Enantioselective manipulation of single chiral nanoparticles using optical tweezers: supplementary material

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This supplement contains an expanded theoretical description of optical tweezers of particles with a chiral shell.

We consider a circularly-polarized Gaussian beam at the objective entrance port, with $\sigma = \pm 1$ denoting left-handed/righthanded polarization. The dimensionless optical force efficiency (see main text for definition in terms of the optical force) is written as

$$\mathbf{Q}(\rho,\phi,z) = \mathbf{Q}_s(\rho,\phi,z) + \mathbf{Q}_e(\rho,\phi,z). \tag{1}$$

The extinction term $\mathbf{Q}_e$ accounts for the rate of momentum removal from the incident beam, while the scattering contribution $\mathbf{Q}_s$ corresponds to the negative of the rate of momentum carried away by the scattered field. The explicit expressions for their cylindrical components are given below as sums over multipoles of the form

$$\sum_{\ell m}(...) \equiv \sum_{\ell=1}^{\infty}\sum_{m=-\ell}^{\ell}(...)$$

- Scattering axial component

$$Q_{sz}(\rho,\phi,z) = -\frac{8\gamma^2}{AN}\mathrm{Re}\sum_{\ell m}\frac{\sqrt{\ell(\ell+2)(\ell+m+1)(\ell-m+1)}}{\ell+1}\left[(A_\ell A_{\ell+1}^* + B_\ell B_{\ell+1}^*)G_{\ell,m}^{(\sigma)}G_{\ell+1,m}^{(\sigma)*}\right]$$

$$-\frac{8\gamma^2}{AN}\sigma\mathrm{Re}\sum_{\ell m}\frac{(2\ell+1)}{\ell(\ell+1)}mA_\ell B_\ell^*|G_{\ell,m}^{(\sigma)}|^2$$

- Scattering radial component

$$Q_{s\rho}(\rho,\phi,z) = \frac{4\gamma^2}{AN}\sum_{\ell m}\frac{\sqrt{\ell(\ell+2)(\ell+m+1)(\ell+m+2)}}{\ell+1}\mathrm{Im}\Big\{(A_\ell A_{\ell+1}^* + B_\ell B_{\ell+1}^*)$$

$$\left[G_{\ell,m}^{(\sigma)}G_{\ell+1,m+1}^{(\sigma)*} + G_{\ell,-m}^{(\sigma)}G_{\ell+1,-(m+1)}^{(\sigma)*}\right]\Big\} - \frac{8\gamma^2}{AN}\sigma\sum_{\ell m}\frac{(2\ell+1)}{\ell(\ell+1)}\sqrt{(\ell-m)(\ell+m+1)}\left[\mathrm{Re}(A_\ell B_\ell^*)\mathrm{Im}(G_{\ell,m}^{(\sigma)}G_{\ell,m+1}^{(\sigma)*})\right]$$

- Scattering azimuthal component

$$Q_{s\phi}(\rho,\phi,z) = -\frac{4\gamma^2}{AN}\sum_{\ell m}\frac{\sqrt{\ell(\ell+2)(\ell+m+1)(\ell+m+2)}}{\ell+1}\mathrm{Re}\Big\{(A_\ell A_{\ell+1}^* + B_\ell B_{\ell+1}^*)\times$$

$$\left[G_{\ell,m}^{(\sigma)}G_{\ell+1,m+1}^{(\sigma)*} - G_{\ell,-m}^{(\sigma)}G_{\ell+1,-(m+1)}^{(\sigma)*}\right]\Big\} + \frac{8\gamma^2}{AN}\sigma\sum_{\ell m}\frac{(2\ell+1)}{\ell(\ell+1)}\sqrt{(\ell-m)(\ell+m+1)}\times\mathrm{Re}(A_\ell B_\ell^*)\mathrm{Re}(G_{\ell,m}^{(\sigma)}G_{\ell,m+1}^{(\sigma)*})$$

- Extinction axial component

$$Q_{ez}(\rho,\phi,z) = \frac{4\gamma^2}{AN}\mathrm{Re}\sum_{\ell m}(2\ell+1)G_{\ell,m}^{(\sigma)}\left[(A_\ell + B_\ell)G_{\ell,m}^{\prime(\sigma)*}\right], \tag{2}$$

- Extinction radial component

$$Q_{e\rho}(\rho,\phi,z)=\frac{2\gamma^2}{AN}\mathrm{Im}\sum_{\ell m}(2\ell+1)G^{(\sigma)}_{\ell,m}\left[(A_\ell+B_\ell)\left(G^{(\sigma)-}_{\ell,m+1}-G^{(\sigma)+}_{\ell,m-1}\right)^*\right]$$

