Enantioselection and chiral sorting of single microspheres using optical pulling forces

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We put forward a novel, twofold scheme that enables at the same time all-optical enantioselection and sorting of single multipolar chiral microspheres based on optical pulling forces exerted by two non-collinear, non-structured, circularly-polarized light sources. Our chiral resolution method can be externally controlled by varying the angle between their incident wavevectors, allowing for a fine-tuning of the range of chiral indices for enantioselection. Enantioselectivity is achieved by choosing angles such that only particles with the same handedness of the light sources are pulled. This proposal allows one to achieve all-optical sorting of chiral microspheres with arbitrarily small chiral parameters, thus outperforming current optical methods. © 2021 Optical Society of America

To circumvent these limitations we put forward a novel enantioselective method for chiral particles with sizes comparable to the incident radiation, composed of two non-collinear, non-structured plane waves, using pulling optical forces. These forces pull a particle all the way towards the source without an equilibrium point as a result of a backward scattering force due to the interference of radiation multipoles, with many applications [28–35]. By calculating the optical force acting on a chiral sphere beyond the dipolar approximation using Mie theory, we demonstrate that it strongly depends on the chiral parameter $k$ and radius, allowing for enantioselection and chiral sorting. We also show that the optical force changes sign as a function of the angle between the two incident wave vectors, from pushing to pulling, demonstrating that one can externally control the enantioselective mechanism. We show that this mechanism can selectively resolve particles with arbitrarily small chiral indexes by suitably adjusting the angle between the two propagation directions. This is in contrast to recent proposals that demonstrate enantioselection and chiral sorting of particles of sizes of the order of wavelength, in which the chiral parameter must be sufficiently large $k \gtrsim 0.1$ thus restricting the range of potential applications [31].

We consider two circularly polarized laser beams (helicity $\sigma = \pm 1$) positioned at an angle $\Theta$ with respect to the common-axis ($z$-axis) propagating in a non-absorbing, non-magnetic medium, as illustrated by Fig 1(a). We model the (paraxial) laser beams as plane waves as their waists are typically much larger than the particle size, which we consider to be in the micrometer range. The total electric field incident on the particle can then be expressed as the superposition

$$E_{\text{in}} = \sum_{j=1}^{2} E_0 e^{i\omega t} \hat{\sigma}_j (\theta_j + i\sigma_j \hat{\phi}_j) e^{i\mathbf{k}_j \cdot \mathbf{r} - \omega t}. \quad (1)$$

The wave vectors $\mathbf{k}_j(\theta_j, \phi_j)$ are such that propagation is on the $xz$ plane. Hence we take $\theta_1 = 0$ and $\phi_2 = \pi$. The beams share the same amplitude $E_0$ and angular frequency $\omega : \mathbf{k}_1 = n_h \frac{\omega}{c} \{\sin(\Theta), 0, \cos(\Theta)\}$ and $\mathbf{k}_2 = n_h \frac{\omega}{c} \{-\sin(\Theta), 0, \cos(\Theta)\}$, where $n_h$ is the refractive index of the host medium and $c$ is the speed of light.

We solve the Mie scattering by the spherical particle by expanding the electromagnetic fields in terms of Debye po-
\[ \kappa \text{ particle with chiral parameter } \kappa = -0.3 \text{ as function of } \Theta \text{ in the case of right circular (dash), transverse electric (solid) and transverse magnetic (points) polarizations.} \]

\[ E = \nabla \times \nabla \times \left( r \Pi^E \right) + i \omega \mu_0 \nabla \times \left( r \Pi^M \right) \quad (5) \]

\[ H = \nabla \times \nabla \times \left( r \Pi^M \right) - i \omega \epsilon_0 \epsilon_0 \nabla \times \left( r \Pi^E \right) , \quad (6) \]

Finally, we determine the force using the Maxwell stress tensor:

\[ F = \lim_{r \to \infty} \left[ -\frac{r}{2} \int \left( \epsilon_1 \epsilon_0 \epsilon_2 E^2 + \mu_0 H^2 \right) d\Omega \right] \quad (7) \]

Since the incident plane waves have the same amplitude and polarization, the optical force will point along the z axis by symmetry.

