# Pulling extremely anisotropic lossy particles using light without intensity gradient

Andrey Novitsky[1] and Cheng-Wei Qiu[2]

[1]*Department of Theoretical Physics and Astrophysics, Belarusian State University, Nezavisimosti Ave. 4, 220030 Minsk, Belarus*

[2]*Department of Electrical and Computer Engineering, National University of Singapore, 4 Engineering Drive 3, Singapore 117583, Singapore*

(Received 28 June 2014; revised manuscript received 16 August 2014; published 7 November 2014)

We study the effect of pulling optical force acting on a nonmagnetic anisotropic bead in electromagnetic fields without intensity gradient. Extreme anisotropy can be realized by a hyperbolic metamaterial made of metal-dielectric multilayers. We find that a passive anisotropic Rayleigh particle cannot be pulled by the electromagnetic beam without intensity gradient and the nonparaxial incident beams can exert backward negative force acting on anisotropic dipole spheres. We investigate the validity of the dipole approximation and establish the conditions for pulling hyperbolic-metamaterial particles. It is important to note that the loss in hyperbolic metamaterial does not suppress the effect of pulling force. We notice that the nonradial components of the permittivity tensor strongly affect the optical force and propose the way of material engineering to ensure the optical pulling.

DOI: 10.1103/PhysRevA.90.053815 PACS number(s): 42.50.Wk, 81.05.Xj, 42.25.Fx

## I. INTRODUCTION

Optical force can be divided in two parts: gradient and nonconservative force. The force of the first type as $\mathbf{F} = \nabla V(\mathbf{r})$ does not perform work in a closed loop and, therefore, is also called conservative. It has been widely used in optical manipulation for microparticles, e.g., in optical tweezers [1,2]. Gradient forces are able to shift objects towards either the intensity minimum or maximum depending on the relation between the refractive indices of the object and ambient medium. Hence the particles can be moved along any direction giving a proper three-dimensional (3D) control [3]. This control is realized using the continuous tuning of the field maximum (minimum) by a computer hologram. A single diffraction-free light beam is an easier solution resulting in the pulling force within a long distance like a tractor beam. The intensity of the diffraction-free beam (Bessel beam [4] and Airy beam [5]) does not change along the direction of propagation; thus the gradient force is absent in this direction.

Light field without intensity gradient usually creates the pushing force (light pressure). This well-known effect was discovered experimentally a hundred years ago by Lebedev [6]. The pulling force $F < 0$ using the light pressure is not obvious. It was predicted recently for a superposition of plane waves having the same value of the longitudinal wave number (nondiffracting beam) in optics [7–9] and acoustics [10,11]. It can be explained in two ways. First, if the forward-scattered electromagnetic field results in the great field momentum, then particle's recoil momentum is directed towards the light source [7,12,13]. Second, the interaction of multipole momenta induced by the incident light in the particle can originate the negative backward optical force [7,8,14].

It was shown preciously for isotropic particles that nonparaxial light beams should be used to obtain pulling force [7–9] and the force in dipole approximation can be negative for a wide range of material and size parameters of the particle [15]. The present paper generalizes the previous results on the case of pulling rotationally symmetric anisotropic spherical particles. Special attention is paid to the influence of different components of the permittivity tensor on the optical force. We investigate (i) the light scattering in the case of anisotropic (including hyperbolic-medium) particles, (ii) paraxiality criterion for pulling effect by the light without intensity gradient, (iii) possibilities of pulling anisotropic Rayleigh particles, and (iv) applicability of the dipole approximation.

The paper consists of seven sections, the first of which is the Introduction. In Sec. II we formulate the problem to be solved, demonstrate the realization of particle's anisotropy, and propose the energy-based criterion for choosing an adequate solution inside a spherical particle. In the third and fourth sections we show for the general nondiffracting beam that pulling force is impossible for passive Rayleigh particles and nonparaxial beams are necessary. In Secs. V and VI we analyze the pulling force effect for dipole beads and Mie spheres in nonparaxial Bessel beams. Section VII concludes the paper.

## II. LIGHT SCATTERING BY ANISOTROPIC DIELECTRIC BEADS

We consider an anisotropic spherical particle in vacuum characterized by the radius $R$, dielectric permittivity tensor

$$\hat{\varepsilon} = \varepsilon_r \mathbf{e}_r \otimes \mathbf{e}_r + \varepsilon_t (\mathbf{e}_\theta \otimes \mathbf{e}_\theta + \mathbf{e}_\varphi \otimes \mathbf{e}_\varphi), \tag{1}$$

and magnetic permeability $\mu = 1$. Here $\mathbf{e}_r$, $\mathbf{e}_\theta$, and $\mathbf{e}_\varphi$ are the basis vectors in spherical coordinates. Real parts of the radial $\varepsilon_r$ and transverse $\varepsilon_t$ permittivities can be positive and negative. The case $\mathrm{Re}(\varepsilon_t)\mathrm{Re}(\varepsilon_r) < 0$ corresponds to the hyperbolic-metamaterial particles [16,17]. Unusual permittivity with extreme values of $\varepsilon_r$ and $\varepsilon_t$ can exist only for the composite media. Nevertheless, particle's material is assumed to be a continuous medium in this paper.

Let us discuss how continuous-medium permittivity tensor Eq. (1) could appear. We consider a multilayer spherical particle shown in the inset of Fig. 1, which consists of a core (permittivity $\varepsilon_1$ and radius $R_1$) and $2N$ alternating spherical layers of the same thickness (permittivities $\varepsilon^{(1)}$ and $\varepsilon^{(2)}$; thicknesses $h^{(1)}$ and $h^{(2)}$). When the discrete-layer system is homogenized, it can be replaced by the homogeneous anisotropic cladding with effective permittivity tensor in Eq. (1), the components of which are calculated as follows [17]:

$$\varepsilon_t = \frac{\varepsilon^{(1)}h^{(1)} + \varepsilon^{(2)}h^{(2)}}{h^{(1)} + h^{(2)}}, \quad \varepsilon_r = \frac{\varepsilon^{(1)}\varepsilon^{(2)}(h^{(1)} + h^{(2)})}{\varepsilon^{(1)}h^{(1)} + \varepsilon^{(2)}h^{(2)}}. \tag{2}$$

 

