The interaction of optical vortices with chiral matter

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ABSTRACT

In recent years, twisted laser beams and optical vortices have attracted considerable interest, in terms of both their fundamental quantum properties and also their potential technical applications. Here we examine what novel features might emerge from the interaction of such beams with chiral matter. In this connection we assess the possible scope for exploiting similarities between the angular momentum properties of circularly polarised light and optical vortices – both with regard to their mechanical torque and also the associated spectroscopic selection rules. Twisted beams have generally been studied only in their interactions with achiral matter, with the theory largely developed for electric dipole coupling. In chiral systems, the low symmetry enables many optical transitions to be allowed under the selection rules for both electric and magnetic multipoles, and the entanglement of spin and orbital photon angular momentum requires careful extrication. Specific issues to be addressed are: what new features, if any, can be anticipated when such beams are used to interrogate a chiral system, and whether in such cases enantiomeric specificity can be expected. To this end we develop theory that goes to a higher order of multipole expansion, also engaging magnetic dipole and electric quadrupole transitions. Finally, we study the response of nematic liquid crystal media to the throughput of twisted laser light. Specific attention is focused on the evolution of the liquid crystal director.

Keywords: Optical vortices, twisted beams, photon angular momentum, chiral optics, liquid crystals

1. INTRODUCTION

In the field of laser photonics, two of the areas attracting significantly heightened recent interest concern optical vortices (beams with a helically twisted wave-front[1]) and chiral optics (such as chirally sculptured materials[2]). The intriguing possibility of engaging twisted wave-front chirality with chiral matter is a subject that has so far eluded detailed analysis – though at the molecular level, the chiral interplay of light and matter is very well known and exemplified by the circular dichroism that characterizes the handedness of sugars. In the former connection one of the key issues is the potential manifestation, through interaction with chiral matter, of the sense of helicity in the orbital angular momentum characterizing twisted laser beams. In particular it needs to be established to what extent this might parallel the engagement of spin angular momentum associated with circularly polarized light. In this paper we examine what novel features might emerge from the interactions of optical vortices with chiral matter, with particular regard to two topics: (i) the operation of fundamental symmetry principles as determinants of spectroscopic selection rules, and (ii) the effects of mechanical torque. For the former analysis, in Section 2 we develop theory that extends beyond the electric dipole approximation usually employed, for simplicity, in describing twisted beam interactions. The key results are given in Section 3. We then report in Section 4 the first results of a simulation of the optomechanical response in nematic liquid crystal media.

2. ELECTRIC AND MAGNETIC DIPOLE COUPLING

For representation of the interactions of twisted optical beams, the theory that is generally applied is cast in terms of electric dipole coupling. Whilst entirely appropriate for most achiral systems, the low symmetry of chiral systems enables many optical transitions to be allowed under the selection rules for both electric and magnetic dipole coupling[3]. Molecular chirality signifies a structural handedness associated with variance under spatial inversion, or a combination of inversion and rotation, equivalent to the usually stated criterion of a lack of any improper (rotation-reflection) axes. Chiroptical phenomena commonly engage circularly polarized light, for even optical rotation is generally interpreted by describing the plane polarized state as a superposition of circular polarizations with opposite handedness. In the case of circularly polarized light, the left- and right- forms designate the sign of intrinsic spin angular momentum, $\pm\hbar$, and also the helicity of the locus described by the associated electromagnetic field vectors. For this reason its interactions with

matter are enantiomerically specific, i.e. the ensuing signals elicit the helicity of molecular conformation. Hence, in considering the chirality of twisted beams endowed with orbital angular momentum, for which each photon is associated with an angular momentum $l\hbar$ with positive or negative integer l, it is appropriate to begin by establishing corresponding principles relating to the interplay of quantized spin and orbital angular momentum. The aim is to develop a framework in which conventional chirality can be depicted, with regard to molecular symmetry and electric/magnetic properties, and then to extend the arguments to address light with a twisted wave-front. For simplicity, materials with a unique enantiomeric specificity are assumed – signifying a chirality that is intrinsic and common to all molecular components. Results for systems of this kind will also apply to single-molecule studies. The longer-range translational-orientational order that can also engender chirality, as in twisted nematic crystals, is considered in the context of mechanical effects in Section 4.

