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On radiation forces acting on a transparent nanoparticle in the field of a focused laser beam

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Abstract. **Radiation forces acting on a transparent spherical nanoparticle in the field of a focused Gaussian laser beam are studied theoretically in the Rayleigh scattering regime. Expressions are derived for the scattering force and Cartesian components of the gradient force. The resultant force acting on a nanoparticle located in the centre of a laser beam is found. The parameters of the focused beam and optical properties of the nanoparticle for which the longitudinal component of the gradient force exceeds the scattering force are determined. Characteristics of the transverse gradient force are discussed.**

Keywords: *nanoparticle, radiation forces, light scattering, polarisability, permittivity, focal length, Gaussian beam, waist.*

1. Introduction

Ashkin's pioneering works [1, 2] on optical levitation and manipulation of small plastic particles by laser radiation stimulated interest in the study of the impact of radiometric forces (forces of light pressure) on particles of different materials (such as transparent plastic and absorbing metal ones). The influence of radiation forces on small particles in laser beams with a relatively small power has interesting prospects for practical use in various technologies, medical and biological research (see, e.g., review [3]). Optical transport of small particles by the radiation forces is a noninvasive technique with a wide practical application for the manipulation of viruses, bacteria, blood cells and yeast; in addition, it can be used in other studies, such as the propagation and scattering of light in aerosols. Radiation forces allow sorting and capturing small particles according to their size and optical properties [4].

One of the promising applications of radiometric forces is the study of nonlinear optical phenomena in liquid suspensions of transparent dielectric particles, the concentration of which is modulated by laser light due to the action of these forces. The possibility of using such suspensions (heterogeneous media) as a nonlinear optical material was pointed out by Palmer [5]. The authors of [6] recorded dynamic gratings in a concentrated liquid suspension of polystyrene microspheres by counterpropagating beams of cw laser radiation. An experiment on four-wave mixing in an aqueous suspension of latex microspheres using an argon laser to determine the optical Kerr coefficient n_2 was carried out in [7]. The Kerr coefficient measured in [7] at a concentration of the microspheres, $N_0 = 6.5\times10^{10}$ cm^{-3}, was 10^5 times greater than in CS_2. Note that, although each of the components of the suspension is an optically linear medium, i.e. the permittivities of the components are independent of the action of radiation, the suspension itself exhibits highly efficient optical nonlinearity. Having a large Kerr coefficient n_2, a liquid suspension of transparent microspheres can serve as a broadband nonlinear medium for low-intensity radiation of long duration. The authors of [8, 9] developed a theory of concentration four-wave mixing in a liquid suspension of transparent microspheres.

In experiments on manipulation of small particles by radiation forces, use is typically made of focused beams of cw laser radiation [1]. Despite a large number of works on the subject, theoretical studies of radiation forces in focused laser beams, to our knowledge, have not been conducted. In this paper, we consider radiation forces acting on a transparent spherical nanoparticle in the field of a focused Gaussian laser beam.

2. Basic relations

In a spatially inhomogeneous laser beam, two radiation forces act on a transparent particle: $\boldsymbol{F}_{\rm scat}$, i.e. the force caused by radiation scattering and acting along the direction of the beam propagation, and $\boldsymbol{F}_{\rm grad}$, i.e. the gradient force associated with the inhomogeneity of the radiation intensity ($\boldsymbol{F}_{{\rm grad}\,x}$ and $\boldsymbol{F}_{{\rm grad}\,y}$ are the force components operating across the beam, and $\boldsymbol{F}_{{\rm grad}\,z}$ is component acting along the beam).

The input amplitude of a Gaussian beam with a plane wavefront in the Cartesian coordinate system in the plane $z = 0$ can be represented as

$$E(x, y, 0) = E_0\exp\left(-\frac{x^2+y^2}{2r_0^2}\right), \quad (1)$$

where E_0 and r_0 are, respectively, the amplitude and beam radius. In this problem under study, the laser beam (1) is focused by a thin lens into a cell with a liquid with an immersed nanoparticle. The scheme of beam focusing by a lens with a focal length f is shown in Fig. 1.

