Realization of optical pulling forces using chirality

Kun Ding,[1,2] Jack Ng,[3,*] Lei Zhou,[2] and C. T. Chan[1,†]

[1]*Department of Physics and Institute for Advanced Study, The Hong Kong University of Science and Technology, Clear Water Bay, Hong Kong*

[2]*Department of Physics, Fudan University, Shanghai, China*

[3]*Department of Physics and Institute of Computational and Theoretical Studies, Hong Kong Baptist University, Kowloon Tong, Kowloon, Hong Kong*

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The optical force acting on a chiral particle is qualitatively different from that acting on an achiral particle due to chirality-dependent forces which couple mechanical linear momentum with optical spin angular momentum. We show that such chirality-induced coupling can serve as a mechanism to realize optical pulling forces that can be predicted analytically and are also observed in full wave simulations for chiral structures.

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I. INTRODUCTION

The majority of research in optical micromanipulation focuses on the interaction of light with isotropic spherical particles, such as those that make up polystyrene, glass, or metallic beads [1–6]. These simple spherical particles serve as prototypes for gaining an intuitive understanding of light-matter interactions [7–9]. However, particles in nature typically have lower symmetries and this can give rise to phenomena that are qualitatively different [10–19]. In practical applications, we often deal with aggregates of particles with complicated shapes and geometry [20–22]. The optical forces acting on such particles or their aggregates are not well understood, although it is generally assumed that the physics is not that different from that concerning a perfectly spherical entity. In this paper, we consider the optical micromanipulation of a class of chiral particles. In particular, we show analytically and numerically that the chirality-induced effects allow light to attract a chiral particle.

We shall first analytically derive the optical force expression for a small anisotropic chiral particle within the dipole approximation. The force expression is then derived for the special case of an isotropic chiral particle, where the analytic description is much simpler to interpret. We shall see that chirality-dependent forces that couple the mechanical linear momentum of photons with the angular momentum of photons will emerge, giving rise to counterintuitive phenomena such as optical pulling forces owing to the chirality of the particles. The possibility of realizing optical pulling forces through chirality is then confirmed by accurate numerical simulations based on scattering theory.

The chirality of a particle is manifested only when the particle interacts with another chiral entity. Electromagnetic waves can have a chiral character, and for a monochromatic wave, the time-averaged chirality flux may be defined as [23–26]

$$\langle \boldsymbol{\Psi} \rangle = \langle \boldsymbol{\Psi}_e \rangle + \langle \boldsymbol{\Psi}_m \rangle\,, \tag{1}$$

where

$$\langle \boldsymbol{\Psi}_e \rangle = \frac{\varepsilon_0 \omega}{4}\mathrm{Im}\{\mathbf{E}\times\mathbf{E}^*\} \quad \text{and} \quad \langle \boldsymbol{\Psi}_m \rangle = \frac{\mu_0 \omega}{4}\mathrm{Im}\{\mathbf{H}\times\mathbf{H}^*\} \tag{2}$$

are the portions of the chirality flux associated with the electric and magnetic fields, respectively. We note that $\langle \boldsymbol{\Psi}_e \rangle$ and $\langle \boldsymbol{\Psi}_m \rangle$ are proportional to the time-averaged spin density in the paraxial limit.

The left- and right-circularly polarized plane waves have opposite chirality and will interact with a chiral particle in different ways. This can lead to different or even completely opposite optical forces. In the case of optical forces acting on achiral particles, additional chirality-dependent terms must be added to the typical expressions. Our task is to highlight the unique role of these chirality-dependent terms in optical pulling.

II. FORCE FORMULA

To illustrate the essence of the physics, let us consider the dipolar limit in which a small chiral particle is characterized by an induced electric dipole moment $\mathbf{p}$ and a magnetic dipole moment $\mathbf{m}$ given by [27]

$$\mathbf{p} = \overleftrightarrow{\boldsymbol{\alpha}}^{ee}\mathbf{E} + \overleftrightarrow{\boldsymbol{\alpha}}^{em}\mathbf{B}, \quad \mathbf{m} = (-\overleftrightarrow{\boldsymbol{\alpha}}^{em})^T\mathbf{E} + \overleftrightarrow{\boldsymbol{\alpha}}^{mm}\mathbf{B}, \tag{3}$$

where the superscript T denotes transpose, and $\overleftrightarrow{\boldsymbol{\alpha}}^{ee}$, $\overleftrightarrow{\boldsymbol{\alpha}}^{mm}$, and $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$ are respectively the electric, magnetic, and chiral polarization tensors of the particle. A chiral particle features a nonzero $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$, due either to the chirality of the constituting molecules or the chiral shape of the particle. While the chirality of naturally occurring substances is usually weak, that of artificial nanostructures or microstructures may be enhanced by resonance [28–31]. Substituting (3) into the time-averaged optical force expression in Ref. [32] gives (see Appendix A)

$$\begin{aligned}\langle \mathbf{F} \rangle = {} & -\boldsymbol{\nabla}\langle U_f \rangle + \frac{1}{4}\mathrm{Re}\{\mathbf{T}^{ee}:\overleftrightarrow{\boldsymbol{\alpha}}^{ee} + \mathbf{T}^{em}:\overleftrightarrow{\boldsymbol{\alpha}}^{em} \\ & + \mathbf{T}^{mm}:\overleftrightarrow{\boldsymbol{\alpha}}^{mm} + \mathbf{T}^{me}:\overleftrightarrow{\boldsymbol{\alpha}}^{em,T}\} - \frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{\tilde{\mathbf{L}} + \tilde{\mathbf{S}}\},\end{aligned} \tag{4}$$

where $\langle f \rangle$ denotes the time-averaged value of f, and

$$\langle U_f \rangle = -\tfrac{1}{4}\mathrm{Re}(\mathbf{E}\cdot\mathbf{p}^*) - \tfrac{1}{4}\mathrm{Re}(\mathbf{B}\cdot\mathbf{m}^*) \tag{5}$$

is a scalar which can be interpreted as the time-averaged free energy. The tensors $\mathbf{T}^{ee}$, $\mathbf{T}^{em}$, $\mathbf{T}^{me}$, and $\mathbf{T}^{mm}$ have components

