Efficiency of excitation of the spatial resonant configurations of the internal optical field of spherical microparticles by focused laser beams

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The problem is considered on the efficiency of excitation of spherical dielectric microparticles by laser beams. Numerical investigations have been carried out of the resonance excitation of the optical field in microspheres when illuminating them by focused Gaussian–Hermitian beams at central and side incidence of radiation. Configurations have been determined of the most optimal transformation of the laser beam energy into the field of the resonator modes.

Introduction

As known, transparent microparticles with the size much greater than the wavelength of incident radiation can be considered as optical resonators with high Q-factor values and are of some interest when studying various nonlinear optical interactions in a small volume of a substance. The resonance excitation of the optical field in such systems is yet a problem of primary importance, especially in view of a wide use of microresonators as optical devices for the purposes of aerosol spectroscopy and optical microelectronics.[1] A number of experimental and theoretical investigations carried out during the past two decades (the review is presented in Ref. 1) has shown that a sharp increase is observed in the efficiency of nonlinear relation between the waves that interact inside the particle, if resonance conditions for pumping wave (the so-called "input" resonance) has been satisfied. In particular, it leads to a significant decrease in the energy threshold of the effects of stimulated light scattering (SRS, SSMB, and stimulated fluorescence).

As it follows from the Mie theory, the necessary and sufficient condition of obtaining the resonance configurations of optical field at illumination of a spherical particle by a plane wave is the existence of a certain correspondence between the value of the particle diffraction parameter $x_a = 2\pi a_0/\lambda$ (a_0 is the particle radius and λ is the wavelength of pump radiation) and its refractive index m_a. Then one of the terms of the expansion series of the internal electromagnetic field over special partial waves (spatial frequencies) starts to dominate that leads to the transformation of the spatial structure of the field and its concentration in the ring zone near the particle surface. Thus generated resonance oscillation modes are called in the scientific literature the whispering modes which are characterized by high values of the Q-factor ($Q \sim 10^5$–10^8). However, in practice, one usually deals not with the plane waves, but with focused beams. The diameter of the beam waist can be about particle diameter as small or even smaller. Spatial structure of the optical field inside the particle in this case is also different than that in the case of a plane wave. The internal field is located according to the beam profile along the direction of its incidence on the particle and takes maximum value at the principal diameter of the particle. Moreover, the light beam can enter the particle not along its diameter, but a little bit asides that leads to the appearance of the sharp asymmetry of the internal field distribution, first, in the azimuth.

Therefore, these peculiarities can lead to the fact that even if the aforementioned necessary conditions hold, no resonance in particles is observed with the sharply focused beams. The illustration of this fact is presented in Fig. 1 that shows a 2D distribution of the relative intensity of the optical field (in xy plane) inside a liquid drop the radius of which corresponds to the TE^1_{60} mode resonance at illumination by a Gaussian beam with the half-width $w_0 = a_0/2$ and different displacement of the beam axis relative to the particle center along the coordinate y. It is seen that at the central incidence of light beam ($y_0 = 0$, Fig. 1*b*) the field in the particle is not resonance contrary to the case of a plane wave incidence under the same conditions (Fig. 1*a*). The ring structure of the field characteristic of the resonance appears only at the displacement of the beam toward the particle edge (Fig. 1*c*) and complete correspondence to the case with a plane wave is reached at $y_0 > a_0$ (Fig. 1*d*). Experimental study in Refs. 2 and 3 was the first to pay due attention to this fact and the theoretical basis of the phenomenon was laid in Refs. 4–6.

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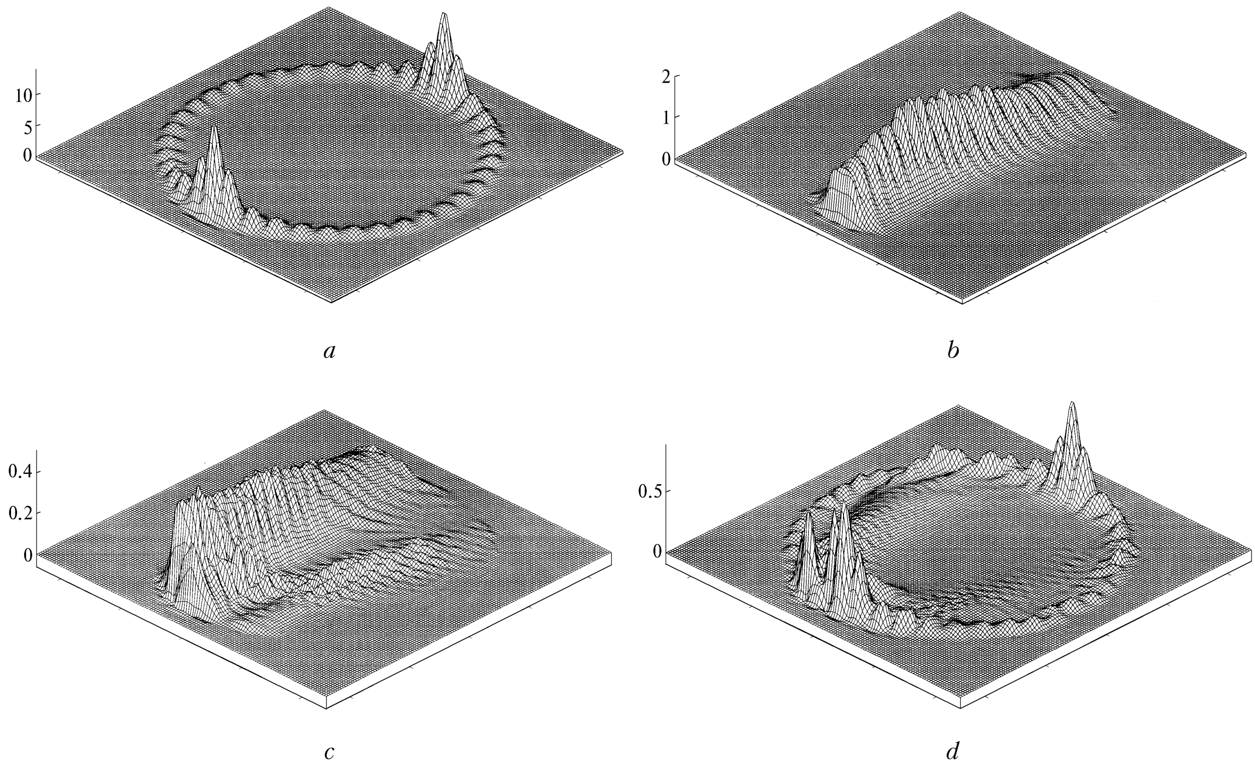


Fig. 1. Spatial distribution of the relative intensity of optical field inside a water drop (x_a = 49.8983) at its illumination by a plane wave (*a*) with the wavelength λ = 0.65 μm and by a Gaussian beam (*b*–*d*) with the parameter w_0/a_0 = 0.5 as a function of the beam position relative to the particle center y_0/a_0 = 0 (*b*), 0.5 (*c*), and 1.12 (*d*).

In this connection, when considering the resonance excitation of optical fields in a particle by a focused beams, it is necessary to take into account the spatial profile of the light beam and the geometry of its incidence in addition to the diffraction parameter and the refractive index of the particulate matter. This paper presents a theoretical investigation of the effect of these parameters on the efficiency of excitation of the resonance in spherical particles.