Chirality signifies a local breaking of parity, thereby permitting an interference of electric and magnetic interactions. In the framework of quantum electrodynamics, the system Hamiltonian comprises unperturbed operators for the chiral centers and the radiation, and also the interaction Hamiltonian, whose role as perturbation operator leads to optical transitions. Each component of the Hamiltonian is necessarily of even parity with respect to space inversion and also even with respect to time reversal. The multipolar interaction Hamiltonian[4] entails a linear coupling of the molecular polarisation field (accommodating all electric multipoles En) with the transverse electric field $\mathbf{e}$ of the radiation, and also a linear coupling of the molecular magnetisation field (all magnetic multipoles Mn) with the magnetic induction field $\mathbf{b}$ of the radiation. Here it is sufficient to restrict consideration to the E1 and M1 interactions whose interplay is mostly associated with manifestations of chirality. The electric field has an odd signature for space parity and even for time, as do the dipoles $\boldsymbol{\mu}$; the magnetic field and also the magnetic dipoles $\mathbf{m}$ are even in space and odd in time. Note, however, that these symmetry properties for the electric and magnetic fields are only necessarily true only for the entirety of the electromagnetic radiation field; they need not necessarily apply to individual radiation modes such as the twisted modes at issue below.

The quantum amplitude M_{fi} for a specific optical interaction in a single chiral centre ξ, as constructed from time-dependent perturbation theory, generally entails a linear combination of scalars of which each is the inner product of two rank r tensors: $\mathbf{S}^{(r)}$ (comprising an outer product of electric and magnetic field components) and $\mathbf{T}^{(r)}$ (an outer product of molecular multipole components)[5];

$$M_{fi}^{\xi} = \exp\left(i\Delta\mathbf{k}\cdot\mathbf{R}_{\xi}\right)\sum_{e,m=0}^{n}\mathbf{S}^{(r)}_{e;\,m;\,n-e-m}\otimes\mathbf{T}^{(r)}_{e;\,m;\,n-e-m}\,. \tag{1}$$

Here, $\Delta\mathbf{k}$ is the mismatch between the summed wave-vectors of all input and any output photons involved in the process at a chiral centre ξ located at $\mathbf{R}_{\xi}$. To identify the tensors $\mathbf{S}^{(r)}$ and $\mathbf{T}^{(r)}$, three labels are used; (e; m; q), corresponding to the number of electric-dipole, magnetic-dipole and electric quadrupole interactions, respectively: the sum of these equals the number of photon interactions n involved in the process. The rank of the tensors, r, is determined by the simple relation $r = e + m + 2q$ which signifies that the rank is between n and $2n$. The molecular tensor $\mathbf{T}^{(r)}$ can be written as a product of one or more molecular transition integrals, determined by the number of photons involved. In order for the result not to vanish identically, the triple product of the group theoretic representations for the initial and final state wavefunctions with that of the interaction must contain the totally symmetric representation of the molecular point group.

For chiral systems, the quantum amplitude associated with a particular optical interaction may include contributions of both positive and negative spatial parity. The rate observable, Γ, emerges from an ensemble sum of contributions;

$$\Gamma \propto \left|M_{fi}\right|^2 = \sum_{\xi,\xi'}^{N} M_{fi}^{\xi}\bar{M}_{fi}^{\xi'}\,, \tag{2}$$

and the result includes the quantum interference of couplings with odd parity for both the radiation and molecule. However it is not only molecular symmetry, but also macroscopic symmetry that determines the nature and extent of any chiroptical response; for a fluid or other microscopically disordered medium, the isotropic symmetry of the bulk comes into play. It is apparent from the exp($i\Delta\mathbf{k}.\mathbf{R}$) factor in equation (1) that each of these contributions generally has