*jacktfng@hkbu.edu.hk

†phchan@ust.hk

of the following form,

$$T^{ee}_{n,ji} = [(\partial_n|E_i|)|E_j| - (\partial_n|E_j|)|E_i| - i|E_i||E_j|\partial_n(\Theta_i + \Theta_j)] \times e^{i(\Theta_j - \Theta_i)},$$
$$T^{em}_{n,ki} = [(\partial_n|E_i|)|B_k| - (\partial_n|B_k|)|E_i| - i|E_i||B_k|\partial_n(\Theta_i + \Phi_k)] \times e^{i(\Phi_k - \Theta_i)}, \quad (6)$$

$$T^{mm}_{n,ji} = [(\partial_n|B_i|)|B_j| - (\partial_n|B_j|)|B_i| - i|B_i||B_j|\partial_n(\Phi_i + \Phi_j)] \times e^{i(\Phi_j - \Phi_i)},$$
$$T^{me}_{n,ki} = [(\partial_n|E_k|)|B_i| - (\partial_n|B_i|)|E_k| + i|B_i||E_k|\partial_n(\Phi_i + \Theta_k)] \times e^{i(\Theta_k - \Phi_i)}, \quad (7)$$

where Θ_j and Φ_j are the phases of the electric and magnetic fields defined by $\mathbf{E} = |E_j|e^{i\Theta_j}\hat{x}_j$ and $\mathbf{B} = |B_j|e^{i\Phi_j}\hat{x}_j$, respectively, and

$$\tilde{\mathbf{L}} = \mathbf{E}\cdot(\overleftrightarrow{\boldsymbol{\alpha}}^{em*} \times \overleftrightarrow{\boldsymbol{\alpha}}^{ee})^T \cdot \mathbf{E}^* - \mathbf{B}\cdot(\overleftrightarrow{\boldsymbol{\alpha}}^{mm\dagger} \times \overleftrightarrow{\boldsymbol{\alpha}}^{em})^T \cdot \mathbf{B}^*,$$
$$\tilde{\mathbf{S}} = \mathbf{B}\cdot(\overleftrightarrow{\boldsymbol{\alpha}}^{em*} \times \overleftrightarrow{\boldsymbol{\alpha}}^{em})^T \cdot \mathbf{E}^* - \mathbf{E}\cdot(\overleftrightarrow{\boldsymbol{\alpha}}^{mm\dagger} \times \overleftrightarrow{\boldsymbol{\alpha}}^{ee})^T \cdot \mathbf{B}^*. \quad (8)$$

It is clear that all terms that depend on $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$ are related to the chirality of the particle. For a plane wave and for propagation-invariant beams which propagate in the z direction, we have $\partial_z|B_i| = \partial_z|E_k| = 0$. If we assume that $\mathrm{Re}(\alpha^{em}_{ij}) \ll \mathrm{Im}(\alpha^{em}_{ij})$, then the general force expression in the z direction becomes

$$\langle\mathbf{F}\rangle_z = \frac{1}{4}\mathrm{Re}\{\mathbf{T}^{ee} : \overleftrightarrow{\boldsymbol{\alpha}}^{ee} + \mathbf{T}^{mm} : \overleftrightarrow{\boldsymbol{\alpha}}^{mm}\}_z - \frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{\tilde{\mathbf{L}} + \tilde{\mathbf{S}}\}_z. \quad (9)$$

Let us now analyze the force formula (9) from the symmetry point of view. Let the incident beam propagate in the z direction, and we consider the sign of the optical force $\langle\mathbf{F}\rangle_z$ terms in the direction of the beam as we change the chirality of the particle inside the beam from left handed to right handed. If we change the chirality of a particle inside the incident beam from left handed to right handed, $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$ changes sign but not $\tilde{\mathbf{S}}$ as it contains the square of $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$. The first term in Eq. (9) also does not change sign. As these forces are proportional to the projection of the linear momentum along the z axis, they generally "push" the particle forward. On the other hand, the $\tilde{\mathbf{L}}$ term derived from the spin angular momentum has opposite signs for particles of different handedness as it is first order in $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$. In addition, the optical force induced by $\tilde{\mathbf{L}}$ does not depend on the sign of k_z. As such, it can potentially give rise to a force in the opposite direction to the beam.

To see the physics more clearly, we consider the special case of a small isotropic chiral particle. The rather formal Eq. (4) then takes a form that makes individual terms easier to interpret and consequently the physics of chirality-induced optical forces easier to understand. We consider a particle with isotropic polarizabilities: $(\overleftrightarrow{\boldsymbol{\alpha}}^{ee})_{ij} = \alpha_{ee}\delta_{ij}$, $(\overleftrightarrow{\boldsymbol{\alpha}}^{em})_{ij} = \alpha_{em}\delta_{ij}$, and $(\overleftrightarrow{\boldsymbol{\alpha}}^{mm})_{ij} = \alpha_{mm}\delta_{ij}$. With these, Eq. (4) is simplified to

$$\begin{aligned}\langle\mathbf{F}\rangle =& -\nabla\langle U_f\rangle + (C_{\mathrm{ext}} + C_{\mathrm{recoil}})c^{-1}\langle\mathbf{S}\rangle \\ &- \mathrm{Im}(\alpha_{ee}\alpha^*_{mm} - \alpha_{em}\alpha^*_{em})\mathrm{Im}(\mathbf{E}\times\mathbf{B}^*) \\ &+ \nabla\times\left[\mathrm{C}^{\mathbf{p}}_{\mathrm{ext}}c\langle\mathbf{L}^{\mathbf{p}}_s\rangle + \mathrm{C}^{\mathbf{m}}_{\mathrm{ext}}c\langle\mathbf{L}^{\mathbf{m}}_s\rangle + \mu_0(\mathrm{Re}\alpha_{em})\langle\mathbf{S}\rangle\right] \\ &+ \left[2\omega^2\mu_0(\mathrm{Re}\alpha_{em}) - \frac{k^5}{3\pi\varepsilon_0}\frac{\mathrm{Im}(\alpha_{ee}\alpha^*_{em})}{\varepsilon_0}\right]\langle\mathbf{L}^{\mathbf{p}}_s\rangle \\ &+ \left[2\omega^2\mu_0(\mathrm{Re}\alpha_{em}) - \frac{k^5\mu_0}{3\pi\varepsilon_0}\mathrm{Im}(\alpha_{mm}\alpha^*_{em})\right]\langle\mathbf{L}^{\mathbf{m}}_s\rangle, \end{aligned} \quad (10)$$