The paper is organized as follows. The principles of description of the electromagnetic field of Gaussian–Hermitian focused beams are briefly considered in the first part that is a review, following the original papers.[4,9–14] The relationships for amplitudes of spherical components of the electric vector of the internal optical field in a spherical particle at its illumination by a focused beam are also presented here. The second part of the paper is aimed at the study of the efficiency of excitation of the resonance of the internal optical field in the particle as a function of type and characteristics of the beam. Analytical formulas for the parameter of the beam axis displacement and its half-width providing the optimal conditions for transformation of the pumping radiation energy into the field of oscillation modes of a particle are derived based on the beam shape coefficients.[13]

1. Peculiarities of distribution of the internal optical field of particles at illumination of them by focused Gaussian beams

As known, the classical Mie theory is used for describing diffraction of a plane electromagnetic wave on a dielectric sphere. In the case of not plane but spatially limited light beams with an arbitrary distribution of the intensity over its cross section one can also use the results of this theory, if preliminary generalized to this class of beams. Many investigations were devoted to this problem, among which we should note Refs. 7 and 8. The central point of the generalized Mie theory is representation of the electromagnetic field of a light beam incident on the particle in the form of series expansion over partial waves (spherical harmonics) analogous to how it is done in the case with a plane wave. As a result, two sets of coefficients appear, $(g_n^m)_{TE}$ and $(g_n^m)_{TH}$, which describe the amplitude and phase of each partial wave and are called the beam shape coefficients (BSC) for the partial waves of TE and TH polarization, respectively.[8] These coefficients do not depend on spatial coordinates and also are determined by the specific profile of the beam and geometry of its incidence on the particle.

Let us consider briefly the description of the field of a focused Gaussian beam using Davis terminology.[9,10] Let us introduce the coordinate system $(x'y'z')$ the origin of which is at the center of the beam caustic of the half-width w_0 (Fig. 2). Let us suppose that a linearly polarized (along x axis) Gaussian beam propagates along the z' axis. The second coordinate system (xyz) is usually related to the center of a spherical particle and is used for the series expansion over partial waves. Position of the origin of coordinates $(x'y'z')$ with respect to the origin of the coordinate system (xyz) is set by the coordinates (x_0, y_0, z_0). To simplify the derivations, let us consider the case when $x_0 = y_0 = z_0 = 0$. Generalization to the case of an arbitrary x_0, y_0, z_0 is obvious but leads to rather cumbersome relationships.

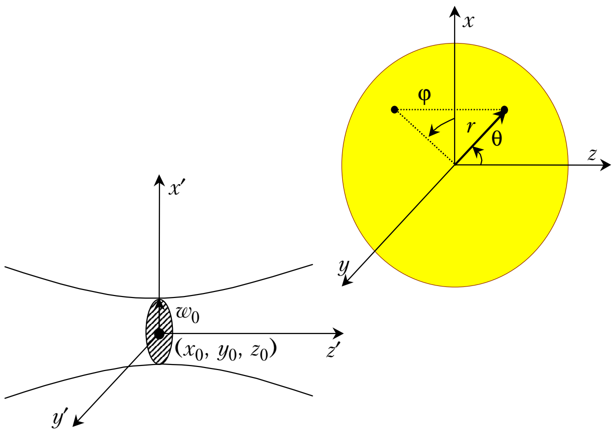


Fig. 2. Coordinate systems in the problem of diffraction of a focused light beam on a spherical particle.

Let us describe the polarized laser beam in the coordinate system (xyz) by the vector potential $\mathbf{A} = (A_x, 0, 0)$ where the non-zero component A_x is determined by the following formula:

$$A_x = \Psi(x, y, z) \exp(-ikz). \quad (1)$$

The function $\Psi(x, y, z)$ should satisfy, by definition, the Helmholtz equation

$$\nabla^2 \mathbf{A}(x, y, z) + k^2 \mathbf{A}(x, y, z) = 0. \quad (2)$$

Obviously, such a description of the beam field requires differentiation with respect to all coordinates. However, the scale of variation of the coordinates x and y is small as compared with the scale of variation in the lengthwise direction z, which is related to the diffraction length $l_z = kw_0^2$. So it is necessary to introduce the dimensionless coordinates (ξ, η, ζ) defined as

$$\xi = x/w_0; \quad \eta = y/w_0; \quad \zeta = z/l_z.$$

Then, substituting expression (1) into Eq. (2) we obtain the equation for the function $\Psi(\xi, \eta, \zeta)$:

$$\left(\frac{\partial^2}{\partial \xi^2} + \frac{\partial^2}{\partial \eta^2}\right)\Psi(\xi, \eta, \zeta) - 2i\frac{\partial \Psi(\xi, \eta, \zeta)}{\partial \zeta} + s^2 \frac{\partial^2 \Psi(\xi, \eta, \zeta)}{\partial \zeta^2} = 0. \quad (3)$$

The small parameter s is introduced in this equation, which has the meaning of the dimensionless parameter of the beam focusing:

$$s = w_0/l_z = 1/(kw_0). \quad (4)$$

This parameter is equal to zero for a plane wave ($w_0 \to \infty$), and usually $s \sim 10^{-2}$–10^{-3} for actual beams.

To solve Eq. (3) the function $\Psi(\xi, \eta, \zeta)$ is expanded into a series over the small parameter s^2:

$$\Psi(\xi, \eta, \zeta) = \Psi_0(\xi, \eta, \zeta) + s^2\,\Psi_2(\xi, \eta, \zeta) + s^4\,\Psi_4(\xi, \eta, \zeta) + \ldots\,. \quad (5)$$

The first term in Eq. (5) describes the solution for the fundamental mode of a Gaussian beam TEM_{00}:

$$\Psi_0(\xi, \eta, \zeta) = iQ_\zeta \exp[-iQ_\zeta(\xi^2 + \eta^2)], \quad (6)$$

where $Q_\zeta = (i + 2\zeta)^{-1}$; $|Q_\zeta|$ is the dimensionless beam width.

If the function Ψ_0 is known, the high order corrections Ψ_{2n} can be determined by substitution of Eq. (6) into the initial equation (3):

$$\left(\frac{\partial^2}{\partial \xi^2} + \frac{\partial^2}{\partial \eta^2} - 2i\frac{\partial}{\partial \zeta}\right)\Psi_{2n+2}(\xi, \eta, \zeta) = -\frac{\partial^2 \Psi_{2n}(\xi, \eta, \zeta)}{\partial \zeta^2}; \quad n \ge 0. \quad (7)$$

Electric **E** and magnetic **H** vectors of the field are related to the potential **A** by obvious relationships:

$$\mathbf{E} = \frac{-ic}{k}\nabla(\nabla\mathbf{A}) - i\omega\mathbf{A}; \quad \mathbf{H} = 1/\mu(\nabla\times\mathbf{A}).$$

These formulas allow one to classify the beams. The beam of nth order (by Davis) is obtained from Eq. (7) by omitting all terms, the order of which is higher than s^n. Thus, the field of the first order beam depends only on Ψ_0, the beam of the third order depends on Ψ_0 and Ψ_2, and so on. Let us note that, generally speaking, none of the beams does not exactly satisfy Maxwell equations, only at $n \to \infty$ or $s \to \infty$.