The amplitude of the focused Gaussian beam at an arbitrary point $z > 0$ is given by (see, e.g., [10])

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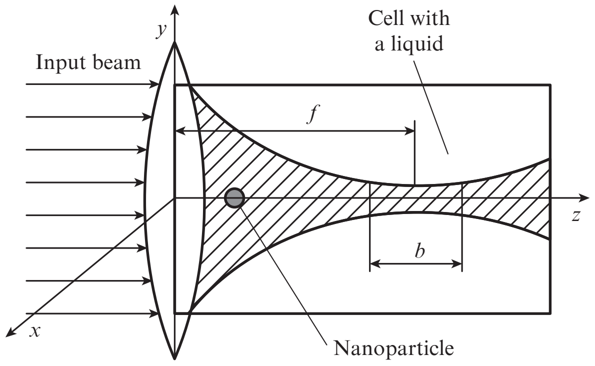

Figure 1. Scheme of beam focusing by a thin lens with a focal length f to a cell with a liquid with an immersed nanoparticle; b is the confocal parameter.

$$E(x,y,z) = \frac{E_0}{\sqrt{(1-z/f)^2+(z/z_d)^2}} \times \exp\left\{-\frac{x^2+y^2}{2r_0^2[(1-z/f)^2+(z/z_d)^2]} + i\psi(x,y,z)\right\}, \quad (2)$$

where $z_d = kr_0^2$ is the diffraction length of the beam; $k = (\omega/c)n$ is the wave number; ω is the radiation frequency; n is the refractive index of the liquid; and $\psi(x,y,z)$ is the phase of the beam amplitude: $\psi(x,y,0) = 0$.

Accordingly, from (2) we find the relation for the beam intensity

$$I(x,y,z) = \frac{I_0}{(1-z/f)^2+(z/z_d)^2} \times \exp\left\{-\frac{x^2+y^2}{r_0^2[(1-z/f)^2+(z/z_d)^2]}\right\}. \quad (3)$$

In the Rayleigh approximation [11], when the size of the particle is small compared with the radiation wavelength, expressions for the forces $\boldsymbol{F}_{scat}$ and $\boldsymbol{F}_{grad}$ can be written as

$$\boldsymbol{F}_{scat} = \boldsymbol{z}\frac{8}{3}\pi\frac{n}{c}k^4\alpha^2 I(x,y,z), \quad (4)$$

$$\boldsymbol{F}_{grad} = \boldsymbol{z}F_{grad\,z} + \boldsymbol{x}F_{grad\,x} + \boldsymbol{y}F_{grad\,y} = 2\pi\frac{n}{c}\alpha\left(\boldsymbol{z}\frac{\partial}{\partial z} + \boldsymbol{x}\frac{\partial}{\partial x} + \boldsymbol{y}\frac{\partial}{\partial y}\right)I(x,y,z), \quad (5)$$

where

$$\alpha = a^3\frac{\overline{m}^2-1}{\overline{m}^2+2} \quad (6)$$

is the polarisability of a spherical nanoparticle of radius a ($ka \ll 1$); $\overline{m} = n_0/n$; n_0 is the refractive index of the particle; and $\boldsymbol{x}$, $\boldsymbol{y}$, $\boldsymbol{z}$ are the unit vectors of the Cartesian coordinate axes. From (3)–(5) for the scattering force $\boldsymbol{F}_{scat}$ and the Cartesian components of the gradient force $\boldsymbol{F}_{grad}$ we find the expression

$$\boldsymbol{F}_{scat} = \boldsymbol{z}F_s^0\frac{I_0}{(1-z/f)^2+(z/z_d)^2} \times \exp\left\{-\frac{x^2+y^2}{r_0^2[(1-z/f)^2+(z/z_d)^2]}\right\}, \quad (7)$$

$$\boldsymbol{F}_{grad\,z} = \boldsymbol{z}F_\nabla^0\frac{2I_0(1-z/z_w)}{f[(1-z/f)^2+(z/z_d)^2]^2} \times \left\{1-\frac{x^2+y^2}{r_0^2[(1-z/f)^2+(z/z_d)^2]}\right\} \times \exp\left\{-\frac{x^2+y^2}{r_0^2[(1-z/f)^2+(z/z_d)^2]}\right\}, \quad (8)$$