where

$$\langle U_f\rangle = -\tfrac{1}{4}\mathrm{Re}(\alpha_{ee})|\mathbf{E}|^2 - \tfrac{1}{4}\mathrm{Re}(\alpha_{mm})|\mathbf{B}|^2 + \tfrac{1}{2}\mathrm{Im}(\alpha_{em})\mathrm{Im}(\mathbf{B}\cdot\mathbf{E}^*) \quad (11)$$

is the free energy,

$$C_{\mathrm{ext}} = C^{\mathbf{p}}_{\mathrm{ext}} + C^{\mathbf{m}}_{\mathrm{ext}} \quad (12)$$

is the extinction cross section,

$$C^{\mathbf{p}}_{\mathrm{ext}} = \frac{k\mathrm{Im}\,(\alpha_{ee})}{\varepsilon_0} \quad (13)$$

is the extinction cross section through the electric dipole channel,

$$C^{\mathbf{m}}_{\mathrm{ext}} = k\mu_0\mathrm{Im}\,(\alpha_{mm}) \quad (14)$$

is the extinction cross section through the magnetic dipole channel,

$$C_{\mathrm{recoil}} = -\frac{k^4\mu_0\mathrm{Re}(\alpha_{ee}\alpha^*_{mm})}{6\pi\varepsilon_0} - \frac{k^4\mu_0}{6\pi\varepsilon_0}\mathrm{Re}(\alpha_{em}\alpha^*_{em}) \quad (15)$$

is directly related to the recoil force [32],

$$\langle\mathbf{L}^{\mathbf{m}}_S\rangle = \frac{\mu_0}{4\omega i}\mathbf{H}\times\mathbf{H}^*, \quad (16)$$

and

$$\langle\mathbf{L}^{\mathbf{p}}_s\rangle = \frac{\varepsilon_0}{4\omega i}\mathbf{E}\times\mathbf{E}^*. \quad (17)$$

The chirality flux

$$\begin{aligned}\langle\boldsymbol{\Psi}\rangle &= \frac{\omega}{4}\mathrm{Im}[\varepsilon_0\mathbf{E}^*\times\mathbf{E} + \mu_0\mathbf{H}^*\times\mathbf{H}] \\ &= \omega^2\mathrm{Re}\{\langle\mathbf{L}^{\mathbf{p}}_s\rangle + \langle\mathbf{L}^{\mathbf{m}}_s\rangle\}\end{aligned} \quad (18)$$

is directly related to Eqs. (16) and (17), indicating a close physical relation between chirality and optical forces. For paraxial beams or plane waves, $\mathbf{k} \parallel \mathbf{E}\times\mathbf{E}^*$, where $\mathbf{k}$ is the propagation direction of the incident wave. Then

$$\begin{aligned}\langle\mathbf{L}^{\mathbf{m}}_s\rangle &= \frac{\mu_0}{4\omega i}\mathbf{H}\times\mathbf{H}^* = \frac{\varepsilon_0}{4\omega i}\hat{\mathbf{k}}[\hat{\mathbf{k}}\cdot(\mathbf{E}\times\mathbf{E}^*)] \\ &= \langle\mathbf{L}^{\mathbf{p}}_S\rangle \equiv \langle\mathbf{L}_s\rangle.\end{aligned} \quad (19)$$

Using Eq. (19) and assuming $|\mathrm{Re}\alpha_{em}| \ll |\mathrm{Im}\alpha_{em}|$, Eq. (10) can be expressed in an even more compact form,

$$\begin{aligned}\langle\mathbf{F}\rangle =& -\nabla\langle U_f\rangle + (C_{\mathrm{ext}} + C_{\mathrm{recoil}})c^{-1}\langle\mathbf{S}\rangle + C_{\mathrm{ext}}c\nabla\times\langle\mathbf{L}_s\rangle \\ &- \frac{k^5}{3\pi\varepsilon_0^2}\mathrm{Im}\left[\alpha^*_{em}\left(\alpha_{ee} + \frac{\alpha_{mm}}{c^2}\right)\right]\langle\mathbf{L}_s\rangle.\end{aligned} \quad (20)$$

Note that in (20), we have thrown away the small terms that are proportional to $\mathrm{Re}\{\alpha_{em}\}$ or $\mathrm{Im}(\alpha_{ee}\alpha^*_{mm} - \alpha_{em}\alpha^*_{em})$. Note that the first and last terms of Eq. (20) both depend on chirality, and that neither exists in the expression for a nonchiral particle [33,34]. Given the same electromagnetic field characterized by the same chirality flux, the force of the particle depends linearly on $\mathrm{Im}(\alpha_{em})$, which has opposite signs for particles with opposite chirality. The implication of the spin angular momentum of light on optical force or optical radiation pressure has been considered recently by several groups [35–40]. For example, using chiral light to sort particles has been addressed theoretically [35] and experimentally [36]. The force and torque on an electric dipole by circular polarized light [37] and optical force on a chiral molecule [38] has been considered theoretically. Using circular polarized light to induce a lateral force on chiral objects has been discussed [39]. The magnetoelectric effects in light-matter interactions have been discussed [40]. Our work has a different emphasis from previous work as we are specifically concerned with the possibility of using chirality-induced coupling to induce light to pull a chiral object.