As the stress tensor is quadratic in the total electric E and magnetic H fields, the optical force \( F_z = F_{zz} + F_{zz} \) has two distinct contributions: the extinction term \( F_{zz} \) arises from cross terms of the form \( E_n \times E_n^*(\text{and likewise for the magnetic field}) \) and represents the rate of linear momentum removal from the incident fields. Part of this momentum is carried away by the scattered fields, and the rest is transferred to the particle as an optical force. Thus, the second contribution to the optical force \( F_{zz} \), which is quadratic in \( E_z \) and \( H_z \), represents the negative of the rate of linear momentum contained in the scattered electromagnetic fields. We find

\[ F_{zz} = \frac{\epsilon_1 \epsilon_0 \epsilon_2 \epsilon_0}{k^2} \sum_{l,m \neq -l} \left( \frac{(\ell + 2)}{2\ell + 1} \right) \left( \frac{(\ell + m)(\ell + m + 1)}{(2\ell + 1)(2\ell + 3)} \right) \times \left[ \gamma_{\ell,m}^{E_{l+1,n}} + \gamma_{\ell,m}^{M_{l+1,n}} \right] \times \frac{\gamma_{\ell,m}^{E_{l-1,n}} + \gamma_{\ell,m}^{M_{l-1,n}}}{4\pi} \left( \sigma_1 Y_{\ell,m}^{E_{l-1,n}} + \sigma_2 Y_{\ell,m}^{M_{l-1,n}} \right) \times \left( \sigma_1 Y_{\ell,m}^{E_{l+1,n}} + \sigma_2 Y_{\ell,m}^{M_{l+1,n}} \right) \right] \quad (8) \]

\[ F_{zz} = \frac{2\pi \epsilon_1 \epsilon_0 \epsilon_2 \epsilon_0}{k^2} \sum_{l,m} \left( \frac{(\ell + 1)(2\ell + 1)}{4\pi} \right) \left[ \gamma_{\ell,m}^{E_{l-1,n}} + \gamma_{\ell,m}^{M_{l-1,n}} \right] \left( \sigma_1 Y_{\ell,m}^{E_{l-1,n}} + \sigma_2 Y_{\ell,m}^{M_{l-1,n}} \right) \times \left( \sigma_1 Y_{\ell,m}^{E_{l+1,n}} + \sigma_2 Y_{\ell,m}^{M_{l+1,n}} \right) \right] \quad (9) \]

We have defined the coefficients

\[ G_{l,m}^{E_{l-1,n}} = (i)^l \cos \Theta d_{m,n}^{E_{l-1,n}}(\Theta) \]  \[ G_{l,m}^{M_{l-1,n}} = (i)^l (-1)^m \cos \Theta d_{m,n}^{M_{l-1,n}}(\Theta) \]  \[ G_{l,m}^{E_{l+1,n}} = (i)^l \cos \Theta d_{m,n}^{E_{l+1,n}}(\Theta) \]  \[ G_{l,m}^{M_{l+1,n}} = (i)^l (-1)^m \cos \Theta d_{m,n}^{M_{l+1,n}}(\Theta) \]  (10)

We consider that the laser beams have wavelength \( \lambda_0 = 1064 \text{ nm} \). They impinge on a chiral microsphere with relative permittivity \( \epsilon_r = 2.5 \) and chiral parameter \( \kappa \) ranging from -0.5 to +0.5. The host medium is an aqueous solution with refractive index \( n_h = 1.332 \). In the following discussion, we normalize the optical force by the intensity \( I_0 = \frac{\epsilon_1 \epsilon_0 \epsilon_0}{\mu_0 \epsilon_0^2 / 2} \) of each incident beam.

In addition to the circular polarization polarization in Eq. (1), we also consider linearly-polarized beams where either the electric (TE) or magnetic (TM) fields are transverse to the plane defined by the wave vector. In Fig. 1(b), we plot the axial force \( F_z \) acting on a microsphere of radius 930 nm and chiral parameter \( \kappa = -0.3 \) as a function of \( \Theta \) in the case of TE (solid) and TM (dashed) linear polarizations, as well as for right circular polarization (RCP, dash), which corresponds to \( \sigma = -1 \). Figure 1(b) reveals that the axial force is positive (pushing) for \( \Theta \lesssim 82^\circ \),

| Angle (deg) | Fz/I0 (pN/mW\(^{-1}\)µm\(^2\)) |
|------------|------------------------|
| 0          |                       |
| 10         |                       |
| 20         |                       |
| 30         |                       |
| 40         |                       |
| 50         |                       |
| 60         |                       |
| 70         |                       |
| 80         |                       |
| 90         |                       |
exhibiting an oscillatory behaviour as a function of \( \Theta \) and eventually changes sign for \( 82^\circ \lesssim \Theta < 90^\circ \) for all polarisation states. At a large incident angle, the interference between them leads to maximally scattered momentum along the forward direction, which is the reason for a strong recoiling force along backward direction. This crossover from pushing (force along the propagation direction) to pulling forces can be externally controlled by varying \( \Theta \) and results from the excitation and interference between electric and magnetic multipole terms. It is worth mentioning that circularly-polarized plane waves exert a stronger overall force in comparison to the case of linear polarization.