- Extinction azimuthal component

$$Q_{e\phi}(\rho,\phi,z)=-\frac{2\gamma^2}{AN}\mathrm{Re}\sum_{\ell m}(2\ell+1)G^{(\sigma)}_{\ell,m}\left[(A_\ell+B_\ell)\left(G^{(\sigma)+}_{\ell,m-1}+G^{(\sigma)-}_{\ell,m+1}\right)^*\right]$$

In addition to the multipole coefficients $G^{(\sigma)}_{\ell m}(\rho,\phi,z)$ defined in Eq. (3) of the main letter, we also define

$$G'^{(\sigma)}_{\ell,m}(\rho,\phi,z)=\int_0^{\theta_0}d\theta\sin\theta\cos\theta_w\sqrt{\cos\theta}\,T(\theta)\,e^{-\gamma^2\sin^2\theta}d^{\ell}_{m,\sigma}(\theta_w)\,J_{m-\sigma}\left(k\rho\sin\theta\right)e^{i[\Phi_w(\theta)+k_w\cos\theta_w z]},$$

$$G^{(\sigma)\pm}_{\ell,m}(\rho,\phi,z)=\int_0^{\theta_0}d\theta\sin\theta\sin\theta_w\sqrt{\cos\theta}\,T(\theta)\,e^{-\gamma^2\sin^2\theta}d^{\ell}_{m\pm1,\sigma}(\theta_w)\,J_{m-\sigma}\left(k\rho\sin\theta\right)e^{i[\Phi_w(\theta)+k_w\cos\theta_w z]},$$

with $\theta_w=\sin^{-1}(\sin\theta/N)$ and $N=n_w/n_g$. The phase Φ_w accounts for the spherical aberration arising from the refractive index mismatch at the glass-water interface:

$$\Phi_w(\theta)=k\left(-L/N\cos\theta+NL\cos\theta_w\right),$$

where L represents the distance between the paraxial focal plane and the glass slide. We also take $T(\theta)=\frac{2\cos\theta}{\cos\theta+N\cos\theta_w}$ for the Fresnel transmission amplitude (neglecting polarization dependence since $N\approx1$).

The factor

$$A=16\gamma^2\int_0^{s_0}ds\,s\exp(-2\gamma^2s^2)\frac{\sqrt{(1-s^2)(N^2-s^2)}}{(\sqrt{1-s^2}+\sqrt{N^2-s^2})^2}$$

is the fraction of beam power transmitted into the sample chamber, with $s_0=\mathrm{NA}/n_g$.

The optical force components also depend on the effective external Mie coefficients A_ℓ and B_ℓ which we derive as follows:

$$\begin{aligned}A_\ell&=a_\ell+i\sigma d_\ell\\B_\ell&=b_\ell-i\sigma c_\ell.\end{aligned}\qquad(3)$$

A_ℓ and B_ℓ are convenient for a number of applications involving circularly-polarized fields scattered by chiral media. The coefficients a_ℓ, b_ℓ, c_ℓ and d_ℓ are the external Mie coefficients for a sphere made of chiral material [1]. For a core-shell nanosphere, their explicit expressions [2] are given in terms of the size parameters $\alpha=k_wa$ and $v=k_wr$ corresponding to the core and outer radii a and r. The refractive indexes of the chiral shell (with respect to the host) are $N_{L/R}=(\sqrt{\epsilon\mu}\pm\kappa)/n_w$, where κ is the chirality parameter (see main text). We also need the relative refractive index of the core N_{I} with respect to the host medium of index n_w. Finally, $N_{\mathrm{II}}=(N_L+N_R)/2$ is the average relative index of the chiral shell.

$$\begin{aligned}a_\ell&=-\Delta_\ell^{-1}(A_{R\ell}W_{L\ell}+A_{L\ell}W_{R\ell})\\b_\ell&=-\Delta_\ell^{-1}(B_{L\ell}V_{R\ell}+B_{R\ell}V_{L\ell})\\c_\ell&=i\Delta_\ell^{-1}(A_{L\ell}V_{R\ell}-A_{R\ell}V_{L\ell})\\d_\ell&=i\Delta_\ell^{-1}(B_{R\ell}W_{L\ell}-B_{L\ell}W_{R\ell})\end{aligned}$$

