Electronic Supplementary Information (ESI)

The contribution of the scatering force relative to the gradient force in the Rayleigh regime

The gradient force and the scattering force can expressed as,[16]

$$\vec{F}_{grad} = -\hat{x}\alpha E_{x0}^2\left[\frac{\xi}{\omega_0} - \frac{1}{2}\frac{K^2\xi}{\omega_0} + \frac{K^2\xi^3}{\omega_0}\right]$$

$$\vec{F}_{scat} = \hat{z}\left(\frac{n_m^2\sigma_p\varepsilon_0}{2}\right)E_{x0}^2 + \hat{y}\left(\frac{K^2\xi^2 n_m^2\sigma_p\varepsilon_0}{2}\right)E_{x0}^2.$$

We note that $\alpha = 4\pi n_m^2\varepsilon_0 a^3\left(\frac{m^2-1}{m^2+2}\right)$ is the polarizability of a spherical particle, in which $m = \frac{n_0 + \tilde{n}_2 I}{n_m}$, the effective refractive index of a weakly absorbing sphere in a medium with $n_m = 1.338$, $\varepsilon_0 = 8.85\times10^{-12}$ (in unit of F/m) is the permittivity of free space, and $a \approx 5.00\times10^{-8}$ (in unit of m) is the radius of particles. We further note that $\sigma_p = \frac{8\pi^3|\alpha|^2}{3\lambda^4 n_m^4\varepsilon_0^2}$ (in unit of m^2) is the scattering cross-section of particles, $\xi = 0.56$ is the normalized coordinate that results in a maximal gradient force, $K \approx \frac{1.6}{\pi}$ is a constant for the use of an objective lens with NA 0.9, and $E_{x0}^2 = \frac{4P_{avg}}{\pi\omega_0^2 n_m\varepsilon_0 c} \approx 4.57\times10^{14}$ (in unit of V^2/m^2) is the field strength, in which $P_{avg} = 0.4$ (in unit of W) is the average of laser power, $c = 3\times10^8$ (in unit of m/s) is the speed of light in vacuum, and $\omega_0 = 5.6\times10^{-7}$ (in unit of m) is the beam waist.

Accordingly, the ratio of the magnitude of the gradient force relative to that of the scattering force can be expressed as,

$$\frac{\left|\vec{F}_{grad}\right|}{\left|\vec{F}_{scat}\right|} = \frac{\alpha E_{x0}^2\left[\frac{\xi}{\omega_0} - \frac{1}{2}\frac{K^2\xi}{\omega_0} + \frac{K^2\xi^3}{\omega_0}\right]}{\sqrt{\left(\frac{n_m^2\sigma_p\varepsilon_0}{2}E_{x0}^2\right)^2 + \left(\frac{K^2\xi^2 n_m^2\sigma_p\varepsilon_0}{2}E_{x0}^2\right)^2}} = \frac{\frac{\alpha\xi E_{x0}^2}{\omega_0}\left[1 - 0.5K^2 + K^2\xi^2\right]}{\sqrt{\left(\frac{n_m^2\sigma_p\varepsilon_0 E_{x0}^2}{2}\right)^2\left[1 + \left(K^2\xi^2\right)^2\right]}}$$

$$=\frac{\dfrac{\alpha\xi E_{x0}^2}{\omega_0}\left[1-0.5K^2+K^2\xi^2\right]}{\dfrac{n_m^2\sigma_p\varepsilon_0 E_{x0}^2}{2}\sqrt{1+\left(K^2\xi^2\right)^2}}=\frac{\dfrac{\alpha\xi}{\omega_0}\left[1-0.5K^2+K^2\xi^2\right]}{\dfrac{n_m^2\sigma_p\varepsilon_0}{2}\sqrt{1+\left(K^2\xi^2\right)^2}}=\frac{2\alpha\xi}{\omega_0 n_m^2\sigma_p\varepsilon_0}\times\frac{\left[1-0.5K^2+K^2\xi^2\right]}{\sqrt{1+\left(K^2\xi^2\right)^2}}$$

$$=\frac{2\alpha\xi}{\omega_0 n_m^2\left(\dfrac{8\pi^3|\alpha|^2}{3\lambda^4 n_m^4\varepsilon_0^2}\right)\varepsilon_0}\times\frac{\left[1-0.5K^2+K^2\xi^2\right]}{\sqrt{1+\left(K^2\xi^2\right)^2}}=\frac{2\alpha\xi\cdot 3\lambda^4 n_m^4\varepsilon_0^2}{\omega_0 n_m^2\cdot 8\pi^3|\alpha|^2\varepsilon_0}\times\frac{\left[1-0.5K^2+K^2\xi^2\right]}{\sqrt{1+\left(K^2\xi^2\right)^2}}$$

