



Enantioselective optical gradient forces using 3D structured vortex light

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ABSTRACT

Here we highlight enantioselective optical gradient forces present in 3D (non-paraxial) structured optical vortex tweezing systems. One chiral force originates from the circular polarization of the light, while remarkably the other is independent of the input polarization - even occurring for unpolarized light - and is not present in 2D structured light nor propagating plane waves. This latter chiral sorting mechanism allows for the enantioselective trapping of chiral particles into distinct rings in the transverse plane through conservative radial forces.

1. Introduction

Light conveys energy and momentum - the basis for optical manipulation of matter [1]. Optical trapping encapsulates techniques involving one or more lasers that enable the three-dimensional control of particles via non-contact optical forces [2–4]. The *non-conservative* radiation pressure force stemming from the transfer of optical linear momentum acts to push particles in the direction of propagation (often referred to as the scattering force, though in terms of photons it is absorption and non-forward scattering); the *conservative* optical gradient force causes particles to be attracted to regions of high intensity (which is in fact the forward elastic scattering of photons); optical binding forces exist *between* particles [5].

An important class of laser modes are structured modes [6], and more specifically those that carry orbital angular momentum (OAM), known as optical vortices or twisted light [7]. Applications of optical vortices are widespread and diverse, including optical communications, quantum entanglement, nanofabrication techniques, astronomy, microscopy and imaging, etc. [8,9]. Optical tweezers using twisted light has seen much activity [10–12], predominantly because they can transfer their OAM and induce mechanical motion of trapped particles. This transfer of OAM to cause rotational motion is completely distinct from the gradient trapping force itself; optical angular momentum is conserved in the gradient force. Until now, as far as we are aware, no optical gradient force trapping scheme has utilized the OAM in a direct manner, i.e., a non-mechanical way.

The OAM of optical vortices is quantified in discrete units of $\ell\hbar$ per photon where $\ell \in \mathbb{Z}$, and the sign of ℓ designates what direction the helical wavefront twists: for $\ell > 0$ it is left-handed, while $\ell < 0$ is right-handed. This chirality of the wavefront in optical vortices is analogous to the well known optical chirality of circularly polarized

light (CPL), where the quantity often referred to as the helicity $\sigma = +1$ corresponds to left-handed CPL and $\sigma = -1$ right-handed CPL, and the spin angular momentum (SAM) of CPL is given by $\sigma\hbar$ per photon. The chirality of optical vortices has witnessed a profusion of research activity, with the field recently surveyed [13]. An object is chiral if it can be distinguished from its mirror image, this manifests most obviously in material objects like human hands but equally applies to molecules and nanostructures [14]. An enantiomer is one of either the right- or left-handed stereoisomer in the enantiomeric pair. Generally, each one of the enantiomeric pair interacts differentially with other chiral objects, and chiroptical techniques involve using the chirality of light to probe this chirality of matter [15–18]. The question addressed in this paper is whether the OAM, both its sign (handedness) and magnitude, can influence the optical gradient trapping force when applied to Rayleigh-sized chiral particles. Specifically, we are concerned with whether an optical vortex can produce enantioselective trapping forces (i.e. chiral trapping forces which act to separate the enantiomeric pair) which depend on ℓ . Previous studies looking at enantioselective optical trapping forces were specifically concerned with circularly polarized unstructured or plane wave light interacting with small chiral particles [19–22]. Being able to optically sort chiral molecular enantiomers using all-optical methods at the nanoscale would have a profound impact on the drug and pharmaceutical industry. Beyond natural molecules, chiral trapping forces have been employed in a number of different setups, predominantly involving chiral nanoparticles and plasmonic tweezing systems [23–36], though there have been some studies involving structured laser light [31,37–41].

2. Conservative chirality-sorting forces

Quantum electrodynamics (QED) [42] is employed to describe the light-matter interactions that correspond to optical gradient forces

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in this paper. Truncated to the dipole approximation, the Power-Zienau-Woolley (PZW) multipolar interaction Hamiltonian describes this coupling as [43]

$$H_{\text{int}}(\xi) = -\epsilon_0^{-1} \mu_i(\xi) d_i^\perp(\mathbf{R}_\xi) - m_i(\xi) b_i(\mathbf{R}_\xi), \quad (1)$$

