

## Fundamental limits of optical force and torque

A. Rahimzadegan,<sup>1,\*</sup> R. Alaei,<sup>1,2,†</sup> I. Fernandez-Corbaton,<sup>1,3</sup> and C. Rockstuhl<sup>1,3</sup>

<sup>1</sup>*Institute of Theoretical Solid State Physics, Karlsruhe Institute of Technology, Karlsruhe 76131, Germany*

<sup>2</sup>*Max Planck Institute for the Science of Light, Erlangen 91058, Germany*

<sup>3</sup>*Institute of Nanotechnology, Karlsruhe Institute of Technology, Karlsruhe 76021, Germany*

(Received 24 May 2016; revised manuscript received 12 December 2016; published 3 January 2017)

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, and nanorobotics up to ambitious projects such as laser-propelled spaceships.

DOI: [10.1103/PhysRevB.95.035106](https://doi.org/10.1103/PhysRevB.95.035106)

**Introduction.** Optical scattering, extinction, and absorption cross sections characterize the strength of light-matter interaction. They quantify the fraction of power a particle scatters, extincts, or absorbs [1,2]. 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 [1,3]. 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  $(2j+1)\lambda^2/2\pi$  [4,5]. The maximum  $C_{\text{sca}}$  is attained when the particle operates in the *overcoupling* ( $\gamma_r \gg \gamma_{\text{nr}}$ ) regime, i.e., the radiative (scattering) loss ( $\gamma_r$ ) is much larger than the nonradiative (Ohmic) loss ( $\gamma_{\text{nr}}$ ). 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  $(2j+1)\lambda^2/8\pi$  [6]. It occurs if the particle operates in the *critical coupling* ( $\gamma_{\text{nr}} = \gamma_r$ ) regime, i.e., the nonradiative ( $\gamma_{\text{nr}}$ ) 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  $\gamma_{\text{nr}} = \gamma_r$ ].

Although optical cross sections are important in studying light-matter interaction at the nanoscale [7–9], the optical force and torque are further key quantities [10–13], 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 optomechanical manipulation [14–17], molecular or particle optical sorting [18], or nanorobotics [19,20]. Optical force and torque are also important in studying the angular momentum of light [21–24]. Moreover, developing ambitious projects like laser-propelled spaceships would benefit from an understanding of these limits [25].

In this paper, based on the multipole expansion in scattering theory [2], 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 the Supplemental Material Ref. [26]. 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 [10,27]

$$\mathbf{F}_p = \frac{1}{2} \text{Re}(\nabla \mathbf{E}^* \cdot \mathbf{p}) = \frac{k I_0}{c} \text{Im}[\alpha(\omega)] \mathbf{e}_z = F_p \mathbf{e}_z, \quad (1)$$

where  $\mathbf{p} = \epsilon_0 \alpha \mathbf{E}$  is the induced Cartesian electric dipole moment,  $\alpha$  is the electric polarizability of the particle,  $I_0 =$

\*Corresponding author: aso.rahimzadegan@kit.edu

†Corresponding author: rasoul.alaei@mpl.mpg.de

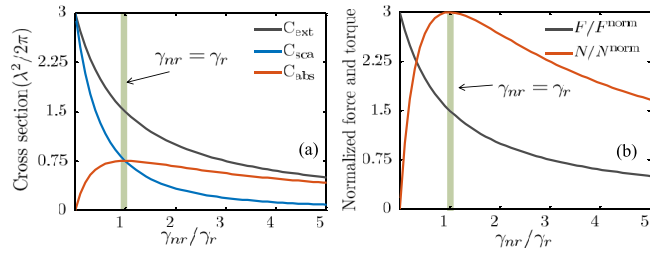


FIG. 1. (a) Maximal scattering, extinction, and absorption cross sections of; and (b) optical force and torque exerted on an isotropic dipolar particle as a function of the loss factor, i.e.,  $\gamma_{nr}/\gamma_r$ . The particle is illuminated by an arbitrarily polarized plane wave (for the cross sections and the force) or a circularly polarized plane wave (only for the torque) at the resonance of a spectrally isolated dipole MC.