a different phase associated with spatial variance in the registration of light at each centre. For optical processes which are non-parametric ($\Delta\mathbf{k} \neq 0$), there is a net destructive interference of quantum amplitudes from different centers in any isotropic distribution – except in certain systems with significantly different mesoscopic symmetry, such as those comprising nanoparticles[6]. For these incoherent processes, no role is played by any orientational order characterizing *long*-range chirality (important exceptions arise for parametric optical processes, $\Delta\mathbf{k} = 0$). Here we focus on incoherent processes since processes of this category are involved in the most common manifestations of chirality such as circular dichroism; also, where optical vortices are concerned, it is known that parametric processes generally entail conservation of orbital angular momentum by the radiation field, such that chirality cannot be engaged[7,8]. In the quantum interferences which are odd in parity for both the matter and radiation – and which can only arise in the case of chiral systems – handedness is apparent in two respects. On performing space inversion on the molecule (signifying a change to the opposite enantiomeric form) but not the radiation (signifying retention of photon circularity), these interference terms change sign. The same is true if the radiation changes handedness, but the molecule retains its enantiomeric form: all contributions to the signal are invariant to inversion of the whole system. This is why chiral interactions must involve handed radiation. Plane polarizations are invariant under space inversion; consequently, applying space inversion to a molecular system interacting with plane polarized light has the same effect as applying it to both the molecule and the radiation, and no chiral specificity can emerge. Optical rotation is an exception since it is a parametric process.

Further conditions need to be satisfied for the interference terms supporting chiral selectivity not to vanish identically. In any component of a non-parametric signal – in particular the odd-parity quantum interference terms – rotational averaging effects a disentanglement of the radiation and molecular fields, $\mathbf{S}^{(r)}$ and $\mathbf{T}^{(r)}$ respectively. This averaging procedure results in a product of scalars (or, for tensors of rank four or more, a linear combination of such scalars), one scalar for the radiation and one for the molecule. Each scalar is derived by contracting the tensor, $\mathbf{S}^{(r)}$ or $\mathbf{T}^{(r)}$, with an isotropic tensor of the same rank. For example, in the E1-M1 interference term for photon absorption by a chiral molecule, the molecular tensor $\mathbf{T}^{(2)} = [\boldsymbol{\mu}][\mathbf{m}^*]$, on contraction with the isotropic tensor of rank 2 (the Kronecker delta), yields the scalar $T = (\boldsymbol{\mu}.\mathbf{m}^*)$, signifying that the electric and magnetic transition moments must not be orthogonal. Equally $\mathbf{S}^{(2)} = [\mathbf{e}][\mathbf{b}^*]$ yields the scalar $S = (\mathbf{e}.\mathbf{b}^*)$; this determines that chiral resolution vanishes when plane polarized light is employed, for then the field polarizations are real and the orthogonality of the electric and magnetic vectors gives $S = 0$. But for any circular (or even elliptical) polarizations, S is non-zero and it also changes sign on reversal of circularity;

$$S = \left(\mathbf{e}\cdot\mathbf{b}^*\right) \xrightarrow{\mathscr{I}} \left((-\mathbf{e})\cdot\mathbf{b}^*\right) = -S \ . \tag{3}$$

Consequently, chiral specificity is manifest. Equally, performing space inversion $\mathscr{I}$ on the molecular system, it is apparent that T takes opposite signs for each member of an enantiomeric pair, since;

$$T = (\boldsymbol{\mu}\cdot\mathbf{m}^*) \xrightarrow{\mathscr{I}} ((-\boldsymbol{\mu})\cdot\mathbf{m}^*) = -T \ . \tag{4}$$

Thus the rate of absorption of left-handed circularly polarized radiation, for example, is different for left- and right-handed enantiomers – though only marginally, because of the relative weakness of the salient interference terms compared to the dominant (usually electric dipole) diagonal contributions to the signal. Equally, each enantiomer exhibits a slightly different rate of absorption for left and for right-handed circular input. This is the physical origin of circular dichroism. Similar remarks apply to optically more intricate processes involving more than one photon, where the quantum amplitude itself comprises products of multipolar couplings, each with a definite spatial parity, and the key interference terms arise from products of these terms with opposite inversion symmetry. Again, for a chirally specific signal to emerge, it is necessary that the resultant radiation and molecular scalars do not vanish identically.

