Cancellation of non-conservative scattering forces in optical traps by counter-propagating beams

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Non-conservative forces in optical tweezers generate undesirable behavior, such as particle loss due to radiation pressure and the preclusion of the thermodynamic equilibrium. Here, we rigorously derive criteria for the elimination of non-conservative forces, and describe how these criteria can be met by a large class of counter-propagating, focused optical beams.

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The optical trapping of particles has applications in a wide range of scientific domains, including biochemistry [1], fundamental thermodynamics [2,3], and metrology [4,5], and could enable the study of macroscopic quantum physics [6,7]. Particle trapping is accomplished by forces that arise from electromagnetic field gradients. These trapping forces generally include a non-conservative component known as the scattering force [8,9]. While this non-conservative component is often neglected in experiments, it can have dramatic effects, such as preventing the trapping of small particles [10–12] or generating the nontrivial dynamics of the trapped object and thereby preventing thermodynamic equilibrium [13,14]. Fundamentally, it prohibits the force on trapped particles from being described using a simple scalar potential. Therefore, it is important to understand and potentially limit the effects of the scattering force.

Experimentally, counter-propagating trapping beams are often used to reduce the scattering force [15–17]. While this approach obviously reduces radiation pressure, which is a component of a non-conservative force [18,19], it does not necessarily cancel the scattering force entirely. In this Letter, we consider a trapped particle that is small compared to the wavelength of the trapping beam; that is, in the Rayleigh regime. We present a generalized set of criteria for optical trapping beams under which the scattering force is completely eliminated. We also highlight how these criteria can be met through the use of counter-propagating beams, as depicted in Fig. (1).

The force acting on a small particle whose velocity is much smaller than the speed of light (relativistic limit) can be expressed as [9,20]

$$\vec{F}(\vec{r},\omega) = \frac{\alpha'(\omega)}{2}\mathrm{Re}\left\{\left\langle \dot{\nabla}\left[\vec{E}^{\dagger}(\vec{r},\omega)\dot{\vec{E}}(\vec{r},\omega)\right]\right\rangle\right\} + \frac{\alpha''(\omega)}{2}\mathrm{Im}\left\{\left\langle \dot{\nabla}\left[\vec{E}^{\dagger}(\vec{r},\omega)\dot{\vec{E}}(\vec{r},\omega)\right]\right\rangle\right\}, \quad (1)$$

where $\vec{E}(\vec{r},\omega)$ is the electric field column vector at position $\vec{r}$ with frequency ω, $\alpha'(\omega)$ and $\alpha''(\omega)$ are the real and imaginary parts of the (retarded) polarizability of the particle, the gradient operator $\dot{\nabla}$ acts only on terms with an overdot [20], † indicates conjugate-transpose, and the angle brackets represent an ensemble average over the monochromatic field realizations. The term that includes the imaginary part embodies the scattering force and is canceled under the condition [9]

$$\mathrm{Im}\left\{\left\langle \dot{\nabla}\left[\vec{E}^{\dagger}(\vec{r},\omega)\dot{\vec{E}}(\vec{r},\omega)\right]\right\rangle\right\} = \vec{0}. \quad (2)$$

We proceed with the intention of finding nontrivial fields that satisfy Eq. (2). In a particle-trapping experiment, the fields of Eqs. (1) and (2) are typically those in the focus of one or several microscope objectives. The electric field near the focus of a high Fresnel number objective is given in terms of the field $\vec{E}_\infty$ in the pupil plane at infinity by [9]

$$\vec{E}(\vec{r},\omega) = -\frac{\mathrm{i}kf e^{-\mathrm{i}kf}}{2\pi}\int_0^{\pi/2}\int_0^{2\pi}\vec{E}_\infty(\theta,\phi)K(\vec{r},\theta,\phi)\mathrm{d}\phi\mathrm{d}\theta, \quad (3)$$

where $K(\vec{r},\theta,\phi) \coloneqq \sin\theta\exp[\mathrm{i}k\hat{u}^{\mathsf{T}}\vec{r}]$, $\hat{u}(\theta,\phi) \coloneqq \sin\theta\cos\phi\hat{x} + \sin\theta\sin\phi\hat{y} + \cos\theta\hat{z}$, T indicates the matrix transpose, f is the focal length of the objective, $k = n\omega/c$ is the wavenumber, n is the refractive index of the medium in the focal region, θ and ϕ are, respectively, the polar and azimuthal spherical coordinates, each vector $\hat{a}(a = x, y, z)$ is a Cartesian unit column vector, and $\vec{r}$ is a position vector measured from the origin/geometric focus (see Fig. 1). If multiple beams are to be used, it is necessary to describe a rotation and/or translation of the