III. OPTICAL PULLING

Next, we give an example to illustrate the potential of the chirality-dependent forces to pull a chiral particle towards the source of light. Recently, there has been increasing interest in the so-called "optical tractor beam," which is a forward propagating beam that can pull a particle backward [32–34,41–47]. Examples of optical tractor beams include the optical solenoid beam [46,47] and the optical pulling force [32–34,41–47]. The backward force of optical solenoid beams originates from the gradient force [46], whereas that of the optical pulling force is a *negative* scattering force arising from the recoil force of the scattered photons [32]. The optical pulling force is typically realized by exciting the electric dipole moment and the magnetic dipole moment or the electric quadrupole moment simultaneously so that their interference produces strong forward scattering. We shall see that for a chiral particle, a different mechanism for realizing the optical pulling force emerges from the coupling of linear momentum with angular momentum via chirality.

The force terms containing $\tilde{\mathbf{L}}$ in Eq. (4) are proportional to the chirality fluxes $\langle \mathbf{\Psi}_e \rangle$ and $\langle \mathbf{\Psi}_m \rangle$. We shall refer to such forces as the chirality forces, as they originate from chirality fluxes of the field and the chirality of the particle (α_{em}). Light waves with different chirality will induce opposite chirality forces on the same particle. When the magnitude of the chirality forces is greater than the summation of the first three terms in Eq. (4), an optical pulling force can be realized.

The above analytical results are valid for a small particle. In the following, we use the Maxwell stress tensor and multiple scattering theory [48] to show that the forces induced by chirality flux can in turn induce an optical pulling force on a chiral object that is not optically small. Such a formulation is "exact" within classical electrodynamics, and subject only to numerical truncation errors. Consider the chiral structure shown in Fig. 1(a). This structure is a collection of metallic spheres ($\varepsilon = -5 + 0.13i$ of gold at wavelength $\lambda = 337$ nm [49]) whose center sits on a left-handed spiral (the black line) [50]. The details of the structure are given in the figure caption. The incident waves consist of two incoherent plane waves propagating along $+z$ (right-circularly polarized, RCP) and $-z$ (left-circularly polarized, LCP), with a somewhat arbitrarily chosen $\lambda = 337$ nm. In such a configuration, the first three terms in Eq. (4) are eliminated due to the counterpropagation of the two plane waves, but the chirality fluxes for the two plane waves add up, resulting in a nonzero chiral force. We

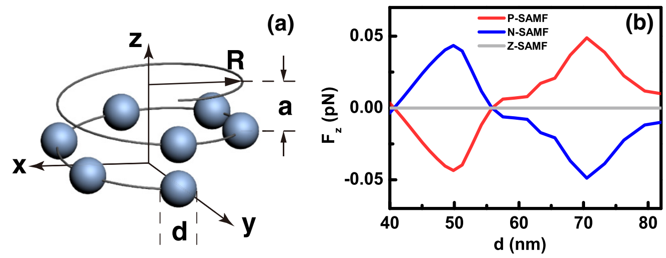

FIG. 1. (Color online) (a) Schematic picture of the prototypical chiral chain made up of 25 metallic spheres ($\varepsilon_r = -5 + 0.13i$) arranged on a spiral marked by the black line and parametrized by $(R \sin 2\pi t, R \cos 2\pi t, at)$, with $0 < t < 4$, and $R = 1.05d$, and pitch $a = 1.05d$, where d is the diameter of the sphere. The spiral's height is $4a$ and each loop has six spheres evenly spaced on the spiral. (b) Calculated optical forces acting on the spiral chains for two counterpropagating incoherent plane waves with $\lambda = 337$ nm propagating in the $+z$ and $-z$ directions, each with an intensity of 10^9 W/m^2. Positive spin angular momentum flux (P-SAMF) (with $\langle \Phi_e \rangle || \mathbf{k}$) can induce both positive and negative optical forces, as shown by the red line (gray line). A negative force here means that the beam attracts the object. The blue line (black line) shows the force due to a negative spin angular momentum flux (N-SAMF).

show in Fig. 1(b) the optical forces calculated for spheres with diameters falling in the range of 40 nm $< d <$ 80 nm. The total length of the spiral is set to scale with the size of the spheres. The z component of the force (F_z) induced by positive spin angular momentum flux (P-SAMF, $\hat{z} \cdot \langle L_s \rangle > 0$) and that induced by negative spin angular momentum flux (N-SAMF, $\hat{z} \cdot \langle \vec{L}_s \rangle < 0$) are equal and opposite. In particular, F_z associated with P-SAMF can be in the same or opposite direction to $\langle \vec{L}_s \rangle$, implying that the chirality force can be along $+\mathbf{k}$ or $-\mathbf{k}$ depending on the size of the spiral structure. This clearly demonstrates that the chirality force can potentially induce a pulling force in the $-\mathbf{k}$ direction. The parameters used here, such as the radius and pitch of the spiral and the size of the particles, are chosen somewhat arbitrarily. But since chirality is a symmetry property, the same physics should be valid for a broad range of parameters.

As described above, the chirality force can induce an optical pulling force. Figure 2 shows the optical force for a specific chiral system illuminated by linearly (LP-BB, solid light gray line), left-circularly (LCP-BB, solid blue line), and right-circularly (RCP-BB, solid red line) polarized nondiffractive Bessel beams. It can be seen that both RCP-BB and LCP-BB can induce a negative force. When the particles and the entire spiral structure are both small, the chiral features cannot be resolved by the incident wave and no optical pulling force is observed. But as the particle and the spiral structure increase in size, Fig. 2 shows that circularly polarized beams can induce optical forces with opposite signs (see, e.g., the forces for sphere diameters of 55 and 70 nm), leading to an optical pulling force for one of the circular polarizations. What we have realized here is an optical pulling force induced by chirality, which is very different from that induced on nonchiral dielectric particles [32]. The optical pulling force in the latter case arises from the simultaneous excitation of multipoles and their interference. In order to illustrate the importance of chirality, we also calculate the optical force for planar

