Optical force and torque provide unprecedented control on the spatial motion of small particles. A valid scientific question, that has many practical implications, concerns the existence of fundamental upper bounds for the achievable force and torque exerted by a plane wave illumination with a given intensity. Here, while studying isotropic particles, we show that different light-matter interaction channels contribute to the exerted force and torque; and analytically derive upper bounds for each of the contributions. Specific examples for particles that achieve those upper bounds are provided.

We study how and to which extent different contributions can add up to result in the maximum optical force and torque. Our insights are important for applications ranging from molecular sorting, particle manipulation, nanorobotics up to ambitious projects such as laser-propelled spaceships.

Optical scattering, extinction, and absorption cross sections characterize the strength of light-matter-interaction. They quantify the fraction of power a particle scatters, extends, or absorbs. To describe the interaction of light with a particle, the incident and scattered fields can be expanded into vector spherical wave functions (VSWFs). VSWFs are the eigenfunctions of the vectorial wave equation in spherical coordinates. For an isotropic, i.e., a rotationally symmetric particle, the amplitudes of the VSWFs expanding the incident and scattered fields are linked by the Mie coefficients. Each coefficient describes a channel for the light-matter-interaction and is uniquely specified by the total angular momentum (AM) number \( j \) and the parity of the fields involved in the scattering process in the respective Mie channel (MC). Depending on \( j \) and the parity, these MCs are referred to as either electric (\( a_j \)) or magnetic (\( b_j \)).

If an isotropic particle is illuminated by a plane wave in a frequency interval where only a single MC is significant, the maximum scattering cross section \( C_{\text{sca}} \) (at resonance) is \( (2j+1)\lambda^2/2\pi \). The maximum \( C_{\text{sca}} \) is attained when the particle operates in the over-coupling \((\gamma_r \gg \gamma_m)\) regime, i.e. the radiative (scattering) loss \((\gamma_r)\) is much larger than the non-radiative (Ohmic) loss \((\gamma_m)\). For a particle with a single electric dipole MC (i.e., electric dipolar particle), the maximum scattering and consequently extinction cross section \( C_{\text{ext}} \) (at resonance) corresponds to \( 3\lambda^2/2\pi \) [Fig. 1 (a)]. Similarly, the maximum absorption cross section \( C_{\text{abs}} \) (at resonance) is \((2j+1)\lambda^2/8\pi \). It occurs if the particle operates in the critical coupling \((\gamma_m = \gamma_r)\) regime, i.e. the non-radiative \((\gamma_m)\) and radiative \((\gamma_r)\) loss are equal. For an electric dipolar particle, the maximum absorption is \( 3\lambda^2/8\pi \) [Fig. 1 (a)] when \( \gamma_m = \gamma_r \).

Although optical cross sections are important in studying light-matter interaction at the nanoscale, the optical force and torque are further key quantities for which upper bounds have not yet been well studied. This is surprising considering the important applications and implications of the optical force and torque in many areas. Examples are the opto-mechanical manipulation of molecules or particles, optical sorting, or nanorobotics. Optical force and torque are also important in studying the angular momentum of light. Moreover, developing ambitious projects like laser-propelled spaceships, would benefit from an understanding of these limits.

In this Letter, based on the multipole expansion in scattering theory, we identify and analyze different terms that contribute to the exerted optical force and torque on isotropic particles; and derive upper bounds for each contribution. Next, considering these contributions, the maximum of the total optical force and torque is calculated. Contrary to the optical cross sections, the force and torque, in a general direction, contain terms that are the result of interference among different MCs.

We start by analyzing particles that are characterized by a single dipole MC. Afterwards, we consider homogeneous dielectric spheres supporting multiple MCs and distinguish different terms contributing to the force and torque. Finally, considering more general isotropic particles, the maximum optical force and torque as a function of the maximum non-negligible multipolar order is calculated. Examples for particles that maximize each of the contributions as well as the force/torque are given. The detailed derivations of the relations, supplementary figures, and more information on the theoretical background are given in a supplemental material (SM). Here, we concentrate on the presentation of the results and the discussion of the physical implications. The force and torque values are all time averaged.

**Fundamental limits on optical force (electric dipole):** An arbitrarily polarized, time harmonic plane wave, propagating in the +z direction, illuminates an isotropic electric dipolar particle. The exerted force is:

$$ F_p = \frac{1}{2} \Re(\nabla E^* \cdot p) = \frac{kI_0}{e} \Im(\alpha(\omega)) e_z = F_p e_z, $$

(1)
where $p = \epsilon_0 \alpha \mathbf{E}$ is the induced Cartesian electric dipole moment, $\alpha$ is the electric polarizability of the particle, $I_0 = \epsilon_0 |E_0|^2 / 2$ is intensity of the illumination, $k$ is the wavenumber, $\epsilon_0$ is the free space permittivity, and $c$ is the speed of light. Alternatively, based on the definition of the extinction cross section of an electric dipolar particle, 

$$C_{\text{ext,p}} = k \text{Im} (\alpha)^2 / \pi,$$

$F_p$ can be rewritten as $(I_0/c) C_{\text{ext,p}}$. The dispersion of $\alpha$ near a resonance can be expressed by a Lorentzian line-shape as\textsuperscript{29}:

$$\alpha (\omega) = \frac{\alpha_0}{\omega^2_{\text{res}} - \omega^2 - k^2 (\gamma_{\text{res}} + \gamma_r)}, \quad (2)$$

where $\omega_{\text{res}}$ is the resonance frequency, $\alpha_0$ is the resonance strength, and $\gamma_r = \alpha_0 k^2 / 6 \pi \epsilon_0$ is the radiative loss of the particle, respectively. The maximum force in Eq. 1 occurs when the particle is non-absorptive and at resonance (i.e., $\text{Im} [\alpha (\omega)]_{\text{max}} = 6 \pi k^2 / \lambda^2$), where the extinction cross section is maximized $(=3 \lambda^2 / 2 \pi)$, and reads as:

$$(F_p)_{\text{max}} = 3 F^{\text{norm}}, \quad F^{\text{norm}} = \frac{I_0 \lambda^2}{c 2 \pi}. \quad (3)$$

$(F_p)_{\text{max}}$ is the fundamental limit for the force an arbitrarily polarized plane wave can exert on an isotropic electric dipole particle. Due to the symmetry of Maxwell’s equations, the same bound can be attained for an isotropic magnetic dipolar particle. $F^{\text{norm}}$ is used as the normalization factor further on. It is important to note that $(F_p)_{\text{max}}$ depends on the resonance wavelength $(\propto \lambda^2)$ of the MC. Therefore, the longer the resonance wavelength, the larger the maximum force.

**Fundamental limits on optical torque (electric dipole):** If a circularly polarized plane wave $\mathbf{E} = E_0 e^{i k z} (\mathbf{e}_z + \sigma \mathbf{e}_y) / \sqrt{2}$, with handedness $\sigma = \pm 1$, impinges on the particle, the optical torque reads as\textsuperscript{11}:

$$N_p = \frac{1}{2} \left[ \Re (\mathbf{p} \times \mathbf{E}^*) - \frac{k^3}{6 \pi \epsilon_0} \Im (\mathbf{p}^* \times \mathbf{p}) \right]$$

$$= \sigma I_0 \frac{k \Im [\alpha (\omega)] - \frac{k^4}{6 \pi} [\alpha (\omega)]^2}{\omega} \mathbf{e}_z = N_p \mathbf{e}_z. \quad (4)$$

Based on the definition of the absorption cross section of an electric dipolar particle, $C_{\text{abs,p}} = k \text{Im} (\alpha) - k^4 |\alpha|^2 / 6 \pi^2$, $N_p$ can be rewritten as $\sigma (I_0 / \omega) C_{\text{abs,p}}$. Therefore, the torque is maximized at the maximum of the absorption cross section $(=3 \lambda^2 / 8 \pi)$ and is equal to:

$$(N_p)_{\text{max}} = 3 \sigma N^{\text{norm}}, \quad N^{\text{norm}} = \frac{I_0 \lambda^2}{\omega 8 \pi}. \quad (5)$$

$(N_p)_{\text{max}}$ is the fundamental limit on the torque exerted on an isotropic electric dipolar particle by a circularly polarized plane wave. The same bound is attained for an isotropic magnetic dipolar particle. $N^{\text{norm}}$ will be later used for normalization. Note that for an isotropic particle, it can be easily deduced from Eq. 4 that a linearly polarized plane wave exerts no torque $N = 0$.

Figure 1 (b) shows the maximal force and torque at resonance exerted by a plane wave as a function of the loss factor, i.e., $\gamma_{\text{res}} / \gamma_r$. In the over-coupling regime, $C_{\text{ext}}$, and consequently the force is maximized. On the other hand, in the critical coupling regime, the $C_{\text{abs}}$ and consequently the torque is maximized.