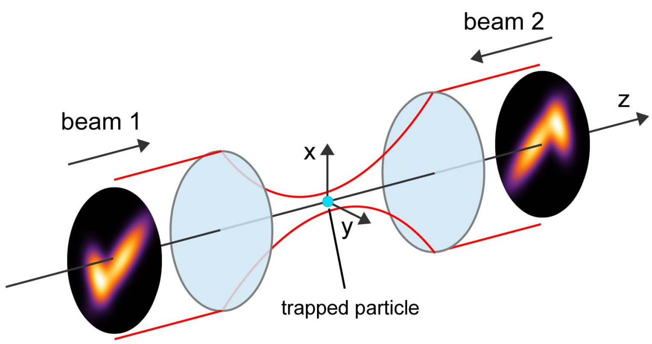

Fig. 1. Illustration of counter-propagating beams that form an optical trap. The two beams, represented by intensity cross-sections and focused by lenses, must satisfy Eq. (7) in order for the non-conservative scattering force acting on the trapped Rayleigh particle to cancel.

vector field given by Eq. (3). It is possible to describe a rotation followed by a translation of a vector function $\vec{f}(\vec{r})$ with respect to the coordinate axes in the following way. Let $\mathbf{R}$ be a rotation matrix, and let $\vec{r}_0$ be the translation vector. Then, under rotation followed by translation, $\vec{f}(\vec{r}) \to \mathbf{R}\vec{f}(\mathbf{R}^\mathsf{T}\vec{r} - \vec{r}_0)$. The field $\vec{E}$ in the focus of N microscope objectives, allowing for rotation and translation with respect to a fixed origin, is therefore given by

$$\vec{E}(\vec{r},\omega) = \frac{\mathrm{i}k}{2\pi}\sum_{p=1}^{N} c_p \int_0^{\pi/2}\int_0^{2\pi} \mathbf{R}_p \vec{E}_{\infty,p}(\theta,\phi) \times K(\mathbf{R}_p^\mathsf{T}\vec{r} - \vec{r}_p, \theta, \phi)\mathrm{d}\phi\mathrm{d}\theta, \tag{4}$$

where $c_m := -f_m e^{-\mathrm{i}kf_m}$. Here, f_m, $\vec{E}_{\infty,m}$, $\mathbf{R}_m$, and $\vec{r}_m$ represent, respectively, the focal length, angular field spectrum, rotation matrix, and translation vector associated with the mth microscope objective. Using Eq. (4), we find $\langle \dot{\nabla}(\vec{E}^\dagger(\omega)\dot{\vec{E}}(\omega))\rangle$

$$= \frac{k^2}{(2\pi)^2}\sum_{p,q}^{N} c_p^* c_q \iint \langle \vec{E}^\dagger_{\infty,p}(\Omega)\mathbf{R}_p^\mathsf{T}\mathbf{R}_q\vec{E}_{\infty,q}(\Omega')\rangle \times K^*(\mathbf{R}_p^\mathsf{T}\vec{r} - \vec{r}_p, \Omega)\nabla K(\mathbf{R}_q^\mathsf{T}\vec{r} - \vec{r}_q, \Omega')\mathrm{d}\Omega'\mathrm{d}\Omega$$
$$= \frac{\mathrm{i}k^3}{(2\pi)^2}\sum_{p,q}^{N} c_p^* c_q \iint \mathbf{R}_q\hat{u}(\Omega')\langle \vec{E}^\dagger_{\infty,p}(\Omega)\mathbf{R}_p^\mathsf{T}\mathbf{R}_q\vec{E}_{\infty,q}(\Omega')\rangle \times K^*(\mathbf{R}_p^\mathsf{T}\vec{r} - \vec{r}_p, \Omega)K(\mathbf{R}_q^\mathsf{T}\vec{r} - \vec{r}_q, \Omega')\mathrm{d}\Omega'\mathrm{d}\Omega, \tag{5}$$

where we have condensed the integration variables of Eq. (4) into Ω.

