}(\mathbf{r}),$$

where N is the total number of unit cells in the crystal. The integrand is split thanks to the facts that, in the length of the unit cell a, $\mathbf{A}_{\mathbf{q}l\sigma}(\mathbf{r})$ and $\exp[-i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{r}]$ are almost constant and $u(\mathbf{r})$ is periodic. The integral is the momentum matrix element $\mathbf{p}_{\lambda'\mathbf{k}'\lambda\mathbf{k}}$. The sum is handled using the Jacobi-Anger identity, which allows us to separate the angular from the radial dependences in $e^{-i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_c}$, and then applying the orthogonality relation for Bessel functions [17]. The final expression for the interaction Hamiltonian is

$$H_I = -(-i)^l \frac{QA_0}{m}\frac{1}{L} e^{-i\omega t} \sum_{\mathbf{k}\mathbf{k}'} \frac{\delta_{\kappa_\parallel q_\parallel}}{q_\parallel}\delta_{\kappa_z q_z} \times e^{i\theta l}\left(\boldsymbol{\epsilon}_\sigma\cdot\mathbf{p}_{c\mathbf{k}'v\mathbf{k}}\right) a^\dagger_{c\mathbf{k}'} a_{v\mathbf{k}} + \mathrm{h.c.}, \tag{2}$$

with $\boldsymbol{\kappa} = \mathbf{k}' - \mathbf{k}$ having cylindrical coordinates $\{\kappa_\parallel, \theta, \kappa_z\}$.

In order to understand the effect that a twisted-light beam has on the ground state of a semiconductor, we focus our attention on the positive part $H_I^{(+)}$ of the interaction Hamiltonian —first term of eq. (2). The action of $H_I^{(+)}$ on the full ground state of the N-electron semiconductor can be shown to yield an eigenvector with expectation value of the orbital angular momentum equal to $\hbar l$. Here we present a simplified version of the theory where we show the action of the Hamiltonian on a single-particle valence-band electron state. The analysis is restricted to optical excitations that couple electron wave-numbers near $\mathbf{k} = 0$, a typical situation in semiconductor optics. We define $\xi = -(-i)^l (QA_0/m)(\boldsymbol{\epsilon}_\sigma\cdot\mathbf{p}_{c0v0})$ independent of the wave vectors (see chapt. 5 of ref. [16]), transform variables $\{\mathbf{k},\mathbf{k}'\} \to \{\mathbf{k},\boldsymbol{\kappa}\}$, fix the value of $\mathbf{k}$ and take the continuum limit for $\boldsymbol{\kappa}$. In the coordinate representation and rotating frame, the action of the Hamiltonian on the valence band single-particle initial state gives

$$H_I^{(+)}\varphi_{v\mathbf{k}}(\mathbf{r}) = \frac{\xi}{2\pi}\int \mathrm{d}\boldsymbol{\kappa}\frac{\delta(\kappa_\parallel - q_\parallel)}{q_\parallel} \times\delta(\kappa_z - q_z) e^{i\theta l}\varphi_{c\mathbf{k}+\boldsymbol{\kappa}}(\mathbf{r}),$$

which makes evident that a "cone"-like transition occurs producing a linear superposition of conduction-band states with varying phases, as depicted in fig. 1. Exploiting the delta functions, using $\varphi_{\lambda\mathbf{k}+\boldsymbol{\kappa}}(\mathbf{r}) \simeq e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}\varphi_{\lambda\mathbf{k}}(\mathbf{r})$, and applying the Jacobi-Anger identity, we get

$$\begin{aligned} H_I^{(+)}\varphi_{v\mathbf{k}}(\mathbf{r}) &= \xi\left[i^l e^{iq_z z} J_l(q_\parallel r_\parallel) e^{i\phi l}\right]\varphi_{c\mathbf{k}}(\mathbf{r}) \\ &\doteq \xi f(\mathbf{r})\varphi_{c\mathbf{k}}(\mathbf{r}). \end{aligned} \tag{3}$$

The dimensionless function $f(\mathbf{r}) = i^l e^{iq_z z} J_l(q_\parallel r_\parallel) e^{i\phi l}$ contains all the relevant information that distinguishes