$\epsilon_0 c |E_0|^2 / 2$  is intensity of the illumination,  $k$  is the wave number,  $\epsilon_0$  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,  $C_{\text{ext,p}} = k \text{Im}(\alpha)$  [28],  $F_p$  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 [29]

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

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

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

$(F_p)_{\text{max}}$  is the fundamental limit for the force an arbitrarily polarized plane wave can exert on an isotropic electric dipolar 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  $\mathbf{E} = E_0 e^{ikz}(\mathbf{e}_x + \sigma \mathbf{i} \mathbf{e}_y) / \sqrt{2}$ , with handedness  $\sigma = \pm 1$ , impinges on the particle, the optical torque reads as [11]

$$\begin{aligned} \mathbf{N}_p &= \frac{1}{2} \left[ \text{Re}(\mathbf{p} \times \mathbf{E}^*) - \frac{k^3}{6\pi\epsilon_0} \text{Im}(\mathbf{p}^* \times \mathbf{p}) \right] \\ &= \frac{\sigma I_0}{\omega} \left\{ k \text{Im}[\alpha(\omega)] - \frac{k^4}{6\pi} |\alpha(\omega)|^2 \right\} \mathbf{e}_z = N_p \mathbf{e}_z. \end{aligned} \quad (4)$$

Based on the definition of the absorption cross section of an electric dipolar particle,  $C_{\text{abs,p}} = k \text{Im}(\alpha) - k^4 |\alpha|^2 / 6\pi$  [28],

$N_p$  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.,  $\gamma_{nr}/\gamma_r$ . In the overcoupling 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 multipolar expansion, the incident and scattered fields are expanded as [1,3,30]

$$\begin{aligned} \mathbf{E}_{\text{inc}} &= - \sum_{j=1}^{\infty} \sum_{m=-j}^j E_{jm} \left( p_{jm} \mathbf{N}_{jm}^{(1)} + i q_{jm} \mathbf{M}_{jm}^{(1)} \right), \\ \mathbf{E}_{\text{sca}} &= \sum_{j=1}^{\infty} \sum_{m=-j}^j E_{jm} \left( a_{jm} \mathbf{N}_{jm}^{(3)} + i b_{jm} \mathbf{M}_{jm}^{(3)} \right), \end{aligned} \quad (6)$$

where  $[\mathbf{M}_{jm}^{(1)}(\mathbf{r}; \omega), \mathbf{N}_{jm}^{(1)}(\mathbf{r}; \omega)]$  are the regular and  $[\mathbf{M}_{jm}^{(3)}(\mathbf{r}; \omega), \mathbf{N}_{jm}^{(3)}(\mathbf{r}; \omega)]$  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  $\mathbf{J}^2$ , and  $m$  is the eigenvalue of the  $z$  component of the AM  $\mathbf{J}_z$ .  $E_{jm}$  is a normalizing factor (Supplemental Material Ref. [26]).

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 Supplemental Material Ref. [26]. Finding the scattered field amplitudes is not a trivial task. However, Mie theory provides analytical solutions for an isotropic particle [3,31]. 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 [3]. Each Mie coefficient has a spectral profile with certain resonance peaks. 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 nonabsorbing particle they are equal to unity [32].

*Fundamental limits on optical force.* The multipolar description of the force on a particle by an arbitrary illumination has been presented in [33,34] (Supplemental Material Ref. [26]). Considering the contributions of different MCs, the force

TABLE I. Fundamental limits on optical force constituents.

Term	$F/F^{\text{norm}}$	$(F)_{\text{max}}/F^{\text{norm}}$	Maximum contribution at	Example (overcoupling regime)
$F_{j^e}$	$\frac{2\pi}{\lambda^2} C_{\text{ext},j^e} = (2j+1)\text{Re}(a_j)$	$(2j+1)$	$ a_j $ resonance	Dielectric sphere (Fig. 2)
$F_{j^m}$	$\frac{2\pi}{\lambda^2} C_{\text{ext},j^m} = (2j+1)\text{Re}(b_j)$	$(2j+1)$	$ b_j $ resonance	Dielectric sphere (Fig. 2)
$F_{j^e j^m}$	$-\frac{2(2j+1)}{j(j+1)} \text{Re}(a_j b_j^*)$	$-\frac{2(2j+1)}{j(j+1)}$	Simultaneous $ a_j $ and $ b_j $ resonance	Dual dielectric sphere (Fig. S3)
$F_{j^e(j+1)^e}$	$-\frac{2j(j+2)}{(j+1)} \text{Re}(a_{j+1} a_j^*)$	$-\frac{2j(j+2)}{(j+1)}$	Simultaneous $ a_j $ and $ a_{j+1} $ resonance	Dielectric core multishell (Fig. S4)
$F_{j^m(j+1)^m}$	$-\frac{2j(j+2)}{(j+1)} \text{Re}(b_{j+1} b_j^*)$	$-\frac{2j(j+2)}{(j+1)}$	Simultaneous $ b_j $ and $ b_{j+1} $ resonance	Dielectric core multishell

on an isotropic particle in along  $+z$  is derived as

$$\begin{aligned}
 F &= \sum_{j=1}^{\infty} \{F_{j^e} + F_{j^m} + F_{j^e j^m} + F_{j^e(j+1)^e} + F_{j^m(j+1)^m}\} \\
 &= [F_p + F_{Q^e} + F_{O^e} + \dots] + [F_m + F_{Q^m} + F_{O^m} + \dots] \\
 &\quad + [F_{p^m} + F_{Q^e Q^m} + F_{O^e O^m} + \dots] \\
 &\quad + [F_{p^e Q^e} + F_{Q^e O^e} + \dots] + [F_{m^e Q^m} + F_{Q^m O^m} + \dots]. \quad (7)
 \end{aligned}$$