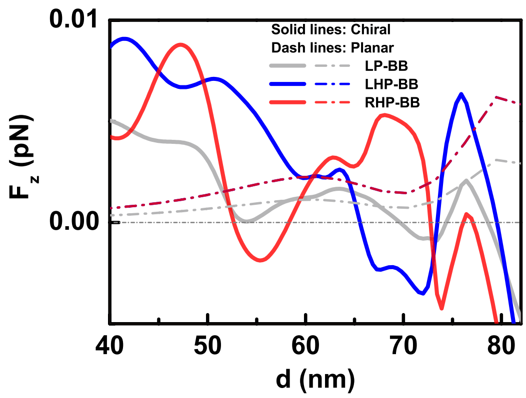

FIG. 2. (Color online) The z component of the optical forces acting on the chiral chain shown in Fig. 1(a), induced by a Bessel beam with $\alpha = 87^\circ$ and $l = 0$ (see Appendix B for the expression of the Bessel beam) with different polarizations. The solid light gray, blue (black), and red (gray) lines are for Bessel beams with linear (LP-BB), left-circular (LCP-BB), and right-circular polarization (RCP-BB), respectively. The intensity of each beam is 10^9 W/m^2 and $\lambda = 337$ nm. For comparison, the dashed lines show the optical forces acting on planar structures with the sphere positions projected onto the xy plane. The results show that circularly polarized Bessel beams can induce negative forces on chiral structures (solid lines) but not on nonchiral structures (dashed lines).

structures (with six evenly spaced spheres sitting on a circle on the xy plane) illuminated by Bessel beams with different polarization, as shown by the dashed lines in Fig. 2. Clearly, no optical pulling force is observed and there is no difference between RCP-BB and LCP-BB.

IV. DISCUSSION

These results show that the idea of deriving an optical pulling force from a light chirality force is robust. This can be seen in the analytic results derived for the dipole limit [Eq. (4)] for a small hypothetical chiral particle as well as

TABLE I. Far-field mode expansion coefficients of a chiral structure. $|a_n|$ and $|b_n|$ indicate electric and magnetic normal mode coefficients, respectively, with $n = 1$ corresponding to dipole, $n = 2$ corresponding to quadrupole, and so on. The integration surface is taken at $\rho/\lambda = 16.67$, and the sphere diameter is set to $d = 56$ nm.

n	$\lvert a_n \rvert$	$\lvert b_n \rvert$
	$\vec{k} \parallel \hat{z},\ \vec{E} \parallel \hat{x}$	
1	0.380	0.905
2	0.109	0.145
3	0.036	0.045
	$\vec{k} \parallel \hat{z},\ \vec{E} \parallel \hat{y}$	
1	0.903	0.386
2	0.144	0.107
3	0.049	0.043
	$\vec{k} \parallel \hat{y},\ \vec{E} \parallel \hat{z}$	
1	0.876	0.405
2	0.186	0.143
3	0.085	0.076

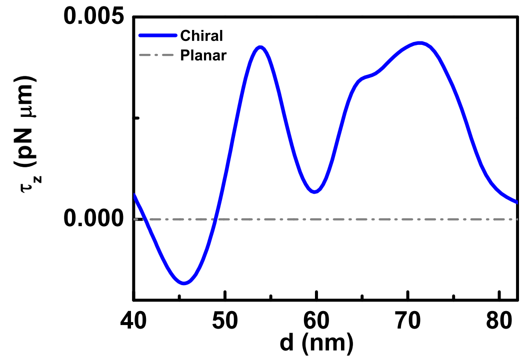

FIG. 3. (Color online) The z component of the total optical torque about the z axis for the chiral chain system shown in Fig. 1(a), illuminated by an x-polarized plane wave (solid blue line). The intensity of the beam is 10^9 W/m^2. The total optical torque acting on a nonchiral planar structure, i.e., when all spheres are sitting on the xy plane, is shown by the dashed line for comparison.

a wavelength-scale spiral object (numerically demonstrated in Figs. 1 and 2). The spiral object depicted in the inset of Fig. 1 is already too complex to derive analytically, but we can use numerical field projection methods [51] to show that the responses are still mainly due to dipolar excitations. We apply multiple scattering theory to calculate the scattered electric field $\mathbf{E}_{sc}$ in the far-field regime, and then compute the overlap between $\mathbf{E}_{sc}$ and individual multipole field [52]. Within the framework of the Maxwell tensor formalism, the fields in the far field give the exact optical force. The results for the first few most important multipoles are tabulated in Table I. As the far-field projections are dominated by an $n = 1$ contribution, it is still appropriate to discuss the physics qualitatively based on the analytic results.

This chirality force can be viewed as a consequence of the coupling of linear momentum with angular momentum. In principle, the "reverse" effect is also allowed, namely, that the illumination by a linearly polarized beam should also induce an optical torque on the chiral particle [19], due to the same coupling effect between linear momentum and angular momentum. This is illustrated in Fig. 3, which demonstrates the existence of optical torque (about the central axis of the spiral) acting on the structure shown in Fig. 1(a) illuminated by a linearly polarized plane wave. The corresponding optical torque acting on planar structures is also shown. It is zero as there is no chirality.

V. CONCLUSION

In conclusion, we have shown that chirality-dependent optical forces enable a mechanism to realize the optical pulling force via the coupling of the linear momentum of a chiral particle with the spin angular momentum of light. We have also performed numerical calculations for wavelength-scale spirals employing the multiple scattering method and the Maxwell stress tensor to supplement the analytic theory predicting the optical pulling force.