It is of practical interest to be able to solve Eq. (2) in terms of the incoming beams just before the objectives. Each spectrum $\vec{E}_{\infty,m}$ can be expressed in terms of the incoming paraxial field $\vec{E}^{\,i}_m(\theta,\phi)$ (with Cartesian components) in the back focal plane of the mth objective by the following matrix relation [21,22]:

$$\vec{E}_{\infty,m}(\theta,\phi) = \mathbf{Q}_m(\theta,\phi)\vec{E}^{\,i}_m(\theta,\phi). \tag{6}$$

In the matrix $\mathbf{Q}_m(\theta,\phi) := a_m(\theta)\mathbf{M}^\mathsf{T}(\phi)\mathbf{I}_m(\theta)\mathbf{L}(\theta)\mathbf{M}(\phi)$, $a_m(\theta) = (\cos\theta)^{1/2}\Theta(-\theta + \vartheta_m)$ under the Abbe sine condition, $\Theta(x)$ is the Heaviside unit step function, and $\vartheta_m := \arcsin(\mathrm{NA}_m/n)$. NA_m is the numerical aperture of the mth objective, $\mathbf{M}(\phi) := \cos\phi(\hat{x}\hat{x}^\mathsf{T} + \hat{y}\hat{y}^\mathsf{T}) + \sin\phi(\hat{x}\hat{y}^\mathsf{T} - \hat{y}\hat{x}^\mathsf{T}) + \hat{z}\hat{z}^\mathsf{T}$ is a rotation matrix that decomposes the field into its s- and p-polarized components, $\mathbf{I}_m(\theta) := t^s_m(\theta)\hat{y}\hat{y}^\mathsf{T} + t^p_m(\theta)(\hat{x}\hat{x}^\mathsf{T} + \hat{z}\hat{z}^\mathsf{T})$ is the matrix that describes the generally complex transmission coefficients of the mth objective for s- and p-polarization, and $\mathbf{L}(\theta) := \cos\theta(\hat{x}\hat{x}^\mathsf{T} + \hat{z}\hat{z}^\mathsf{T}) + \sin\theta(\hat{x}\hat{z}^\mathsf{T} - \hat{z}\hat{x}^\mathsf{T}) + \hat{y}\hat{y}^\mathsf{T}$ is the matrix that determines the rotation of an incoming ray.

It is important to note that each paraxial beam is defined as originally propagating along the z-axis such that the transverse field distribution corresponds to the field in the xy-plane (see e.g., Fig. 2). The rotation matrix $\mathbf{R}_m$ then acts to rotate the vector field of the mth beam around the origin. The spherical coordinates θ and ϕ are related to x and y by $x = f\sin\theta\cos\phi$ and $y = f\sin\theta\sin\phi$ (see [9], Section 3.5).

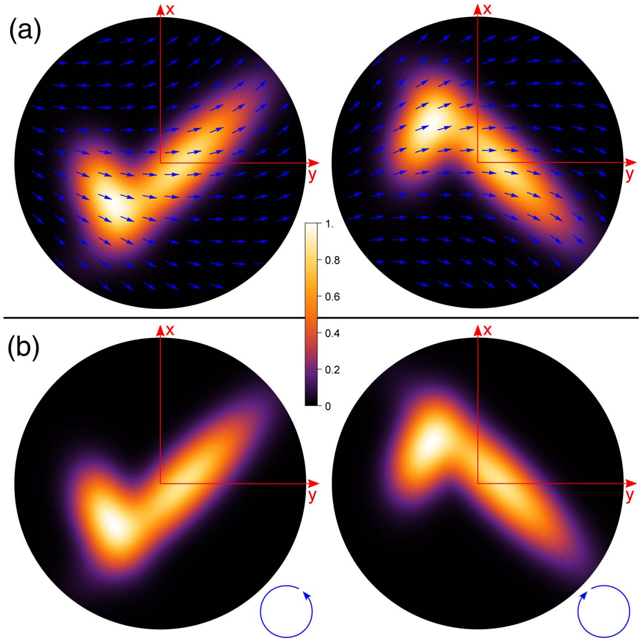

Fig. 2. Arbitrarily chosen examples of collimated beam profiles that fulfill Eqs. (14) and (15). (a) A linearly polarized beam pair where the polarization direction and intensity vary across each beam. (b) A circularly polarized beam pair where the intensity varies across each beam. The arrows indicate the direction or handedness of the polarization. The colored bar indicates the normalized intensity of the beams.