Following the standard procedure of Mie theory, let us represent the scalar function $\Psi(\xi, \eta, \zeta)$ in the form of expansion over partial waves in the spherical coordinate system[11]:

$$\Psi_{TE}(r, \theta, \varphi) = -\frac{-E_0}{kr} \times \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} (g_n^m)_{TE}\,\Psi_n(kr)\,P_n^m(\cos\theta)\exp(im\varphi);$$

$$\Psi_{TH}(r,\ \theta,\ \varphi) = -\frac{-E_0}{kr}\times$$

$$\times \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} (g_n^m)_{TH} \Psi_n(kr)\, P_n^m(\cos\theta) \exp(im\varphi). \quad (8)$$

Here $P_n^m(\cos\theta)$ are the Legendre polynomials and the shape coefficients $(g_n^m)_{TE}$ and $(g_n^m)_{TH}$ can be determined as 2D integrals of the radial components E_r and H_r of the initial beam field:

$$(g_n^m)_{TE} = -\frac{1}{4\pi}(i^{n-1})\frac{(kr)^2}{\Psi_n(kr)}\frac{(n-|m|)!}{(n-|m|)!}\times$$

$$\times\int_0^{\pi} \sin\theta\, d\theta \int_0^{2\pi} d\varphi\, P_n^m(\cos\theta)\exp(-im\varphi)\frac{cH_r(r,\theta,\varphi)}{nH_0};$$

$$(g_n^m)_{TH} = -\frac{1}{4\pi}(i^{n-1})\frac{(kr)^2}{\Psi_n(kr)}\frac{(n-|m|)!}{(n-|m|)!}\times$$

$$\times\int_0^{\pi} \sin\theta\, d\theta \int_0^{2\pi} d\varphi\, P_n^m(\cos\theta)\exp(-im\varphi)\frac{cE_r(r,\ \theta,\varphi)}{nE_0}. \quad (9)$$

Radial components of electromagnetic field are related to its Cartesian coordinates by the known relationship:

$$E_r = E_x \sin\theta\cos\varphi + E_y \sin\theta\sin\varphi + E_z \cos\theta.$$

Thus, for example, we obtain for the beam of the first order (in Davies approximation) at arbitrary $(x_0,\ y_0,\ z_0)$, according to Eq. (6):

$$E_r = E_0\ iQ_\zeta \exp(ikz_0)\exp(-\ ikr\cos\theta)\times$$

$$\times\exp[-\ (skr\sin\theta)^2\ iQ_\zeta]\exp[-\ iQ_\zeta(\xi_0^2+\eta_0^2)]\times$$

$$\times\exp[2s\ iQ_\zeta\ kr\sin\theta\ (\xi_0\cos\varphi+\eta_0\sin\varphi)]\times$$

$$\times\{\sin\theta\cos\varphi\ (1-2s^2kr\ Q_\zeta\cos\theta)+2s\ \xi_0\ Q_\zeta\cos\theta\}, \quad (10)$$

where $\xi_0 = x_0/w_0$; $\eta_0 = y_0/w_0$; $\zeta_0 = z_0/l_z$. The expression has analogous form if one replaces $\cos\varphi$ by $\sin\varphi$ and ξ_0 by η_0 in the last row of Eq. (10).

Supposing that the BSC have been determined with the required accuracy, the expansions of the spherical components of the initial light beam follow from Eq. (1) and (8). For example, for the electric field

$$E_r = -\frac{iE_0}{(kr)^2}\sum_{n=1}^{\infty}(-i)^n\ (2n+1)\ \Psi_n(kr)\times$$

$$\times\sum_{m=-n}^{n}(g_n^m)_{TH}\ \pi_n^{|m|}(\theta)\exp(im\varphi);$$

$$E_\theta = -\frac{E_0}{kr}\sum_{n=1}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}\times$$

$$\times\left\{\Psi_n(kr)\sum_{m=-n}^{n}(g_n^m)_{TE}\ im\ \pi_n^{|m|}(\theta)\exp(im\varphi)+\right.$$

$$\left.+\ \Psi_n'(kr)\sum_{m=-n}^{n}(g_n^m)_{TH}\ \tau_n^{|m|}(\theta)\exp(im\varphi)\right\};$$

$$E_\varphi = -\frac{E_0}{kr}\sum_{n=1}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}\times$$

$$\times\left\{-\ \Psi_n(kr)\sum_{m=-n}^{n}(g_n^m)_{TE}\ \tau_n^{|m|}(\theta)\exp(im\varphi)+\right.$$

$$\left.+\ i\ \Psi_n'(kr)\sum_{m=-n}^{n}(g_n^m)_{TH}\ im\ \pi_n^{|m|}(\theta)\exp(im\varphi)\right\}. \quad (11)$$

The angular functions $\pi_n^{|m|}$ and $\tau_n^{|m|}$ are defined as

$$\pi_n^{|m|}(\theta) = \frac{1}{\sin\theta}P_n^{|m|}(\cos\theta);\quad \tau_n^{|m|}(\theta) = \frac{d}{d\theta}P_n^{|m|}(\cos\theta).$$

Calculation of BSC $(g_n^m)_{TE}$ and $(g_n^m)_{TH}$ for the specific type of beam is a separate problem and was considered, for example, in Refs. 8, 12–14. Historically first method for calculating the coefficients was the method of direct integration of Eq. (9) using the quadrature formulas. The method of finite series[8] and the methods based on the Van de Hulst localization principle for different classes of beams[13,14] were used later. One should consider in detail one of them, the method of integral localized approximation,[14] because, in our opinion, it is the most flexible and effective from the standpoint of numerical calculations. We use it in our studies of the efficiency of excitation of resonance.

Following the extended treatment of the principle of localization,[5] the light beam incident on a spherical particle with the impact parameter r can be replaced by a partial wave with the number n so that the translation $kr \to n + 1/2$; $\theta \to \pi/2$; $\exp(-\ ikr\cos\theta) \to 1$ is correct. Then, if we introduce the so-called operator of localization $\mathfrak{I}$ as

$$\mathfrak{I}[f(kr;\ \theta)] = f(n + 1/2;\ \pi/2), \quad (12)$$

and apply it to the formulas for the radial components E_r and H_r of the Gaussian beam field, the corresponding formulas for BSC g_n^m take the form:

$$(g_n^m)_{TE} = -\frac{Z_n^m}{2\pi H_0}\int_0^{2\pi} d\varphi\ \mathfrak{I}[H_r(r,\theta,\varphi)]\exp(-\ im\varphi);$$

$$(g_n^m)_{TH} = -\frac{Z_n^m}{2\pi H_0}\int_0^{2\pi} d\varphi\ \mathfrak{I}[E_r(r,\theta,\varphi)]\exp(-\ im\varphi), \quad (13)$$

where $Z_n^0 = i2n(n+1)/(2n+1)$; $Z_n^m = [-2i/(2n+1)]^{|m|-1}$, $m \neq 0$. Thus, to calculate the sets of BSC it is necessary to know the formulas for radial components of the beam field only in the focal plane $z = 0$.