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APPENDIX A: THE MULTIPOLE EXPANSION OF OPTICAL FORCE ACTING ON A CHIRAL PARTICLE

1. Basic formulations

Let us consider a small chiral particle that may be described by an electric dipole moment $\mathbf{p}$ and a magnetic dipole moment $\mathbf{m}$, and the induced moments are given by [27]

$$\mathbf{p} = \overleftrightarrow{\boldsymbol{\alpha}}^{ee}\mathbf{E} + \overleftrightarrow{\boldsymbol{\alpha}}^{em}\mathbf{B}, \quad \mathbf{m} = (-\overleftrightarrow{\boldsymbol{\alpha}}^{em})^T\mathbf{E} + \overleftrightarrow{\boldsymbol{\alpha}}^{mm}\mathbf{B}. \tag{A1}$$

Here $\overleftrightarrow{\boldsymbol{\alpha}}^{ee}$, $\overleftrightarrow{\boldsymbol{\alpha}}^{mm}$, and $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$ are the electric, magnetic, and chiral polarization tensors, $\mathbf{A}^T$ denotes the transpose of $\mathbf{A}$, and $\overleftrightarrow{\boldsymbol{\alpha}}^{me} = -\overleftrightarrow{\boldsymbol{\alpha}}^{emT}$ for a reciprocal medium. The multipole expansion of the time-averaged optical force (referred to as the optical force hereafter) for a particle illuminated by a monochromatic field is given in Ref. [32]. Here, we assume that the particle is small compared to the incident wavelength. Therefore we keep only the electric and magnetic dipole moments. In this case, the optical force is given by

$$\langle \mathbf{F}\rangle = \mathbf{F_p} + \mathbf{F_m} + \mathbf{F_{p\times m}}, \tag{A2}$$

where

$$\mathbf{F_p} = \tfrac{1}{2}\mathrm{Re}\{(\nabla\mathbf{E}^*)\cdot\mathbf{p}\} \tag{A3}$$

is the electric dipole force,

$$\mathbf{F_m} = \tfrac{1}{2}\mathrm{Re}\{(\nabla\mathbf{B}^*)\cdot\mathbf{m}\} \tag{A4}$$

is the magnetic dipole force, and

$$\mathbf{F_{p\times m}} = -\frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{\mathbf{p}\times\mathbf{m}^*\} \tag{A5}$$

is the recoil force. We shall rewrite Eq. (A2) term by term in the following sections, and transform it into a compact form.

2. Electric dipole terms

The electromagnetic field can be written in Cartesian coordinates,

$$\mathbf{E} = |E_j|e^{i\Theta_j}\hat{\mathbf{x}}_j, \quad \mathbf{B} = |B_j|e^{i\Phi_j}\hat{\mathbf{x}}_j, \tag{A6}$$

where $|E_j|$ and $|B_j|$ are the magnitudes of the field's jth Cartesian component, Θ_j and Φ_j are the phases of the corresponding fields, and $\hat{\mathbf{x}}_j$ is the jth unit vector in the Cartesian coordinate system. We adopt the summation convention. Substituting Eq. (A6) into Eq. (A3) yields

$$\begin{aligned}
\mathbf{F_p} &= \tfrac{1}{2}\mathrm{Re}\{(\nabla\mathbf{E}^*)\cdot\mathbf{p}\} = \tfrac{1}{2}\mathrm{Re}\{p_i\nabla E_i^*\}\\
&= \tfrac{1}{2}\mathrm{Re}\big\{\big(\alpha_{ij}^{ee}E_j + \alpha_{ik}^{em}B_k\big)\nabla E_i^*\big\}\\
&= \tfrac{1}{2}\hat{e}_n\mathrm{Re}\big\{(\partial_n|E_i|e^{-i\Theta_i})\big(\alpha_{ij}^{ee}|E_j|e^{i\Theta_j} + \alpha_{ik}^{em}|B_k|e^{i\Phi_k}\big)\big\}\\
&= \tfrac{1}{2}\hat{e}_n\big\{(\partial_n|E_i|)|E_j|\mathrm{Re}\big[\alpha_{ij}^{ee}e^{i(\Theta_j-\Theta_i)}\big]\\
&\quad + (\partial_n|E_i|)|B_k|\mathrm{Re}\big[\alpha_{ik}^{em}e^{i(\Phi_k-\Theta_i)}\big]\\
&\quad + |E_i||E_j|(\partial_n\Theta_i)\mathrm{Im}\big[\alpha_{ij}^{ee}e^{i(\Theta_j-\Theta_i)}\big]\\
&\quad + |E_i||B_k|(\partial_n\Theta_i)\mathrm{Im}\big[\alpha_{ik}^{em}e^{i(\Phi_k-\Theta_i)}\big]\big\},
\end{aligned} \tag{A7}$$

where α_{ij}^{ee} is the ij component of $\overleftrightarrow{\boldsymbol{\alpha}}^{ee}$, and similar notations are employed for the other polarizabilities. The first two terms originate from the inhomogeneity of the field amplitude (i.e., $\partial_n|E_i|$), whereas the last two terms arise from the inhomogeneity of the field's phases (i.e., $\partial_n\Theta_i$).

After some manipulations, Eq. (A7) can be rewritten in a compact form,

$$\mathbf{F_p} = -\nabla\langle U_\mathbf{p}\rangle + \tfrac{1}{4}\mathrm{Re}\{\mathbf{T}^{ee} : \overleftrightarrow{\boldsymbol{\alpha}}^{ee} + \mathbf{T}^{em} : \overleftrightarrow{\boldsymbol{\alpha}}^{em}\}, \tag{A8}$$

where

$$\langle U_\mathbf{p}\rangle = -\tfrac{1}{4}\mathrm{Re}(\mathbf{E}\cdot\mathbf{p}^*), \tag{A9}$$

and the auxiliary quantities are given by

$$\begin{aligned}
T_{n,ji}^{ee} &= [(\partial_n|E_i|)|E_j| - (\partial_n|E_j|)|E_i| - i|E_i||E_j|\partial_n(\Theta_i + \Theta_j)]\\
&\quad \times e^{i(\Theta_j-\Theta_i)},\\
T_{n,ki}^{em} &= [(\partial_n|E_i|)|B_k| - (\partial_n|B_k|)|E_i| - i|E_i||B_k|\partial_n(\Theta_i + \Phi_k)]\\
&\quad \times e^{i(\Phi_k-\Theta_i)}.
\end{aligned} \tag{A10}$$

Here $\mathbf{T}^{ee}$ and $\mathbf{T}^{em}$ are rank-3 tensors that depend only on the field, whereas $\overleftrightarrow{\boldsymbol{\alpha}}^{ee}$ and $\overleftrightarrow{\boldsymbol{\alpha}}^{em}$ depend only on the particle, i.e., the contributions from the field and from the particle are separated.