**Multipole expansion:** To extend our analysis to include multiple MCs, we go beyond the single channel dipole approximation. In the multipole expansion, the incident and scattered fields are expanded as\textsuperscript{1,3,29}:

$$E_{\text{inc}} = - \sum_{j=1}^{\infty} \sum_{m=-j}^{j} E_{jm} \left( p_{jm} N_{jm}^{(1)} + i q_{jm} M_{jm}^{(1)} \right),$$

$$E_{\text{ sca}} = \sum_{j=1}^{\infty} \sum_{m=-j}^{j} E_{jm} \left( a_{jm} N_{jm}^{(3)} + i b_{jm} M_{jm}^{(3)} \right), \quad (6)$$

where $[M_{jm}^{(1)} (r; \omega), N_{jm}^{(1)} (r; \omega)]$ are the regular and $[M_{jm}^{(3)} (r; \omega), N_{jm}^{(3)} (r; \omega)]$ the outgoing vector spherical wave functions (VSWFs). $(p_{jm}, q_{jm})$ and $(a_{jm}, b_{jm})$ are the amplitudes of the VSWFs expanding the incident and scattered fields. $j(j+1)$ is the eigenvalue of the AM squared $J^2$, and $m$ is the eigenvalue of the $z$-component of the AM $J_z$. $E_{jm}$ is a normalizing factor (SM).

The incident fields are assumed to be known. Their VSWF amplitudes can be calculated using orthogonality relations. The VSWF amplitudes of a plane wave are given in the SM. Finding the scattered field amplitudes is not a trivial task. However, Mie theory provides analytical solutions for an isotropic particle.\textsuperscript{3,30} The VSWF amplitudes of the scattered and incident fields are related by $a_{jm} = a_j p_{jm}$, and $b_{jm} = b_j q_{jm}$, with $a_j$ and $b_j$ being the electric and magnetic Mie coefficients.\textsuperscript{3} Each Mie coefficient has a spectral profile with certain resonance peaks. However, for any of these channels (Mie channels) the angular momentum and parity of the fields are preserved and no energy cross-coupling occurs among different MCs. Based on energy conservation, the Mie coefficients are always smaller than unity and at resonance of a non-absorbing particle they are equal to unity.\textsuperscript{31}

![](http://example.com/image.png)
Fundamental limits on optical force: The multipolar description of the force on a particle by an arbitrary illumination has been presented in \(^{22,33}\) (SM). Considering the contributions of different MCs, the force on an isotropic particle in along +z is derived as:

\[
F = \sum_{j=1}^{\infty} \left\{ F_{j^e} + F_{j^m} + F_{j^e,j^m} + F_{j^e(j+1)^e} + F_{j^m(j+1)^m} \right\}
\]

\[
= \left[ F_p + F_{Q^e} + F_{Q^m} + \ldots \right] + \left[ F_m + F_{Q^m} + F_{O^m} + \ldots \right] + \left[ F_{pm} + F_{Q^e Q^m} + F_{Q^m O^m} + \ldots \right] + \left[ F_{Q^e Q^m} + F_{Q^m O^m} + \ldots \right] + \left[ F_{mQ^m} + F_{Q^m O^m} + \ldots \right]. \quad (7)
\]

\(F_{j^e} (F_{j^m})\) is the force due to an individual electric (magnetic) MC. \(F_{j^e,j^m}\) is the force due to the spectral interference of two MCs with identical \(j\) but opposite character. \(F_{j^e(j+1)^e} (F_{j^m(j+1)^m})\) is due to the spectral interference of two electric (magnetic) MCs with the same character and \(j\) and \(j + 1\) total AM number.

Assuming an arbitrarily polarized plane wave, propagating in +z direction and illuminating an isotropic particle, we have derived the expression for different force terms and the conditions to maximize their individual contribution (SM). The results are shown in Table I. Specific examples are mentioned that maximize each term. Figure 2 shows different non-zero contributions to the optical force exerted on a non-absorbing dielectric sphere up to the multipole order \(j = 3\) (In Fig. S2 the same is considered for an absorbing particle, which shows a significant damping in the force near resonance). For all the upcoming figures, the maximum contribution of each force term is shown by a same color dashed line. Let us now focus on maximizing the total optical force. For a homogeneous sphere like the one in Fig. 2, at least for the lower size parameter values, where the spectral overlap of MCs is small, the interference terms are small and the optical force can be approximated by the individual contribution of MCs. Therefore, in this case, the maximal total optical force is well approximated by the maximum of the individual contribution of the MCs, \((2j + 1)F^{\text{norm}}\).

### Table I: Fundamental limits on optical force constituents

| Term | \(F/F^{\text{norm}}\) | \((F)_{\text{max}}/F^{\text{norm}}\) | Maximum at (over-coupling regime) | Example |
|------|-----------------|----------------------------|-------------------------------|----------|
| \(F_{j^e}\) | \(\frac{\sigma_{j^e}}{2\pi} C_{\text{ext},j^e} = (2j + 1) \Re (a_j)\) | \((2j + 1)\) | \(|a_j|\) resonance | dielectric sphere (Fig. 2) |
| \(F_{j^m}\) | \(\frac{\sigma_{j^m}}{2\pi} C_{\text{ext},j^m} = (2j + 1) \Re (b_j)\) | \((2j + 1)\) | \(|b_j|\) resonance | dielectric sphere (Fig. 2) |
| \(F_{j^e,j^m}\) | \(-\frac{2(2j+1)}{\gamma(j+1)} \Re (a_j b_j^*\) | \(-\frac{2(2j+1)}{\gamma(j+1)}\) | simultaneous \(|a_j|\) and \(|b_j|\) resonance | dual dielectric sphere (Fig. S3) |
| \(F_{j^e(j+1)^e}\) | \(-\frac{2j(j+2)}{(j+1)} \Re (a_j a_{j+1}^*)\) | \(-\frac{2j(j+2)}{(j+1)}\) | simultaneous \(|a_j|\) and \(|a_{j+1}|\) resonance | dielectric core-multishell (Fig. S4) |
| \(F_{j^m(j+1)^m}\) | \(-\frac{2j(j+2)}{(j+1)} \Re (b_j b_{j+1}^*)\) | \(-\frac{2j(j+2)}{(j+1)}\) | simultaneous \(|b_j|\) and \(|b_{j+1}|\) resonance | dielectric core-multishell |

### Table II: Fundamental limits on optical torque constituents

| Term | \(N/N^{\text{norm}}\) | \((N)_{\text{max}}/N^{\text{norm}}\) | Maximum at (critical-coupling regime) | Example |
|------|-----------------|----------------------------|-------------------------------|----------|
| \(N_{j^e}\) | \(\frac{\sigma_{j^e}}{\omega} C_{\text{abs},j^e} = 4\sigma (2j + 1) \Re (a_j - |a_j|^2)\) | \((2j + 1)\) | \(|a_j|\) resonance | dielectric sphere (Fig. S5) |
| \(N_{j^m}\) | \(\frac{\sigma_{j^m}}{\omega} C_{\text{abs},j^m} = 4\sigma (2j + 1) \Re (b_j - |b_j|^2)\) | \((2j + 1)\) | \(|b_j|\) resonance | dielectric sphere (Figs. 4-5) |

**FIG. 2:** Non-absorbing dielectric sphere: (a) Optical force exerted by an arbitrarily polarized plane wave on a non-absorbing sphere \((\epsilon_r = (3.5)^2, \mu_r = 1)\) depending on the sphere’s size parameter (solid line). Contributions of the non-interference terms (dashed line). (b) The individual contributions of the dipole, quadrupole, and octopole electric and (c) magnetic MCs. (d) The partial contribution of the interference of dipole-dipole, quadrupole-quadrupole and octopole-octopole electric and magnetic MCs. (e) The interference of dipole-quadrupole and quadrupole-octopole, electric and magnetic MCs.