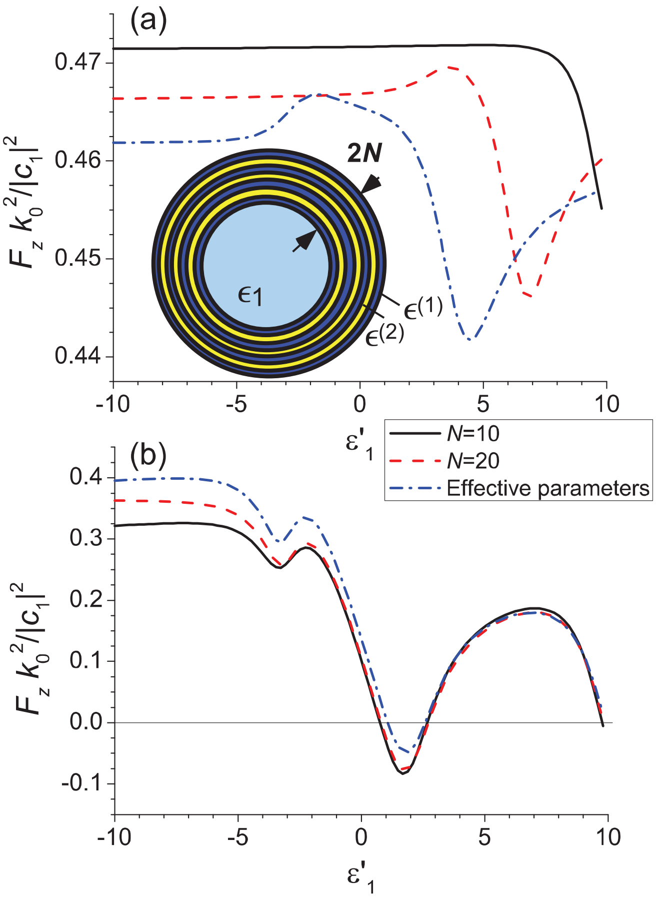


FIG. 1. (Color online) Dimensionless optical force $F_z k_0^2/|c_1|^2$ acting on a multilayer spherical particle in nonparaxial Bessel beam in Eq. (21) versus permittivity of the core $\varepsilon_1 = \varepsilon_1' + i0$ for two types of hyperbolic-metamaterial coatings formed by $2N$ alternating spherical layers with permittivities $\varepsilon^{(1)}$ and $\varepsilon^{(2)}$: (a) $\varepsilon^{(1)} = -17.2 + 0.8i$, $\varepsilon^{(2)} = 2.59$ (effective parameters of the periodic structure $\varepsilon_r = 6.1 + 0.05i$ and $\varepsilon_t = -7.3 + 0.4i$); size parameter of the core $2\pi R_1/\lambda = 0.25$, $\alpha = 70^\circ$; (b) $\varepsilon^{(1)} = -4 + 0.16i$, $\varepsilon^{(2)} = 20 + 0.04i$ (effective parameters $\varepsilon_r = -10 + 0.5i$ and $\varepsilon_t = 8 + 0.1i$), $2\pi R_1/\lambda = 1.1$, and $\alpha = 80^\circ$. Parameters: size parameter of the particle $2\pi R_2/\lambda = 2$, Bessel beam's order $m = 1$, and beam's amplitude $c_2 = ic_1$ ($\lambda$ is the radiation wavelength).

Hyperbolic metamaterial can be realized as a periodic metal-dielectric structure having $\mathrm{Re}(\varepsilon_t)\mathrm{Re}(\varepsilon_r) < 0$. Thus radial anisotropy of the form (1) appears in the model of spherical multilayer systems.

In Fig. 1, we compare the optical forces acting on a multilayer particle and a core-shell particle consisting of a core and anisotropic coating with effective transverse and radial permittivities in Eq. (2). The force is calculated using the integration of the Maxwell stress tensor over a sphere around the bead, while the scattered electric and magnetic fields are found from the Mie theory (for multilayer particles we apply the method described in Ref. [18]). Effective parameters in Eq. (2) are well applicable, when $N$ is great. However, smaller $N$ results in the reasonable values of the optical force, too. Optical force in Fig. 1(a) is positive (pushing) and corresponds to the hyperbolic metamaterial with $\mathrm{Re}(\varepsilon_r) > 0$ and $\mathrm{Re}(\varepsilon_t) < 0$. For the second type of the hyperbolic metamaterials $\mathrm{Re}(\varepsilon_r) < 0$ and $\mathrm{Re}(\varepsilon_t) > 0$ and the force can be negative (pulling). Parameters $\varepsilon^{(1)}$ and $\varepsilon^{(2)}$ in Fig. 1 are exemplary.

When an anisotropic sphere in vacuum is illuminated with an electromagnetic wave of angular frequency $\omega = 2\pi c/\lambda$ ($c$ is the speed of light; $\lambda$ is the wavelength), the scattered electric and magnetic fields are computed using the Mie coefficients

$$a_l = \frac{n^2 j_{\nu-1/2}(nx)[x j_l(x)]' - j_l(x)[nx j_{\nu-1/2}(nx)]'}{n^2 j_{\nu-1/2}(nx)\big[x h_l^{(1)}(x)\big]' - h_l^{(1)}(x)[nx j_{\nu-1/2}(nx)]'},$$
$$b_l = \frac{j_l(nx)[x j_l(x)]' - j_l(x)[nx j_l(nx)]'}{j_l\big[x h_l^{(1)}(x)\big]' - h_l^{(1)}(x)[nx j_l(nx)]'}, \qquad (3)$$

where $j_l(x) = \sqrt{\pi/2x}\,J_{l+1/2}(x)$ and $h_l^{(1)}(x) = \sqrt{\pi/2x}\,H_{l+1/2}^{(1)}(x)$ are the spherical Bessel function and Hankel function of the first kind of order $l$, respectively [$J_{l+1/2}(x)$ and $H_{l+1/2}^{(1)}(x)$ are the Bessel and Hankel functions], $j_l'(y)$ stands for the derivative with respect to function's argument $y$, $n = \sqrt{\varepsilon_t}$, $x = k_0 R$ is the size parameter, $k_0 = \omega/c$ is the wave number in vacuum, and $\nu = \sqrt{l(l+1)\varepsilon_t/\varepsilon_r + 1/4}$. In the case of isotropic media $\varepsilon_r = \varepsilon_t$ and we arrive at the ordinary Mie coefficients ($\nu = l + 1/2$) [19].

From the comparison of the Mie series truncated at $l = 1$ and radiation of the dipole (electric and magnetic dipole moments are $\mathbf{p} = \alpha_e \mathbf{E}$ and $\mathbf{m} = \alpha_m \mathbf{H}$, respectively), one can link the polarizabilities of the bead $\alpha_{e,m}$ with the Mie coefficients $a_1$ and $b_1$ as

$$\alpha_e = \frac{3ia_1}{2k_0^3}, \quad \alpha_m = \frac{3ib_1}{2k_0^3}. \qquad (4)$$

When the size parameter $x = k_0 R \ll 1$, the spherical Bessel and Hankel functions approximately equal

$$j_m(x) \approx \sqrt{\frac{\pi}{2}}\frac{2^{-m-1/2}}{\Gamma(m+3/2)}x^m,$$
$$h_m^{(1)}(x) \approx \sqrt{\frac{\pi}{2}}\left[\frac{2^{-m-1/2}}{\Gamma(m+3/2)}x^m + \frac{im}{\pi}2^{m+1/2}\Gamma(m+1/2)x^{-m-1}\right], \qquad (5)$$

where $\Gamma(m)$ is the gamma function. By substituting these functions into the Mie coefficients, we have the polarizabilities

$$\alpha_{e,m} = \frac{\alpha_{e,m}^{(0)}/k_0^3}{1 - 2i\alpha_{e,m}^{(0)}/3}, \qquad (6)$$

where the normalized static electric and magnetic polarizabilities

$$\alpha_e^{(0)} = \frac{\varepsilon_t - \nu_1/2 - 1/4}{\varepsilon_t + \nu_1 + 1/2}x^3, \quad \alpha_m^{(0)} = 0 \qquad (7)$$

are introduced, and $\nu_1 = \sqrt{2\varepsilon_t/\varepsilon_r + 1/4}$ is the quantity $\nu$ at $l = 1$. It should be noted that $\alpha_m = 0$ only for Rayleigh particles, whose radiuses are much smaller than the wavelength ($x \ll 1$). For bigger dipole spheres, magnetic polarizability $\alpha_m$ should be described by Eq. (4) as it was discussed in Ref. [20] in the case of nonmagnetic isotropic particles.

Incident light induces electric and magnetic fields inside an isotropic bead. Generally these fields can be described by a couple of independent solutions described by the Bessel spherical functions of the first $j_n(x)$ and second $y_n(x)$ kind. However, since $y_m(x)$ becomes infinite at the center of the particle, there exists only one solution inside the sphere [21].

For anisotropic particles, both independent solutions can result in the singular fields at the center. In this case the energy-based criterion should be used,

$$\int |\mathbf{E}|^2 dV < \infty. \tag{8}$$

From the criterion one concludes (see the Appendix A) that the appropriate solution of the Maxwell equations expressed by means of the spherical Bessel functions $j_n(x)$ should satisfy condition $\mathrm{Re}(\nu_1) > 0$.