3. Magnetic dipole terms

Similar to the electric dipole force $\mathbf{F_p}$, the magnetic dipole force $\mathbf{F_m}$ can be obtained by substituting (A6) into (A4):

$$\begin{aligned}
\mathbf{F_m} &= \frac{1}{2}\mathrm{Re}\{(\nabla\mathbf{B}^*)\cdot\mathbf{m}\} = \frac{1}{2}\mathrm{Re}\{m_i\nabla B_i^*\}\\
&= \frac{1}{2}\mathrm{Re}\big\{\big(\alpha_{ij}^{mm}B_j - \alpha_{ik}^{em,T}E_k\big)\nabla B_i^*\big\}\\
&= \frac{\hat{\mathbf{x}}_n}{2}\mathrm{Re}\big\{(\partial_n|B_i|e^{-i\Phi_i})\big(\alpha_{ij}^{mm}|B_j|e^{i\Phi_j} - \alpha_{ik}^{em,T}|E_k|e^{i\Theta_k}\big)\big\}\\
&= \frac{\hat{\mathbf{x}}_n}{2}\big\{(\partial_n|B_i|)|B_j|\mathrm{Re}\big[\alpha_{ij}^{mm}e^{i(\Phi_j-\Phi_i)}\big]\\
&\quad - (\partial_n|B_i|)|E_k|\mathrm{Re}\big[\alpha_{ik}^{em,T}e^{i(\Theta_k-\Phi_i)}\big]\\
&\quad + |B_i||B_j|(\partial_n\Phi_i)\mathrm{Im}\big[\alpha_{ij}^{mm}e^{i(\Phi_j-\Phi_i)}\big]\\
&\quad - |B_i||E_k|(\partial_n\Phi_i)\mathrm{Im}\big[\alpha_{ik}^{em,T}e^{i(\Theta_k-\Phi_i)}\big]\big\}.
\end{aligned} \tag{A11}$$

Upon performing similar derivations as in the electric dipole term, we arrive at

$$\mathbf{F_m} = -\nabla\langle U_\mathbf{m}\rangle + \tfrac{1}{4}\mathrm{Re}\{\mathbf{T}^{mm} : \overleftrightarrow{\boldsymbol{\alpha}}^{mm} + \mathbf{T}^{me} : \overleftrightarrow{\boldsymbol{\alpha}}^{em,\mathrm{T}}\}, \tag{A12}$$

where

$$\langle U_\mathbf{m}\rangle = -\tfrac{1}{4}\mathrm{Re}(\mathbf{B}\cdot\mathbf{m}^*), \tag{A13}$$

and the auxiliary quantities are given by

$$\begin{aligned}
T_{n,ji}^{mm} &= [(\partial_n|B_i|)|B_j| - (\partial_n|B_j|)|B_i| - i|B_i||B_j|\partial_n(\Phi_i + \Phi_j)]\\
&\quad \times e^{i(\Phi_j-\Phi_i)},\\
T_{n,ki}^{me} &= [(\partial_n|E_k|)|B_i| - (\partial_n|B_i|)|E_k| + i|B_i||E_k|\partial_n(\Phi_i + \Theta_k)]\\
&\quad \times e^{i(\Theta_k-\Phi_i)}.
\end{aligned} \tag{A14}$$

The expression (A12) is similar to its electric counterpart, Eq. (A8).

4. The p × m term

Using Eq. (A6), the recoil force term Eq. (A5) can be written as

$$\mathbf{F}_{\mathbf{p}\times\mathbf{m}} = -\frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{\mathbf{p}\times\mathbf{m}^*\}$$
$$= -\frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{(\overleftrightarrow{\alpha}^{ee}\mathbf{E}+\overleftrightarrow{\alpha}^{em}\mathbf{B})$$
$$\times(-\overleftrightarrow{\alpha}^{em\dagger}\mathbf{E}^*+\overleftrightarrow{\alpha}^{mm*}\mathbf{B}^*)\},\tag{A15}$$

where $\mathbf{A}^\dagger$ denotes the Hermitian conjugate of $\mathbf{A}$. Using the mathematical identity

$$(\overleftrightarrow{\mathbf{A}}\cdot\vec{\mu})\times(\overleftrightarrow{\mathbf{B}}\cdot\vec{\nu}) = [(\overleftrightarrow{\mathbf{A}}\cdot\vec{\mu})\times\overleftrightarrow{\mathbf{B}}]\cdot\vec{\nu} = -[\overleftrightarrow{\mathbf{B}}^T\times(\overleftrightarrow{\mathbf{A}}\cdot\vec{\mu})]^T\cdot\vec{\nu}$$
$$= -[(\overleftrightarrow{\mathbf{B}}^T\times\overleftrightarrow{\mathbf{A}})\cdot\vec{\mu}]^T\cdot\vec{\nu}$$
$$= -\vec{\mu}^T\cdot(\overleftrightarrow{\mathbf{B}}^{\mathrm{T}}\times\overleftrightarrow{\mathbf{A}})^T\cdot\vec{\nu},\tag{A16}$$