The individual contribution of a MC is directly related to the extinction cross section (SM). Although the \((2j + 1)\) factor is bigger for higher multipoles, the resonance wavelengths of the channels are lower and hence in general, for an isotropic particle, the achievable maximum force...
To calculate the maximum total optical force, the contribution of interference terms should be considered. For an isotropic particle with finite volume and finite permeability and permittivity (positive or negative), which is illuminated by a plane wave at a given frequency, avoiding pathological cases, the optical response to any order of accuracy is assumed to be describable with a finite number of multipole moments and hence the total optical force is always finite. For a non-absorbing particle, each MC coefficient is modeled by a simple formula \( a_j = \cos \alpha_j \exp i \alpha_j, \quad b_j = \cos \beta_j \exp i \beta_j \) with a single real-valued variable \( \alpha_j \) or \( \beta_j \). Based on this model and Eq. 7, assuming that the resonance of the MCs can be optimally engineered (i.e. assuming all \( \alpha_j \) and \( \beta_j \) to be independent), the maximum force can be calculated as a function of the maximum non-negligible multipole order \( j_{\text{max}} \). For a smaller number of Mie channels the maximal force can be derived analytically, i.e. up to \( j_{\text{max}} = 3 \). For a larger number of Mie channels a genetic algorithm has been used (SM). The results are shown in Fig. 3(a). For a dipolar particle, the maximum of the optical force is 3.375\( F_{\text{norm}} \). This upper bound for the force is not met at the resonance due to the interference contribution. Using particle swarm optimization (PSO)\(^{36} \), we have optimized a dielectric core-multishell particle that receives the maximum optical force in dipolar approximation at \( \lambda = 1 \mu m \) [Fig. 3(c)] (SM). This demonstrates the applicability of the present formalism.

**Fundamental limits on optical torque:** In a similar approach, the z-component of the optical torque exerted on an isotropic particle by an arbitrary illumination, presented in Ref. 32, can be rewritten as:

\[
N = \sum_{j=1}^{\infty} \left\{ N_{j^e} + N_{j^m} \right\} = [N_p + N_{Q^e} + N_{O^e} + ...] + [N_m + N_{Q^m} + N_{O^m} + ...],
\]

where \( N_{j^e} \) (\( N_{j^m} \)) is the contribution of an electric (magnetic) MC to the optical torque. The contribution of an electric MC to the torque is:

\[
N_{j^e} = \frac{\lambda^3 I_0}{8 \pi^2 c} \Re \left( a_j - |a_j|^2 \right) \sum_{m=-j}^{j} m |p_{jm}|^2 = \frac{\sigma I_0 C_{\text{abs,j}^e}}{\omega}. \quad (9)
\]

The exerted torque is directly related to the averaged total \( A \) of the incident wave in the z-direction. For a circularly polarized plane wave propagating along z, the contribution of an electric MC to the torque simplifies to:

\[
N_{j^e} = 4\sigma (2j + 1) \Re \left( a_j - |a_j|^2 \right) N_{\text{norm}}. \quad (10)
\]

For an absorbing isotropic particle at the critical coupling regime, \( \Re \left( a_j - |a_j|^2 \right) \) is maximum and equal to 0.25. Therefore, the maximum torque is derived as:

\[
(N_{j^e})_{\text{max}} = \sigma (2j + 1) N_{\text{norm}}. \quad (11)
\]

**FIG. 3:** (a) Maximum total optical force and (b) torque as a function of the maximum non-negligible multipole moment order \( j_{\text{max}} \). (c) Optical force exerted on an optimized isotropic non-absorbing core-shell particle, illuminated by an arbitrarily polarized plane wave. The figure also illustrates the contributions of the individual dipolar electric and magnetic MCs and their interference. Parameters of the particle: \( r_1 = 88 \text{ nm}, \quad r_2 = 179 \text{ nm}, \quad \epsilon_1 = 4^2 \quad \text{and} \quad \epsilon_2 = (2.64)^2 \).

**FIG. 4:** Absorbing dielectric sphere: (a) Optical torque on an absorbing sphere \( [\epsilon_2 = (3.5 + 0.16)i, \quad \mu_2 = 1] \), by a circularly polarized plane wave as a function of the sphere’s size parameter. The individual contribution of the dipole, quadrupole, and octopole electric and (b) magnetic MCs.

is smaller for higher order multipoles.

When extending our analysis to more general isotropic particles, interference terms come into play. Figure S3 considers the optical force on a dual \(^{34} \) dielectric sphere. Due to the simultaneous resonance of the electric \( (a_j) \) and magnetic \( (b_j) \) MCs, the contribution of the interference term \( F_{j^e,j^m} \) is maximized. In Fig. S4, a core-multishell particle is analyzed, where the electric dipole-quadrupole and quadrupole-octopole interference force contributions are maximized \( (F_{Q^e} = -3F_{\text{norm}}, \quad F_{Q^m} \approx -5.3F_{\text{norm}}) \). Note that the maximum contribution of the interference terms is always negative. However, as proven in SM, the maximum positive contribution of an interference term (at off-resonance) is 8 times smaller than the maximum negative contribution (at resonance).
A similar relation can be derived for the contribution of a magnetic MC. Table II summarizes the two contributions and the conditions for maximizing them. To maximize the total optical torque, $C_{\text{abs}}$ should be maximized, i.e. the critical coupling condition should be satisfied. The critical coupling is met at a single frequency. In Fig. 4, the optical torque is critically coupled at the magnetic dipole MC resonance.

Unlike the force, the torque along $+z$, does not have interference terms and contribution of MCs directly add up. Therefore, for a dual (identical $a_j$ and $b_j$) sphere that is critically coupled, from Table II, the torque is $(N_{j'}\max + N_{j}\max) = 2(2j+1)N_{\text{norm}}$. This is shown in Fig. S5. As a simple extrapolation, it can be concluded that the maximum optical torque on a critically coupled particle occurs when all the multipole moments overlap resonantly. Therefore, the maximum optical torque as a function of $j_{\max}$ is $\sum_{j=1}^{j_{\max}} 2(2j+1)N_{\text{norm}} = 2j_{\max}(j_{\max} + 2)$. This relation is plotted in Fig. 3(b).

For completeness, we have calculated the optical force and torque on a silver nanosphere (Fig. S6) to compare our results with a realistic absorbing particle. Our results can be used to design super-acceleratable and -rotatable particles by engineering the spectral resonance of the MCs. The designed particles can in turn be used in opto-nanorobots.

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Supplemental Material for:

“Fundamental Limits of Optical Force and Torque”

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S2
I. MULTIPOLE EXPANSION

A macroscopically homogeneous scatterer is illuminated by an arbitrary incident electromagnetic wave $\mathbf{E}_{\text{inc}}$ and scatters the field $\mathbf{E}_{\text{sca}}$. Based on the multipole theory of electromagnetics, the scattered and incident fields can be expanded in vector spherical wave functions (VSWF) as [1, 2]:

$$\mathbf{E}_{\text{sca}}(r; \omega) = \sum_{j=1}^{\infty} \sum_{m=-j}^{j} E_{jm} \left[ a_{jm}(\omega) \mathbf{N}_{jm}^{(3)}(r; \omega) + i b_{jm}(\omega) \mathbf{M}_{jm}^{(3)}(r; \omega) \right],$$

(S1)

$$\mathbf{E}_{\text{inc}}(r; \omega) = - \sum_{j=1}^{\infty} \sum_{m=-j}^{j} E_{jm} \left[ p_{jm}(\omega) \mathbf{N}_{jm}^{(1)}(r; \omega) + i q_{jm}(\omega) \mathbf{M}_{jm}^{(1)}(r; \omega) \right],$$

where $\mathbf{M}^{(f)}$ and $\mathbf{N}^{(f)}$ are the vector spherical wave functions (VSWFs) defined as:

$$\mathbf{M}_{jm}^{(f)}(r; \omega) = \frac{z_{j}^{(f)}(kr) \exp(\text{i}m\phi)}{\sqrt{j(j+1)}} \left[ \text{i} \pi_{jm}(\cos \theta) \mathbf{e}_{\theta} - \tau_{jm}(\cos \theta) \mathbf{e}_{\phi} \right],$$

(S2)

$$\mathbf{N}_{jm}^{(f)}(r; \omega) = \sqrt{j(j+1)} \frac{z_{j}^{(f)}(kr)}{kr} P_{jm}(\cos \theta) \exp(\text{i}m\phi) \mathbf{e}_{r} + \frac{1}{kr} \frac{d}{dr} \left[ r z_{j}^{(f)}(kr) \right] \frac{\exp(\text{i}m\phi)}{\sqrt{j(j+1)}} \left[ \tau_{jm}(\cos \theta) \mathbf{e}_{\theta} + \text{i} \pi_{jm}(\cos \theta) \mathbf{e}_{\phi} \right],$$

(S3)

where $\pi_{jm}(\cos \theta) = m P_{jm}(\cos \theta) / \sin \theta$, $\tau_{jm}(\cos \theta) = d P_{jm}(\cos \theta) / d\theta$. $z_{j}^{(1)}(kr) = j_{j}(kr)$ and $z_{j}^{(3)}(kr) = h_{j}^{(1)}(kr)$, are the spherical Bessel and Hankel functions of first kind, respectively. The choice of $f$ should respect the physical meaningfulness of the waves at the origin ($kr \to 0$) and infinity ($kr \to \infty$). $P_{jm}$ are the associated Legendre polynomials defined as:

$$P_{jm}(x) = \sqrt{(j+m)! / (j-m)!} \sqrt{(j-|m|)! / (j+|m|)!} \left[ \text{sign}(m) \right]^{m} (-1)^{|m|} (1-x^{2})^{\frac{|m|}{2}} \frac{d^{m}|m|}{dx^{m}} P_{j}(x),$$

(S4)

where $P_{j}$ are the unassociated Legendre polynomials. $a_{jm}$ and $b_{jm}$ are the amplitudes of the VSWFs expanding the scattered field. $p_{jm}$ and $q_{jm}$ are the amplitudes of the VSWFs expanding the incident field. The subscripts $j$ and $m$ are the total and $z$ component of the total angular momentum number of the interacting field. $k$ is the wavevector, and:

$$E_{jm} = |E_{0}| \sqrt{\frac{2j+1}{4\pi}} \sqrt{\frac{(j-m)!}{(j+m)!}} (-1)^{m},$$

S3
where $|E_0|$ is the magnitude of the expanded wave.