The above considerations bring us to the heart of the issue, addressing the possibility for engaging the chirality of a twisted beam with that of matter. The helicity of optical vortices, manifest in their wave-front structure, is exhibited in additional phase factors in the positive and negative frequency components. In particular, the analytic signal for the electric field of a Laguerre-Gaussian (LG) mode propagating in the z-direction is[9];

$$\mathbf{e}(\mathbf{r}) = \hat{\boldsymbol{\varepsilon}}\, f_{lp}(r) \exp\left[i(kz - l\varphi)\right] \ , \tag{5}$$

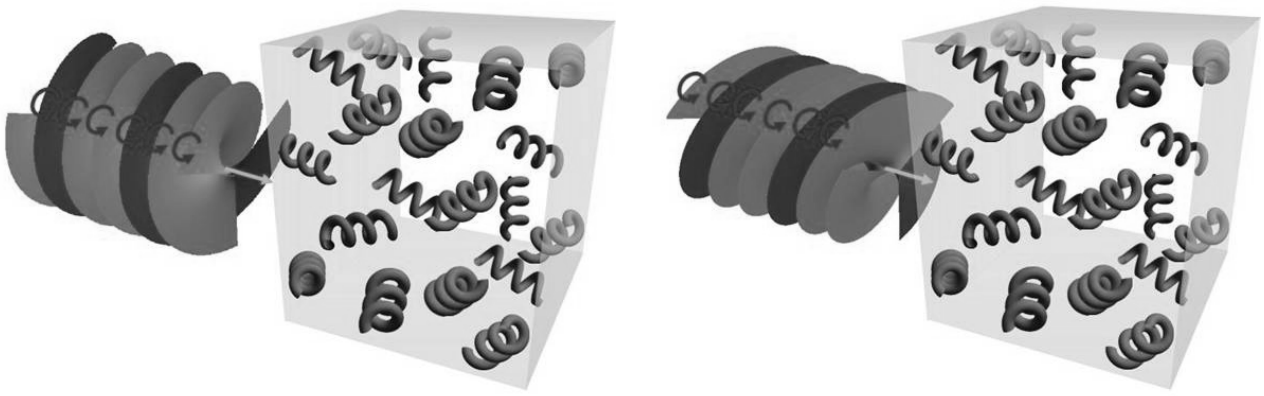
where $\hat{\boldsymbol{\varepsilon}}$ is the polarization vector (for either plane or circularly polarized light) and $f_{lp}(r)$ is a radial distribution function; l is the orbital angular momentum quantum number, for which a positive sign denotes left helicity and a negative sign, right. Of the two space-dependent terms in the field phase, the first is the origin of the wave-vector mismatch factor in equation (1), and the second is an azimuthal phase factor characteristic of twisted beams. The first component of the phase in (5) changes sign on space inversion just as the circular polarisation vectors behave. The second phase factor changes by addition of π on space inversion. The magnetic field for a twisted beam has an exactly similar form[7].

No difference should be expected between the behavior of right and left forms of any optical vortex if the photons it comprises are plane polarized and the material they interact with is achiral, in accordance with the dictates of parity. This is equivalent to the case of circularly polarized, planar wave-front (e.g. Gaussian) modes, interacting with achiral material. However, there are several differences between LG photons and those associated with planar wave-fronts. In the present context a significant difference is that plane waves have no restriction in the direction of propagation, while the symmetry of LG photons designates propagation in one specified (z) direction. This indicates that mirror inversion along a plane containing the z-axis is the relevant symmetry element. Arguing symmetry on these grounds is sufficient to establish that there is no differentiation between twisted beams of opposite handedness when achiral material is interrogated.

3. CHIRALITY, POLARIZATION CIRCULARITY, AND WAVEFRONT HELICITY

Consider first the simpler case of a vortex beam comprising plane polarized photons, interrogating a system of chiral molecules. Mirror inversion of either part of the system (molecule or radiation) gives a system different from the original, and a change in signal signifying chiral specificity might be anticipated. Representing in brackets the circularity of the radiation and the matter respectively, both cases require identity of (L, L) and (R, R), and so too (L, R) and (R, L). However (L, L) and (L, R) differ. More succinctly, denoting either handedness L/R by a circularity C and the reverse by C^*, we have (C,C) = (C^*,C^*) which may or may not be equal to (C,C^*). For the twisted beam case it is thus necessary to ascertain whether there is a mechanism, by means of which signals that are *permissibly* different in these terms may *in fact* differ. As discussed above, the observable for a fluid is obtained as the ensemble sum of contributions from all relevant centers. As in the case of plane waves, the factor $\exp(i\Delta\mathbf{k}.\mathbf{R})$ continues to play an important role and represents any net (longitudinal) optical phase shift associated with a process mediated by a chiral center ξ located at $\mathbf{R}_\xi = (r, \varphi, z)$. However another (azimuthal) phase factor of similar form, $\exp(-i\Delta l\varphi)$, now appears alongside it and also enters M_{fi}^{ξ} through the radiation tensor $\mathbf{S}^{(r)}$ (here $\Delta l\hbar$ represents any mismatch between the orbital angular momentum sums of all input and any output photons involved in the process). For both types of phase factor, any component that persists in the observable must average to zero. First, if the optical process is parametric, then $\Delta l = 0$ and the phase factor disappears; hence any corresponding dependence on the sign of the orbital angular momentum of the radiation is lost. The transition rate, proportional to $\left|\left\langle M_{fi}^{\xi}\right\rangle\right|^2$, has no dependence on the sign of the orbital angular momentum l and is therefore not a chiral discriminator. For a *non*-parametric process, even if $\Delta l \neq 0$, the incoherent character means that the ensemble rate is proportional to $\left\langle\left|M_{fi}^{\xi}\right|\right\rangle^2$ and the azimuthal phase factor cancels at each and every centre ξ; once again the observable transition rate does not involve the orbital angular momentum or its sign. Hence the circularity of the optical vortex is immaterial.