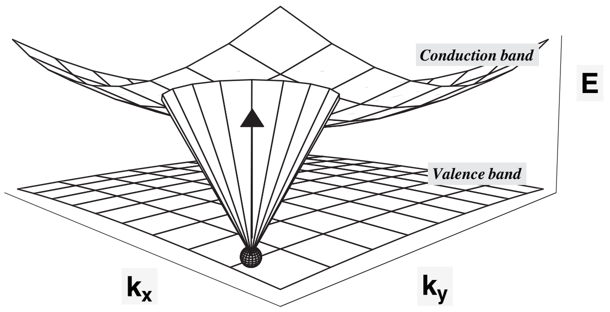

Fig. 1: Schematic representation of interband optical excitation with twisted light. A twisted-light beam excites a valence band electron into a superposition of states of the conduction band in a “cone”-like fashion. The laser parameter $q_\|$ equals the cone radius at the conduction band.

the action of a twisted beam from its counterpart, the plane wave. Because of the presence of $f(\mathbf{r})$, the RHS of eq. (3) is *not* a conduction band eigenstate. For the expectation value of the z-component of the orbital-angular-momentum operator $L_z = -i\hbar\partial_\phi$ in the state of eq. (3) we obtain $\langle L_z\rangle = \hbar l$, since $\langle L_z\rangle = \hbar l$ for the state $e^{i\phi l}$ and $\langle L_z\rangle = 0$ for the states $u_{c\mathbf{k}}(\mathbf{r})$ and $e^{i\mathbf{k}\cdot\mathbf{r}}$. Thus, for short times, the evolution leads to a well-defined transfer of orbital angular momentum $\hbar l$ from the light beam to the electron.

This net transfer of orbital angular momentum to the photo-excited electrons is expected to result in electric currents and associated magnetic fields. The latter would be an experimentally detectable signature of the type of optical excitation described here, and may also lead to opto-electronic applications. An estimate of the total current/magnetic field produced by all electrons can be obtained by calculating the total number of electrons excited by the field, then determining the current/magnetic field of a single electron, and, finally, multiplying the single-electron current/magnetic field by the number of photo-excited electrons. We note that this is a single-particle calculation which does not take into account the electron-electron Coulomb interaction. This is justified in the present study by the fact that the main correction introduced by the Coulomb interaction would be the renormalization of the single-particle energies [18,19]. This mean-field effect would be small since we are working in the regime of low excitation (the density of photogenerated electrons and holes is small), and, furthermore, by its nature, it does not affect the qualitative picture drawn in this work. Beyond the mean-field approximation, the Coulomb interaction introduces complex and possibly interesting scattering effects [18,20], which are beyond the scope of our present analysis, and are left for future study. Again, however, these effects are expected to be small in the low-excitation regime.

The number of electrons excited in a volume $\mathrm{d}V$ and a time interval t is $\mathrm{d}N_{ex} = \alpha I(\mathbf{r})t/\hbar\omega$, with α the absorption coefficient. $I(\mathbf{r}) = I_0 e^{-\alpha z}$ is the intensity of the light field, which is related to the vector potential by $2I_0 = v\epsilon\omega^2 A_0^2$, where v is the speed of light in the medium. Then, the total number of electrons that are excited in $V = L^3$ is

$$N_{ex} = \pi\frac{L^2 t I_0}{\hbar\omega} f(q_\| L),$$

with $f(q_\| L)$ a function containing $J_{l-1}(x)$, $J_l(x)$, $J_{l+1}(x)$, and $e^{-\alpha L}$.

In order to calculate the electric current, we first determine the single photo-excited electron state. For short times such that $t(\omega + E_g/\hbar) < 1$ the evolution of the state under the action of the laser field is, in the Schrödinger picture,

$$\begin{aligned}\psi(\mathbf{r},t) &= U(t)\varphi_{v\mathbf{k}}(\mathbf{r})\\ &\simeq \varphi_{v\mathbf{k}}(\mathbf{r}) - e^{-iE_g t/\hbar}(it/\hbar)\xi f(\mathbf{r})\varphi_{c\mathbf{k}}(\mathbf{r}),\end{aligned} \quad (4)$$