It follows from Eq. (13) that all coefficients g_n^m for a plane wave (linearly polarized along the x axis) are equal to zero, except $(g_n^{\pm 1})_{\rm TE} = \mp(i/2)$ and $(g_n^{\pm 1})_{\rm TH} = 1/2$, because $E_r = E_x \sin\theta \cos\varphi$ and $H_r = H_y \sin\theta \sin\varphi$. Then for a Gaussian beam of the first order of approximation we obtain:

$$\Im[E_r(r,\theta,\varphi)] = E_0 = iQ_\zeta \exp(ikz_0) \times$$

$$\times \exp[-iQ_\zeta(\xi_0^2 + \eta_0^2)] \exp[-iQ_\zeta s^2(n+1/2)^2] \times$$

$$\times \exp[2s(n+1/2)\, iQ_\zeta(\xi_0 \cos\varphi + \eta_0 \sin\varphi)] \cos\varphi;$$

$$(g_n^m)_{\rm TH} = 1/2\,(-i)^{|m|-1}\, s^{|m|-1} \exp[-(\xi_0^2 + \eta_0^2)] \times$$

$$\times \exp\,(i\,(k\,w_0)^2 \zeta_0)\, \frac{(\xi_0 - i\eta_0)^{|m|-1}}{(m-1)!} \times$$

$$\times \{1 - 2iskw_0\zeta_0[m - (\xi_0^2 + \eta_0^2)]\}\,. \qquad (14)$$

The relationships of reciprocity between the coefficients are valid[11]:

$$(g_n^{-m}(\xi_0, \eta_0, \zeta_0))_{\rm TH} = [g_n^m\,(\xi_0, -\eta_0, \zeta_0)]_{\rm TH};$$

$$(g_n^{-m}(\xi_0, \eta_0, \zeta_0))_{\rm TE} = -\,[g_n^m(\xi_0, -\eta_0, \zeta_0)]_{\rm TE};$$

$$(g_n^m(\xi_0, \eta_0, \zeta_0))_{\rm TE} = (-i)^m\,[g_n^m(\eta_0, -\xi_0, \zeta_0)]_{\rm TH}, \quad m \geq 0.$$

The majority of the results presented above are related to the fundamental mode of the Gaussian beam $\rm TEM_{00}$. At the same time, it seems to be important to consider the principles of theoretical description of a more wide class of Gaussian–Hermit beams of high order, denoted in literature as TEM_{nm} with generally arbitrary set of indices n and m. It was shown[7,15] that electromagnetic field of such beams can be described based on the $\rm TEM_{00}$ mode field by means of cross differentiation with respect to the coordinates, i.e., in the following symbolic form:

$$\mathrm{TEM}_{nm} = \frac{\partial^n \partial^m [\mathrm{TEM}_{00}]}{\partial\xi^n \partial\eta^m}\,. \qquad (15)$$

Following this procedure, we obtain for the $\mathrm{TEM}_{10}^{(x)}$ mode of the Gaussian beam polarized along the x axis:

$$E_\zeta^{10} = -\,2iQ_\zeta\,(\xi - \xi_0)\,E_\zeta^{00};$$

$$E_\zeta^{10} = -\,2Q_\zeta\, s[1 - 2iQ_\zeta\,(\xi - \xi_0)^2]\,E_\zeta^{00};$$

$$H_\eta^{10} = -\,2iQ_\zeta\,(\xi - \xi_0)\,H_\eta^{00};$$

$$H_\zeta^{10} = -\,4iQ_\zeta^2\,(\xi - \xi_0)\,(\eta - \eta_0)\,H_\eta^{00}. \qquad (16)$$

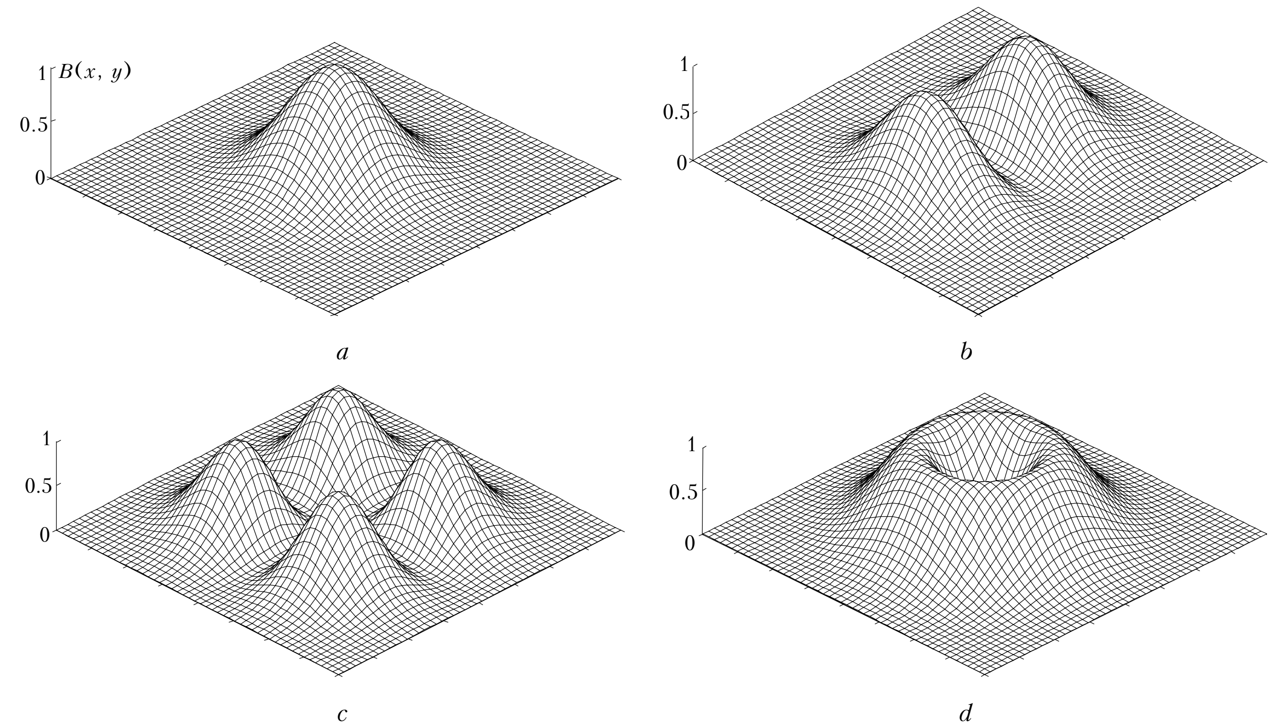


Fig. 3. Spatial distribution of the normalized intensity (in the focal plane $z = 0$) of the light beams $\rm TEM_{00}$ (a), $\rm TEM_{10}$ (b), $\rm TEM_{11}$ (c), and $\mathrm{TEM}_{dn}^{(\mathrm{hel})}$ (d).