The amplitudes of the VSWFs expanding the incident field

If the incident field is known, then, their VSWF amplitudes can be calculated, using orthogonality relations, by integrating the field across a closed spherical surface of arbitrary radius $r$ as:

$$p_{jm}(\omega) = -\frac{\int_0^{2\pi} d\phi \int_0^\pi E_{\text{inc}}(r, \theta, \phi; \omega) \cdot \left[N^{(1)}_{jm}(r, \theta, \phi; \omega)\right]^* \sin \theta d\theta d\phi}{E_{jm} \int_0^{2\pi} d\phi \int_0^\pi \left|N^{(1)}_{jm}(r, \theta, \phi; \omega)\right|^2 \sin \theta d\theta},$$

(S5)

$$q_{jm}(\omega) = -\frac{\int_0^{2\pi} d\phi \int_0^\pi E_{\text{inc}}(r, \theta, \phi; \omega) \cdot \left[M^{(1)}_{jm}(r, \theta, \phi; \omega)\right]^* \sin \theta d\theta d\phi}{iE_{jm} \int_0^{2\pi} d\phi \int_0^\pi \left|M^{(1)}_{jm}(r, \theta, \phi; \omega)\right|^2 \sin \theta d\theta}.$$  

(S6)

For an incident linearly $x$- and $y$-polarized plane wave propagating in the $z$ direction, based on the derivation of [2], and renormalization to the convention we have used in this letter, the field VSWF amplitudes are as follows:

$$p_{jm}^{\text{pw},x}(\omega) = imq_{jm}^{\text{pw},x}(\omega) = -i^{j+1} m_\delta |m| \sqrt{\pi (2j + 1)},$$

(S7)

$$p_{jm}^{\text{pw},y}(\omega) = imq_{jm}^{\text{pw},y}(\omega) = -i^j \sigma |m| \sqrt{\pi (2j + 1)},$$

(S8)

where $\delta$ is the delta Kronecker. For a circularly polarized plane wave propagating in the $z$ direction $E_{\text{inc}}(r; \omega) = E_0 e^{ikz} (e_x + i\sigma e_y) / \sqrt{2}$, the incident field VSWF amplitudes are as follows ($\sigma$ is the handedness of the wave):

$$p_{jm}^{\text{cp}}(\omega) = imq_{jm}^{\text{cp}}(\omega) = \frac{p_{jm}^{\text{pw},x}(\omega) + i\sigma p_{jm}^{\text{pw},y}(\omega)}{\sqrt{2}} = -i^{j+1} \sigma m_\delta \sqrt{2\pi (2j + 1)}.$$  

(S9)

For an arbitrarily polarized plane wave, propagating along $z$, the VSWF amplitudes can be derived by a linear combination of the two linearly $x$- and $y$-polarized polarized plane wave (Should be normalized).
The amplitudes of the VSWFs expanding the scattered field and Mie theory

Finding the scattered field amplitudes are not a trivial task and normally Maxwell equation solvers are required to calculate the scattered field and derive the amplitudes through orthogonality relation, similar to the case for the incident field VSWF amplitudes. However, for an isotropic particle, analytical solutions exist, which is normally refereed to as the Mie theory [3]. The equations relating the scattered field VSWF amplitudes to that of the incident field is:

\[
a_{jm} (\omega) = a_j (\omega) p_{jm} (\omega), \quad b_{jm} (\omega) = b_j (\omega) q_{jm} (\omega),
\]

(S10)

where \(a_j\) and \(b_j\) are known as the Mie coefficients. The value of these coefficients for an isotropic sphere and core-multishell particle can be found in [2, 4]. The spectrum of the Mie coefficients consists of several resonance profiles, which are the interaction channels for the light and the particle. Throughout this letter, we refer to Mie coefficients as the Mie channels (MCs). If the particle does not absorb light, at maximum light-particle interaction (i.e. resonance), the Mie coefficients achieve the maximum value of unity.

From now on, for simplicity, we ignore the angular argument \(\omega\).

Spherical to Cartesian multipole conversion

By comparison of the far field expressions of multipole moments and the expanded fields of Eq. S1, the induced electric and magnetic Cartesian dipole moments can be related to the scattered
field VSWF amplitudes by [1]:

\[ p = \begin{pmatrix} p_x \\ p_y \\ p_z \end{pmatrix} = C_0 \begin{pmatrix} a_{11} - a_{1-1} \\ i(a_{11} + a_{1-1}) \\ -\sqrt{2}a_{10} \end{pmatrix}, \]  

\[ \mathbf{m} = \begin{pmatrix} m_x \\ m_y \\ m_z \end{pmatrix} = cC_0 \begin{pmatrix} b_{11} - b_{1-1} \\ i(b_{11} + b_{1-1}) \\ -\sqrt{2}b_{10} \end{pmatrix}, \]  

where \( C_0 = 2\sqrt{3}\pi |E_0| k/cZ_0 \), \( c \) is the speed of light, and \( Z_0 \) is the impedance of the free space.

II. OPTICAL CROSS SECTIONS

Scattering, extinction, and absorption cross sections are defined as [5]:

\[ C_{\text{sca}} = k^{-2} \sum_{n=1}^{\infty} \sum_{m=-n}^{j} \left( |a_{jm}|^2 + |b_{jm}|^2 \right), \]  

\[ C_{\text{ext}} = k^{-2} \sum_{j=1}^{\infty} \sum_{m=-j}^{j} \Re \left( a_{jm}p_{jm}^* + b_{jm}q_{jm}^* \right), \]  

\[ C_{\text{abs}} = C_{\text{ext}} - C_{\text{sca}}. \]

Below the individual contribution of an electric MC \( a_j \) to the optical cross sections of an isotropic particle is calculated.

**Scattering cross section**

The contribution of \( a_j \) to the scattering cross section of the particle when illuminated by an arbitrary illumination is:

\[ C_{\text{sca},j^e} = k^{-2} \sum_{m=-n}^{n} |a_{jm}|^2 = k^{-2} |a_j|^2 \sum_{m=-j}^{j} |p_{jm}|^2. \]
If a z-propagating arbitrarily polarized plane wave (Eqs. S7-S9) illumination is assumed, it simplifies to:

$$C_{\text{sca},j^e} = (2j + 1) \frac{\lambda^2}{2\pi} |a_j|^2 .$$  \hspace{1cm} (S17)

To maximize the above expression, the particle should be non-absorptive and the frequency of the light should be tuned for the resonance of the MC, where $a_j = 1$. Therefore, the maximum contribution is equal to [6]:

$$[C_{\text{sca},j^e}]_{\text{max}} = (2j + 1) \frac{\lambda^2}{2\pi} .$$  \hspace{1cm} (S18)

**Extinction cross section**

The contribution of $a_j$ to the extinction cross section of the particle when illuminated by an arbitrary illumination is:

$$C_{\text{ext},j^e} = k^{-2} \sum_{m=-j}^{j} \Re (a_j m p_{jm}^*) = k^{-2} \Re (a_j) \sum_{m=-j}^{j} |p_{jm}|^2 .$$  \hspace{1cm} (S19)

If a z-propagating arbitrarily polarized plane wave (Eqs. S7-S9) illumination is assumed, it simplifies to:

$$C_{\text{ext},j^e} = (2j + 1) \frac{\lambda^2}{2\pi} \Re (a_j) .$$  \hspace{1cm} (S20)

Therefore, the maximum contribution occurs at the MC resonance of a non-absorbing particle and is equal to:

$$[C_{\text{ext},j^e}]_{\text{max}} = (2j + 1) \frac{\lambda^2}{2\pi} .$$  \hspace{1cm} (S21)