$$=\frac{3\xi\lambda^4 n_m^2\varepsilon_0\alpha}{4\omega_0\pi^3|\alpha|^2}\times\frac{\left[1-0.5K^2+K^2\xi^2\right]}{\sqrt{1+K^4\xi^4}}$$

$$=\frac{3\times 0.56\times\left(1064\times10^{-9}\right)^4\times(1.338)^2\times\left(8.85\times10^{-12}\right)}{4\times5.6\times10^{-7}\times\pi^3}\times\frac{\alpha}{|\alpha|^2}\times\frac{\left[1-0.5(1.6/\pi)^2+(1.6/\pi)^2(0.56)^2\right]}{\sqrt{1+(1.6/\pi)^4(0.56)^4}}$$

$$=4.91\times10^{-31}\times\frac{\alpha}{|\alpha|^2}\times0.9485\quad=4.66\times10^{-31}\times\frac{\alpha}{|\alpha|^2}\quad=4.66\times10^{-31}\times\frac{4\pi n_m^2\varepsilon_0 a^3\left(\dfrac{m^2-1}{m^2+2}\right)}{\left|4\pi n_m^2\varepsilon_0 a^3\left(\dfrac{m^2-1}{m^2+2}\right)\right|^2}$$

$$=\frac{4.66\times10^{-31}}{4\pi n_m^2\varepsilon_0 a^3}\times\frac{\left(\dfrac{m^2-1}{m^2+2}\right)}{\left|\dfrac{m^2-1}{m^2+2}\right|^2}\quad=\frac{4.66\times10^{-31}}{4\times\pi\times(1.338)^2\times\left(8.85\times10^{-12}\right)\times\left(5\times10^{-8}\right)^3}\times\frac{\left(\dfrac{m^2-1}{m^2+2}\right)}{\left|\dfrac{m^2-1}{m^2+2}\right|^2}$$

$$=\frac{4.66\times10^{-31}}{2.49\times10^{-32}}\times\frac{\left(\dfrac{m^2-1}{m^2+2}\right)}{\left|\dfrac{m^2-1}{m^2+2}\right|^2}\quad=18.7\times\frac{\left(\dfrac{m^2-1}{m^2+2}\right)}{\left|\dfrac{m^2-1}{m^2+2}\right|^2}.$$

We then simplify the term, $\dfrac{m^2-1}{m^2+2}$, as following,

$$\frac{m^2-1}{m^2+2}=\frac{\left(\dfrac{n_0+\tilde{n}_2 I}{n_m}\right)^2-1}{\left(\dfrac{n_0+\tilde{n}_2 I}{n_m}\right)^2+2}=\frac{\left(\dfrac{n_0+\tilde{n}_2 I}{n_m}\right)^2-\dfrac{n_m^2}{n_m^2}}{\left(\dfrac{n_0+\tilde{n}_2 I}{n_m}\right)^2+\dfrac{2n_m^2}{n_m^2}}=\frac{\dfrac{(n_0+\tilde{n}_2 I)^2}{n_m^2}-\dfrac{n_m^2}{n_m^2}}{\dfrac{(n_0+\tilde{n}_2 I)^2}{n_m^2}+\dfrac{2n_m^2}{n_m^2}}=\frac{\dfrac{1}{n_m^2}\left[(n_0+\tilde{n}_2 I)^2-n_m^2\right]}{\dfrac{1}{n_m^2}\left[(n_0+\tilde{n}_2 I)^2+2n_m^2\right]}$$

$$=\frac{(n_0+\tilde{n}_2 I)^2-n_m^2}{(n_0+\tilde{n}_2 I)^2+2n_m^2}.$$

Note that the doped nanobeads employed in this work comprise dielectric materials (polystyrene) and dopants (dye molecules), with the dielectric materials presumably dominant relative to the dopant. Because the abundant dielectric material is resonant with neither the fundamental nor the 2nd harmonic of the optical frequency of the laser whereas the less abundant dopant is resonant with the 2nd harmonic of the laser, the effective refractive index can then be expressed as $n_0 + \tilde{n}_2 I$, in which n_0 and $\tilde{n}_2$ is the linear and nonlinear refractive index of the spherical nanobeads, respectively. In general, n_0 is a real quantity while $\tilde{n}_2$ is a complex quantity and is appreciable at the fundamental wavelength of the laser provided that the two-photon resonant condition is energetically fulfilled.[21] We next assume that the nonlinear term, which includes the laser intensity, can be simply defined as $\tilde{n}_2 I = A + iB$, where A and B are the magnitude of the real and imaginary components of the complex nonlinear refractive index, respectively.