$$\boldsymbol{F}_{grad\,x} = -\boldsymbol{x}F_\nabla^0\frac{2I_0 x}{r_0^2[(1-z/f)^2+(z/z_d)^2]^2} \times \exp\left\{-\frac{x^2+y^2}{r_0^2[(1-z/f)^2+(z/z_d)^2]}\right\}, \quad (9a)$$

$$\boldsymbol{F}_{grad\,y} = -\boldsymbol{y}F_\nabla^0\frac{2I_0 y}{r_0^2[(1-z/f)^2+(z/z_d)^2]^2} \times \exp\left\{-\frac{x^2+y^2}{r_0^2[(1-z/f)^2+(z/z_d)^2]}\right\}, \quad (9b)$$

where

$$F_s^0 = \frac{8}{3}\pi\frac{n}{c}k^4\alpha^2;\quad F_\nabla^0 = 2\pi\frac{n}{c}\alpha;\ \text{and}$$

$$z_w = \frac{f}{1+(f/z_d)^2} \quad (10)$$

is the coordinate of the beam waist.

Note that the scattering force $F_{scat} \propto a^6$, and the gradient force $F_{grad} \propto a^3$. Therefore, only for relatively small radii of spherical nanoparticles, as will be shown below, the longitudinal component of the gradient force $F_{grad\,z}$ may exceed the scattering force F_{scat}. This form of the expressions for the radiation forces in a Gaussian beam without focusing is given in [12]. The above expression (7)–(9) transform into the corresponding formula of this work at $f \to \infty$.

3. Results and discussion

When a nanoparticle rests on the beam axis ($x = y = 0$), it will be subjected to the scattering force $\boldsymbol{F}_{scat}$ and the longitudinal component of the gradient force $\boldsymbol{F}_{grad\,z}$. In this case, for $\overline{m} > 1$, the transverse components of the gradient force $\boldsymbol{F}_{grad\,x}$ and $\boldsymbol{F}_{grad\,y}$ will contribute to the retention of the particle on the z axis. Note that in the case of $\overline{m} < 1$, implemented by Ashkin [2] for air particles in a mixture of glycerol and water, their presence on the beam axis is unstable because when the particles are displaced from the beam centre, transverse gradient forces will divert them to its periphery.

From (7) and (8) for the resultant force $\boldsymbol{F}_z = \boldsymbol{F}_{scat} + \boldsymbol{F}_{grad\,z}$ we find the expression

$$\boldsymbol{F}_z = \boldsymbol{z}F_\nabla^0\frac{I_0}{f[(1-z/f)^2+(z/z_d)^2]}\left[\Re + \frac{2(1-z/z_w)}{(1-z/f)^2+(z/z_d)^2}\right], \quad (11)$$

where

$$\Re = \frac{F_s^0}{F_\nabla^0}f = \frac{4}{3}k^4\alpha f.$$

It follows from (11) that for $\alpha > 0$ ($\overline{m} > 1$) the longitudinal component of the gradient force $F_{grad\,z}$ in the region $z < z_w$ acts in one direction with the scattering force F_{scat}. At the waist point $z = z_w$ it is zero, while in the region $z > z_w$ it starts to act in the reverse direction. Note that in the absence of focusing (at $f \to \infty$) due to diffraction spreading of the beam, the force $F_{\text{grad}\,z}$ is always directed opposite to the force F_{scat} [12]. It is easy to show that in the region

$$z_w + \frac{f}{\Re}(1 - \sqrt{1 - \Gamma^2}) \leqslant z \leqslant z_w + \frac{f}{\Re}(1 + \sqrt{1 - \Gamma^2}) \qquad (12)$$

at $\Gamma = \Re(z_w/z_d) < 1$, the resultant force $F_z < 0$, i.e. acts in the reverse direction with respect to the direction of the beam propagation. Note that in the region $F_z < 0$, localisation of a nanoparticle is possible. When $\Gamma = 1$, i.e. at

$$a = a_0 = \sqrt[3]{\frac{3z_d(\overline{m}^2 + 2)}{4k^4 z_w f(\overline{m}^2 - 1)}} \sim \left(\frac{r_0}{f}\right)^{2/3}, \qquad (13)$$

$F_z = 0$ at point $\hat{z}_0 = z_w + f/\Re$. It is clear that for $\Gamma > 1$ ($a > a_0$), region (12) is absent and, therefore, the force F_z for all z acts only in the direction of the beam propagation. Thus, formula (13) determines the maximum radius of nanoparticles for their possible localisation by the resultant force.