Extinction cross section characterizes the total interaction of the particle with the light, i.e. the scattering plus the absorption cross section. When the non-radiative loss is very small, then, at resonance the extinction cross section maximizes. We refer to this regime as the over-coupling regime. In the over-coupling region, the absorption cross section vanishes and the scattering and extinction cross sections are both equal.
Absorption cross section

Finally, the contribution of $a_j$ to the absorption cross section of the particle is:

$$C_{\text{abs},j} = C_{\text{ext},j} - C_{\text{sca},j} = \frac{\Re (a_j) - |a_j|^2}{k^2} \sum_{m=-j}^{j} |p_{jm}|^2 = (2j + 1) \frac{\lambda^2}{2\pi} \left[ \Re (a_j) - |a_j|^2 \right]. \quad (S22)$$

For a non-absorbing particle, the cross section vanishes and $\Re (a_j) = |a_j|^2$. It can be shown that the maximum of $\Re (a_j - |a_j|^2)$ is at the channel resonance and is equal to 0.25. Therefore, the maximum absorption cross section is $[7]$:

$$[C_{\text{abs},j}]_{\text{max}} = \frac{(2j + 1) \lambda^2}{4 \frac{2\pi}{2\pi}}. \quad (S23)$$

We refer to this regime as the critical coupling, where non-radiative loss ($\gamma_{\text{nr}}$) is equal to the radiative loss ($\gamma_r$). In critical coupling regime, the scattering and absorption cross section are equal.

Similar results derive for the individual contribution of a magnetic MC $b_j$ to the optical cross sections:

$$C_{\text{sca},j} = (2j + 1) \frac{\lambda^2}{2\pi} |b_j|^2, \quad (S24)$$

$$C_{\text{ext},j} = (2j + 1) \frac{\lambda^2}{2\pi} \Re (b_j), \quad (S25)$$

$$C_{\text{abs},j} = (2j + 1) \frac{\lambda^2}{2\pi} \left[ \Re (b_j) - |b_j|^2 \right], \quad (S26)$$

$$[C_{\text{sca},j}]_{\text{max}} = \frac{(2j + 1) \lambda^2}{2\pi}, \quad (S27)$$

$$[C_{\text{ext},j}]_{\text{max}} = \frac{(2j + 1) \lambda^2}{2\pi}, \quad (S28)$$

$$[C_{\text{abs},j}]_{\text{max}} = \frac{(2j + 1) \lambda^2}{4 \frac{2\pi}{2\pi}}. \quad (S29)$$

Note that the above-mentioned upper bounds that are vastly used in the literature are only valid given the assumptions: isotropy of the particle, individual contribution of a MC and plane wave illumination. Breaking any of these assumptions might change the upper bounds. For example a
non-absorbing dipolar dual sphere can scatter twice as the upper bound derived in Eq. S18. This is because the magnetic and electric MCs overlap spectrally and the total cross section is the sum of the two. The total optical cross section is an incoherent sum of individual MC contributors and no interference terms appear if multiple MCs profiles overlap spectrally.

III. OPTICAL FORCE

Spherical multipole expansion

The optical force exerted on an arbitrary particle by an arbitrary illumination, expanded into spherical multipoles, is as follows (the formula is what [8, 9] have derived, but re-normalized to be consistent with other formulas used in this letter) (SI units):

\[
F = F_x \mathbf{e}_x + F_y \mathbf{e}_y + F_z \mathbf{e}_z, \tag{S30}
\]

\[
F_z = -\frac{I_0}{c} \frac{\lambda^2}{4\pi^2} \sum_{j=1}^{\infty} \sum_{m=-j}^{j} (j+2) \frac{(j+1)^2 - m^2}{(2j+1)(2j+3)} (2a_{j+1,m}a_{jm}^* - b_{j+1,m}a_{jm}^* + 2b_{j+1,m}b_{jm}^* - b_{j+1,m}q_{jm}^* - q_{j+1,m}b_{jm}^*) \\
+ \frac{m}{j(j+1)} (2a_{jm}b_{jm}^* - a_{jm}q_{jm}^* - p_{jm}b_{jm}^*), \tag{S31}
\]

\[
F_x + iF_y = -\frac{I_0}{c} \frac{\lambda^2}{4\pi^2} \sum_{j=1}^{\infty} \sum_{m=-j}^{j} \left\{ \sqrt{\frac{(j+m+1)(j+m+2)(j+2)}{(2j+1)(2j+3)(j+1)}} \left[ 2a_{jm}a_{j+1,m+1}^* - p_{jm}a_{j+1,m+1}^* + 2b_{jm}b_{j+1,m+1}^* - b_{jm}q_{j+1,m+1}^* \right] \\
- a_{j+1,m-1}p_{jm}^* - p_{j+1,m-1}a_{jm}^* + 2b_{j+1,m-1}b_{jm}^* - b_{j+1,m-1}q_{jm}^* \\
- q_{j+1,m-1}b_{jm}^* - \sqrt{\frac{(j+m+1)(j-m)}{j(j+1)}} \left[ -2a_{jm}b_{j,m+1}^* + 2b_{jm}a_{j,m+1}^* \right] \\
+ a_{jm}q_{j,m+1}^* - b_{jm}p_{j,m+1}^* - q_{jm}a_{j,m+1}^* + p_{jm}b_{j,m+1} \right\}. \tag{S32}
\]
For an isotropic particle, and z-propagating plane wave illumination, lateral force is always zero, i.e., \( F_x = F_y = 0 \). Therefore, only the parallel force \( F_z \) is relevant and throughout the letter, it is referred to as the exerted optical force \( F \). Assuming isotropy of the particle and rearranging the formula with respect to the Mie coefficients, the exerted optical force by an arbitrary illumination can be written as:

\[
F = \sum_{j=1}^{\infty} \left[ F_{je} + F_{jm} + F_{je,jm} + F_{je(j+1)e} + F_{jm(j+1)m} \right]. \tag{S33}
\]

**Fundamental bounds for the single constituents of the total optical force**

The optical force consists of various terms, showing different MC contributions. The individual contribution of an electric \((F_{je})/\) magnetic \((F_{jm})\) MC, the contribution of the interference of the electric and magnetic MCs \((F_{je,jm})\), the contribution of the interference of two electric MCs with adjacent \( j \) numbers \([F_{je(j+1)e}]\), and the contribution of the interference of two magnetic MCs with adjacent \( j \) numbers \([F_{jm(j+1)m}]\).

The normalizing force factor, that will be used throughout the letter, is defined as:

\[
F_{\text{norm}} = \frac{I_0 \lambda^2}{c 2\pi}. \tag{S34}
\]

Below each of the terms are analytically calculated and the upper bounds for their contribution are derived.

*Individual contribution of an electric \((F_{je})/\)magnetic \((F_{jm})\) MC:* Ignoring the interference terms, the individual contribution of an induced electric multipole to the exerted optical force
by an arbitrary illumination is derived as:

\[
F_{je} = \frac{I_0}{c} \frac{\lambda^2}{4\pi^2} \sum_{m=-j}^{j} \Im \left[ \frac{(j+2)}{(j+1)} \sqrt{\frac{(j+1)^2 - m^2}{(2j+1)(2j+3)}} p_{j+1,m} a^*_{jm} \right. \\
+ \frac{(j-1)}{j} \sqrt{\frac{(j^2 - m^2)}{(4j^2 - 1)}} p_{jm} p_{j-1,m} + \frac{m}{j(j+1)} a^*_jm q^*_jm \left. \right] \\
= \frac{I_0}{c} \frac{\lambda^2}{4\pi^2} \sum_{m=1,-1} \Im \left[ \frac{(j+2)}{(j+1)} \sqrt{\frac{(j+1)^2 - m^2}{(2j+1)(2j+3)}} p_{j+1,m} (a^*_p a^*_m) \right. \\
+ \frac{(j-1)}{j} \sqrt{\frac{(j^2 - m^2)}{(4j^2 - 1)}} p_{jm} p_{j-1,m} + \frac{m}{j(j+1)} a^*_jm q^*_jm \left. \right] \\
= \frac{I_0}{c} \frac{\lambda^2}{4\pi^2} \Im \left\{ a_j \sum_{m=-j}^{j} p_{jm} \left[ \frac{(j+2)}{(j+1)} \sqrt{\frac{(j+1)^2 - m^2}{(2j+1)(2j+3)}} p^*_{j+1,m} \right. \\
+ \frac{(j-1)}{j} \sqrt{\frac{(j^2 - m^2)}{(4j^2 - 1)}} p^*_{j-1,m} + \frac{m}{j(j+1)} q^*_jm \right\}. \tag{S35}
\]