By combining Eqs. (2), (5), and (6), while noting that the inner product of two vectors is the matrix trace of their outer product, and by the invariance of the matrix trace under cyclic permutations, we find

$$\vec{0} = \int_0^{\pi/2}\int_0^{2\pi}\int_0^{\pi/2}\int_0^{2\pi} \mathrm{Re}\Big\{\sum_{p,q}^{N} c_p^* c_q \times \mathrm{Tr}[\mathbf{Q}_q^\mathsf{T}(\theta',\phi') \times \mathbf{R}_q^\mathsf{T}\mathbf{R}_p\mathbf{Q}_p(\theta,\phi)\mathbf{W}_{pq}(\theta,\phi,\theta',\phi')] \times K^*(\mathbf{R}_p^\mathsf{T}\vec{r} - \vec{r}_p, \theta,\phi)K(\mathbf{R}_q^\mathsf{T}\vec{r} - \vec{r}_q, \theta',\phi') \times [\mathbf{R}_q\hat{u}(\theta',\phi')]\Big\}\mathrm{d}\phi'\mathrm{d}\theta'\mathrm{d}\phi\mathrm{d}\theta, \tag{7}$$

where $\mathbf{W}_{mn}(\theta,\phi,\theta',\phi') := \langle \vec{E}^{i*}_m(\theta,\phi)\vec{E}^{i\mathsf{T}}_n(\theta',\phi')\rangle$ is the cross-spectral density matrix between the mth and nth paraxial input beams. Equation (7) is an important result—it allows us to recast the problem in terms of the coherence between the input beams.

We proceed by finding sufficient conditions under which multiple beams can be used to cancel scattering forces. We begin by noticing that if the quantity within the braces in Eq. (7) is purely imaginary for all values of $(\vec{r},\theta,\phi,\theta',\phi')$, then the integrand is always zero and the equation is satisfied. Let $\mathbf{A}_{mn}(\theta,\phi,\theta',\phi') := \mathbf{Q}_n^\mathsf{T}(\theta',\phi')\mathbf{R}_n^\mathsf{T}\mathbf{R}_m\mathbf{Q}_m(\theta,\phi)$ and let

$$\vec{h}_{mn}(\vec{r},\theta,\phi,\theta',\phi') := \mathbf{R}_n\hat{u}(\theta',\Phi'_{mn})\mathrm{Tr}[\mathbf{A}_{mn}(\theta,\Phi_{mn},\theta',\Phi'_{mn}) \times \mathbf{W}_{mn}(\theta,\Phi_{mn},\theta',\Phi'_{mn})] \times K^*(\mathbf{R}_m^\mathsf{T}\vec{r},\theta,\Phi_{mn})K(\mathbf{R}_n^\mathsf{T}\vec{r},\theta',\Phi'_{mn}),$$

where $\Phi'_{mn}(\phi') := a'_{mn}\phi' + b'_{mn}$, $\Phi_{mn}(\phi) := a_{mn}\phi + b_{mn}$, each a_{mn} and a'_{mn} takes a value of 1 or −1, and each b_{mn} and b'_{mn} is a scalar. These variable changes from ϕ/ϕ' to Φ_{mn}/Φ'_{mn} are allowed by the fact that each function of ϕ/ϕ' in Eq. (7) is 2π-periodic. In the following, we are interested in the values of a'_{mn}, a_{mn}, b'_{mn}, and b_{mn}, which lead to a cancellation of the scattering force. Then, assuming identical objectives and overlapping foci, it is sufficient to find solutions for each of the N^2 matrices $\mathbf{W}_{mn}$, which satisfy the following equation:

$$\vec{0} = \mathrm{Re}\left\{\sum_{p,q}^{N} \vec{h}_{pq}(\vec{r}, \theta, \phi, \theta', \phi')\right\}. \tag{8}$$

The solutions provided hereafter will be those that satisfy Eq. (8).