From (7), (8) for the ratio of the forces acting along the z axis we find the expression

$$R(z) = \frac{F_{\text{grad}\,z}}{F_{\text{scat}}} = \frac{2}{\Re}\frac{1 - z/z_w}{(1 - z/f)^2 + (z/z_d)^2}. \qquad (14)$$

It can be shown that the coefficient $R(z)$ reaches extreme values

$$R_{\text{extr}} = \mp\frac{1}{\Re}\left(\frac{z_d}{z_w}\right) = \mp\frac{1}{\Gamma} \qquad (15)$$

at points $z_{\mp} = z_w(1 \pm f/z_d)$ from region (12) (at $\Gamma = 1$, $z_+ = \hat{z}_0$). It is easy to make sure that the distance between these points $\Delta z = z_+ - z_-$ is less than the confocal parameter $b = 2f^2/z_d$.

Estimates for a spherical latex nanoparticle in water (n_0 = 1.58 and n = 1.33) at f = 5 cm, r_0 = 0.1 cm and $k = 10^5$ cm^{-1} show that $\Gamma = 1$ for a radius a_0 = 13.6 nm. Consequently, the resultant force $\boldsymbol{F}_z$ at the given parameters can change the sign for nanoparticles with a radius $a < a_0$.

Figures 2 and 3 shows the dependences of the normalised forces F_{scat} and $F_{\text{grad}\,z}$ on the reduced coordinate $\bar{z} = z/f$ for three radii of the nanoparticle. One can see from Fig. 2 that $F_{\text{scat}}(\bar{z})$ has the form of a symmetrical curve with a maximum at the waist point ($\bar{z} \approx 1$). From (7) it follows that the maximum force F_{scat} is

$$F_{\text{scat}}^{\max}(\bar{z} \approx 1) = F_s^0 I_0 \frac{z_d^2}{z_w f}, \qquad (16)$$

because at point $z = z_w$ the beam intensity on the z axis reaches its maximum value [for the above-given parameters, $I(z_w) = 4\times10^4 I_0$], and its radius has a minimum defined by the expression

$$r(z_w) = r_0\frac{\sqrt{z_w f}}{z_d}. \qquad (17)$$

The dependence of the longitudinal component of the gradient force $F_{\text{grad}\,z}(\bar{z})$ has the form of a dispersion curve (see Fig. 3) and, as follows from (8), $F_{\text{grad}\,z}$ reaches extreme values

$$F_{\text{grad}\,z}^{\text{extr}} = \mp\frac{9}{8\sqrt{3}}F_\nabla^0 I_0 z_d\left(\frac{z_d}{z_w f}\right)^2 \qquad (18)$$

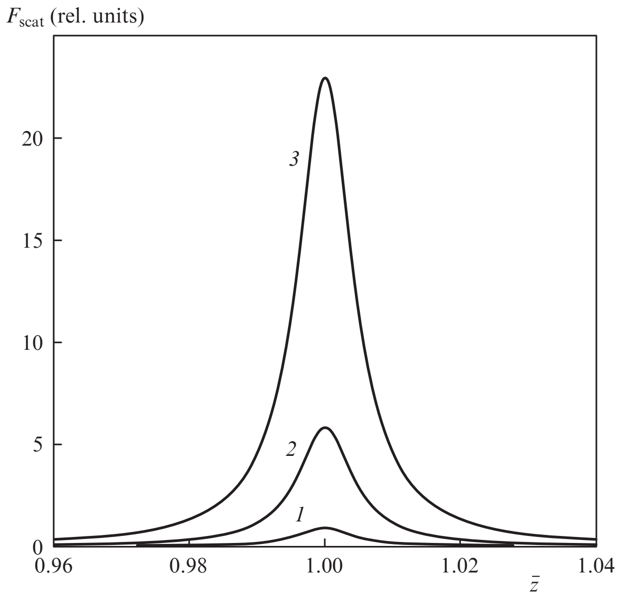

Figure 2. Normalised scattering force F_{scat} as a function of $\bar{z} = z/f$ for nanoparticle radii a = (*1*) 10, (*2*) 13.6 and (*3*) 17.1 nm.