$F_{j^e}$  ( $F_{j^m}$ ) 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 (Supplemental Material Ref. [26]). The results are shown in Table I. Specific examples are mentioned that maximize each term. Figure 2 shows different nonzero contributions to the optical force exerted on a nonabsorbing 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,  $(2j+1)F^{\text{norm}}$ . The individual contribution of a MC is directly related to the extinction cross section (Supplemental Material Ref. [26]). 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 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 [35] 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 ( $F_{p^e Q^e} = -3F^{\text{norm}}$ ,  $F_{Q^e O^e} \approx -5.3F^{\text{norm}}$ ). Note that the maximum contribution of the interference terms is always negative. However, as proven in Supplemental Material Ref. [26], the maximum positive contribution of an interference term (at off-resonance) is eight times smaller than the maximum negative contribution (at resonance).

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 where classical electrodynamics is not applicable, we assume

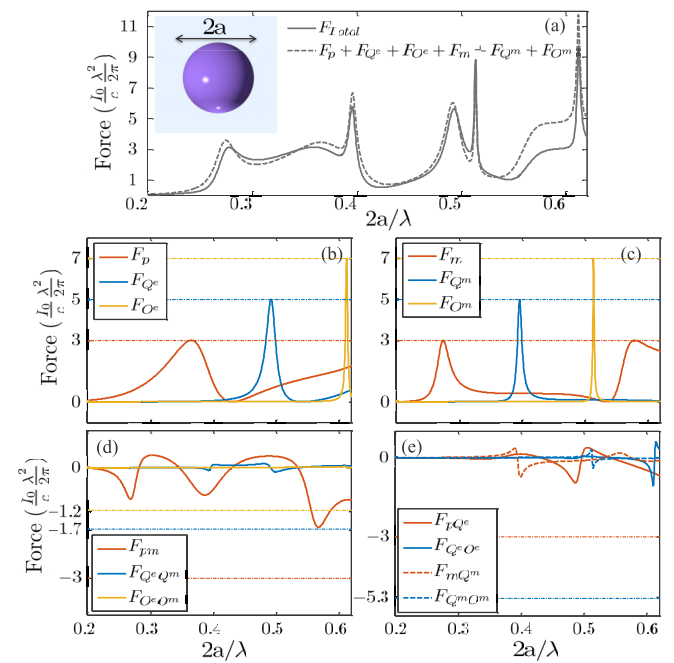


FIG. 2. *Nonabsorbing dielectric sphere.* (a) Optical force exerted by an arbitrarily polarized plane wave on a nonabsorbing sphere [ $\epsilon_r = (3.5)^2$ ,  $\mu_r = 1$ ] depending on the sphere's size parameter (solid line). Contributions of the noninterference terms (dashed line). (b) The individual 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.

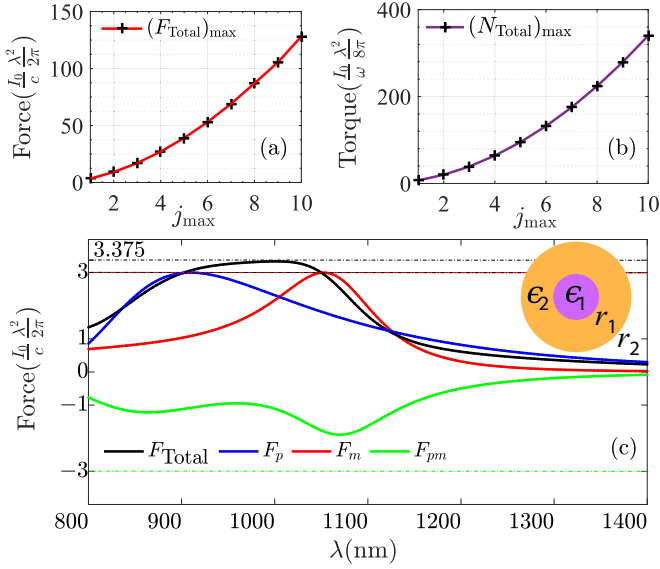


FIG. 3. (a) Maximum total optical force and (b) torque as a function of the maximum non-negligible multipole moment order  $j_{\max}$ . (c) Optical force exerted on an optimized isotropic nonabsorbing core-shell particle, illuminated by an arbitrarily polarized plane wave. The figure also illustrates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