Accordingly, $\frac{m^2-1}{m^2+2}$ can be expressed as,

$$\frac{m^2-1}{m^2+2} = \frac{(n_0 + A + iB)^2 - n_m^2}{(n_0 + A + iB)^2 + 2n_m^2} = \frac{[(n_0 + A) + iB]^2 - n_m^2}{[(n_0 + A) + iB]^2 + 2n_m^2} = \frac{(n_0 + A)^2 + 2(n_0 + A)(iB) + (iB)^2 - n_m^2}{(n_0 + A)^2 + 2(n_0 + A)(iB) + (iB)^2 + 2n_m^2}$$

$$= \frac{(n_0 + A)^2 - B^2 - n_m^2 + i[2(n_0 + A)(B)]}{(n_0 + A)^2 - B^2 + 2n_m^2 + i[2(n_0 + A)(B)]}.$$

For convenience, we define $X = (n_0 + A)^2 - B^2 - n_m^2$, $Y = (n_0 + A)^2 - B^2 + 2n_m^2$ and $Z = 2(n_0 + A)(B)$. As a result,

$$\frac{m^2-1}{m^2+2} = \frac{X + iZ}{Y + iZ} = \frac{X + iZ}{Y + iZ} \times \frac{Y - iZ}{Y - iZ} = \frac{XY + iZY - iXZ + Z^2}{Y^2 + Z^2} = \frac{XY + Z^2}{Y^2 + Z^2} + i\frac{(Y - X)Z}{Y^2 + Z^2}$$

$$= \frac{1}{Y^2 + Z^2}\left[(XY + Z^2) + i(Y - X)Z\right],$$

and

$$\left|\frac{m^2-1}{m^2+2}\right|^2 = \left|\frac{1}{Y^2 + Z^2}\left[(XY + Z^2) + i(Y - X)Z\right]\right|^2$$

$$= \frac{1}{(Y^2 + Z^2)^2}\left[(XY + Z^2) + i(Y - X)Z\right]\left[(XY + Z^2) - i(Y - X)Z\right]$$

$$= \frac{(XY + Z^2)^2 + (Y - X)^2 Z^2}{(Y^2 + Z^2)^2} = \frac{X^2Y^2 + 2XYZ^2 + Z^4 + Y^2Z^2 - 2XYZ^2 + X^2Z^2}{(Y^2 + Z^2)^2}$$

$$= \frac{X^2Y^2 + Z^4 + Y^2Z^2 + X^2Z^2}{(Y^2 + Z^2)^2} = \frac{X^2Y^2 + Y^2Z^2 + X^2Z^2 + Z^4}{(Y^2 + Z^2)^2}$$

$$= \frac{Y^2(X^2 + Z^2) + Z^2(X^2 + Z^2)}{(Y^2 + Z^2)^2} = \frac{(Y^2 + Z^2)(X^2 + Z^2)}{(Y^2 + Z^2)^2} = \frac{X^2 + Z^2}{Y^2 + Z^2}.$$

Particularly, the real part of the term, $\frac{m^2 - 1}{m^2 + 2}$, which is $\frac{XY + Z^2}{Y^2 + Z^2}$, is responsible for the gradient force. Therefore, the ratio of the magnitude of the gradient force relative to that of the scattering force becomes:

$$\frac{\left|\vec{F}_{grad}\right|}{\left|\vec{F}_{scat}\right|} = 18.7 \times \frac{\left(\frac{m^2 - 1}{m^2 + 2}\right)}{\left|\frac{m^2 - 1}{m^2 + 2}\right|^2} = 18.7 \times \frac{\left(\frac{XY + Z^2}{Y^2 + Z^2}\right)}{\left(\frac{X^2 + Z^2}{Y^2 + Z^2}\right)} = 18.7 \times \frac{XY + Z^2}{Y^2 + Z^2} \times \frac{Y^2 + Z^2}{X^2 + Z^2} = 18.7 \times \frac{XY + Z^2}{X^2 + Z^2}.$$

Because $X = (n_0 + A)^2 - B^2 - n_m^2$ and $Y = (n_0 + A)^2 - B^2 + 2n_m^2$, hence $Y > X$. Accordingly, $\frac{XY + Z^2}{X^2 + Z^2} > 1$, and $\frac{\left|\vec{F}_{grad}\right|}{\left|\vec{F}_{scat}\right|} = 18.7 \times \frac{XY + Z^2}{X^2 + Z^2} > 18.7$. In other word, the magnitude of the gradient force is much greater than that of scattering force and the contribution of the scattering force is estimated to be less than 18.7^{-1} or 5.35%. Accordingly, we conclude that the contribution of the scattering force is much less than that of the gradient force as long as the particle is in the Rayleigh regime regardless of non-resonant or resonant conditions.