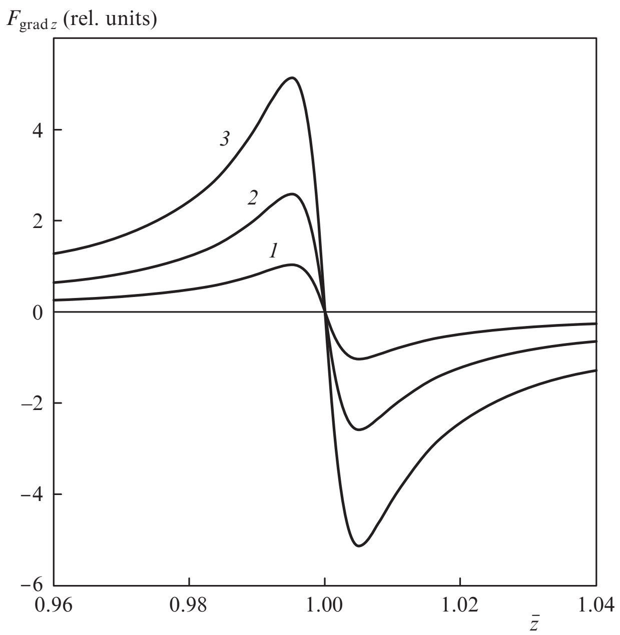

Figure 3. Normalised longitudinal component of the gradient force $F_{\text{grad}\,z}$ as a function of $\bar{z} = z/f$ for nanoparticle radii a = (*1*) 10, (*2*) 13.6 and (*3*) 17.1 nm.

at points $z_\pm^\nabla = z_w[1 \pm f/(\sqrt{3}\,z_d)]$. It can be shown that at these points

$$F_{\text{scat}}(z_\pm^\nabla) = \frac{3}{4}F_s^0 I_0\frac{z_d^2}{z_w f} = \frac{3}{4}F_{\text{scat}}^{\max} \qquad (19)$$

and therefore

$$R(z_\pm^\nabla) = \mp\frac{3\sqrt{3}}{8k^3\alpha}\frac{r_0^2}{z_w f} = \mp\frac{\sqrt{3}}{2}R_{\text{extr}}. \qquad (20)$$

Figure 4 shows the dependences of the normalised resultant force F_z on the reduced coordinate $\bar{z}$ for the same param-

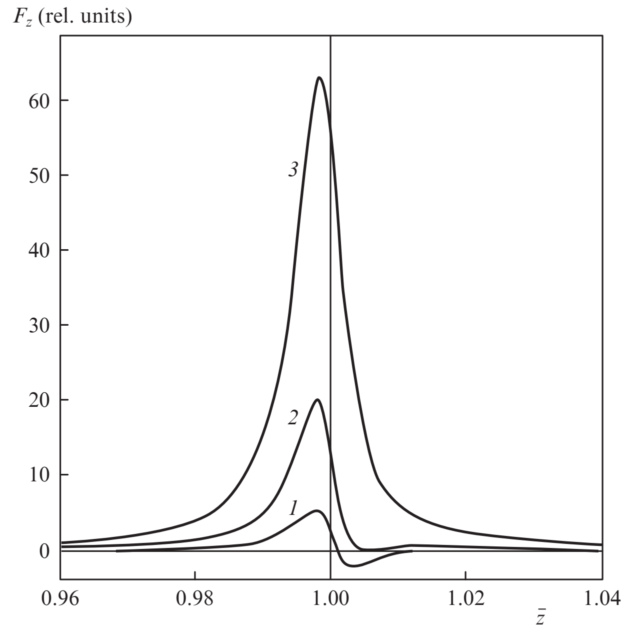

Figure 4. Normalised resultant force F_z as a function of $\bar{z} = z/f$ for nanoparticle radii a = (*1*) 10, (*2*) 13.6 and (*3*) 17.1 nm.

eters as for Figs 2 and 3. One can see from Fig. 4 that when $a = a_0 = 13.6$ nm, in accordance with (12) the resultant force F_z at $\bar{z} = \hat{z}_0/f$ is equal to zero, and at $a < a_0$ the force $F_z < 0$ in the region defined by inequalities (12). Unlike $F_{\rm scat}$, the impact of $F_{{\rm grad}\,z}$ causes the displacement of the maximum F_z to the left of point $\bar{z} = 1$.