where $\mu_i(\xi)$ and $m_i(\xi)$ are the electric and magnetic dipole operators, respectively; $d_i^\perp(\mathbf{R}_\xi)$ and $b_i(\mathbf{R}_\xi)$ are the transverse (with respect to the Poynting vector) electric displacement field and magnetic field mode operators, respectively, acting on a particle ξ at the location \mathbf{R}_ξ ; Einstein summation of repeated tensor indices is assumed throughout, i.e. $a_i b_i = \mathbf{a} \cdot \mathbf{b}$. The first term in (1) is the electric dipole coupling (E1) and the second magnetic dipole coupling (M1). For input circularly-polarized Laguerre–Gaussian (LG) modes the electromagnetic free field expansion operators truncated to first-order in the paraxial parameter $k w_0$ may be given by [44,45]

$$d_i^\perp(\mathbf{r}) = \sum_{k,\sigma,\ell,p} \Omega \left[\left\{ e_i^{(\sigma)} + \frac{i}{\sqrt{2}k} \left(\frac{\partial}{\partial r} - \frac{\ell}{r} \right) e^{i\sigma\phi} \hat{z}_i \right\} \times f_{|\ell|,p}(r) a_{|\ell|,p}^{(\sigma)}(k\hat{\mathbf{z}}) e^{i(kz+\ell\phi)} - H.c. \right] \quad (2)$$

and

$$b_i(\mathbf{r}) = \sum_{k,\sigma,\ell,p} \frac{\Omega}{\epsilon_0 c} \left[\left\{ b_i^{(\sigma)} + \frac{1}{\sqrt{2}k} \left(\sigma \frac{\partial}{\partial r} - \frac{\ell}{r} \right) e^{i\sigma\phi} \hat{z}_i \right\} \times f_{|\ell|,p}(r) a_{|\ell|,p}^{(\sigma)}(k\hat{\mathbf{z}}) e^{i(kz+\ell\phi)} - H.c. \right], \quad (3)$$

where $\Omega = i(\hbar c k \epsilon_0 / 2 A_{\ell,p}^2 V)^{1/2}$ is the normalization constant for LG modes, $e_i^{(\sigma)}(b_i^{(\sigma)}) = \sqrt{2}^{-1}(\hat{x}(\hat{y}) \pm i\sigma\hat{y}(\hat{x}))_i$ represents the electric (magnetic) polarization vector for circularly polarized light with $\sigma = \pm 1$ the helicity, with V the quantization volume; $a_{|\ell|,p}^{(\sigma)}(k\hat{\mathbf{z}})$ is the annihilation operator; $\exp i(kz + \ell\phi)$ is a phase term; and $H.c.$ stands for Hermitian conjugate. The terms that depend on $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are the transverse components of the fields while those that depend on $\hat{\mathbf{z}}$ are the longitudinal components. 2D structured light possesses only the transverse components, whilst 3D structured light also has the longitudinal component [6]. Furthermore, whilst the polarization state of 2D structured light is adequately described by 2D polarization theory, e.g. the four Stokes parameters, due to the fact all three spatial components of the field vectors (2) and (3) generally play a role in 3D structured light, the theory of polarization has been extended to the 3D case [46–49]. In this work we describe light by its source 2D polarization state and make clear whether it is paraxial (2D structured) or non-paraxial (3D structured). To generate significant longitudinal field components, a 2D structured laser source is tightly focused using a high numerical aperture (NA) lens, producing 3D structured light at the focal plane. $f_{|\ell|,p}(r)$ is a radial distribution function given as

$$f_{|\ell|,p}(r) = \frac{C_p^{|\ell|}}{w_0} \left(\frac{\sqrt{2}r}{w_0} \right)^{|\ell|} e^{-\frac{r^2}{w_0^2}} L_p^{|\ell|} \left[\frac{2r^2}{w_0^2} \right], \quad (4)$$

where $C_p^{|\ell|} = \sqrt{2p!/\pi(p+|\ell|)!}$ is a normalization constant, w_0 is the minimum transverse extent of the beam around $z = 0$ (beam waist), and $L_p^{|\ell|}$ is the generalized Laguerre polynomial, p being the radial index. The potential energy responsible for the optical gradient trapping force originates in forward Rayleigh scattering. In photonic terms, an input laser photon is annihilated at the particle and then an identical photon (same mode) is created at the same particle. This two photon interaction requires second-order perturbation theory to yield the leading order contribution to the potential energy [1]:

$$U = \text{Re} \sum_R \frac{\langle I | H_{\text{int}} | R \rangle \langle R | H_{\text{int}} | I \rangle}{E_R - E_I}, \quad (5)$$

where the initial state of the system is given by $|I\rangle = |E_0(\xi); n(k, \sigma, \ell, p)\rangle$: the particle is in the ground state and the radiation field consists of n photons in the mode (k, σ, ℓ, p) ; the final state of the system is identical to the initial; the virtual intermediate state is given by $|R\rangle = |E_\alpha(\xi); (n-1)(k, \sigma, \ell, p)\rangle$.