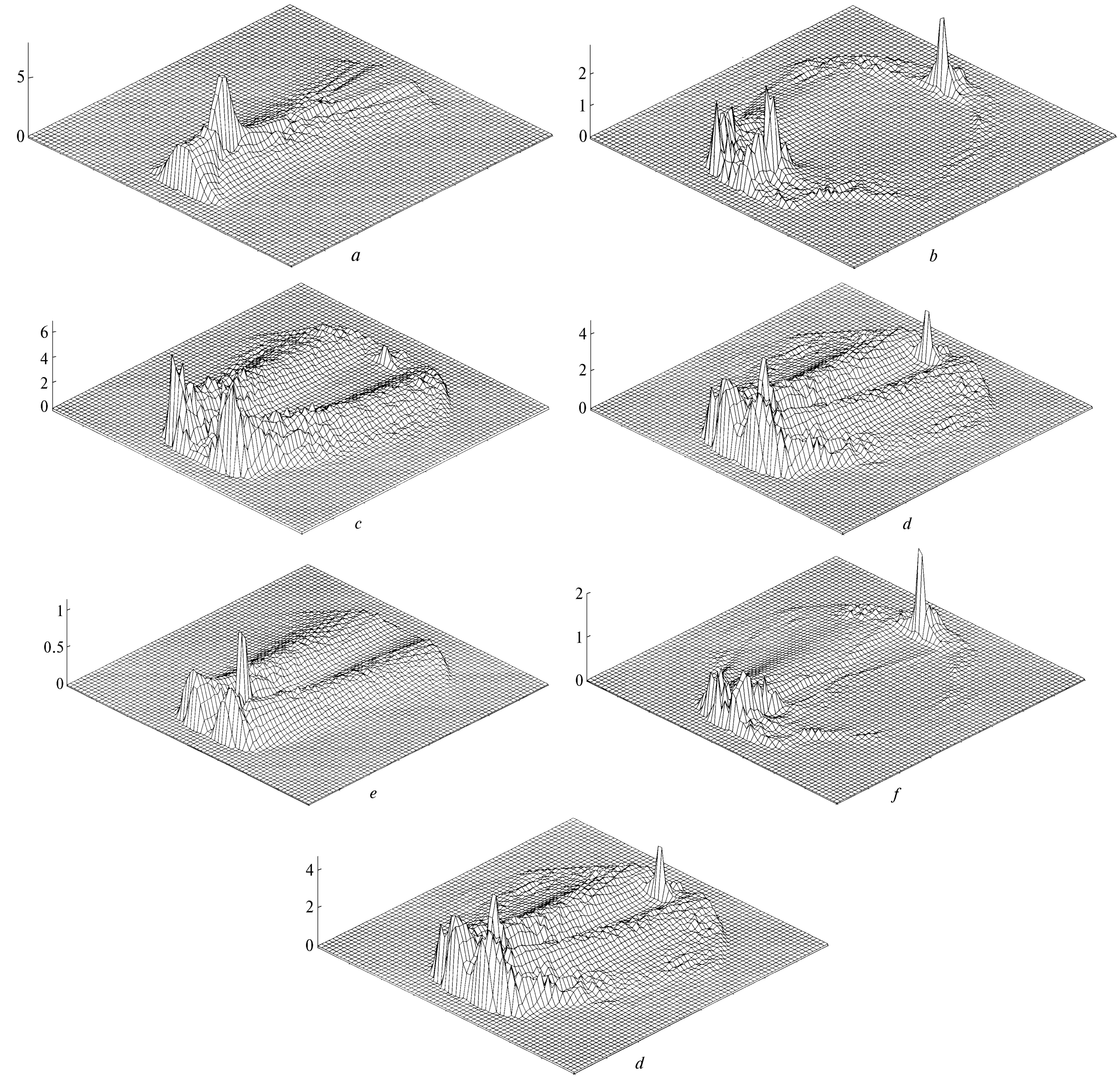


Fig. 4. Distribution of the factor of the light field inhomogeneity B in the YZ plane inside water drop with the radius of 10 μm at illumination of it by focused beam ($\xi_0 = \zeta_0 = 0$) with $\lambda = 0.532$ μm: TEM_{00}, $\eta_0 = 0$ (*a*); TEM_{00}, $\eta_0 = 0.8$ (*b*); TEM_{11}, $\eta_0 = 0$ (*c*); TEM_{11}, $\eta_0 = 0.3$ (*d*); $\mathrm{TEM}_{dn}^{(\mathrm{hel})}$, $\eta_0 = 0$ (*e*); and $\mathrm{TEM}_{dn}^{(\mathrm{hel})}$, $\eta_0 = 0.3$ (*f*).

Correspondingly, for the ring beam with circular polarization $\mathrm{TEM}_{dn}^{(\mathrm{hel})} = (\mathrm{TEM}_{10}^{(x)} + i\,\mathrm{TEM}_{01}^{(x)})/\sqrt{2}$:

$$E_\xi^{dn} = H_\eta^{dn} = -\sqrt{2}Q_\zeta[i(\xi - \xi_0) - (\eta - \eta_0)]\, E_\xi^{00};$$

$$E_\zeta^{dn} = -\sqrt{2}Q_\zeta\, s[1 - 2iQ_\zeta(\xi - \xi_0)^2 +$$

$$+ 2Q_\zeta(\xi - \xi_0)\,(\eta - \eta_0)]\, E_\xi^{00};$$

$$H_\zeta^{dn} = i\,\sqrt{2}Q_\zeta\, s[2Q_\zeta(\xi - \xi_0)\,(\eta - \eta_0) +$$

$$+ 2iQ_\zeta(\eta - \eta_0)^2 - 1]\, H_\eta^{00}. \qquad (17)$$

The components of the fundamental mode field E_ξ^{00} and H_η^{00} in Eqs. (16)–(17) are equal to:

$$E_\xi^{00} = H_\eta^{00} = E_0\, iQ_\zeta \exp\,(ikz_0)\, \times$$

$$\times \exp\,\{-iQ_\zeta\,[(\xi - \xi_0)^2 + (\eta - \eta_0)^2]\}.$$

The distribution of the normalized intensity of the beams TEM_{00}, TEM_{10}, TEM_{11}, and $\mathrm{TEM}_{dn}^{(\mathrm{hel})}$ in the cross section $z = 0$ is shown in Fig. 3. They are normalized to the value equal to the intensity of plane wave in vacuum ($cE_0^2/8\pi$).

Let us return to the problem of scattering of light wave on a spherical particle. The aforementioned reasoning show that the components of the internal electric field inside the particle in the case of incidence of a spatially limited focused beam on the particle can be written as follows:

$$E_r = -\frac{iE_0}{(kr)^2}\sum_{n=1}^{\infty} n(n+1)\,\Psi_n(kr)\sum_{m=-n}^{n} c_n^m\,\pi_n^{|m|}(\theta)\exp(im\varphi),$$

$$E_\theta = -\frac{E_0}{kr}\sum_{n=1}^{\infty}\{\Psi_n(kr)\sum_{m=-n}^{n} d_n^m\, im\,\pi_n^{|m|}(\theta)\exp(im\varphi) +$$

$$+\, i\,\Psi_n'(kr)\sum_{m=-n}^{n} c_n^m\,\tau_n^{|m|}(\theta)\exp(im\varphi)\};$$

$$E_\varphi = -\frac{E_0}{kr}\sum_{n=1}^{\infty}\{-\,\Psi_n(kr)\sum_{m=-n}^{n} d_n^m\,\tau_n^{|m|}(\theta)\exp(im\varphi) +$$

$$+\, i\,\Psi_n'(kr)\sum_{m=-n}^{n} c_n^m\, im\,\pi_n^{|m|}(\theta)\exp(im\varphi)\}. \quad (18)$$

As is seen, it is the generalized analog of the relevant formula for plane wave taking into account the modification of amplitude coefficients c_n and d_n by the beam shape coefficients:

$$c_n^m = c_n\,(g_n^m)_{\mathrm{TH}};\quad d_n^m = d_n\,(g_n^m)_{\mathrm{TE}}.$$

Some examples of numerical calculations of the relative intensity of the internal optical field in water drops illuminated by Gaussian beams are shown in Figs. 4*a–d*.