with $U(t)$ the evolution operator (see eq. (3)). The electric-current density of this state is calculated using the quantum-mechanical expression $\mathbf{j} = (Q\hbar/m)\Im[\psi^*\boldsymbol{\nabla}\psi] - \frac{Q^2}{m}\Re[\psi^*\mathbf{A}\psi]$. After a lengthy calculation we arrive at the electric current and separate the terms in powers of A_0 (hidden in ξ). The zero-order term does not give new phenomena, and so it is not presented. The first-order and second-order terms are

$$\mathbf{j}_1(\mathbf{r},t) = \frac{2Q}{mL^3}\Im\left\{\left(\frac{t\xi}{\hbar}\right)\mathbf{p}_{v0c0}e^{-iE_g t/\hbar}f(\mathbf{r})\right\}, \quad (5)$$

$$\mathbf{j}_2(\mathbf{r},t) = \left(\frac{t|\xi|}{\hbar}\right)^2\frac{Q\hbar}{mL^3}\Im\left\{f(\mathbf{r})^*\nabla f(\mathbf{r})\right\}. \quad (6)$$

Let us comment on the main characteristics of these two currents. $\mathbf{j}_1(\mathbf{r},t)$ contains the vector momentum matrix element $\mathbf{p}_{v0c0}$, which reappears in the calculation of the current due to the mixing of the two terms of $\psi(\mathbf{r},t)$. Thus, as $\mathbf{p}_{v0c0}$ itself, this component of the macroscopic current has a microscopic origin. This current is somewhat analogous to the optical polarization in the standard interband optical transitions induced by plane waves. Note that this current has no equivalent in atomic physics [21]. In fig. 2 we show the current vector field for the cases $l = 1$ to 4. A clear feature of the current for $l = 1$ is that it displays a net circulation around the z-axis, which will give rise to a sizable magnetic field. For all other values of l the current shows net circulations around off-centered axes, like in the cases $l = 2$ to 4 seen in fig. 2, but no net circulation around the center of the beam. Finally, without explicitly showing it here, we mention that the current field behaves like a travelling wave in the z-direction, and therefore, at a given time, clockwise and counterclockwise circulations alternate as one moves along the z-axis. The second-order term $\mathbf{j}_2(\mathbf{r},t)$ comes from just the second term in $\psi(\mathbf{r},t)$. As can be seen in eq. (6), $\mathbf{j}_2(\mathbf{r},t)$ stems directly from the function $f(\mathbf{r})$, defined after eq. (3), whose spatial dependence mimics closely that of laser beam and varies significantly only on a macroscopic scale. Thus, we

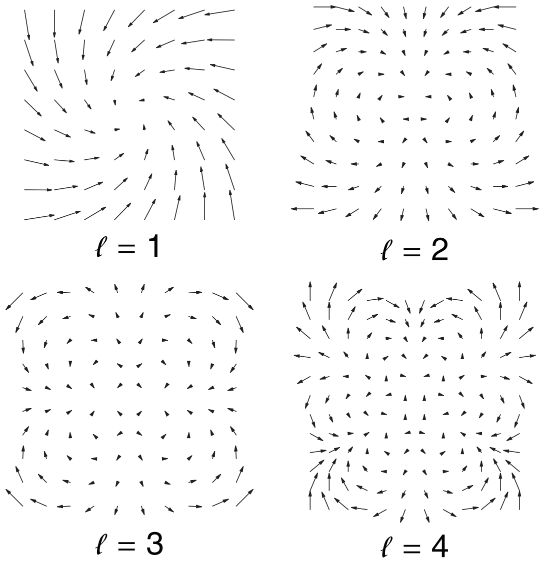

Fig. 2: First-order electric current for OAM $l = 1$ to 4. The center of each plot coincides with the center of the twisted-light laser beam.

interpret that this current has a macroscopic origin and is analog to the motion induced by twisted light on atomic systems.