For chiral molecules interacting with twisted light comprising *circular* photons, there are eight possibilities for combinations of different handedness; each molecule, each photon and the vortex itself can be of either handedness, as illustrated in Figure 1. In a sense the problem reduces to that which applies to other beams of circular polarization – but now there is a mechanism for the associated signals to actually differ. Again the helicity of the wave-front is immaterial so far as chiral specificity is concerned. Consider for example a left-handed twisted beam comprising left-handed circular photons interacting with a system of left-handed molecular enantiomers – and the simplest optical process in which chirality can be manifest, namely circular dichroism. The quantum interference terms responsible for chiral discrimination change sign if either the circularity of the photons or the isomeric form of the molecules is changed; the interference terms are invariant to a change of both. No component of the signal changes if, for example, the beam is modified to a right-handed vortex but the circular sense of the photons and molecular handedness is unchanged.



(a) (b)

Figure 1. Interaction of left-circularly polarized radiation (represented by rotary arrows) with left-handed (randomly oriented) chiral molecules: (a) left-handed Laguerre-Gaussian beam, $l = 3$, and (b) right-handed Laguerre-Gaussian beam, $l = -3$. In each case the radiation is represented by surfaces of constant phase.

It is of interest to note one exceptional case, where certain of the above assumptions are unwarranted, concerning second harmonic generation by reflection at a chiral surface. This system has long-range orientational order, and local chirality is modified as the space within which surface molecules reside is no longer isotropic. The process has parametric character, because surface reflection satisfies a reduced requirement for wave-vector matching – one that applies only to the surface-parallel components of the two input and single output photon wave-vectors. As has been shown[10], a chirally sensitive response then emerges through electric dipole coupling alone.

4. NEMATIC LIQUID CRYSTALS

Nematic liquid crystals represent a prime example of systems in which long-range orientational order can engender chirality beyond the dimensions of individual molecules. As such the mechanical interactions of such systems with circularly polarized light, due to the intrinsic spin angular momentum of the latter, are well attested[11,12]. More recently, the utilization of optical torque to effect orientational control in nanoscale systems has also come under intense scrutiny[13,14]. Now that the principles determining the forces exerted by twisted optical beams have been established[15], it is timely to consolidate these concepts with a detailed appraisal of effects resulting from the throughput of twisted optical beams in nematic liquid crystals. Already it has been experimentally demonstrated that the transfer of spin and/or orbital angular momentum results in a rotation of the constituent molecules results and hence a modification of the local direction of alignment[16], through a process of stimulated light scattering.

The physical effect of primary interest in the interaction of an Laguerre-Gaussian beam with a nematic liquid crystal concerns the angular distribution of the director $\hat{\mathbf{n}}(\mathbf{r})$ in the region illuminated by the beam. Figure 2 schematically depicts a case in which the liquid crystal occupies the half-space z/ge_0 and the LG beam is assumed to be incident upon the system in such a manner that the beam waist coincides with the plane $z = 0$. In the absence of the light, assuming isotropic elastic properties for the liquid crystal, the free energy of the system takes the simple form;