3. Efficiency of excitation of resonance of the internal field

Let us judge on the efficiency of excitation of resonance in particle from the value of mean energy of the internal optical field W_i accumulated during one period. As known, the formula for it is as follows:

$$W_i = \frac{1}{8\pi}\int_{V_a}\varepsilon_a\,\mathbf{E}_i\,\mathbf{E}_i^*\,\mathrm{d}\mathbf{r}' = \frac{E_0^2\,\varepsilon_a}{8\pi}\int_{V_a} B_i(\mathbf{r}')\,\mathrm{d}\mathbf{r}'\,, \quad (19)$$

where integration is made over the particle volume. Let us use the expansions of the internal optical field over partial waves and, for certainty, let us consider only the TE_{nm} modes. Then

$$B_i(\mathbf{r}) = \frac{1}{E_0^2}\,[E_\theta(\mathbf{r})\,E_\theta^*(\mathbf{r}) + E_\varphi(\mathbf{r})\,E_\varphi^*(\mathbf{r})] =$$

$$= \frac{1}{(k_a r)^2}\,|d_n^m|^2\,|\Psi_n(k_a r)|^2\sum_{m=-n}^{n}\sum_{m_1=-n}^{n}(g_n^m)_{\mathrm{TE}}\,(g_n^{m_1})^*_{\mathrm{TE}}\,\times$$

$$\times\exp[i\,(m-m_1)\varphi]\{mm_1\pi_n^{|m|}(\theta)\pi_n^{|m_1|}(\theta) + \tau_n^{|m|}(\theta)\tau_n^{|m_1|}(\theta)\}.$$

Integrating with respect to spherical coordinates and taking into account mutual orthogonality of the angular functions $\pi_n^{|m|}(\theta)$, $\tau_n^{|m|}(\theta)$, and $\exp(im\varphi)$, we obtain for the accumulated energy the following expression:

$$W_i = \frac{E_0^2\,a_0\,\varepsilon_a}{8k_a^2}\,\frac{2n(n+1)}{2n+1}\,\times$$

$$\times\,|d_n^m|^2\,\{|\Psi_n(k_a a_0)|^2 + |\Psi_{n+1}(k_a a_0)|^2 -$$

$$-\,\frac{2n+1}{k_a a_0}\,\Psi_n(k_a a_0)\,\Psi_{n+1}^*(k_a a_0)\}\,\times$$

$$\times\sum_{m=-n}^{n}|(g_n^m)_{\mathrm{TE}}|^2\,\frac{(n+|m|)!}{(n-|m|)!}\,. \quad (20)$$

Similarly, for the TH_{nm} modes we have:

$$W_i = \frac{E_0^2\,a_0\,\varepsilon_a}{8k_a^2}\,\frac{2n(n+1)}{2n+1}\,\times$$

$$\times\,|c_n^m|^2\,\{|\Psi_n(k_a a_0)|^2 + |\Psi_{n+1}(k_a a_0)|^2 +$$

$$+\,\frac{2(n+1)}{(k_a a_0)^2}|\Psi_n(k_a a_0)|^2 - \frac{2n+3}{k_a a_0}\Psi_n(k_a a_0)\,\Psi_{n+1}^*(k_a a_0)\}\,\times$$

$$\times\sum_{m=-n}^{n}|(g_n^m)_{\mathrm{TE}}|^2\,\frac{(n+|m|)!}{(n-|m|)!}\,. \quad (21)$$

As is seen from Eqs. (20)–(21), the dependence of the amount of accumulated light energy on the beam type and geometry of its incidence on the particle is completely presented by the factor

$$K_n = \sum_{m=-n}^{n}|(g_n^m)|^2\,\frac{(n+|m|)!}{(n-|m|)!} \quad (22)$$

and is determined only by the sum of the series over BSC. Using Eq. (13), we obtain for the incidence of plane wave $(s \to 0)$ on aspherical particle that in this case the factor K_n takes its maximum value equal to

$$K_n(s \to 0) = 1/2\; n(n+1). \quad (23)$$

It is the evidence of the fact that the beams that are appropriate for excitation of corresponding resonance configurations of the optical field are always present in a plane wave, because its width is infinite. On the other hand, if one estimates this process from the standpoint of energy loss from the exciting beam, then, obviously, plane wave has the smallest efficiency just because of its infinite width. So let us consider only spatially limited light beams.

Let us perform theoretical analysis assuming that $\xi_0 = \zeta_0 = 0$ and arbitrary η_0 and w_0. Thus, it is prescribed that the beam axis moves relative to the particle center along the coordinate y (see Fig. 2), the width of the light beam also can vary. For certainty, let us study excitation of only the TE_{nm} resonance modes in a particle.

Let us first consider the case of a Gaussian beam and the fundamental mode TEM_{00}. According to Eqs. (13) and (14), one can write

$$\Im[H_r(r,\theta,\varphi)] \sim \exp(-iQ_\zeta\eta_0^2)\times$$

$$\times\exp[2s(n+1/2)\, iQ_\zeta\eta_0 \sin\varphi]\sin\varphi\,;$$

$$(g_n^m)_{\mathrm{TE}} = -\frac{Z_n^m}{2\pi H_0}\int_0^{2\pi} \mathrm{d}\varphi\, \Im[H_r(r,\theta,\varphi)]\exp(-im\varphi) \sim$$

$$\sim\frac{1}{2i}\exp(-iQ_\zeta\eta_0^2)\left\{\int_0^{2\pi}\mathrm{d}\varphi\exp[iP_\eta\sin\varphi - i\varphi(m-1)] -\right.$$

$$\left.-\int_0^{2\pi}\mathrm{d}\varphi\exp[iP_\eta\sin\varphi - i\varphi(m+1)]\right\}, \qquad (24)$$

where $P_\eta = 2s(n+1/2)\, iQ_\zeta\eta_0$. Noting that

$$\int_0^{2\pi}\mathrm{d}\varphi\exp[iP_\eta\sin\varphi - i\varphi(m-1)] =$$

$$= 2\pi(-i)^{m-1} I_{m-1}(iP_\eta),$$

where $I_m(z)$ is the modified Bessel function, let us rewrite Eq. (24) in the form

$$(g_n^m)_{\mathrm{TE}} \sim \exp(-\eta_0^2)\,[I_{m-1}(R_\eta) + I_{m+1}(R_\eta)];$$

$$R_\eta = iP_\eta. \qquad (25)$$

It is necessary to study the right-hand side of this formula to reveal an extremum in its dependence on η_0.