In Fig. 2, the optical force is calculated as a function of both the chirality parameter \( \kappa \) and sphere radius for \( \Theta = 85^\circ \). We consider (a) TE, (b) TM and (c) RCP waves. The regions in color indicate the parameters yielding an optical pulling force while white regions correspond to pushing forces. It is clearly demonstrated in Fig. 2(a) and Fig. 2(b) that the linearly polarized waves exert identical forces on both enantiomers with opposite chirality parameters \( \kappa \), as expected from symmetry. In the case of TE polarization, pulling takes place regardless of chirality and results from the excitation and interference between dipolar limit, which hence can be exploited as a free parameter for the sorting of the particles with radius \( 1.50 \mu \text{m} \) (solid), \( 1.24 \mu \text{m} \) (dash) and \( 1.50 \mu \text{m} \) (solid).

Increasing the size of the particle, because a larger number of electric and magnetic multipoles come into play. When considered as a function of radius, the optical force oscillates with an increasing amplitude, as shown in Fig. 3(a) for \( \kappa = -0.3 \) (red), \( \kappa = 0 \) (dash) and \( \kappa = 0.3 \) (black). In both panels of Fig. 3, we take RCP beams making a half-angle \( \Theta = 85^\circ \). The magnitude of the pulling force peaks in the intermediate size range defined by the value of the laser wavelength \( \lambda_0 = 1.064 \text{ nm} \). As the radius increases further, too many multipoles contribute to the force, making it harder to identify the condition of constructive interference along the forward direction.

Figure 3(b) provides additional information on how the magnitude of the pulling effect decreases as the radius increases past \( \lambda_0 / 2 \). We plot the optical force variation with \( \kappa \) and show that the maximum pulling force for a radius of \( 1.50 \mu \text{m} \) (solid) is indeed smaller than for a radius of \( 1.24 \mu \text{m} \) (dash). More importantly, Fig. 3(b) illustrates the correlation between size and chirality already discussed in connection with Fig. 2(c). Indeed, the interval of chiral parameters selected by the pulling effect clearly depends on the radius. In the example shown in the figure, particles with radius \( 1.50 \mu \text{m} \) are pulled for smaller values of \( |\kappa| \), with the handedness selected by the helicity of the laser beams.

A sought-after scenario for applications is to sort particles with chirality parameters \( \kappa \) within a selected range of values by controlling some external parameter. Since the range of values of \( \kappa \) leading to pulling varies critically on the half-angle \( \Theta \), it is possible to sort particles with a desired chirality by adjusting the directions of the laser beams. In Fig. 4, we plot the normalized optical force \( F_2 / F_0 \) as a function of \( \Theta \) and \( \kappa \) for RCP beams and microsphere radii (a) \( 1.26 \mu \text{m} \), (b) \( 1.27 \mu \text{m} \), (c) \( 1.49 \mu \text{m} \) and (d) \( 1.50 \mu \text{m} \). As in Fig. 2, only pulling forces \( F_2 < 0 \) are shown for clarity. In all cases, the minimum \( |\kappa| \) leading to pulling depends strongly on \( \Theta \) and can be tuned to arbitrarily small values by selecting the appropriate angle for a given radius. For instance, for the radius \( 1.26 \mu \text{m} \), corresponding to panel (a), one can pull particles with axial chirality parameters as low as \( \kappa \approx -4 \times 10^{-3} \) by selecting \( \Theta = 66.5^\circ \). Similar examples can be inferred for the other panels shown in the figure. Such capability of sorting particles with unprecedented, very small chiral parameters clearly outperforms existing chiral resolution methods based on optical forces [31], which are typically limited to chiral particles satisfying \( |\kappa| \gtrsim 0.1 \). Figure 4 also reveals that the maximal angle \( \Theta = 90^\circ \) does not impose a fundamental limit on the values of \( \kappa \) of the particles that can be sorted. Indeed, these values depend on the radius, which hence can be exploited as a free parameter.
to select the desired range of $\kappa$ for chiral resolution. Figure 4 also indicates the maximum value of $\Theta$ that leads to pulling only of particles with the same handedness of the circularly-polarized laser beams. In the example shown in the figure, we have taken RCP beams, so pulling is achieved only for right-handed particles ($\kappa < 0$) as long as $\Theta$ is smaller than the value at the intersection between the colored region and the horizontal dashed lines in Fig. 4.

In summary, we have put forward an all-optical method for chiral resolution and enantioselection of single chiral microspheres based on optical pulling forces or tractor beams. Chiral particles with the same handedness of the circularly-polarized laser beams are optically pulled, whereas those with opposite handedness are flushed by radiation pressure. It is possible to fine-tune the range of chiral parameters by adjusting the angle $2\Theta$ between the laser beams, which can be controlled with a high precision in typical optical setups. Our proposal allows for an all-optical sorting of multipolar chiral microspheres with arbitrarily small chiral parameters, thus outperforming current optical methods. Altogether our findings pave the way for novel applications of optical pulling forces, such as chiral resolution, sorting, and self-assembling of chiral microparticles.

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