For the currents with net circulation around the beam axis, *e.g.* the case $l = 1$ in fig. 2, we estimate the magnetic field b_z at the center the beam. We adopt a semiclassical approach and use Biot-Savart's law with the currents from eqs. (5) and (6) whose azimuthal components are

$$j_{1\phi}(\mathbf{r}, t) = -2\frac{q^2 A_0 p_0^2}{m^2 L^3 \hbar} J_1(q_\| r_\|) \sin(q_z z)\, t, \tag{7}$$

$$j_{2\phi}(\mathbf{r}, t) = l\left(\frac{t|\xi|}{\hbar}\right)^2 \frac{q\hbar}{mL^3}\frac{1}{r_\|} J_l(q_\| r_\|)^2. \tag{8}$$

Then, the total magnetic field is determined by $B_z = N_{ex} b_z$. Our semiclassical approach is an admissible procedure in perturbation theory since the quantum fluctuations of the current are of higher order in the vector potential and thus smaller than their mean values.

As an example, we estimate the magnetic field B_z for a realistic situation. We choose the following typical experimental parameters [12,14,22,23]. For the laser beam: laser with frequency $\nu = 3 \cdot 10^{14}\,\mathrm{s}^{-1}$, repetition rate $F = 100\,\mathrm{MHz}$, power $P = 3\,\mu\mathrm{J\,s}^{-1}$, pulse duration $t = 10\,\mathrm{fs}$, $q_z = 2 \cdot 10^7\,\mathrm{m}^{-1}$, $q_\| = 4 \cdot 10^6\,\mathrm{m}^{-1}$ (spot size $\simeq (0.5\,\mu\mathrm{m})^2$), and $l = 1$. For the semiconductor material we choose GaAs parameters: $\alpha = 10^5\,\mathrm{m}^{-1}$, $a = 0.6\,\mathrm{nm}$, $v = c/3$, $L = 1.3\,\mu\mathrm{m}$ and $|\boldsymbol{\epsilon}_\sigma \cdot \mathbf{p}_{c0v0}| = 6.4 \cdot 10^{-25}\,\mathrm{kg\,m/s}$. Assuming a top-hat laser pulse (which approximates the cw-field of eq. (2) for long enough pulses) these values yield $N_{ex} = 3 \cdot 10^5$ and $(|\xi|t/\hbar) = 0.8$. The resulting z-component of the magnetic field in the center of the beam is $|B_{1z}| = 1\,\mathrm{mT}$ and $|B_{2z}| = 0.5\,\mu\mathrm{T}$ for the first-order and second-order currents, respectively. This estimate is reliable since the build-up of the magnetic field takes place in a time shorter than the typical relaxation times of electrons in the conduction band of semiconductors at low excitation density [18].

Our theory can be put to test by measuring this predicted magnetic field. We suggest the use of time-resolved Faraday rotation. A quick estimate of the polarization rotation angle can be obtained with the Verdet formula $\theta = V B_z \ell$, where V is the Verdet constant and ℓ is the length traversed by the probe beam. Taking $V = 10^{-4}\,\mathrm{rad/(G\,cm)}$ and $\ell = 10\,\mu\mathrm{m}$ one obtains $\theta \simeq 10^{-6}\,\mathrm{rad}$, which is clearly measurable according to ref. [24]. We note that the influence of the orbital and spin angular momenta carried by the photons could be discriminated, for example, by comparing two separate measurements, one with $l = 0$ and another one with $l = 1$ with the same circular polarization.

Let us finally comment on the conservation of the angular momentum in the optical transitions considered here. Naturally, in the optical absorption both the orbital and spin angular momenta of the photon are transferred to the electrons, but from our analysis we can see that they exert their influence in different ways. The photon spin plays a role completely analogous to the one played in the case of non-twisted light, determining which pair of energy bands are connected by the optical excitation. On the other hand, the OAM of the light generates a macroscopic effect, revealed in the generation of currents whose characteristic lengths are much greater than the size of the unit cell.

In conclusion, in this letter we have shown for the first time the effect that light carrying orbital angular momentum has on semiconductors. We started by generalizing the basic theory of photo-excitation with plane waves to the case of Bessel mode beams. We found that the excitation process generates a superposition of conduction band states with a well-defined angular momentum. This superposition state is associated to an electric-current density, which acts as the source of a magnetic field. As an example, the magnitude of this magnetic field has been estimated in a typical experimental situation. The important regime of excitonic generation has not been addressed here and will be the subject of a future work, as well as the inclusion of electron-electron Coulomb interaction effects.

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