one can transform Eq. (A15) into

$$\mathbf{F}_{\mathbf{p}\times\mathbf{m}}$$
$$= -\frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{(\overleftrightarrow{\alpha}^{ee}\mathbf{E}+\overleftrightarrow{\alpha}^{em}\mathbf{B})\times(-\overleftrightarrow{\alpha}^{em\dagger}\mathbf{E}^*+\overleftrightarrow{\alpha}^{mm*}\mathbf{B}^*)\}$$
$$= -\frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{(\overleftrightarrow{\alpha}^{ee}\mathbf{E})\times(-\overleftrightarrow{\alpha}^{em\dagger}\mathbf{E}^*)+(\overleftrightarrow{\alpha}^{ee}\mathbf{E})\times(\overleftrightarrow{\alpha}^{mm*}\mathbf{B}^*)$$
$$+(\overleftrightarrow{\alpha}^{em}\mathbf{B})\times(-\overleftrightarrow{\alpha}^{em\dagger}\mathbf{E}^*)+(\overleftrightarrow{\alpha}^{em}\mathbf{B})\times(\overleftrightarrow{\alpha}^{mm*}\mathbf{B}^*)\}$$
$$= -\frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{\mathbf{E}\cdot(\overleftrightarrow{\alpha}^{em*}\times\overleftrightarrow{\alpha}^{ee})^T\cdot$$
$$\times\mathbf{E}^*-\mathbf{E}\cdot(\overleftrightarrow{\alpha}^{mm\dagger}\times\overleftrightarrow{\alpha}^{ee})^T\cdot\mathbf{B}^*$$
$$+\mathbf{B}\cdot(\overleftrightarrow{\alpha}^{em*}\times\overleftrightarrow{\alpha}^{em})^T\cdot\mathbf{E}^*-\mathbf{B}\cdot(\overleftrightarrow{\alpha}^{mm\dagger}\times\overleftrightarrow{\alpha}^{em})^T\cdot\mathbf{B}^*\}.\tag{A17}$$

Introducing two more auxiliary quantities

$$\tilde{\mathbf{L}} = \mathbf{E}\cdot(\overleftrightarrow{\alpha}^{em*}\times\overleftrightarrow{\alpha}^{ee})^T\cdot\mathbf{E}^*-\mathbf{B}\cdot(\overleftrightarrow{\alpha}^{mm\dagger}\times\overleftrightarrow{\alpha}^{em})^T\cdot\mathbf{B}^*,$$
$$\tilde{\mathbf{S}} = \mathbf{B}\cdot(\overleftrightarrow{\alpha}^{em*}\times\overleftrightarrow{\alpha}^{em})^T\cdot\mathbf{E}^*-\mathbf{E}\cdot(\overleftrightarrow{\alpha}^{mm\dagger}\times\overleftrightarrow{\alpha}^{ee})^T\cdot\mathbf{B}^*,\tag{A18}$$

Eq. (A17) then becomes

$$\mathbf{F}_{\mathbf{p}\times\mathbf{m}} = -\frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{\tilde{\mathbf{L}}+\tilde{\mathbf{S}}\}.\tag{A19}$$

5. Grouping all terms together

Summing (A8), (A12), and (A19), the total optical force is given by

$$\langle\mathbf{F}\rangle = -\nabla\langle U_f\rangle + \frac{1}{4}\mathrm{Re}\{\mathbf{T}^{ee}:\overleftrightarrow{\alpha}^{ee}+\mathbf{T}^{em}:\overleftrightarrow{\alpha}^{em}$$
$$+\mathbf{T}^{mm}:\overleftrightarrow{\alpha}^{mm}+\mathbf{T}^{me}:\overleftrightarrow{\alpha}^{emT}\} - \frac{k^4}{12\pi\varepsilon_0 c}\mathrm{Re}\{\tilde{\mathbf{L}}+\tilde{\mathbf{S}}\},\tag{A20}$$

$$\langle U_f\rangle = \langle U_{\mathbf{p}}\rangle + \langle U_{\mathbf{m}}\rangle = -\tfrac{1}{4}\mathrm{Re}(\mathbf{E}\cdot\mathbf{p}^*) - \tfrac{1}{4}\mathrm{Re}(\mathbf{B}\cdot\mathbf{m}^*).\tag{A21}$$

This is just Eq. (4) in the main text.

APPENDIX B: BESSEL BEAM

In this paper, to eliminate the axial gradient force along the z direction, we consider Bessel beams of the form [53]

$$E_x(\rho,\phi,z) = \tfrac{1}{2}E_0(\alpha_0)e^{ik_z z}e^{il\phi}$$
$$\times\{P_\perp i^{2-l}J_{2-l}(k_\perp\rho)e^{-i2\phi}(-1\pm 1)$$
$$+P_\perp i^{2+l}J_{2+l}(k_\perp\rho)e^{i2\phi}(-1\mp 1)$$
$$+i^{-l}J_{-l}(k_\perp\rho)+i^l J_l(k_\perp\rho)\},\tag{B1}$$

$$E_y(\rho,\phi,z) = \tfrac{1}{2}E_0(\alpha_0)e^{ik_z z}e^{il\phi}$$
$$\times\{P_\perp i^{3-l}J_{2-l}(k_\perp\rho)e^{-i2\phi}(-1\pm 1)$$
$$+P_\perp i^{3+l}J_{2+l}(k_\perp\rho)e^{i2\phi}(1\pm 1)$$
$$+i^{1-l}J_{-l}(k_\perp\rho)+i^{1+l}J_l(k_\perp\rho)\},\tag{B2}$$

$$E_z(\rho,\phi,z) = E_0(\alpha_0)e^{ik_z z}e^{il\phi}P_{||}\{i^{1-l}J_{1-l}(k_\perp\rho)e^{-i\phi}(-1\pm 1)$$
$$+i^{1+l}J_{1+l}(k_\perp\rho)e^{i\phi}(-1\mp 1)\},\tag{B3}$$

where E_0 is the on-axis electric field, J_n is the Bessel function of nth order, (ρ,ϕ,z) are cylindrical coordinates, $\pm(\mp)$ means left- (right-) handed circular polarization, and

$$k_\perp = k\sin\alpha_0,\quad k_z = k\cos\alpha_0,$$
$$P_\perp = \frac{1-\cos\alpha_0}{1+\cos\alpha_0},\quad P_{||} = \frac{\sin\alpha_0}{1+\cos\alpha_0}.\tag{B4}$$

In the calculation of Fig. 2, the parameters we used are $\alpha_0 = 87^\circ$ and $l = 0$.

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