For a z-propagating arbitrarily polarized plane wave illumination, using Eq. S7 and Eq. S20, it can be written as:

\[
F_{je} = \frac{I_0}{c} C_{ext,j^e} = (2j + 1) F_{\text{norm}} \Re (a_j). \tag{S36}
\]

Therefore, at resonance of the electric MC, for a non-absorbing particle, the maximum force contribution is:

\[
[F_{je}]_{\text{max}} = (2j + 1) \frac{I_0}{c} \frac{\lambda^2}{2\pi} = (2j + 1) F_{\text{norm}}. \tag{S37}
\]

Similarity, it can be proven that the maximum individual contribution of a magnetic MC is:

\[
[F_{jm}]_{\text{max}} = \frac{I_0}{c} [C_{ext,j^m}]_{\text{max}} = (2j + 1) \frac{I_0}{c} \frac{\lambda^2}{2\pi} = (2j + 1) F_{\text{norm}}. \tag{S38}
\]

S11
Contribution due to the interference of electric and magnetic MC with the same \( j \) \((F_{j^e,j^m})\): The partial contribution of the interference of \( a_j \) and \( b_j \) is:

\[
F_{j^e,j^m} = -\frac{2\lambda^2 I_0}{4\pi^2 cj (j + 1)} \sum_{m=-j}^{j} m \Im \left( a_{jm}b_{jm}^* \right) = -\frac{I_0 \lambda^2}{2\pi^2 c j (j + 1)} \sum_{m=-j}^{j} m \Im \left( a_j b_j^* p_{jm}q_{jm}^* \right). \tag{S39}
\]

For a z-propagating arbitrarily polarized plane wave illumination, it simplifies to:

\[
F_{j^e,j^m} = -F_{\text{norm}}^\frac{(2j + 1)}{j (j + 1)} \sum_{m=-j}^{j} m^2 \Im \left( a_j b_j^* i \delta_{|m|1} \right) = -\frac{2 (2j + 1)}{j (j + 1)} \Re \left( a_j b_j^* \right) F_{\text{norm}}. \tag{S40}
\]

The maximum contribution is when \( a_j \) and \( b_j \) simultaneously resonate and the particle is non-absorptive:

\[
[F_{j^e,j^m}]_{\text{max}} = -\frac{2 (2j + 1)}{j (j + 1)} F_{\text{norm}}. \tag{S41}
\]

Contribution of the interference of two electric \([F_{j^e(j+1)^e}]\) / two magnetic \([F_{j^m(j+1)^m}]\) MCs with adjacent \( j \): The partial contribution of the interference of \( a_j \) and \( a_{j+1} \) is:

\[
F_{j^e(j+1)^e} = -\frac{I_0 \lambda^2}{c 4\pi^2} \sum_{m=-j}^{j} \Im \left[ \frac{(j + 2)}{(j + 1)} \sqrt{\frac{(j + 1)^2 - m^2}{(2j + 1) (2j + 3)}} 2a_{j+1,m}a_{jm}^* \right] \\
= -\frac{I_0 \lambda^2}{c 2\pi^2} \frac{(j + 2)}{(j + 1)} \sqrt{\frac{j (j + 2)}{(2j + 1) (2j + 3)}} \sum_{m=-j}^{j} \Im \left[ p_{j+1,m}p_{jm}^* a_{j+1,a_j}^* \right]. \tag{S42}
\]

For a z-propagating arbitrarily polarized plane wave illumination, it simplifies to:

\[
F_{j^e(j+1)^e} = -\frac{2j (j + 2)}{(j + 1)} \Re \left( a_{j+1,a_j}^* \right) F_{\text{norm}}. \tag{S43}
\]

The maximum contribution is when an electric MC resonance spectrally overlaps with an electric MC resonance of one order higher and the particle is non-absorptive:

\[
[F_{j^e(j+1)^e}]_{\text{max}} = -\frac{2j (j + 2)}{(j + 1)} F_{\text{norm}}. \tag{S44}
\]
Similarly:

\[ F_{jm(j+1)m} = -\frac{2j(j+2)}{(j+1)} \Re (b_{j+1} b_j^*) F_{\text{norm}}, \quad (S45) \]

and the maximum value can be derived for simultaneous resonance of \( b_j \) and \( b_{j+1} \) for a non-absorbing particle:

\[ \left[ F_{jm(j+1)m} \right]_{\text{max}} = -\frac{2j(j+2)}{(j+1)} F_{\text{norm}}. \quad (S46) \]

**Maximum positive Contribution of the interference terms:** For the non-interference terms, the maximum contribution is always positive and the terms cannot be negative in the whole spectrum. On the other hand, the maximum contribution of the interference terms are always negative. However, as shown in Fig. 2, the interference terms can also be positive at some frequencies. Here, we want to find the maximum positive contribution for the interference terms. All interference terms in the simplified form (isotropic particle, plane wave illumination), can be written as:

\[ F_{jejm} = -\frac{2(2j+1)}{j(j+1)} \Re (a_j b_j^*) F_{\text{norm}}, \quad (S47) \]
\[ F_{j+j+1} = -\frac{2j(j+2)}{(j+1)} \Re (a_{j+1} a_j^*) F_{\text{norm}}, \quad (S48) \]
\[ F_{jm(j+1)m} = -\frac{2j(j+2)}{(j+1)} \Re (b_{j+1} b_j^*) F_{\text{norm}}. \quad (S49) \]

Since in general, the MCs are independent, it will be enough to calculate the extema of \( \Re (uv^*) \), where \( u \) and \( v \) are two arbitrary independent MCs. For the Mie coefficients of a non-absorbing particle, as shown before \( \Re (u) = |u|^2 \). For any complex number \( u = |u|e^{i\alpha} \). Therefore:

\[ \Re (|u|e^{i\alpha}) = |u|^2 \]
\[ |u| \cos \alpha = |u|^2 \]
\[ \cos \alpha = |u|, \quad (S50) \]

S13
where $-\frac{\pi}{2} \leq \alpha \leq \frac{\pi}{2}$. As a result, for any non-absorbing particle, the Mie coefficients can be written in the form [10]:

$$u = \cos \alpha e^{i\alpha}. \quad (S51)$$

We want to find the maximum and minimum of the function $f = \Re (uv^*)$, which appears in the interference contributions to the optical force. It can be written as:

$$f = \Re (uv^*)$$
$$= \Re (\cos \alpha e^{i\alpha} \cos \beta e^{-i\beta})$$
$$= \cos \alpha \cos \beta \cos (\alpha - \beta). \quad (S52)$$

To find the extrema of the function, derivative of the function with respect to $\alpha$ and $\beta$ is required. The maximum of the function is trivial and is equal to 1 (at the simultaneous resonance of the two MCs), the minimum of the function can be derived as $-1/8$ (off-resonance of the two MCs):

$$f(\alpha = \pm \frac{\pi}{3}, \beta = \mp \frac{\pi}{3}) = -\frac{1}{8}. \quad (S53)$$

In summary, the fundamental bounds for the different constituents are:

$$0 \leq F_{je} \leq (2j + 1) F_{\text{norm}}, \quad (S54)$$
$$0 \leq F_{jm} \leq (2j + 1) F_{\text{norm}}, \quad (S55)$$
$$-\frac{2(2j + 1)}{j(j+1)} F_{\text{norm}} \leq F_{je,jm} \leq \frac{(2j + 1)}{4j(j+1)} F_{\text{norm}}, \quad (S56)$$
$$-\frac{2j(j+2)}{(j+1)} F_{\text{norm}} \leq F_{je(j+1)e} \leq \frac{j(j+2)}{4(j+1)} F_{\text{norm}}, \quad (S57)$$
$$-\frac{2j(j+2)}{(j+1)} F_{\text{norm}} \leq F_{jm(j+1)m} \leq \frac{j(j+2)}{4(j+1)} F_{\text{norm}}. \quad (S58)$$