$$\mathcal{F}_0(r) = \tfrac{1}{2} K \left\{ \left[\nabla \cdot \mathbf{n}(r) \right]^2 + \left[\nabla \times \mathbf{n}(r) \right]^2 \right\} , \tag{6}$$

where K is a constant characteristic of the liquid crystal. The symmetry of the problem suggests that the director $\hat{\mathbf{n}}(\mathbf{r})$ can be expressed in terms of the local azimuthal angle $\psi(\mathbf{r})$ such that $\hat{\mathbf{n}}(\mathbf{r}) = (\sin\psi, \cos\psi, 0)$. This permits consideration in terms of $\psi(\mathbf{r})$ and the free energy can be shown to reduce to the simple form $\mathcal{F}_0 = \tfrac{1}{2} K \left[\nabla \Psi \right]^2$. The presence of the twisted light leads to the introduction of an additional interaction term that can be written succinctly in the following

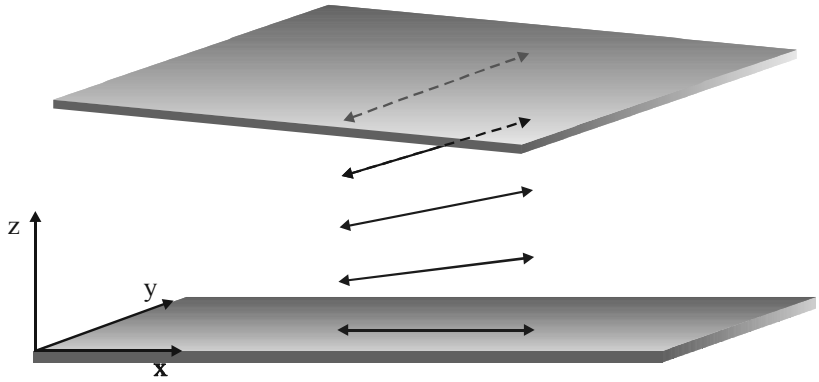


Figure 2. Director orientations in a twisted nematic liquid crystal.

form, where the coupling constant $\Lambda_{l,p}(r)$ depends on a number of parameters involving both the light and the liquid crystal;

$$\mathcal{F}_{\text{int}} = \Lambda_{l,p}(r)\sin^2\Psi(\mathbf{r}) \ . \tag{7}$$

For emphasis, only the dependence on the mode numbers l and p are explicitly indicated. In the situation considered here the spatial dependence of ψ is only on the cylindrical variables r and z and there is no dependence on ϕ. Thus the total free energy takes the form;

$$\mathcal{F} = \mathcal{F}_0 + \mathcal{F}_{\text{int}} = \tfrac{1}{2}K\left[\nabla\Psi(r,z)\right]^2 + \Lambda_{l,p}\sin^2\Psi(r,z) \, . \tag{8}$$

Adopting the standard Landau formalism to minimize the free energy of the system now leads to the following partial differential ψ field equation;

$$K\left\{\frac{\partial^2\Psi}{\partial r^2} + \frac{1}{r}\frac{\partial\Psi}{\partial r} + \frac{\partial^2\Psi}{\partial z^2}\right\} + \Lambda_{l,p}\sin 2\Psi(r,z) = 0 \ . \tag{9}$$

A solution of equation (9) is numerically tractable, and results have been obtained for a variety of systems subject to appropriate boundary conditions,

$$\Psi(r,0) = 0; \ \ \Psi(r,\infty) = 0; \ \ \Psi(\infty, z>0) = 0 \quad . \tag{10}$$

To illustrate the ensuing results, it is instructive to consider an oriented nematic liquid crystal system that has recently been the subject of attention with regard to another form of optically induced reorientation – in that case obliquely incident plane polarized light leading to a secondary instability after the stationary Freedericksz transition[17]. The present analysis focuses on the same system, nematic 5CB (pentylcyanotriphenyl) doped with an anthraquinone derivative dye (AD1), here subject to a Laguerre-Gaussian throughput beam. Numerical results have been secured using equation (9) with the appropriate parameter value, $K = 0.64 \times 10^{-12}$, and for a beam with a realistic intensity of 10^8 W cm^{-2}, wavelength $\lambda = 600$ nm, and beam-waist $w_0 = 35\,\lambda$, also taking the simplest beam structure associated with orbital angular momentum, i.e. $l = 1$, and $p = 0$. As shown in Figure 3, the most prominent mechanical effect of the twisted beam is to significantly modulate the value of ψ along the z-axis, with a radial fall-off that largely reflects the intensity distribution. Note, however, that the minima and maxima are interchanged on moving outwards from the beam center, and that in consequence there is a characteristic radial position at which the principal modulation is entirely absent. Currently ongoing work is further addressing the consequences of changes in the coupling parameters, especially the l value that quantifies the orbital angular momentum.