with

$$\begin{aligned}\Delta_\ell&=W_{L\ell}V_{R\ell}+B_{L\ell}W_{R\ell}\\A_{R\ell}&=X_{R\ell}(-)\eta^{(1)}_\ell(v)-N_{\mathrm{II}}U_{R\ell}(-)j_\ell(v)\\A_{L\ell}&=X_{L\ell}(+)\eta^{(1)}_\ell(v)-N_{\mathrm{II}}U_{L\ell}(+)j_\ell(v)\\B_{L\ell}&=X_{L\ell}(-)N_{\mathrm{II}}\eta^{(1)}_\ell(v)-U_{L\ell}(-)j_\ell(v)\\B_{R\ell}&=X_{R\ell}(+)N_{\mathrm{II}}\eta^{(1)}_\ell(v)-U_{R\ell}(+)j_\ell(v)\\V_{L\ell}&=X_{L\ell}(+)\eta^{(3)}_\ell(v)-N_{\mathrm{II}}U_{L\ell}(+)h^{(1)}_\ell(v)\\V_{R\ell}&=X_{R\ell}(-)\eta^{(3)}_\ell(v)-N_{\mathrm{II}}U_{R\ell}(-)h^{(1)}_\ell(v)\\W_{L\ell}&=X_{L\ell}(-)N_{\mathrm{II}}\eta^{(3)}_\ell(v)-U_{L\ell}(-)h^{(1)}_\ell(v)\\W_{R\ell}&=X_{R\ell}(+)N_{\mathrm{II}}\eta^{(3)}_\ell(v)-U_{R\ell}(+)h^{(1)}_\ell(v)\end{aligned}$$

We have also introduced the functions

$$\begin{aligned}X_{R\ell}(\pm)&=j_\ell(N_Rv)+D_{4\ell}y_\ell(N_Rv)\pm D_{2\ell}y_\ell(N_Lv)\\X_{L\ell}(\pm)&=j_\ell(N_Lv)+D_{1\ell}y_\ell(N_Lv)\pm D_{3\ell}y_\ell(N_Rv)\\U_{R\ell}(\pm)&=\eta^{(1)}_\ell(N_Rv)+D_{4\ell}\eta^{(2)}_\ell(N_Rv)\pm D_{2\ell}\eta^{(2)}_\ell(N_Lv)\\U_{L\ell}(\pm)&=\eta^{(1)}_\ell(N_Lv)+D_{1\ell}\eta^{(2)}_\ell(N_Lv)\pm D_{3\ell}\eta^{(2)}_\ell(N_Rv)\end{aligned}$$

where

$$
\begin{aligned}
D_{1\ell} &= -\Delta_\ell^{-1}[G_\ell(N_R)H_\ell(N_L) + F_\ell(N_R)K_\ell(N_L)] \\
D_{2\ell} &= \Delta_\ell^{-1}[F_\ell(N_R)K_\ell(N_R) - G_\ell(N_R)H_\ell(N_R)] \\
D_{3\ell} &= \Delta_\ell^{-1}[G_\ell(N_L)H_\ell(N_L) - F_\ell(N_L)K_\ell(N_L)] \\
D_{4\ell} &= -\Delta_\ell^{-1}[G_\ell(N_L)H_\ell(N_R) + F_\ell(N_L)K_\ell(N_R)]
\end{aligned}
$$

F, G, H and K are functions of the refractive index variable $N = N_L, N_R$ defined as

$$
\begin{aligned}
F_\ell(N) &= N_{\mathrm{II}} y_\ell(N\alpha)\eta_\ell^{(1)}(N_{\mathrm{I}}\alpha) - N_{\mathrm{I}}\eta_\ell^{(2)}(N\alpha)j_\ell(N_{\mathrm{I}}\alpha) \\
G_\ell(N) &= N_{\mathrm{I}} y_\ell(N\alpha)\eta_\ell^{(1)}(N_{\mathrm{I}}\alpha) - N_{\mathrm{II}}\eta_\ell^{(2)}(N\alpha)j_\ell(N_{\mathrm{I}}\alpha) \\
H_\ell(N) &= N_{\mathrm{I}} j_\ell(N\alpha)\eta_\ell^{(1)}(N_{\mathrm{I}}\alpha) - N_{\mathrm{I}}\eta_\ell^{(1)}(N\alpha)j_\ell(N_{\mathrm{I}}\alpha) \\
K_\ell(N) &= N_{\mathrm{II}} j_\ell(N\alpha)\eta_\ell^{(1)}(N_{\mathrm{I}}\alpha) - N_{\mathrm{II}}\eta_\ell^{(1)}(N\alpha)j_\ell(N_{\mathrm{I}}\alpha)
\end{aligned}
$$

$j_\ell(\rho)$, $y_\ell(\rho)$ are the spherical Bessel functions of the first and second kind, respectively; whereas $h_\ell^{(1)}(\rho)$ is the spherical Hankel function of the first kind [3]. We also define

$$
\begin{aligned}
\eta_\ell^{(1)}(\rho) &= \frac{1}{\rho} d[\rho j_\ell(\rho)]/d\rho \\
\eta_\ell^{(2)}(\rho) &= \frac{1}{\rho} d[\rho y_\ell(\rho)]/d\rho \\
\eta_\ell^{(3)}(\rho) &= \frac{1}{\rho} d[\rho h_\ell^{(1)}(\rho)]/d\rho
\end{aligned}
$$

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[2] C. F Bohren, Scattering of electromagnetic waves by an optically active spherical shell, J. Chem. Phys. **62**, 1566 (1975).

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