S14
Fundamental bounds for the total optical force

Now that for all the contributions to the total optical force, we have found the fundamental bounds, it is time to calculate the bounds for the total optical force itself. For that goal, we will write all Mie coefficients as the form introduced in Eq. S51. For the Mie coefficients $a_j$ and $b_j$, we write:

$$a_j = \cos \alpha_j e^{i \alpha_j}, \quad (S59)$$

$$b_j = \cos \beta_j e^{i \beta_j}. \quad (S60)$$

Therefore the total optical force in Eq. S33 can be written as:

$$F = F_{\text{norm}} \sum_{j=1}^{\infty} \left[ (2j+1) \Re(a_j + b_j) - \frac{2(2j+1)}{j(j+1)} \Re(a_j b_j^*) - \frac{2j(j+2)}{j+1} \Re(a_j a_{j+1}^* + b_j b_{j+1}^*) \right]$$

$$= F_{\text{norm}} \left\{ \sum_{j=1}^{\infty} (2j+1)(\cos^2 \alpha_j + \cos^2 \beta_j) - \frac{2(2j+1)}{j(j+1)} \cos \alpha_j \cos \beta_j \cos(\alpha_j - \beta_j) \right.$$ 

$$- \frac{2j(j+2)}{j+1} \left[ \cos \alpha_j \cos \alpha_{j+1} \cos(\alpha_j - \alpha_{j+1}) + \cos \beta_j \cos \beta_{j+1} \cos(\beta_j - \beta_{j+1}) \right]\} \quad (S61)$$

At a specific frequency, the angles $\alpha_j$ and $\beta_j$ can be assumed to be independent of each other. This independence is assumed based on the degree of freedom that the design of the particle gives (e.g. the permittivity and radii of a core-multishell particle). Here, assuming that the particle is excitable up to the multiple response $j_{\text{max}}$, the total optical force is rewritten as the following form:

$$F = F_{\text{norm}} \sum_{j=1}^{j_{\text{max}}} (2j+1)(\cos^2 \alpha_j + \cos^2 \beta_j) - \frac{2(2j+1)}{j(j+1)} \cos \alpha_j \cos \beta_j \cos(\alpha_j - \beta_j)$$

$$- \frac{2j(j+2)}{j+1} \left[ \cos \alpha_j \cos \alpha_{j+1} \cos(\alpha_j - \alpha_{j+1}) + \cos \beta_j \cos \beta_{j+1} \cos(\beta_j - \beta_{j+1}) \right]. \quad (S62)$$

For the simplest case of a dipolar approximation (i.e. only the electric and magnetic dipole moments), the formula developed above for the total optical force is simplified into:

$$F_{\text{dipole}} = 3F_{\text{norm}}[\cos^2 \alpha_1 + \cos^2 \beta_1 - \cos \alpha_1 \cos \beta_1 \cos(\alpha_1 - \beta_1)]. \quad (S63)$$
Previously the maximum individual contribution of either of the electric or magnetic dipoles are calculated to be $3F_{\text{norm}}$. The total optical force for a dual particle at resonance is also $3F_{\text{norm}}$ (Fig. S3). Can the force on a dipolar particle, surpass those values? The function above can be optimized numerically and analytically. The maximum of the function is calculated to be:

$$(F_{\text{dipole}})_{\text{max}}|_{\alpha_1 = \pm \frac{\pi}{6}, \beta_1 = \mp \frac{\pi}{6}} = \frac{27}{8} F_{\text{norm}}. \quad (S64)$$

In Fig. 3(c), using particle swarm optimization (PSO), we have optimized a core-multishell particle to result in the maximum possible normalized total optical force in dipolar approximation. Details are given in a separate section below. The maximum of the function can be analytically derived up to $j_{\text{max}} = 3$. To solve for higher orders $j_{\text{max}} > 3$, a genetic algorithm is implemented to maximize the function $F/F_{\text{norm}}$ with $2j_{\text{max}}$ variables ($\alpha_j, \beta_j, j = 1, 2, \ldots, j_{\text{max}}$). The maximum normalized total optical force as a function of $j_{\text{max}}$ up to $j_{\text{max}} = 10$ is shown in Fig. 3(a). The maximum values from $j_{\text{max}} = 1$ to $j_{\text{max}} = 10$ are calculated to be:

$$(F_{\text{Total}})_{\text{max}} = \{3.37, 9.18, 17.11, 27.07, 39.05, 53.03, 69.03, 87.02, 105.41, 128.18, \ldots\} F_{\text{norm}}. \quad (S65)$$

The maximum of the total optical force as a function of $j_{\text{max}}$ is plotted up to $j_{\text{max}} = 50$. A basic fitting to a quadratic function results to the following equation:

$$(F_{\text{Total}})_{\text{max}} = (0.97631 j_{\text{max}}^2 + 3.1173 j_{\text{max}} - 0.888333) F_{\text{norm}}, \quad (S66)$$

where the norm of residuals is equal to 1.0699.
Cartesian dipole expansion

The time averaged optical force exerted on a dipolar particle by an arbitrary incident wave, expanded into Cartesian dipole moments, is \([11, 12]\) (SI units):

\[
F = F_p + F_m + F_{pm},
\]

\[
F_p = \frac{1}{2} \Re [(\nabla E^*_{\text{inc}}) \cdot p],
\]

\[
F_m = \frac{1}{2} \Re [(\nabla B^*_{\text{inc}}) \cdot m],
\]

\[
F_{pm} = -\frac{Z_0 k^4}{12\pi} \Re [(p \times m^*)],
\]

where \(F_p, F_m\) and \(F_{pm}\) are the forces due to the induced electric Cartesian dipole moment \(p\), magnetic Cartesian dipole moment \(m\), and the interference of the two, respectively. The gradient of a vector \(A\) is defined as:

\[
\nabla A = \begin{bmatrix}
\partial_x A_x & \partial_x A_y & \partial_x A_z \\
\partial_y A_x & \partial_y A_y & \partial_y A_z \\
\partial_z A_x & \partial_z A_y & \partial_z A_z
\end{bmatrix}.
\]

IV. OPTICAL TORQUE

Spherical multipole expansion

The optical torque exerted on an arbitrary scatterer in free space, using the VSWF amplitudes, is as follows (re-normalized) (notice that there is a missing \(j\) in the reference paper \([8]\), in the
original form) (SI units):

\[ \mathbf{N} = N_x \mathbf{e}_x + N_y \mathbf{e}_y + N_z \mathbf{e}_z, \quad (S72) \]

\[ N_z = -\frac{I_0 \lambda^3}{c 8\pi^3} \sum_{j=1}^{\infty} \sum_{m=-j}^{j} m \left[ |a_{jm}|^2 + |b_{jm}|^2 - R \left( a_{jm} p_{jm}^* + b_{jm} q_{jm}^* \right) \right], \quad (S73) \]

\[ N_x + iN_y = -\frac{I_0 \lambda^3}{c 8\pi^3} \sum_{j=1}^{\infty} \sum_{m=-j}^{j} \left\{ \sqrt{(j + m + 1)(j - m)} \left( a_{jm} a_{j,m+1}^* + b_{jm} b_{j,m+1}^* \right) + \frac{1}{2} \left( a_{jm} p_{jm}^* + a_{j,m+1}^* p_{jm} + b_{j,m} q_{j,m+1}^* + q_{jm} b_{j,m+1}^* \right) \right\}. \quad (S74) \]

For a z propagating plane wave, the only non-zero torque is \( N_z \) and it will be refereed to as the exerted optical torque \( N \).

The exerted optical torque on an isotropic particle by an arbitrary illumination can be rewritten as:

\[ N = \sum_{j=1}^{\infty} (N_{je} + N_{jm}) . \quad (S75) \]

As can be seen, for the optical torque along \( \mathbf{k} \) direction (i.e. the z-direction), no interference term contributes.

We define the normalizing torque factor as:

\[ N_{\text{norm}} = \frac{I_0 \lambda^2}{\omega 8\pi}. \quad (S76) \]

Below each of the two terms are analytically calculated and the upper bounds for their contribution are derived.
Fundamental bounds for the single constituents of the total optical torque

*Contribution of an individual electric (N_{je})/magnetic (N_{jm}) MC:* The individual contribution of an electric MC $a_j$ is:

$$N_{je} = -\frac{I_0}{c} \frac{\lambda^3}{8\pi^3} \sum_{m=-j}^{j} m \left[ |a_{jm}|^2 - \Re (a_{jm} p_{jm}^*) \right]$$

$$= -\frac{I_0}{c} \frac{\lambda^3}{8\pi^3} \sum_{m=-j}^{j} m \left[ |a_j|^2 |p_{jm}|^2 - \Re (a_j p_{jm}^*) \right]$$

$$= -\frac{I_0}{c} \frac{\lambda^3}{8\pi^3} \left[ |a_j|^2 - \Re (a_j) \right] \sum_{m=-j}^{j} m |p_{jm}|^2. \quad \text{(S77)}$$

Considering only an electric dipole MC, the relation simplifies to:

$$N_p = -\frac{I_0}{c} \frac{\lambda^3}{8\pi^3} \left[ |a_1|^2 - \Re (a_1) \right] \left( |p_{11}|^2 - |p_{1-1}|^2 \right), \quad \text{(S78)}$$

which suggests that a circularly polarized plane wave (Eq. S9), where all the power is put into one amplitude, is the optimum illumination to exert maximal torque on an absorbing isotropic particle.