We first consider the special case of two ($N = 2$) identical objectives with overlapping foci. This special case reduces the mathematical complexity and facilitates understanding. At the end, we consider the general case of N beams. In the specific case of two beams, it is sufficient that

$$\vec{h}_{22} = -\vec{h}^*_{11}, \tag{9}$$

and

$$\vec{h}_{21} = -\vec{h}^*_{12}. \tag{10}$$

Without a loss of generality, we can assume that $\mathbf{R}_1 = \hat{x}\hat{x}^\mathsf{T} + \hat{y}\hat{y}^\mathsf{T} + \hat{z}\hat{z}^\mathsf{T}$, which is the identity matrix, that $\mathbf{R}_2(\varphi) = \hat{x}\hat{x}^\mathsf{T} + \cos\varphi(\hat{y}\hat{y}^\mathsf{T} + \hat{z}\hat{z}^\mathsf{T}) + \sin\varphi(\hat{z}\hat{y}^\mathsf{T} - \hat{y}\hat{z}^\mathsf{T})$, which is a rotation around the x-axis, and that $\Phi_{22} = \phi$, $\Phi'_{22} = \phi'$, $\Phi_{21} = \phi$, and $\Phi'_{21} = \phi'$. We also assume that the Fresnel transmission coefficients of the objectives $t^s(\theta)$ and $t^p(\theta)$ are real values such that all values of $\mathbf{A}_{mn}$ are real. Then, the only nontrivial solution of Eq. (9) is found by setting $\varphi = \pi$, $\Phi_{11} = -\phi + \pi$, and $\Phi'_{11} = -\phi' + \pi$, which gives

$$\mathbf{W}_{22}(\theta, \phi, \theta', \phi') = \mathbf{B} \circ \mathbf{W}^*_{11}(\theta, -\phi + \pi, \theta', -\phi' + \pi), \tag{11}$$

where $\circ$ indicates the element-wise (Hadamard) product and $\mathbf{B} := (\hat{x}\hat{x}^\mathsf{T} + \hat{y}\hat{y}^\mathsf{T} + \hat{z}\hat{z}^\mathsf{T} + \hat{z}\hat{y}^\mathsf{T} + \hat{y}\hat{z}^\mathsf{T}) - (\hat{x}\hat{y}^\mathsf{T} + \hat{x}\hat{z}^\mathsf{T} + \hat{y}\hat{x}^\mathsf{T} + \hat{z}\hat{x}^\mathsf{T})$.

Similarly, the only nontrivial solution of Eq. (10) is found by setting $\varphi = \pi$, $\Phi_{12} = -\phi + \pi$, and $\Phi'_{12} = -\phi' + \pi$, which gives

$$\mathbf{W}_{21}(\theta, \phi, \theta', \phi') = \mathbf{B} \circ \mathbf{W}^*_{12}(\theta, -\phi + \pi, \theta', -\phi' + \pi) \tag{12}$$

which is

$$= \mathbf{B} \circ \mathbf{W}^\mathsf{T}_{21}(\theta', -\phi' + \pi, \theta, -\phi + \pi), \tag{13}$$

where in the last line, we have used the fact that $\mathbf{W}_{mn}(\theta, \phi, \theta', \phi') = \mathbf{W}^\dagger_{nm}(\theta', \phi', \theta, \phi)$. It is important to note that we have arbitrarily chosen $\mathbf{R}_2$ to be a rotation around the x-axis. A different choice of rotation axis would lead to different relations.

Equations (11)–(13) provide concise relations for the properties of the two beams which, if fulfilled, would lead to the cancellation of the scattering forces. However, for the sake of example, it is useful to note that Eq. (11) can be rewritten as

$$\begin{aligned}\langle \vec{E}^i_2(\theta, \phi)\vec{E}^{i\dagger}_2(\theta', \phi')\rangle = \langle [\vec{v}^* \circ \vec{E}^{i*}_1(\theta, -\phi + \pi)] \\ \times [\vec{v} \circ \vec{E}^i_1(\theta', -\phi' + \pi)]^\mathsf{T}\rangle,\end{aligned} \tag{14}$$

where $\vec{v} := e^{i\gamma}(\hat{x} - \hat{y} - \hat{z})$ and γ is a random variable that takes values between 0 and 2π. Equation (12) can be similarly written as

$$\begin{aligned}\langle \vec{E}^i_2(\theta, \phi)\vec{E}^{i\dagger}_1(\theta', \phi')\rangle = \langle [\vec{v}^* \circ \vec{E}^{i*}_1(\theta, -\phi + \pi)] \\ \times [\vec{v} \circ \vec{E}^i_2(\theta', -\phi' + \pi)]^\mathsf{T}\rangle.\end{aligned} \tag{15}$$