In conclusion of this section, we have considered three regimes. The first regime (Rayleigh approximation) corresponds to small particle's radius compared with the radiation wavelength $R \ll \lambda$. In the second regime (dipole approximation) we are confined with the electric $\mathbf{p}$ and magnetic $\mathbf{m}$ dipole moments. Particle's radius is compared with the wavelength $R \sim \lambda$. The third regime is the exact calculation using the full Mie series, which includes electric and magnetic dipoles, quadrupoles, octupoles, etc. Materials with extreme anisotropy can be realized in practice as a spherical multilayer system. Then effective parameters are given by Eq. (2). There are two independent solutions of the Maxwell equations in a spherical particle. Both of them can be singular at the center of an anisotropic sphere. In this case we propose the energy-based criterion Eq. (8).

## III. IMPOSSIBILITY OF PULLING PASSIVE RAYLEIGH PARTICLES

For electromagnetic fields without intensity gradient along the $z$ axis, the projection of time-averaged optical force onto this direction equals [15,22]

$$F_z = \frac{k_0\beta}{2}[\mathrm{Im}(\alpha_e)|\mathbf{E}|^2 + \mathrm{Im}(\alpha_m)|\mathbf{H}|^2] - \frac{k_0^4}{3}\mathrm{Re}[\alpha_e\alpha_m^*(\mathbf{E}\times\mathbf{H}^*)_z] \tag{9}$$

in dipole approximation, where $\beta = k_z/k_0$ is the dimensionless longitudinal wave number; $\mathbf{E}$ and $\mathbf{H}$ are the fields at the center of the spherical bead $r = 0$.

Nonmagnetic Rayleigh particles have no magnetic polarizability ($\alpha_m = 0$); hence the sign of the optical force $F_z = (k_0\beta/2)\mathrm{Im}(\alpha_e)|\mathbf{E}|^2$ depends on the sign of $\mathrm{Im}(\alpha_e)$. Let us analyze the imaginary part of electric polarizability

$$\mathrm{Im}(\alpha_e) = \frac{\mathrm{Im}\big(\alpha_e^{(0)}\big) + (2/3)\big[\mathrm{Re}\big(\alpha_e^{(0)}\big)^2 + \mathrm{Im}\big(\alpha_e^{(0)}\big)^2\big]}{k_0^3\big\{\big[1 + (2/3)\mathrm{Im}\big(\alpha_e^{(0)}\big)\big]^2 + (4/9)\mathrm{Re}\big(\alpha_e^{(0)}\big)^2\big\}}. \tag{10}$$

Since the second term in the nominator is positive, the necessary condition of pulling reads $\mathrm{Im}(\alpha_e^{(0)}) < 0$. Rayleigh spheres with small size parameters $x \ll 1$ possess small static polarizability $|\alpha_e^{(0)}| \ll 1$. Having in mind that normally $|\mathrm{Re}(\alpha_e^{(0)})| \gg \mathrm{Im}(\alpha_e^{(0)})$, the optical force is negative (pulling) when

$$\mathrm{Im}\big(\alpha_e^{(0)}\big) + \frac{2}{3}\mathrm{Re}\big(\alpha_e^{(0)}\big)^2 < 0. \tag{11}$$

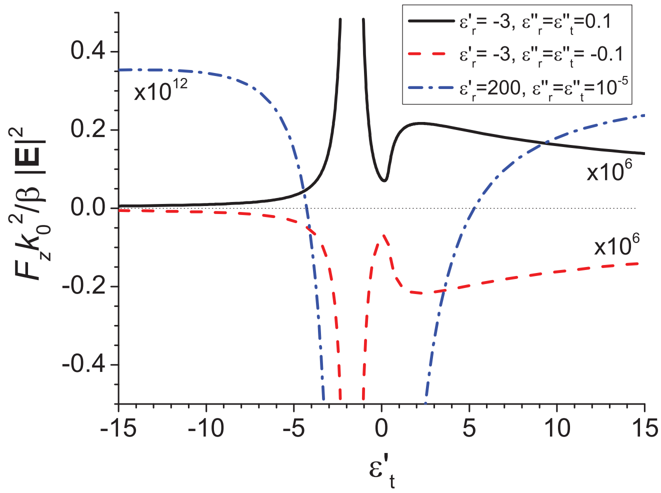


FIG. 2. (Color online) Dimensionless optical force $F_z k_0^2/\beta|\mathbf{E}|^2$ versus transverse permittivity $\varepsilon'_t$ for the beads of size $x = k_0 R = 0.01$.

By considering complex quantities as $\varepsilon_{r,t} = \varepsilon'_{r,t} + i\varepsilon''_{r,t}$ and $\nu = \nu' + i\nu''$, we show some typical dependencies for Rayleigh particles in Fig. 2. For ordinary material parameters, the force is either positive or negative depending on the sign of the imaginary part of permittivity. The force can change from pushing to pulling and vice versa, when the imaginary part of static polarizability is comparable with the squared real part and condition (11) holds (dot-dashed curve). In this case, the material parameters are hardly achievable, because the losses $\varepsilon''_{r,t}$ are extremely small and the permittivity $\varepsilon'_r$ is great.

Now we show that passive anisotropic Rayleigh particles cannot be pulled by the light, because necessary condition $\mathrm{Im}(\alpha_e^{(0)}) < 0$ does not hold. The imaginary part of the static electric polarizability in Eq. (7)

$$\mathrm{Im}(\alpha_e^{(0)}) = x^3\frac{A}{(\varepsilon'_t + \nu'_1 + 1/2)^2 + (\varepsilon''_t + \nu''_1)^2} \tag{12}$$

is negative, if the nominator is negative, i.e.,

$$A = (\varepsilon''_t - \nu''_1/2)(\varepsilon'_t + \nu'_1 + 1/2) - (\varepsilon'_t - \nu'_1/2 - 1/4)(\varepsilon''_t + \nu''_1) < 0 \tag{13}$$

or

$$\nu''_1\varepsilon'_t > \varepsilon''_t(\nu'_1 + 1/2). \tag{14}$$

Let us study when this condition is met in the case $|\varepsilon''_{r,t}| \ll |\varepsilon'_{r,t}|$. We expect that, for greater losses, the pulling is less feasible because of nonelastic momentum transfer from light to particle resulting in pushing force [7,8].

(i) $\varepsilon'_t/\varepsilon'_r > -1/8$. Then $\nu_1 \approx |\nu_1|(1 + i\varphi)$ (note that we choose a solution with $\nu'_1 > 0$ to guarantee the finiteness of the electromagnetic energy), where

$$|\nu_1| = \sqrt{\left|2\frac{\varepsilon'_t}{\varepsilon'_r} + \frac{1}{4}\right|}, \quad \varphi = \frac{\varepsilon''_t\varepsilon'_r - \varepsilon'_t\varepsilon''_r}{\varepsilon'^2_r|\nu_1|}. \tag{15}$$

For passive media $\varepsilon''_{r,t} > 0$. The necessary condition in Eq. (14) presented as

$$\frac{\varepsilon''_r}{\varepsilon''_t} < \frac{1}{p^2}\left(-p - \frac{1}{4} - \frac{1}{2}\sqrt{2p + 1/4}\right) < 0 \tag{16}$$

contradicts with the medium passivity and, therefore, cannot be satisfied for any ratio $p = \varepsilon_t'/\varepsilon_r' > -1/8$. When $\varepsilon_{r,t}'' < 0$, the inequality in Eq. (16) can be satisfied.

(ii) $\varepsilon_t'/\varepsilon_r' < -1/8$. This means that the medium is hyperbolic. In this situation $\nu_1 \approx \pm|\nu_1|(\varphi + i)$. The sign should be chosen to ensure $\nu_1' > 0$: "+" for $\varphi > 0$ and "−" for $\varphi < 0$. Using $\nu_1' > 0$ the necessary condition (14) to have a pulling force takes the form

$$\pm|\nu_1|\varepsilon_t' > \varepsilon_t''/2. \tag{17}$$

We choose sign +, if $\varphi > 0$ or $\varepsilon_t''\varepsilon_r' - \varepsilon_t'\varepsilon_r'' > 0$. For passive media $\varepsilon_{r,t}'' > 0$ the above inequality together with $\varepsilon_t'/\varepsilon_r' < -1/8$ requires $\varepsilon_r' > 0$ and $\varepsilon_t' < 0$. However, in this case the condition in Eq. (17) fails, i.e., $-|\nu_1||\varepsilon_t'| > |\varepsilon_t''|/2$. For any amplifying media $\varepsilon_{r,t}'' < 0$ we derive opposite inequalities $\varepsilon_r' < 0$ and $\varepsilon_t' > 0$, which bring us to identical inequality $|\nu_1||\varepsilon_t'| > -|\varepsilon_t''|/2$ as Eq. (17).