Working strictly in the dipole regime the potential energy of a particle is then a sum of three distinct contributions:

$$U \simeq U_{\text{E1E1}} + U_{\text{E1M1}} + U_{\text{M1M1}}. \quad (6)$$

Chiral effects dependent on material handedness originate in the E1M1 term and so from now on we neglect the pure E1E1 and M1M1 effects as they are independent of material chirality [15].

3. 2D circularly polarized 3D LG input

Using Eq. (1) with the fields Eqs. (2) and (3) in Eq. (5) we produce the following potential energy for a 2D circularly polarized 3D LG input

$$U_{\text{E1M1}}^{\text{Circ}} = -\frac{I}{2\epsilon_0 c^2} \text{Re} \left[\left\{ \bar{e}_i^\sigma f - \frac{i}{k} \left(f' - \frac{\ell}{r} f \right) e^{-i\sigma\phi} \hat{z}_i \right\} \times \left\{ b_j^\sigma f + \frac{1}{k} \left(\sigma f' - \frac{\ell}{r} f \right) e^{i\sigma\phi} \hat{z}_j \right\} G_{ij} + c.c. (i \leftrightarrow j) \right], \quad (7)$$

where we have dropped most dependencies for notational clarity, $c.c. (i \leftrightarrow j)$ stands for taking the complex conjugate and swapping the indices of the expression in brackets, $I = \hbar c^2 k / A_{\ell,p}^2 V$ is the beam intensity, $f' = \partial_r f_{|\ell|,p}(r)$, and the imaginary quantity referred to as the mixed electric–magnetic polarizability tensor (which has different signs for each chiral particle of an enantiomeric pair) is given explicitly as

$$G_{ij}(\omega, -\omega) = \sum_\alpha \left(\frac{\mu_i^{0\alpha} m_j^{\alpha 0}}{E_{\alpha 0} - \hbar\omega} + \frac{m_j^{0\alpha} \mu_i^{\alpha 0}}{E_{\alpha 0} + \hbar\omega} \right), \quad (8)$$

where the transition dipole moments are defined as $\mu_i^{0\alpha} (m_i^{0\alpha}) = \langle E_0 | \mu_i (m_i) | E_\alpha \rangle$ and $E_{\alpha 0} = E_\alpha - E_0$. To account for the randomly oriented nature of chiral particles in the liquid or gas phase the potential energy (7) must be rotationally averaged. Using well known methods [43] in which each particle is decoupled from the space-fixed frame into the laboratory frame we produce

$$\langle U_{\text{E1M1}}^{\text{Circ}} \rangle = -\frac{I}{3\epsilon_0 c^2} \left[\sigma f^2 + \frac{1}{2k^2} \left(\sigma f'^2 - \frac{2\ell}{r} f f' + \frac{\sigma \ell^2}{r^2} f^2 \right) \right] \text{Im} G_{\lambda\lambda}, \quad (9)$$

The optical force \mathbf{F} can be calculated from the energy shift via the well-known relation: $\mathbf{F} = -\text{Re}\nabla U$, its explicit form is given in Supplement 1. The potential energy (9) is plotted in Fig. 1 with the direction of the forces \mathbf{F} overlaid for the parallel $\ell = |\sigma|$ and antiparallel $\ell = -|\sigma|$ cases (further plots can be found in Supplement 1). This enantioselective trapping mechanism is quite similar to that of circularly polarized plane waves or 2D structured light, except in our system we have the spin–orbit-interactions (SOI) which occur in 3D fields [50] and these influence the helicity density distributions. For example, in the antiparallel case we produce an on-axis helicity density for $|\ell| = 1$.

4. 2D polarization independent enantioselective force

While the physics of the mechanism in the previous section is similar to that of previous chiral gradient forces [19,21,51,52], we will now highlight a distinct enantioselective trapping mechanism which is independent of the 2D polarization state. Optical helicity density h for propagating plane waves, 2D structured fields, and evanescent waves is proportional to the degree of 2D circular polarization of the input beam $h \propto \sigma$, being zero for both linearly polarized and unpolarized inputs and taking on its maximum value for circular polarization. For both linearly polarized and unpolarized plane waves there is no possibility for enantioselective gradient forces because $\sigma = 0$ and thus their optical helicity $h = 0$. In comparison, 3D structured LG modes are known to possess a non-zero optical helicity density contribution for 2D linear polarizations [45,53] and it has recently been shown that remarkably this contribution is in fact independent of 2D polarization, persisting even