Let us use asymptotic representation of the function $I_m(R_\eta)$ for not very wide beams, i.e., when the conditions Re $\{R_\eta\} > 12$ has been satisfied[16]:

$$I_m(R_\eta) \approx \frac{\exp(R_\eta)}{(2\pi R_\eta)^{1/2}}\times$$

$$\times\left\{1+\sum_{k=1}^{m+2}\frac{(-1)^k}{k!(8R_\eta)^k}(4m^2-1)(4m^2-9)\ldots[4m^2-(2k-1)]\right\},$$

$$\mathrm{Re}\,\{R_\eta\} > m. \qquad (26)$$

Only the factor before braces in Eq. (26) depends on $R_\eta(\eta_0)$, which is the same for both terms in Eq. (25). So, let us substitute Eq. (26) into Eq. (25) omitting the factors independent of R_η, and use the representation for P_η again:

$$|(g_n^m)_{\mathrm{TE}}|^2 \sim \frac{\exp\{2\eta_0[2s(n+1/2)-\eta_0]\}}{4\pi s(n+1/2)\eta_0}.$$

Simple analysis shows that this function reaches the maximum value at the axis displacement value $\eta_0 = s(n+1/2) \equiv (\eta_0)_{00}$ or, in absolute coordinates:

$$\left(\frac{y_0}{a_0}\right)_{00} = \frac{(n+1/2)}{x_a}. \qquad (27)$$

As the inequality $n > x_a$ is always satisfied for optical resonators with high Q-factor, it is obvious that the beam axis should be outside the particle for the most effective excitation of the resonance field inside the particle. In spite of paradoxicality of this result from the standpoint of geometrical optics, it is the direct consequence of the Van de Hulst localization principle considered above and is an evidence of the fact that the resonance mode field is formed not by the central part of the light beam, but by the field of its edge part decreasing with distance, which forms the subset of geometrical beams incident at grazing angles to the particle surface.

The normalized factor $\bar{K}_n$ is shown in Fig. 5 in relative coordinates as a function of the value of the displacement parameter y_0/a_0 of a Gaussian beam axis (TEM_{00}, $\lambda = 0.65$ μm) for three resonance modes in a water drop: TE_{60}^1 ($x_a = 49.8983$), TE_{60}^2 ($x_a = 54.2559$), TE_{60}^3 ($x_a = 53.9390$). The curves are normalized to the value of the factor K_n for a plane wave (Eq. (23)).

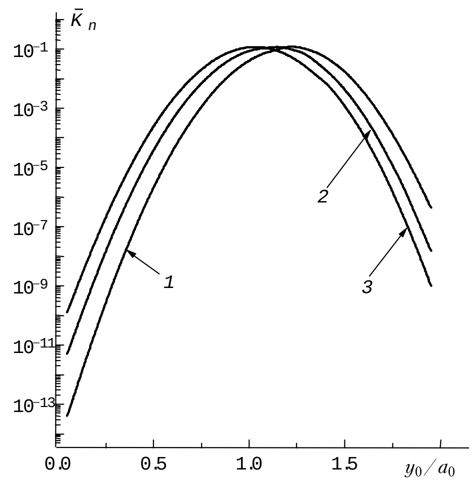


Fig. 5. The normalized factor $\bar{K}_n$ as a function of the displacement y_0/a_0 of the Gaussian beam axis (TEM_{00}, $\lambda = 0.65$) for three resonance modes in a water drop: TE_{60}^1 (*1*), TE_{60}^2 (*2*), TE_{60}^3 (*3*).

It is seen that the maximum of these dependences is reached at the values of the parameter $w_0\,\eta_0/a_0$ determined by Eq. (27). The higher is the order of the resonance mode, the closer is the position of the optimal displacement of the beam to the particle surface.

If we consider the incidence of a Gaussian beam of TEM_{00} mode on a spherical particle along the axis ($x_0 = y_0 = z_0 = 0$), then, using Eq. (14), we obtain:

$$|(g_n^m)_{\mathrm{TE}}|^2 \sim 1/4[1 - 2s^2(n-1)(n-2)], \quad m = \pm 1.$$

Corresponding dependence $K_n(y_0/a_0)$ at different values of the parameter w_0/a_0 is shown in Fig. 6. Thus, the wider the beam (smaller s), or, in other words, the closer it is to the plane wave, the more active is the excitation of resonances.

Let us consider now the ring beams TEM_{dn}. Performing the same calculations as in the previous case, we have:

$$\Im[H_r(r,\theta,\varphi)] \sim$$

$$\sim \exp(-iQ_\zeta\eta_0^2)\exp[2s(n+1/2)\,iQ_\zeta\eta_0\sin\varphi]\sin\varphi\,\times$$

$$\times\left\{is(n+1/2)\cos\varphi - s(n+1/2)\sin\varphi + \eta_0\right\};$$

$$(g_n^m)_{TE} = -\exp(-\eta_0^2)\left\{s(n+1/2)(-i)^m I_{m-1}(R_\eta) -\right.$$

$$- s(n+1/2)(-i)^{m-2} I_{m-2}(R_\eta) +$$

$$\left.+(-i)^{m-2}\eta_0[I_{m-1}(R_\eta)+I_{m+1}(R_\eta)]\right\}. \quad (28)$$

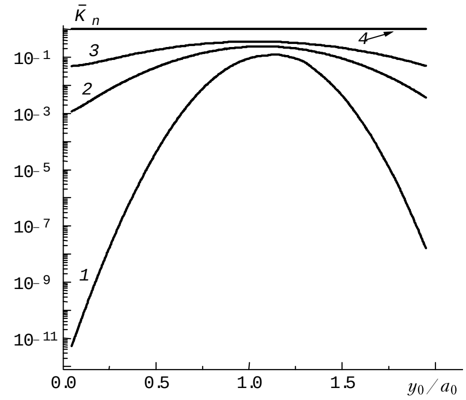


Fig. 6. The normalized factor $\bar{K}_n$ as a function of the relative displacement of the Gaussian beam y_0/a_0 axis (TEM_{00}, $\lambda = 0.65$) at excitation of the TE_{60}^2 mode in a water drop and different relative beam widths $w_0/a_0 = 0.3$ (*1*), 0.6 (*2*), 0.9 (*3*), and ∞ (*4*).

Then we take into account that, according to the asymptotic (26), at Re $\{R_\eta\} \gg 1$, $I_{m+1}(R_\eta) \gg I_m(R_\eta) \gg$ $\gg I_{m-1}(R_\eta)$.

Then, approximately

$$(g_n^m)_{TE}|^2 \sim \exp(-2\eta_0^2)\,\{\pi(-i)^{m-2}\,\eta_0\,I_{m+1}(R_\eta)\}^2 \Rightarrow$$

$$\Rightarrow \exp(-2\eta_0^2)\,\eta_0\exp[4\,\eta_0\,s(n+1/2)]\ .$$

Seeking the maximum of this expression leads to the result $\eta_0 = 0$. In other words, to obtain the best results, the ring beam should be directed toward the particle center. Intuitively, it is understandable, because the structure of the intensity distribution in the beam cross section is the ring with maximum situated at the distance of $\bar{r}_{max} = (\xi^2+\eta^2)^{1/2} = 1/\sqrt{2}$ from the particle center. So, directing the beam to the particle center and selecting its radius w_0 such that the maximum of the intensity of the ring zone is at the distance of $(y_0/a_0)_{00} = \bar{r}_{max}$, one can expect most optimal excitation of the resonance.