The similar reason is valid for the sign −, resulting in the same conclusion that passive spherical beads in Rayleigh approximation cannot be pulled to the light source.

To summarize, passive anisotropic Rayleigh particles are always pushed by the light, while the gain assists pulling force. In the case of transparent particles $\varepsilon_{r,t}'' = 0$ ($\varphi = 0$) we still have two cases.

If $\varepsilon_t'/\varepsilon_r' > -1/8$, $\nu_1$ is a real number, $\mathrm{Im}(\alpha_e^{(0)}) = 0$, and $\mathrm{Im}(\alpha_e) > 0$. This means that the force is always pushing.

If $\varepsilon_t'/\varepsilon_r' < -1/8$ (hyperbolic media), $\nu_1 = \pm i|\nu_1|$ is an imaginary number. Since $\mathrm{Re}(\nu_1) = 0$, the solution with finite electromagnetic energy inside the sphere cannot be found. The case of transparent media is a peculiar situation between two normal cases: medium with an absorption (pushing optical force) and medium with a gain (pulling force). Point $\varepsilon_{r,t}'' = 0$ is the singular point, at which the optical force flips.

## IV. NONPARAXIAL LIGHT BEAMS

In this section we study the correlation between normalized longitudinal wave number $\beta = k_z/k_0$ and pulling force for a particle in dipole approximation. The necessary condition for $\beta$ to obtain optical force $F_z < 0$ reads

$$\beta < \frac{2k_0^3\mathrm{Re}[\alpha_e\alpha_m^*(\mathbf{E}\times\mathbf{H}^*)_z]}{3[\mathrm{Im}(\alpha_e)|\mathbf{E}|^2 + \mathrm{Im}(\alpha_m)|\mathbf{H}|^2]}. \tag{18}$$

Aiming estimation, we assume $|\mathbf{E}|^2 = |\mathbf{H}|^2$ and $\mathrm{Re}(\mathbf{E}\times\mathbf{H}^*)_z \leqslant |\mathbf{E}|^2$, then

$$\beta < g(\varepsilon_r',\varepsilon_t') = \frac{2k_0^3\mathrm{Re}(\alpha_e\alpha_m^*)}{3[\mathrm{Im}(\alpha_e) + \mathrm{Im}(\alpha_m)]}. \tag{19}$$

When $\varepsilon_t'/\varepsilon_r' > -1/8$, the condition for $\beta$ can be derived in the closed form for transparent spheres $\varepsilon_r = \varepsilon_r'$, $\varepsilon_t = \varepsilon_t'$. Indeed,

$$\begin{aligned}\beta &< \frac{k_0^3\mathrm{Re}(\alpha_e\alpha_m^*)}{3\sqrt{\mathrm{Im}(\alpha_e)\mathrm{Im}(\alpha_m)}}\\ &< \frac{y_1y_2 + (4/9)y_1^2y_2^2}{3\sqrt{[1+(4/9)y_1^2][1+(4/9)y_2^2]}},\end{aligned} \tag{20}$$

where static polarizabilities $y_1 = \alpha_e^{(0)}$ and $y_2 = \alpha_m^{(0)}$ are real. The maximum of the function on the right-hand side $f(y_1,y_2)$

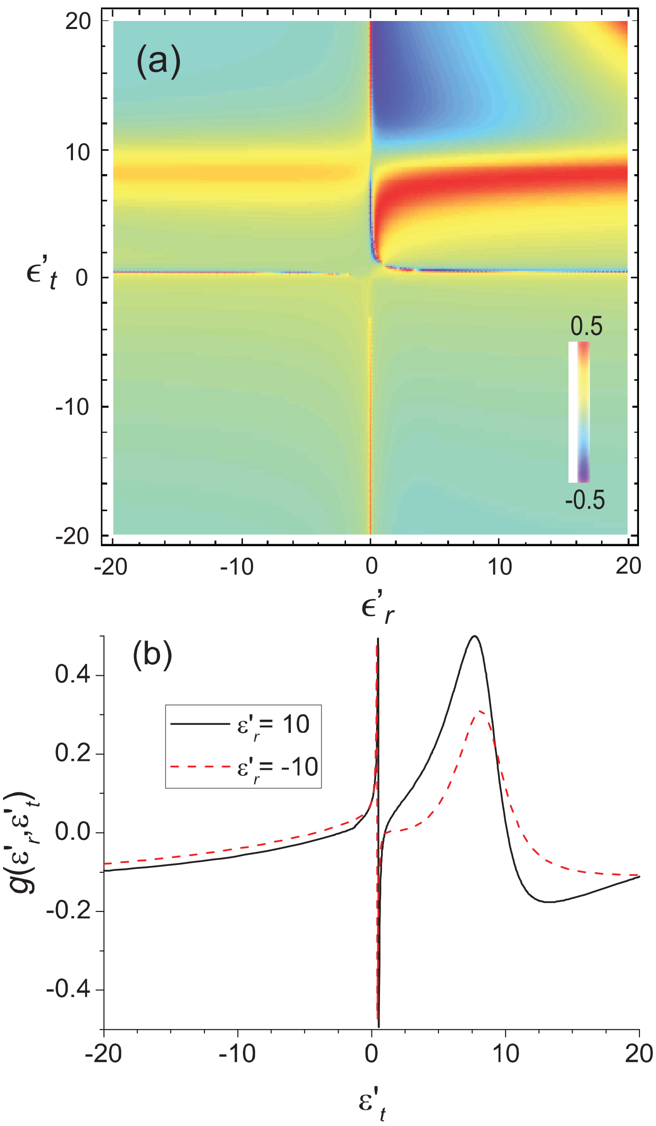


FIG. 3. (Color online) (a) Function $g(\varepsilon_r',\varepsilon_t')$ calculated for a nonmagnetic spherical bead of size parameter $x = k_0R = 1$ ($\varepsilon_{r,t}'' = 10^{-8}$). (b) The cuts of function $g(\varepsilon_r',\varepsilon_t')$ at $\varepsilon_r' = \pm 10$.

follows from solving equations $\partial f/\partial y_1 = 0$ and $\partial f/\partial y_2 = 0$. The maximum requires $y_1 = y_2$ resulting in $\beta < 1/2$. In terms of the cone angle $\alpha = \arccos(\beta)$ for a nonparaxial Bessel beam we get to $\alpha > 60^\circ$ as for isotropic particles [15]. For absorbing spheres, the angles $\alpha$ should be greater.

The situation $\varepsilon_t'/\varepsilon_r' < -1/8$ is basically different, because the static polarizability $\alpha_e^{(0)}$ is a complex number even for transparent spheres. However, we can expect that the condition $\beta < 1/2$ (i.e., $\alpha > 60^\circ$) holds for all dipole particles. To support this assumption, we demonstrate the quantity $g(\varepsilon_r',\varepsilon_t')$ from Eq. (19) in Fig. 3. Since $g(\varepsilon_r',\varepsilon_t')$ is in the interval from $-0.5$ to 0.5, parameter $\beta$ cannot be greater than 0.5. When function $g(\varepsilon_r',\varepsilon_t')$ is greater, the pulling force is more feasible. From the diagram in Fig. 3(a) we conclude that the normal dielectrics ($\varepsilon_r' > 0$, $\varepsilon_t' > 0$) can be pulled towards the light source, while metallic particles ($\varepsilon_r' < 0$, $\varepsilon_t' < 0$) cannot be pulled in dipole approximation. Only one type of hyperbolic metamaterial ($\varepsilon_r' < 0$, $\varepsilon_t' > 0$) can result in the backward optical force. Minimum angle $\alpha$ for hyperbolic media can be estimated to be 70°. The previous conclusion of minimum angles $\alpha$ is invalid beyond the dipole approximation.