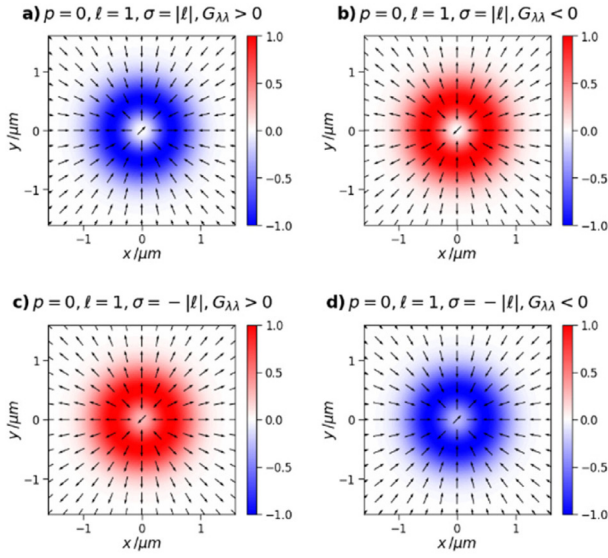


Fig. 1. Individually normalized enantioselective potential energy (9) with forces overlaid (arrows indicate only direction) $w_0 = \lambda$ in all plots.

for unpolarized input light [54]. The rotationally averaged gradient trapping potential energy due to an 2D unpolarized 3D LG beam may be calculated by averaging (9) over the two orthogonal polarizations $\sigma = 1$ and $\sigma = -1$ (see Supplement 1 for further details):

$$\langle U_{\text{EIM1}}^{\text{ind}} \rangle = \frac{I}{3\epsilon_0 c^2} \frac{\ell}{k^2 r} f' f \text{Im} G_{\lambda\lambda}. \quad (10)$$

The gradient forces acting on a chiral particle stemming from this 2D polarization independent potential energy are calculated as

$$\begin{aligned} \langle \mathbf{F}_{\text{EIM1}}^{\text{ind}} \rangle = & \hat{r} \frac{I}{3\epsilon_0 c^2} \left(\frac{C_p^{|\ell|}}{w_0} \right)^2 \left(\frac{\sqrt{2}r}{w_0} \right)^{2|\ell|} e^{-\frac{2r^2}{w_0^2}} \frac{8\ell}{k^2} \\ & \times \left[\left(\frac{|\ell|}{w_0^2 r} - \frac{r}{w_0^4} \right) L_p^{2|\ell|} + \left(\frac{2|\ell|}{w_0^2 r} - \frac{4r}{w_0^4} \right) L_p^{|\ell|} L_{p-1}^{|\ell|+1} \right. \\ & \left. - \frac{2r}{w_0^4} \left(L_{p-1}^{2|\ell|+1} + L_p^{|\ell|} L_{p-2}^{|\ell|+2} \right) \right] \text{Im} G_{\lambda\lambda}. \end{aligned} \quad (11)$$

The potential energy (10) with the direction of the forces (11) overlaid at the focal plane $z = 0$ are shown in Fig. 2. What the graphs show is that for a given sign of ℓ , the right-handed and left-handed chiral particles in an enantiomeric mixture are subject to discriminatory radial trapping forces which act to separate them into distinct rings in the transverse plane. For example, in Fig. 2 the enantiomer with $G > 0$ is pushed away from the beam axis and towards the blue (outer) ring, meanwhile in 2b it shows the $G < 0$ is pushed towards the central spot and away from the outer ring. The results are dependent on the sign of ℓ , and keeping everything else the same, for $-\ell$ the graphs in Fig. 2 are the same but with the signs reversed. For example, for $-\ell$ the 2a $G > 0$ would look exactly like 2b. It is important to make clear that these 2D polarization independent enantioselective forces are identical for either a 2D linearly polarized or 2D unpolarized 3D structured optical vortex. Also crucial is that both (10) and (11) stem purely from the longitudinal fields of (2) and (3) and so the 3D structure of the optical vortex beam generated by a tight focus is essential.