that the optical response is describable with a finite number of multipole moments and hence the total optical force is always finite. For a nonabsorbing particle, each Mie coefficient is modeled by a simple formula ( $a_j = \cos \alpha_j \exp i \alpha_j$ ,  $b_j = \cos \beta_j \exp i \beta_j$ ) with a single real-valued variable  $\alpha_j$  or  $\beta_j$  [36]. Based on this model and Eq. (7), assuming that the resonance of the MCs can be optimally engineered (i.e., assuming all  $\alpha_j$ 's and  $\beta_j$ 's to be independent), the maximum force can be calculated as a function of the maximum non-negligible multipole order  $j_{\max}$ . For a smaller number of Mie channels the maximal force can be derived analytically, i.e., up to  $j_{\max} = 3$ . For a larger number of Mie channels a genetic algorithm has been used (Supplemental Material Ref. [26]). 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) [37], we have optimized a dielectric core-multishell particle that receives the maximum optical force in dipolar approximation at  $\lambda = 1$   $\mu\text{m}$  [Fig. 3(c)] (Supplemental Material Ref. [26]). 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. [33],

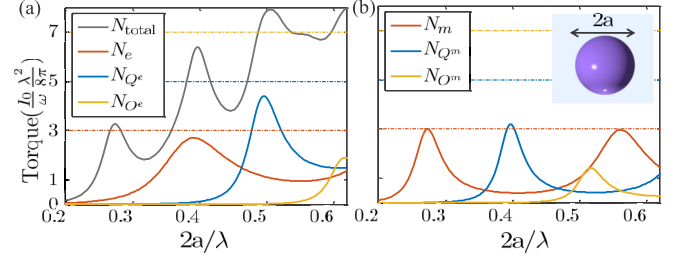


FIG. 4. *Absorbing dielectric sphere.* (a) Optical torque on an absorbing sphere [ $\epsilon_r = (3.5 + 0.16i)^2$ ,  $\mu_r = 1$ ], by a circularly polarized plane wave as a function of the sphere's size parameter. The individual contribution of the dipole, quadrupole, and octopole electric and (b) magnetic MCs.

can be rewritten as

$$N = \sum_{j=1}^{\infty} \{N_{j^e} + N_{j^m}\} = [N_p + N_{Q^e} + N_{O^e} + \dots] + [N_m + N_{Q^m} + N_{O^m} + \dots], \quad (8)$$

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

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

The exerted torque is directly related to the averaged total AM [1] 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

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

For an absorbing isotropic particle at the critical coupling regime,  $\text{Re}(a_j - |a_j|^2)$  is maximum and equal to 0.25 [6]. Therefore, the maximum torque is derived as

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

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,

TABLE II. Fundamental limits on optical torque constituents.

Term	$N/N^{\text{norm}}$	$(N)_{\max}/N^{\text{norm}}$	Maximum contribution at	Example (critical-coupling regime)
$N_{j^e}$	$\frac{\sigma I_0}{\omega} C_{\text{abs},j^e} = 4\sigma(2j+1)\text{Re}(a_j -  a_j ^2)$	$\sigma(2j+1)$	$ a_j $ resonance	Dielectric sphere (Fig. S5)
$N_{j^m}$	$\frac{\sigma I_0}{\omega} C_{\text{abs},j^m} = 4\sigma(2j+1)\text{Re}(b_j -  b_j ^2)$	$\sigma(2j+1)$	$ b_j $ resonance	Dielectric sphere (Figs. 4 and S5)

from Table II, the torque is  $(N_{j^e})_{\max} + (N_{j^m})_{\max} = 2(2j + 1)N^{\text{norm}}$ . 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) and a gold nanopatch (Fig. S7) to compare our results with a realistic absorbing

particle. Our results can be used to design superacceleratable and -rotatable particles by engineering the spectral resonance of the MCs. The designed particles can in turn be used in optonanorobots.

*Acknowledgments.* We thank H. Rahmanpanah and A. Jalali for their help in the optimizations. We acknowledge partial financial support by the Deutsche Forschungsgemeinschaft through Grant No. CRC 1173. A.R. also acknowledges support from the Karlsruhe School of Optics and Photonics (KSOP).

A.R. and R.A. equally contributed to the work.

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