Equations (14) and (15) lead to illuminating examples of beam pairs that give only conservative trapping forces. We consider the case in which the *realizations* of the beams are related in the following way, as suggested by Eqs. (14) and (15):

$$\vec{E}^i_2(\theta, \phi) = \vec{v}^* \circ \vec{E}^{i*}_1(\theta, -\phi + \pi). \tag{16}$$

For instance, if beam 1 is collimated and everywhere linearly polarized, then beam 2 must also be linearly polarized, such that Eq. (16) is satisfied [Fig. 2(a)]. If beam 1 is collimated and everywhere circularly polarized, then beam 2 must also be circularly polarized, but with the opposite handedness [Fig. 2(b)].

Mutually coherent beams give rise to standing waves near the focus, including points where the field is canceled (nulls). Standing waves may be undesirable, especially if there is a phase drift between the beams such that the extrema in the field move over time. However, the depth of the coherence-induced extrema in the field can be reduced or eliminated by controlling the coherence between the beams, which depends on the random phase variable γ inside $\vec{v}$ in Eq. (16). Let $p(\gamma)$ be the probability density function for γ. If $p(\gamma) = \delta(\gamma - \eta)$, where η is a constant and $\delta(x)$ is the Dirac delta distribution, then beams 1 and 2 are coherent and true nulls will be present. If $p(\gamma) = 1/(2\pi)$, $0 \le \gamma < 2\pi$, then beams 1 and 2 are incoherent, and there will be no coherence-induced extrema. Other probability distributions would lead, in general, to partial coherence between the beams. Experimentally, the phase could be controlled by an optical phase modulator, where the modulation frequency is much larger than any oscillation frequency of the particle, in order to generate different probability distributions.

Solutions of Eq. (8) can also be found for $N > 2$. For an even number of beams, a sufficient set of solutions follows immediately from the $N = 2$ case: if the $N = 2$ case holds in a pairwise manner, where members of any particular pair are incoherent with other beam pairs, then Eq. (8) is clearly satisfied. For example, in the $N = 4$ case, it is sufficient that $\mathbf{W}_{13} = \mathbf{W}_{14} = \mathbf{W}_{23} = \mathbf{W}_{24} = \mathbf{0}$ and that beam pairs 1–2 and 3–4 each satisfy Eqs. (9) and (10). In the case of three beams ($N = 3$), by considering all possible rotations of the beams, it can be shown that nondegenerate solutions of Eq. (8) do not exist. That is, the only nontrivial solutions are those in which two of the beams overlap such that there are, essentially, two beams. We expect similar solutions with a greater odd number of beams, where each solution will be degenerate with an even numbered beam solution.

Beyond the cancellation of the scattering forces, there are some additional consequences of the solutions of the form given by Eqs. (11)–(13). First, due to the symmetry of the input beams, the trapping potential will be symmetric through the origin. Second, the optical field will apply no torque to the particle, at least in the coherent case. This can be seen by realizing that in the coherent case, the fields near the focus form a standing wave, which implies that the electric field is purely real. This further implies that the torque on the Rayleigh particle is $\vec{\tau} \propto \vec{E} \times \vec{E}^* = \vec{0}$.

In summary, we have described a set of conditions under which the scattering forces on Rayleigh particles are eliminated in an optical trap. Notably, we have shown how these conditions are met by focusing counter-propagating beams with specific coherence relations. An experimental realization of these conditions would avoid nonequilibrium situations in thermodynamics experiments with optical tweezers. Combined with methods for mapping the non-conservative components of optical forces [23,24], a realization would also constitute an interesting tool for studying the effects of the scattering force on trapped particles.

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