5. Discussion and conclusions

The chiral forces derived here using perturbative QED can alternatively be interpreted as the force arising from the gradient of the optical helicity density h [35]. It is clear that the forces in Figs. 1 and 2 are proportional to $(\text{Im} G_{\lambda\lambda}) \nabla h$, where h is given by Eqs. (14) and (17) in Ref. [45], respectively. We note that our *conservative* discriminatory

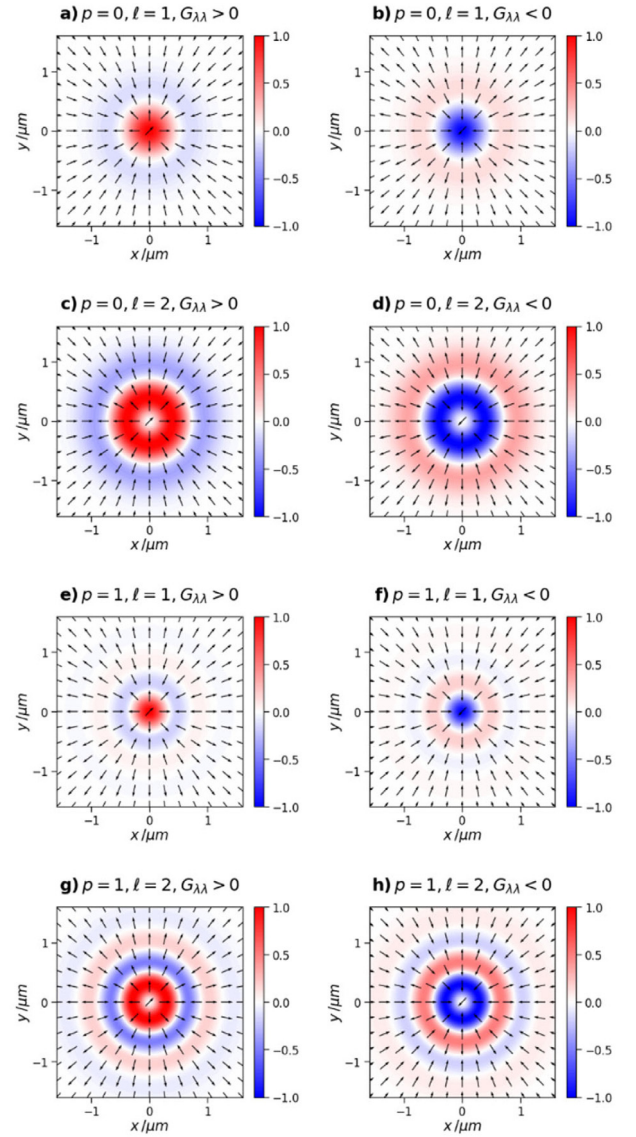


Fig. 2. Individually normalized 2D polarization independent enantioselective potential energy (10) with vector optical force (11). Otherwise as Fig. 1 (arrows indicate only direction).

trapping force derived using QED relates to the $-\omega 4\pi \text{Re} \{ \chi \} \nabla h$ term derived with a classical field.

In Eq. 2 of Ref. [35]. For the propagating plane wave and evanescent wave modelled in that study and elsewhere [55] the helicity density is $h \propto \sigma$ and so for linear or randomly polarized propagating plane or evanescent waves conservative enantioselective forces analogous to our 3D structured vortex 2D polarization independent force (11) do not exist. The lateral *non-conservative* optical forces presented in Ref. [35] are due to transverse spin angular momentum density [56] in evanescent waves. Although the evanescent fields have 2D polarization independent transverse spin density [57], they possess canonical momentum density only in the direction of propagation, and so projecting the transverse spin onto the momentum density in evanescent waves produces zero helicity. We have highlighted a similar chirality-sorting force as that discovered by Hayat et al. [35] but ours is a conservative gradient force in 3D optical vortex tweezing systems, its magnitude is proportional to the tightness of the focus, but also increases for higher p values as it stems from the gradients of the field.

Evidently for natural chiral particles (e.g. molecules) the magnitudes of the forces involved will be small due their small $G_{\lambda\lambda}$ alongside the fact there will be present the dominant chirality-independent trapping potential U_{EIEI} from (6), however they are considered experimentally distinguishable [1,21,51,52], though are yet to be observed. Experimental realization of the fundamental mechanisms outlined here therefore have more potential in systems comprising of nanoparticles or the use of plasmonic enhancement in nanophotonic setups [27,58,59], and in particular the newly proposed method of force detection by Sifatet al. [60] is readily amenable.

This work has provided further evidence of the significant potential of 3D structured light in light-matter interactions and nanophotonics [6]. To be clear, while enantiomer separation schemes utilizing the optical helicity of circularly polarized plane waves in gradient force mechanisms have been put forward [19,21], linearly polarized and unpolarized propagating plane waves possess no longitudinal fields or optical helicity and so the 2D polarization independent discriminatory force (11) could never have been envisaged under the plane wave approximation, nor a 2D structured light field.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.optcom.2022.128197>.

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