Figure 5 shows the dependence of the limiting radius a_0 of a nanoparticle on the focal length f of the lens for different r_0, calculated by formula (13). Obviously, the smaller the f and the larger the input beam radius r_0, the larger nanoparticles can be localised in region (12).

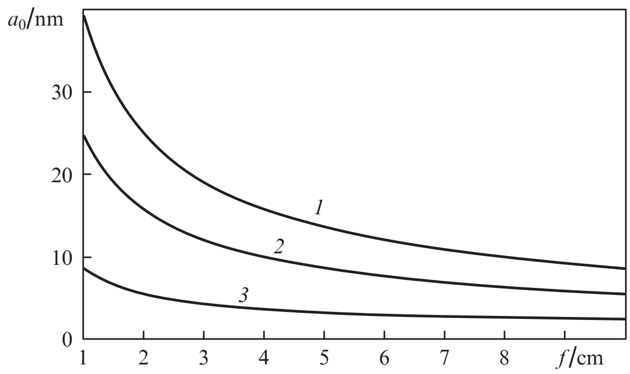

Figure 5. Dependences of the nanoparticle limiting radius a_0 on the focal length f of the lens, calculated from the condition $\Gamma(a_0, f, r_0) = 1$, at which the resultant force $F_z = 0$, for r_0 = (*1*) 0.1, (*2*) 0.05 and (*3*) 0.01 cm.

Let us now briefly discuss the transverse components of the gradient force. Due to the cylindrical symmetry of the focused beam we confine ourselves to one component, i.e. $F_{{\rm grad}\,x}$. It follows from (9a) that $F_{{\rm grad}\,x}$ takes a maximum value

$$F^{\max}_{{\rm grad}\,x}(z) = -\sqrt{\frac{2}{\rm e}}F^0_\nabla \frac{I_0}{r_0[(1 - z/f)^2 + (z/z_{\rm d})^2]^{3/2}} \qquad (21)$$

at points $x_0^\pm = \pm r_z/\sqrt{2}$, displaced from the z axis by a distance that is $\sqrt{2}$ times smaller than the current beam radius $r_z = r_0 \times \sqrt{(1 - z/f)^2 + (z/z_{\rm d})^2}$. At focal points, we have $x_0^\pm = \pm(r_0/\sqrt{2})(f/z_{\rm d})$ and therefore

$$F^{\max}_{{\rm grad}\,x}(z = f) = -\sqrt{\frac{2}{\rm e}}F^0_\nabla I_0\left(\frac{z_{\rm d}}{f}\right)^3. \qquad (22)$$

Thus, the transverse component of the gradient force at the focal point $z = f$ reaches a maximum value near the z axis (when $r_0 = 0.1$ cm, $f = 5$ cm and $k = 10^5$ cm^{-1}, the displacement of $x_0^\pm$ from the z axis is $\sim 3.5\times10^{-4}$ cm).

4. Conclusions

Thus, we have studied theoretically the characteristics of radiation forces acting on a transparent spherical nanoparticle in a focused laser beam with a Gaussian intensity distribution. We have determined the resultant force acting on a nanoparticle, which is in the centre of the laser beam, and have shown that up to point of the beam waist the longitudinal component of the gradient force, as the scattering force, acts in the direction of its propagation. In the region behind the waist point the longitudinal component of the gradient force changes its sign and becomes oppositely directed to the scattering force. In addition, for small nanoparticles [$a < a_0 \sim (r_0/f)^{2/3}$] the resultant force $\boldsymbol{F}_z$ reverses its direction. In this case, the possibility of capture and localisation of nanoparticles as well as spatial separation of nanoparticles having various sizes and optical properties is realised.

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