## V. ANISOTROPIC DIPOLE PARTICLES IN NONPARAXIAL BESSEL BEAMS: PULLING FORCE FOR LOSSY PARTICLES

We consider nonparaxial Bessel beam as an incident light beam illuminating an anisotropic particle in vacuum. In the following analysis, the Bessel beam is used as a model of nondiffracting light beam; however, the beam model itself does not play a crucial role for presented results. Electric and magnetic fields of a nonparaxial Bessel beam in cylindrical coordinates $(r, \varphi, z)$ can be written as [8,15,23]

$$\mathbf{E} = e^{im\varphi + i\beta k_0 z}\left(J_m(qk_0r)c_2\mathbf{e}_z - \frac{c_1}{q}(\mathbf{e}_z \times \mathbf{b}) + \frac{\beta}{q}c_2\mathbf{b}\right),$$
$$\mathbf{H} = e^{im\varphi + i\beta k_0 z}\left(J_m(qk_0r)c_1\mathbf{e}_z + \frac{\beta}{q}c_1\mathbf{b} + \frac{c_2}{q}(\mathbf{e}_z \times \mathbf{b})\right), \tag{21}$$

where $\mathbf{b} = iJ_m'(qk_0r)\mathbf{e}_r - (m/qk_0r)J_m(qk_0r)\mathbf{e}_\varphi$, $J_m'(qk_0r) = [dJ_m/d(qk_0r)]$, $m$ is beam's order, $(\mathbf{e}_r, \mathbf{e}_\varphi, \mathbf{e}_z)$ is the set of basis vectors in cylindrical coordinates, and $\beta = k_z/k_0$ and $q = \sqrt{k_0^2 - k_z^2}/k_0 = \sqrt{1-\beta^2}$ are the dimensionless longitudinal and transverse wave numbers. Complex coefficients $c_1$ and $c_2$ introduce TE-polarized and TM-polarized field contributions for the nonparaxial Bessel beam.

Bessel beam can be treated as a superposition of elementary plane waves, the wave vectors of which occupy the cone with angle $2\alpha$ at the vertex [see the inset in Fig. 4(a)]. Then the longitudinal $\beta$ and transverse $q$ wave numbers can be represented in terms of the angle $\alpha$ as $\beta = \cos\alpha$ and $q = \sin\alpha$. Paraxial Bessel beams are characterized by small angles $\alpha$, while nonparaxial Bessel beams have large $\alpha$ (small longitudinal wave number $\beta = k_z/k_0$).

In the dipole approximation, pulling optical force $F_z < 0$ is predicted using Eq. (9). Approximation certainly works for small size parameters $x = k_0R \ll 1$. But even for greater sizes $x$ the force in dipole approximation is adequate. Color spots in density plots of Fig. 4 illustrate the regions of radial $\varepsilon_r'$ and transverse $\varepsilon_t'$ permittivities, for which the force in dipole approximation is negative, $F_z < 0$. White area is occupied by

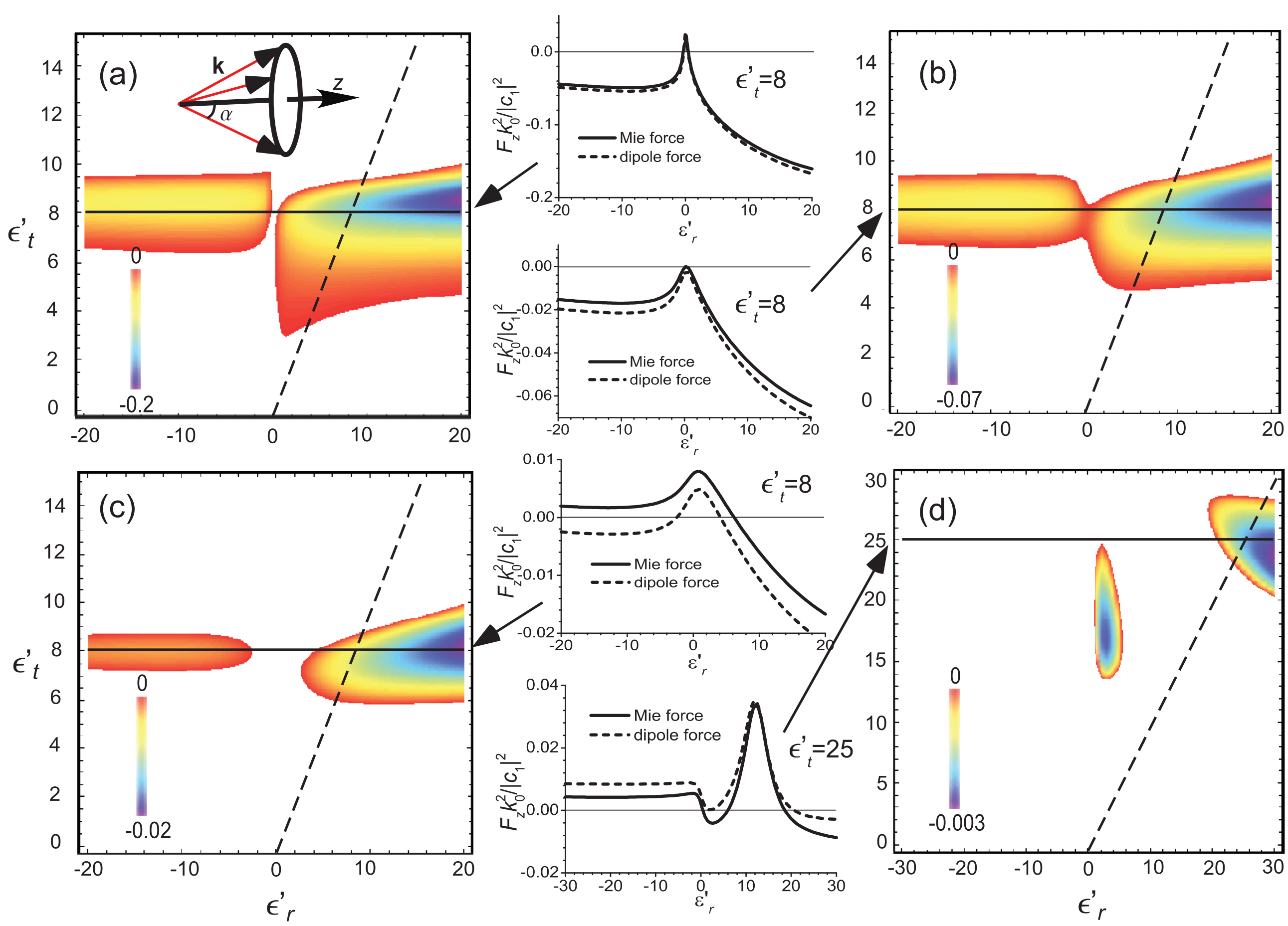


FIG. 4. (Color online) Density plots of the time-averaged pulling optical force $F_z k_0^2/|c_1|^2 < 0$ in dipole approximation exerted by a nonparaxial Bessel beam on an anisotropic particle for (a) Bessel beam's order $m = 1$, particle's loss parameter $\varepsilon_{r,t}'' = 0.1$; (b) $m = 1$, $\varepsilon_{r,t}'' = 1$; (c) $m = 1$, $\varepsilon_{r,t}'' = 2$; (d) $m = -1$, $\varepsilon_{r,t}'' = 1$. Dashed line indicates the case of isotropic permittivity $\varepsilon_r = \varepsilon_t$. In the insets in the middle of the figure, the optical forces in the dipole approximations $F_z$ and beyond $F_z^{\mathrm{Mie}}$ are shown for (a)–(c) $\varepsilon_t' = 8$ and (d) $\varepsilon_t' = 25$. Parameters: particle's size parameter $k_0R = 1$, beam's cone angle $\alpha = 80°$, and beam's amplitude $c_2 = ic_1$.