Considering higher order MCs and assuming a circularly polarized plane wave illumination, using equation Eq. S15, Eq. S77 simplifies to:

$$N_{je} = \sigma 4 (2j + 1) \left[ \Re (a_j) - |a_j|^2 \right] N^{\text{norm}} = \frac{\sigma I_0 C_{\text{abs},j^e}}{\omega}, \quad \text{(S79)}$$

and the maximum torque, occurs for the maximum absorption cross section (Eq. S23) and reads as:

$$[N_{je}]_{\text{max}} = \frac{\sigma I_0 [C_{\text{abs},j^e}]_{\text{max}}}{\omega} = \sigma (2j + 1) N^{\text{norm}}. \quad \text{(S80)}$$
Similarly, for a magnetic MC $b_j$, the following relation derives:

$$[N_{jm}]_{\text{max}} = \sigma (2j + 1) N^{\text{norm}}. \quad (S81)$$

**Fundamental bounds for the total optical torque**

Calculating the maximum of the total optical torque is easier than the force, since no interference term contributes to the torque along the wave propagation. To maximize the torque, it is required to overlap all the MC resonances spectrally and tune the loss for critical coupling at that frequency. Therefore, the maximum total optical torque as a function of $j_{\text{max}}$ is only a summation of all individual contributions $\sum_{j=1}^{j_{\text{max}}} 2(2j + 1)N^{\text{norm}} = 2j_{\text{max}}(j_{\text{max}} + 2)$. This relation is plotted in Fig. 3(b).

**Cartesian dipole expansion**

The time averaged optical torque exerted on a dipolar particle by an arbitrary incident wave, expanded into Cartesian dipole moments, is (SI units) [13]:

$$N = \frac{1}{2} \left\{ \Re (p \times E_{inc}^* + m \times B_{inc}^*) \right. \\
- \frac{k^3}{6\pi} \left[ \frac{1}{\epsilon_0} \Im (p^* \times p) + \mu_0 \Im (m^* \times m) \right] \right\}. \quad (S82)$$

V. PARTICLE SWARM OPTIMIZATION (PSO)

Particle swarm optimization (PSO) is a search-based optimization method, based on motion and intelligence of swarms, in which individuals orient their motion towards the personal ($P_{\text{best}}$)
and overall \((G_{\text{best}})\) best locations imposed by an associated objective function of particles. The modification of particles is defined through the velocity concept as [14]:

\[
\nu_{it}^{i+1} = w_{it}^{it} \nu_{it}^{it} + c_1 \times \text{rand} \times (P_{\text{best}_i} - s_{it}^{it}) + c_2 \times \text{rand} \times (G_{\text{best}_i} - s_{it}^{it}),
\]

\(\text{(S83)}\)

\[
s_{it}^{i+1} = s_{it}^{it} + \nu_{it}^{i+1},
\]

\(\text{(S84)}\)

\[
w_{it}^{it} = w_{\text{max}} - \left( \frac{w_{\text{max}} - w_{\text{min}}}{\text{iter}_{\text{max}}} \right) \times it,
\]

\(\text{(S85)}\)

where \(\nu_{it}^{it}\) and \(s_{it}^{it}\) are the velocity and position of particle \(i\) at PSO iteration \(it\), \(\text{rand}\) is a random number between 0 and 1, and \(w\) is the weighting factor. The values \(c_1 = c_2 = 2\), \(w_{\text{max}} = 0.9\), and \(w_{\text{min}} = 0.4\) are suggested as proper input values, independent of the problem [14]. We have used PSO to maximize the optical force exerted on a core-multishell particle in dipolar approximation. The variables are the three radii of the core and shells and the objective function is \(F/F_{\text{norm}}\). The result is shown in Fig. 3(c).

VI. NUMERICAL RESULTS

Figure S1, explores the effect of the imaginary part of the particle’s material refractive index (related to non-radiative loss) on the maximal force and torque (i.e., at the MC resonance) exerted by an arbitrarily polarized plane wave on a single channel dipolar particle.
Figure S1. a) The normalized maximal force and torque exerted by an arbitrarily polarized plane wave (for the force) or a circularly polarized plane wave (only for the torque) at the resonance of a dipole MC on an isotropic particle, as a function of the imaginary part of the refractive index. Refractive index is $n = n' + in''$. Maximum torque occurs at the critical coupling regime (shown by a green strip), i.e $\gamma_{nr} \approx \gamma_r$. (A linearly polarized plane wave exerts no torque on the particle.)

Figure S2. Time averaged total optical force exerted by an arbitrarily polarized plane wave on a non-absorbing sphere ($\epsilon_r = (3.5)^2$, $\mu_r = 1$) (black) and an absorbing sphere ($\epsilon_r = (3.5 + 0.16i)^2$, $\mu_r = 1$) (red) as a function of the sphere’s size parameter (solid line). The contribution of the non-interference (= $F_{total} - F_{interf}$) terms (dashed line).
Figure S3. *Non-absorbing dual dielectric sphere*: a) The time averaged total optical force exerted on a non-absorbing dual sphere \((\varepsilon_r = \mu_r = 3.5)\), illuminated by an arbitrarily polarized plane wave, as a function of the sphere’s size parameter. The individual contribution of the dipole, quadrupole, and octopole electric and magnetic MCs. b) The partial contribution due to the interference of dipole-dipole, quadrupole-quadrupole, and octopole-octopole electric and magnetic MCs.

In Fig. 2a, we have plotted the total force and the non-interference contributions to the force for a non-absorbing dielectric sphere. To see the effect of absorption, in Fig. S2, we have also plotted the total force and the non-interference terms for an absorbing sphere. The figure helps to compare the effect of absorption on the interference terms and the total force.

In Fig. S4, the optical force exerted by an arbitrarily polarized plane wave on a silver-silicon-silver core-multishell particle is calculated. The metal is characterized by a lossless Drude model \(\varepsilon_{\text{metal}} = 1 - \omega_p^2/\omega^2\), where \(\omega_p\) is the plasma frequency of the silver. The electric dipole-quadrupole and quadrupole-octopole MC interference force contributions are engineered to be maximized by spectrally overlapping the electric dipole \((a_1)\), quadrupole \((a_2)\) and octopole \((a_3)\) MC resonances. The particle is primarily designed for the purpose of super-scattering [15]. Unlike optical cross sections, the total optical force is not derived by adding the individual MC contributions. However,
Figure S4. Non-absorbing core-multishell particle: a) The time averaged total optical force exerted on an isotropic non-absorbing silver-silicon-silver core-multishell particle, illuminated by an arbitrarily polarized plane wave, as a function of the normalized frequency $\omega/\omega_p$ and the non-negligible partial contributions of the individual dipolar, quadrupolar, and octopolar electric MCs; and b) the interference of dipole-quadrupole and quadrupole-octopole, electric MCs. The upper bounds for the contributions are shown by a dashed line of the same color. ($r_1 = 0.4749\lambda_p$, $r_2 = 0.6404\lambda_p$, $r_3 = 0.8249\lambda_p$, $\lambda_p = 2\pi c/\omega_p$, $\epsilon_{\text{silicon}} = 12.96$ [15])

the interference terms contribute to the total optical force significantly. Such that the total force is equal to the individual contribution of a single electric octopole.

In Fig. S5, the optical torque exerted by a circularly polarized plane wave on a dual dielectric sphere is calculated. Due to duality, the electric and magnetic MCs are identical. In the analytical relation for optical torque, no interference term appears. Therefore, the maximum total exerted torque on the dual sphere for one multipole is twice as big as $(N_{je})_{\text{max}}$. This makes dual particles as good candidates for super-rotatable objects.

In Fig. S6, the optical force and torque on a realistic silver nanoparticle is calculated to be able to compare the results with the previous results. The interesting observation is that the electric
Figure S5. *Absorbing dual dielectric sphere*: The time averaged total optical torque exerted on an absorbing dual sphere, illuminated by a circularly polarized plane wave as a function of the sphere’s size parameter. The individual contribution of the dipole, quadrupole, and octopole electric and magnetic MCs. The maximum contribution for each term is shown by a dashed line of the same color.

Figure S6. *Silver sphere*: a) The time averaged total optical force and b) torque exerted on a silver sphere ($a = 45$ nm), illuminated by an arbitrary polarized plane wave (for the force) and a circularly polarized plane wave (only for the torque). The individual contribution of the dipole and quadrupole electric MCs and their interference. The maximum contribution for each term is shown by a line of the same color. (Experimental data from Ref. 16).

The quadrupolar contribution is much more pronounced in the exerted optical torque than in the force. This large torque is due to the fact that the particle absorbs considerably at the electric quadrupole.
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