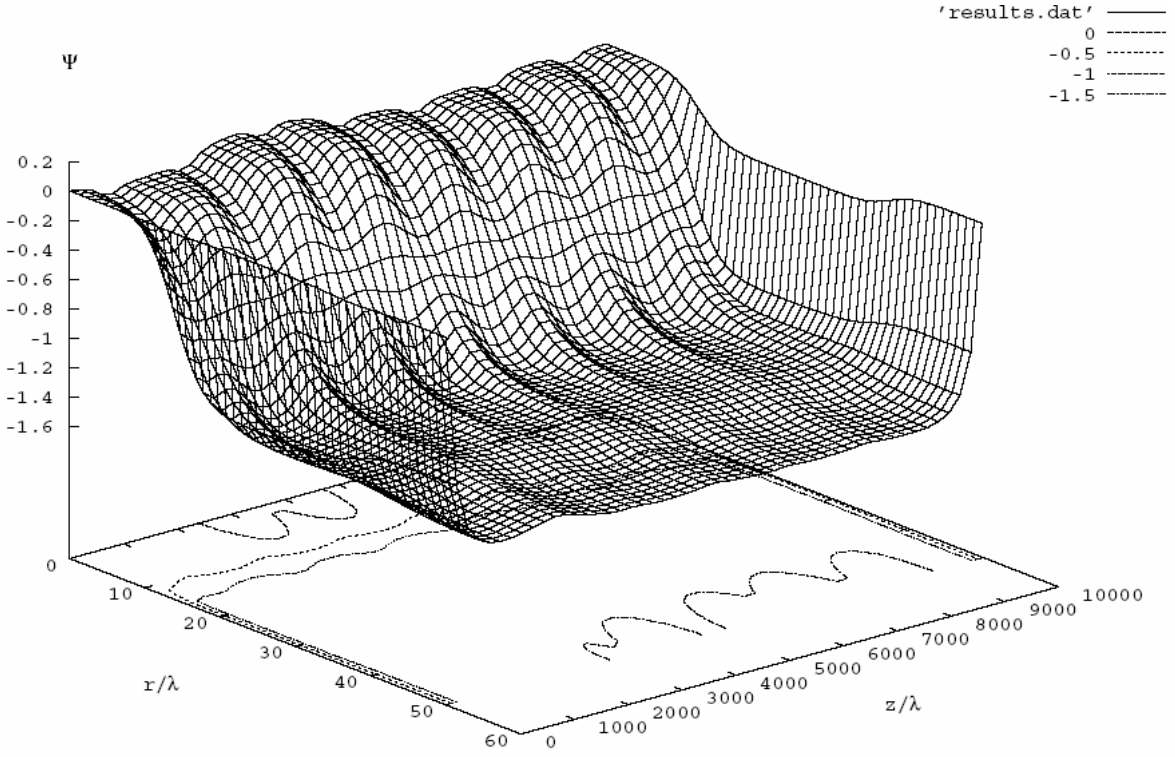


Figure 3. Results of a simulation on a 5CB-AD1 dye-doped nematic liquid crystal, subject to the throughput of a twisted optical beam – parameter values given in the main text.

5. CONCLUSION

This paper reports two main results. First, through an analysis of the grounds for chiroptical behavior involving electric and magnetic dipole interactions, it has been established that the helicity of optical vortices cannot engage through any parametric or non-parametric optical process with the chirality of an isotropic molecular system, other than through circularity of its photons. Thus, the manifestations of orbital angular momentum differ markedly from those associated with photon spin angular momentum, significantly limiting any scope for enantiomerically specific photochemical or spectroscopic exploitation. Secondly, it has been demonstrated how to develop an analytically sound and numerically tractable representation of the mechanical effects of twisted beams on systems with longer-range translational-orientational order, notably twisted nematic liquid crystals. The preliminary results are encouraging for the prospects of new forms of optomechanical control.

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