the values of positive force. Four plots in the middle column of Fig. 4 show the comparison of the optical forces calculated in dipole approximation $F_z$ and beyond $F_z^{\mathrm{Mie}}$ (using the higher-order multipoles as quadrupoles, octupoles, etc.). In Figs. 4(a) and 4(b) the deviation of the dipole force $F_z$ from the exact Mie-theory force $F_z^{\mathrm{Mie}}$ is negligible compared with the values of $F_z$, that is $|F_z^{\mathrm{Mie}} - F_z| \ll |F_z|$. In this case the pulling force can be predicted using the dipole approximation. However, when the values of the force $|F_z|$ are small, the deviation $|F_z^{\mathrm{Mie}} - F_z|$ is comparable with $|F_z|$ and the dipole approximation cannot be used. For instance, in Fig. 4(c) the optical force is positive for $\varepsilon_r' < 0$, though negative force appears in dipole approximation. Contrariwise the pulling optical force for small $\varepsilon_r'$ in Fig. 4(d) is not predicted in dipole approximation. The dipole force is applicable when $\varepsilon'$ and $\varepsilon''$ are not great [Figs. 4(a) and (b)]. Then $|\sqrt{\varepsilon' + i\varepsilon''}|k_0 R$ entering Mie coefficients is quite small.

Real parts of radial and transverse permittivities can be positive and negative numbers. From Figs. 3(a) we notice that the pulling force effect is feasible for the passive hyperbolic-metamaterial particles with $\varepsilon_t' > 0$ and $\varepsilon_r' < 0$. One could expect the pulling in a wide range of $\varepsilon_r'$ for near-transparent particles (small $\varepsilon_{r,t}''$) as in Fig. 4(a). However, it is quite surprising that the backward force exists in a wide range of $\varepsilon_r'$ for particles with large losses [see Fig. 4(b)]. This means that the composite metal-based materials, such as hyperbolic media, can be exploited for optical pulling using gradientless light beams. When the losses further increase [Fig. 4(c)], the hyperbolic-metamaterial sphere is no more attracted by the light source, though the positive $\varepsilon_r'$ and $\varepsilon_t'$ result in $F_z < 0$. The dependence of the force $F_z$ on $\varepsilon_r''$ is weak. Estimating for the metal-dielectric periodic structure of cocentric spherical layers $\varepsilon_t'' = \mathrm{Im}[(h^{(d)}\varepsilon^{(d)} + h^{(m)}\varepsilon^{(m)})/(h^{(d)} + h^{(m)})] \approx \mathrm{Im}[\varepsilon^{(m)}]h^{(m)}/(h^{(d)} + h^{(m)}) \sim A$, we can establish the limitations on the imaginary part of the metal permittivity $\mathrm{Im}[\varepsilon^{(m)}] \sim A(1 + h^{(d)}/h^{(m)})$, where $\varepsilon^{(d)}$ and $\varepsilon^{(m)}$ are the permittivities of dielectric and metal, and $h^{(d)}$ and $h^{(m)}$ are the thicknesses of dielectric and metal. Thus the metal losses can be quite high, if $h^{(d)} > h^{(m)}$. When $\varepsilon_t''$ is great, the pulling force turns into a pushing one as in Fig. 5.

Reaction of the negative-order Bessel beam ($m = -1$) is the same as that of the positive-order Bessel beam ($m = 1$) after the sign change in amplitude $c_2 \to -c_2$. In Fig. 4(d) we show the force $F_z$ in the case of $m = -1$, which cannot be backward either for negative $\varepsilon_r'$ or for small $\varepsilon_t'$. However, for greater cone angles $\alpha$ the pulling optical force is feasible, when $\varepsilon_r' < 0$. Decreasing $\varepsilon_{r,t}''$ below 0.1, one can reduce the cone angle $\alpha$ up to 70°. It should be noted that the negative force in the range $\varepsilon_r' < 0$ is forbidden for $\alpha = 70°$ and $m = 1$.

Anisotropy enhances the force values compared with the case of isotropic particles (isotropic parameters are marked with the dashed lines in Fig. 4). The force minima are shifted towards the greater positive permittivities $\varepsilon_r'$. In the case of the hyperbolic-metamaterial particles the pulling is weaker than for isotropic ones; nevertheless, $F_z < 0$ is realized in a broad range of permittivities $\varepsilon_r'$.

Thus we have justified that the optical force in dipole approximation is applicable for enough small permittivities and losses, when $|\sqrt{\varepsilon}|k_0 R \sim 1$. Particle's loss parameter $\varepsilon_t''$ should not be a small quantity to ensure pulling, but it should be small compared with the permittivity $\varepsilon_t'$. Another permittivity tensor component $\varepsilon_r'$ does not influence much and, therefore, it can take values smaller than $\varepsilon_r''$ as in Fig. 4(b). We have shown how the loss parameters $\varepsilon_{r,t}''$ should be engineered to guarantee the pulling effect. It should be noticed that the above results are valid for other nonparaxial light beams and the Bessel beam is used as one of the possible electromagnetic beam models.

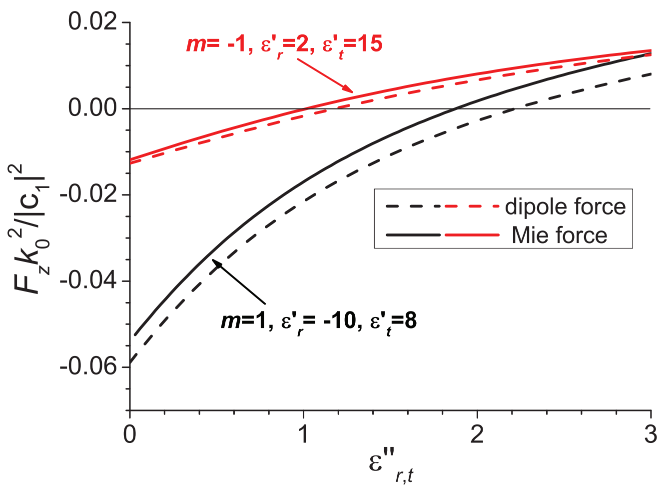


FIG. 5. (Color online) Dimensionless optical force $F_z k_0^2/|c_1|^2$ calculated using the dipole and Mie solutions as function of the loss parameter $\varepsilon_r'' = \varepsilon_t''$ ($k_0 R = 1$, $\alpha = 80°$, and $c_2 = i c_1$).

## VI. ANISOTROPIC MIE PARTICLES IN NONPARAXIAL BESSEL BEAMS: DIPOLE PULLING FORCE BEYOND THE DIPOLE APPROXIMATION

For anisotropic particles of larger radius, the dipole approximation fails and higher-order multipoles should be taken into account. Extending the results obtained in dipole approximation (Fig. 4) one can expect the greatest pulling optical force near transverse permittivity $\varepsilon_t' = 8$. In Fig. 6 the radial permittivity is chosen as $\varepsilon_r' = -10$ (the case of hyperbolic medium $\varepsilon_r'\varepsilon_t' < 0$). Low losses of particle's material $\varepsilon_{r,t}''$ and large cone angle $\alpha$ of the Bessel beam are the beneficial conditions for pulling force (solid curve in Fig. 6). In this case the first strong dip around $k_0 R = 1$ appears due to the interaction of electric and magnetic dipoles [the last term in Eq. (9)]. Weaker dips for greater $k_0 R$ appear as the superposition of interaction terms of dipole and higher-order multipoles. Negative optical force near the first dip is quite stable with respect to the losses, but quickly fades away, when the cone angle $\alpha$ gets smaller. Thus the hyperbolic-metamaterial lossy particles are pulled to the light source mainly for small parameters $k_0 R$ (in dipole approximation).

The first dip in Fig. 7(a) is well described in the dipole approximation, while the following dips require higher-order Mie terms. Nevertheless, all negative-force dips in Fig. 7(a) definitely correspond to the interaction of electric and magnetic dipoles. The quadrupole interaction is able to be the reason for the optical pulling forces, too, but the dipole resonances are stronger than the quadrupole ones in the considered example. Since the quadrupole dips are basically narrower

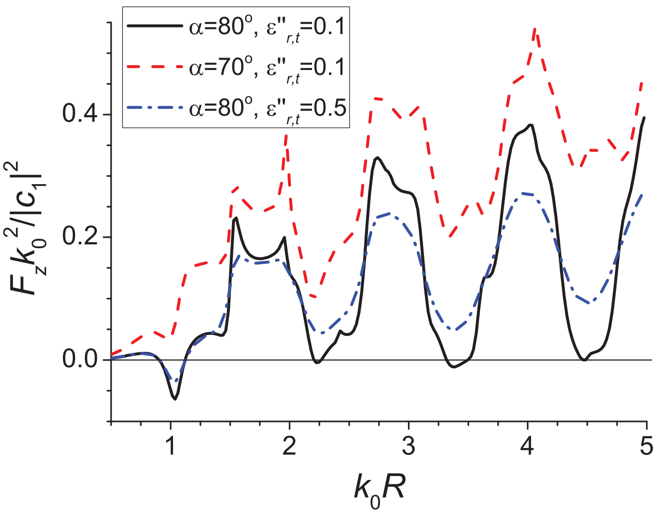


FIG. 6. (Color online) Dimensionless optical force $F_z k_0^2/|c_1|^2$ beyond the dipole approximation versus the size parameter $x = k_0 R$ of the hyperbolic-metamaterial bead ($\varepsilon_r' = -10$, $\varepsilon_t' = 8$, $m = 1$, and $c_2 = ic_1$).

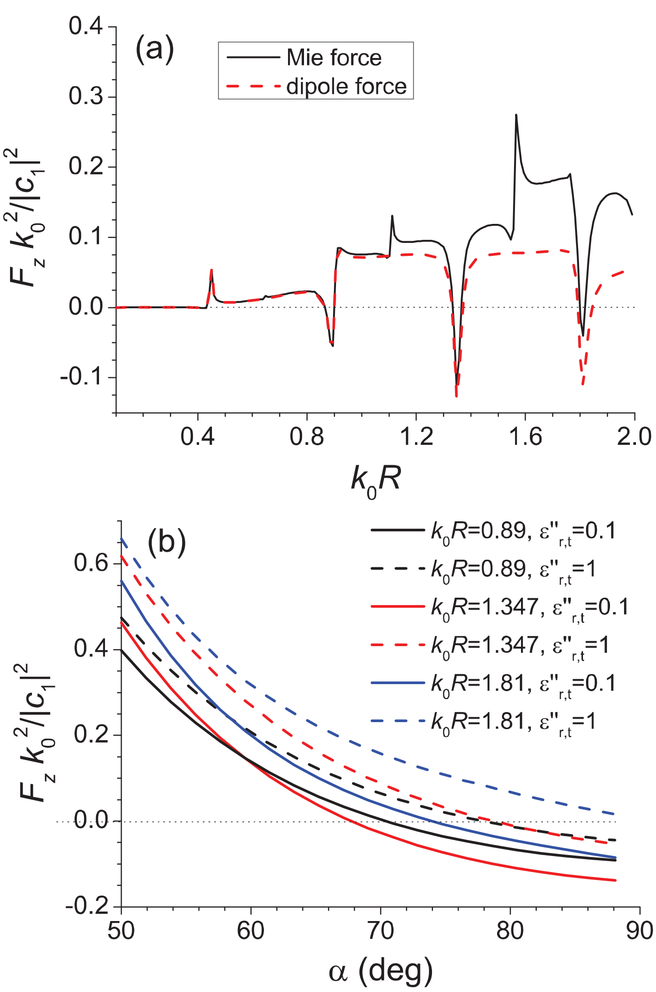


FIG. 7. (Color online) (a) Dimensionless optical force $F_z k_0^2/|c_1|^2$ versus the size parameter $k_0 R$ for a hyperbolic-metamaterial particle ($\varepsilon''_{r,t} = 0.1$, $\alpha = 80^\circ$). (b) Cone angle $\alpha$ dependence of the optical force beyond the dipole approximation. Parameters: $\varepsilon_r' = -10$, $\varepsilon_t' = 48$, $m = 1$, and $c_2 = ic_1$.

than the dipole dips, the losses inevitably destroy the pulling force effect beyond the dipole approximation. So, the dipole interaction is the main mechanism to get the stable pulling forces with respect to the losses. One can propose a technique to engineer material and size of particles, which enables the backward force: (i) ensure the pulling optical force in dipole approximation, and (ii) require the dipole moments are greater than the higher-order multipoles ($\{|a_1|,|b_1|\} \gg \{|a_{l>1}|,|b_{l>1}|\}$).

The dependence of the optical force on the cone angle $\alpha$ is shown in Fig. 7(b) for the three dips found in Fig. 7(a). When $k_0 R = 1.347$, the optical pulling force exists for the cone angles $\alpha$ of the nonparaxial Bessel beam less than 70°. Limitation of 70° is justified by the diagram in Fig. 3(a), but smaller angles are not forbidden, because the size $k_0 R = 1.347$ is beyond the dipole approximation. Smaller losses $\varepsilon''_{r,t}$ mean smaller $\alpha$ is possible [see Fig. 7(b)].

## VII. CONCLUSION

We have revealed that mainly the transverse permittivity component influences the pulling optical force without intensity gradient. Since the radial component can vary in wide ranges resulting in $F_z < 0$, the permittivity tensor can be engineered in a way to reduce the loss parameter $\varepsilon_t''$ and obtain the proper value of $\varepsilon_r'$. This is achieved by judiciously selecting the material parameters and thicknesses of the multilayer system forming a homogeneous anisotropic medium with permittivity tensor components Eq. (2).

We have shown that the dipole approximation is valid for anisotropic particle sizes $k_0 R \sim 1$. Moreover, the dipolar term in the optical force $\sim \mathrm{Re}(\mathbf{p} \times \mathbf{m}^*)_z$ plays an important part beyond the dipole approximation [see Fig. 7(a)]. We have noticed that the dipole-based pulling force is more robust to the losses than the higher-order-multipoles-based pulling force.

We have proved that passive anisotropic Rayleigh particles cannot be pulled by a gradientless light and have given the limitations on the nonparaxiality characteristics (longitudinal wave number $\beta = k_z/k_0$) in dipole approximation. To support our conclusions we have used the optical force calculations for the model of a nonparaxial Bessel beam (no intensity gradient along the direction of beam's propagation).

We have discovered that hyperbolic-metamaterial particles with $\varepsilon_t' > 0$ and $\varepsilon_r' < 0$ can be attracted to the light source. Metal-containing hyperbolic-medium particles should possess significant losses. It is important that the pulling force is stable with respect to the particle's losses, the values of which can achieve $\varepsilon''_{t,r} \sim 1$.

## ACKNOWLEDGMENTS

We acknowledge Professor I. D. Feranchuk and Dr. V. M. Galynsky for helpful discussions. A.N. is grateful for financial support from the Belarusian Republican Foundation for Fundamental Research (Grant No. F13M-014). C.W.Q. acknowledges the support from the National University of Singapore (Grant No. R-263-ODO-678-133).

## APPENDIX: CHOOSING APPROPRIATE SOLUTION INSIDE AN ANISOTROPIC SPHERICAL PARTICLE

Electric field inside an anisotropic sphere with permittivity tensor Eq. (1) can be written as [18]

$$\mathbf{E} = \sum_{l=1}^{\infty}\sum_{m=-l}^{l} F_{lm}(\theta,\varphi)[\mathcal{E}_l^m(j)+\mathcal{E}_l^m(y)],$$
$$\mathcal{E}_l^m(z) = \left(z_l(\sqrt{\varepsilon_t}k_0r)A_1^{(l,m)}\mathbf{e}_\theta + \frac{z_{\nu-1/2}(\sqrt{\varepsilon_t}k_0r)}{r}A_2^{(l,m)}\mathbf{e}_r + \frac{[\sqrt{\varepsilon_t}k_0r z_{\nu-1/2}(\sqrt{\varepsilon_t}k_0r)]'}{r}A_3^{(l,m)}\mathbf{e}_\varphi\right), \quad \text{(A1)}$$

where $F_{lm}(\theta,\varphi)$ is the tensor function of angles in spherical coordinates $\theta$ and $\varphi$, derivative is calculated over the argument $\sqrt{\varepsilon_t}k_0r$ of the spherical function $z$, and $A_1^{(l,m)}$, $A_2^{(l,m)}$, and $A_3^{(l,m)}$ are constant amplitudes. Vector $\mathcal{E}_l^m$ depends on the spherical function $z$, e.g., spherical Bessel functions $j$ and $y$.

Electric field (A1) in the vicinity of the center of the sphere $r=0$ can be approximated by the greatest term in the first sum

$$\left|\mathcal{E}_{l=1}^m(j)\right| \sim \frac{j_{\nu_1-1/2}(\sqrt{\varepsilon_t}k_0r)}{r} \sim r^{\nu_1-3/2} \quad \text{(A2)}$$

and $\mathcal{E}_l^m(y)=0$ as nonphysical singular solution, if $\mathrm{Re}(\nu_1)>0$. Here $\nu_1=\sqrt{2\varepsilon_t/\varepsilon_r+1/4}$ is the quantity $\nu$ for $l=1$. When $\mathrm{Re}(\nu_1)<0$, $\mathcal{E}_l^m(j)=0$ as nonphysical singular solution and $|\mathcal{E}_1^m(y)|\sim r^{-\nu_1-3/2}$.

When $0<\mathrm{Re}(\nu_1)<3/2$, electric field at the center of spherical particle becomes infinite for both spherical Bessel functions $j_n$ and $y_n$. Condition $0<\mathrm{Re}(\nu_1)<3/2$ holds true for a wide class of lossless anisotropic materials with $-1/8<\varepsilon_t/\varepsilon_r<1$ including the hyperbolic metamaterials characterized by $\varepsilon_t\varepsilon_r<0$.

We claim that the criterion for choosing an appropriate solution is the finiteness of the electromagnetic energy stored inside the sphere Eq. (8). Electric field according to Eq. (8) is a quadratically integrable function as a wave function in quantum mechanics. By substituting solution (A2) into Eq. (8) we derive

$$\int_0^R |\mathbf{E}|^2 r^2 dr \sim \int_0^R r^{2\,\mathrm{Re}(\nu_1)-1}dr. \quad \text{(A3)}$$

If $\mathrm{Re}(\nu_1)>0$, the integral always results in the finite energy inside the sphere. If $\mathrm{Re}(\nu_1)<0$, we should use another solution $|\mathcal{E}_1^m(y)|\sim r^{-\nu_1-3/2}$ expressed by means of the spherical Bessel function of the second kind. The only exception is the case $\mathrm{Re}(\nu_1)=0$, which brings us to the logarithmically divergent integral $\int_0^R r^{-1}dr=\ln(r)|_0^R$.

The situation $\mathrm{Re}(\nu_1)=0$ corresponds, e.g., to the model of the lossless hyperbolic metamaterial or, more strictly, anisotropic medium with $\mathrm{Im}(\varepsilon_t/\varepsilon_r)=0$ and $\mathrm{Re}(\varepsilon_t/\varepsilon_r)+1/8<0$. Complex transverse and radial permittivities should be linked using a real coefficient $C$ as follows:

$$\varepsilon_t = C\varepsilon_r, \quad C<-\frac{1}{8}. \quad \text{(A4)}$$

In this case both independent solutions do not ensure finite electromagnetic energy. Electric field $E\sim r^{i\,\mathrm{Im}(\nu_1)-3/2}$ rapidly oscillates. Peculiar case (A4) is a very sharp condition and cannot be realized in practice. For example, it is not feasible to realize perfectly transparent (lossless) anisotropic media. Passive transparent media can be characterized by the permittivities $\varepsilon_t+i\delta_t$ and $\varepsilon_r+i\delta_r$, where $\delta_{t,r}>0$ is a very small quantity. This situation recalls instable equilibrium point of the pendulum, which is not realizable in practice.

The infinite electric (magnetic) field at the center of the spherical particle can be understood as follows. In realistic situation anisotropic particle can be presented as a very small isotropic core covered with anisotropic shell (we believe it is technologically difficult to create the required anisotropy at the center). Then there is a regular solution at the center, while the field in the vicinity of $r=0$ is great, but not infinite.

Thus the appropriate solution $\mathcal{E}_l^m(j)$ [$\mathcal{E}_l^m(y)$] should satisfy condition $\mathrm{Re}(\nu)>0$ [$\mathrm{Re}(\nu)<0$]. In computations, we ordinarily choose positive $\mathrm{Re}(\nu)>0$ and solutions in terms of the spherical Bessel functions $j_n(x)$.


[1] A. Ashkin, Phys. Rev. Lett. **24**, 156 (1970).
[2] A. Ashkin, J. M. Dziedzic, J. E. Bjorkholm, and S. Chu, Opt. Lett. **11**, 288 (1986).
[3] D. G. Grier, Nature (London) **424**, 810 (2003).
[4] J. Durnin, J. Opt. Soc. Am. A **4**, 651 (1987).
[5] G. A. Siviloglou, J. Broky, A. Dogariu, and D. N. Christodoulides, Phys. Rev. Lett. **99**, 213901 (2007).
[6] P. Lebedev, Ann. Phys. (Leipzig) **311**, 433 (1901).
[7] J. Chen, J. Ng, Z. Lin, and C. T. Chan, Nat. Photon. **5**, 531 (2011).
[8] A. Novitsky, C.-W. Qiu, and H. Wang, Phys. Rev. Lett. **107**, 203601 (2011).
[9] S. Sukhov and A. Dogariu, Phys. Rev. Lett. **107**, 203602 (2011).
[10] P. L. Marston, J. Acoust. Soc. Am. **120**, 3518 (2006).
[11] P. L. Marston, J. Acoust. Soc. Am. **122**, 3162 (2007).
[12] A. Dogariu, S. Sukhov, and J. J. Saenz, Nat. Photon. **7**, 24 (2013).
[13] V. Kajorndejnukul *et al.*, Nat. Photon. **7**, 787 (2013).
[14] C.-W. Qiu *et al.*, Nanophotonics **3**, 181 (2014).
[15] A. Novitsky, C.-W. Qiu, and A. Lavrinenko, Phys. Rev. Lett. **109**, 023902 (2012).
[16] D. R. Smith and D. Schurig, Phys. Rev. Lett. **90**, 077405 (2003).
[17] A. Poddubny, I. Iorsh, P. Belov, and Y. Kivshar, Nat. Photon. **7**, 948 (2013).
[18] A. Novitsky and L. Barkovsky, Phys. Rev. A **77**, 033849 (2008).
[19] G. Mie, Ann. Phys. (Leipzig) **330**, 377 (1908).
[20] A. Garcia-Etxarri, R. Gomez-Medina, L. S. Froufe-Perez, C. Lopez, L. Chantada, F. Scheffold, J. Aizpurua, M. Nieto-Vesperinas, and J. J. Saenz, Opt. Express **19**, 4815 (2011).
[21] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1998).
[22] M. Nieto-Vesperinas, J. J. Saenz, R. Gomez-Medina, and L. Chantada, Opt. Express **18**, 11428 (2010).
[23] A. V. Novitsky and D. V. Novitsky, J. Opt. Soc. Am. A **24**, 2844 (2007).