One can obtain the same result directly from Eq. (28) by assuming that $\eta_0 = 0$. Indeed, in this case the only non-zero beam shape coefficients are:

$$(g_n^m)_{TE} \approx s(n+1/2)\exp[s^2(n+1/2)^2]\times$$

$$\times\left\{i\int_0^{2\pi} d\varphi\,(\cos\varphi\sin\varphi)\exp(-im\varphi) -\right.$$

$$\left.-\int_0^{2\pi} d\varphi\,\sin^2\varphi\exp(-im\varphi)\right\} =$$

$$= \pi s(n+1/2)\exp[-s^2(n+1/2)^2],\quad m = \pm 2,\,0. \quad (29)$$

Varying the beam width w_0, we obtain that the coefficients (29) take their maximum values under the following condition:

$$(w_0/a_0)_{dn} = \sqrt{2}\,(n+1/2)/x_a. \quad (30)$$

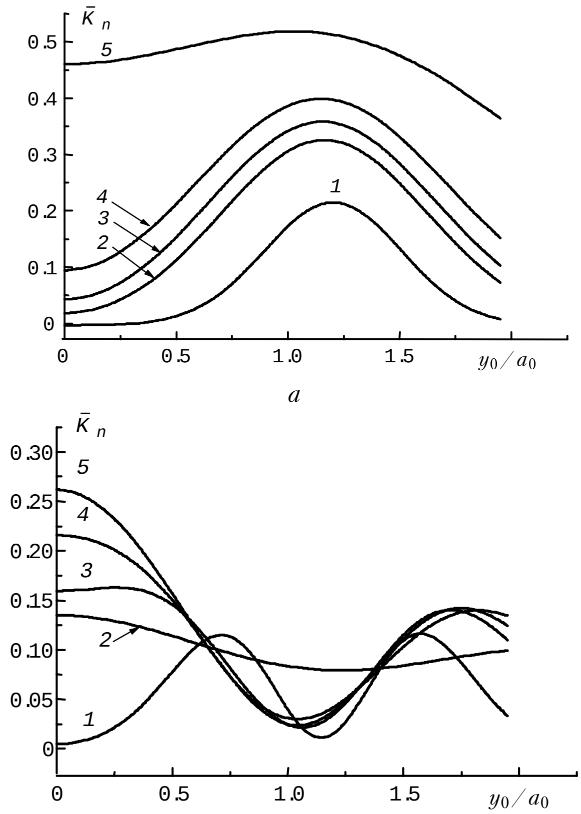
The light beams with cross structure of electromagnetic field corresponding to the TEM_{01} and TEM_{11} modes are also characterized by zero value of the intensity at the beam axis (Fig. 3*b*). Hence, it is expedient to look for a maximum of the coefficients (g_n^m) only in their dependence on w_0 ($\eta_0 = 0$).

Similarly to Eq. (28), we obtain for TEM_{01} mode:

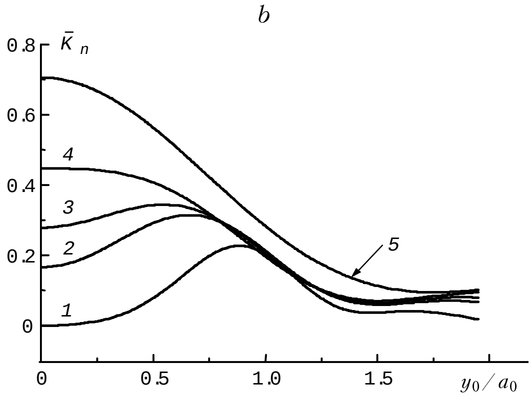
$$\Im[H_r(r,\theta,\varphi)] \sim s(n+1/2)\exp[-s^2(n+1/2)^2]\sin^2\phi;$$

$$(g_n^m)_{TE} \approx \pi s(n+1/2)\exp[-s^2(n+1/2)^2]$$

$$m = \pm 2,\,0. \quad (31)$$



b



c

Fig. 7. The coefficient $\bar{K}_n$ as a function of the relative displacement y_0/a_0 of the Gaussian beam axes of the TEM_{00} (*a*), TEM_{11} (*b*), and TEM_{dn} (*c*) modes relative to the water drop center at different values of the relative beam width $w_0/a_0 = 0.6$ (*1*), 0.9 (*2*), 1.0 (*3*), 1.24 (*3*), and 2.0 (*5*).

The maximum of coefficients (31) is reached at $(w_0/a_0)_{01} = \sqrt{2}\ (n + 1/2)/x_a$.

Correspondingly, for TEM_{11} mode:

$$\Im[H_r(r, \theta, \varphi)] \sim$$

$$\sim s^2(n + 1/2)^2 \exp[-s^2(n + 1/2)^2] \cos\varphi \sin^2\varphi;$$

$$(g_n^m)_{TE} \approx (\pi/2)s^2(n + 1/2)^2 \exp[-s^2(n + 1/2)^2]$$

$$m = \pm 1, \pm 3;$$

$$(w_0/a_0)_{11} = (n + 1/2)/x_a \qquad (32)$$

Figure 7 shows the values of the coefficient $\bar{K}_n$ reflecting, as it was mentioned above, the efficiency of excitation of resonance in a spherical particle, as a function of the relative displacement of the axes of beams of different spatial configuration and at different parameters (w_0/a_0). The calculations were made for a water drop with the radius corresponding to the resonance of TE^1_{100} mode ($x_a = 80.99428$).

Comparison of different plots in this figure shows that in all cases the change of the beam width requires careful selection of the coordinate of its incidence on the particle. The exception is only the fundamental Gaussian mode TEM_{00}. The maximum of the coefficient K_n is observed here at the same value of the beam displacement as determined by Eq. (27). Figure 8 illustrates this conclusion, showing the dependence of the coordinate of optimal displacement of the axes of light beams (y_0/a_0) on their characteristic width.

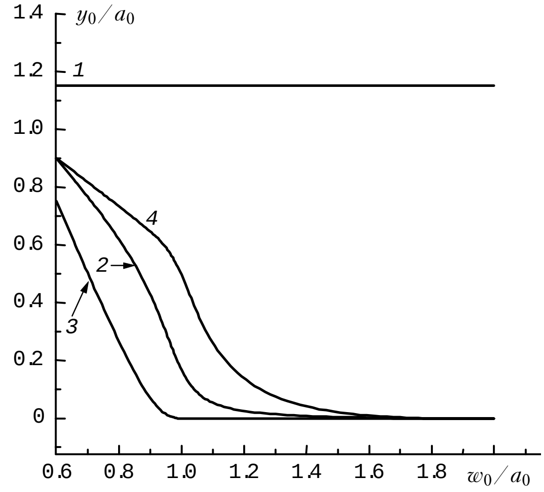


Fig. 8. Coordinate of the optimal displacement of the light beam axis y_0/a_0 as a function of their characteristics width w_0/a_0: TEM_{00} (*1*), TEM_{01} (*2*), TEM_{11} (*3*), and TEM_{dn} (*4*).

The maximum possible values of the coefficient $\bar{K}_n$ for the TE^1_{100} resonance mode, which can be reached for different types of beams, are shown in Fig. 9 as functions of their width. It follows from the figure that the best conditions for excitation of TE_{nm} resonances in spherical particles are realized when illuminating them by TEM_{01} mode of the Gaussian beam. This mode is characterized by the fact that ~ 90% of the total beam energy is concentrated in two maxima (see Fig. 2) with the half-width $d_b \approx 0.42\ w_0$ which lie in the *yz* plane, where the energy of the resonance modes TE_{nm} is predominantly concentrated. It causes the best relation of the light field of the beam with the field of the resonance modes of the particle and, hence, the high values of the coefficient $\bar{K}_n \approx 0.9$ close to the idealized case of incidence of a plane wave.

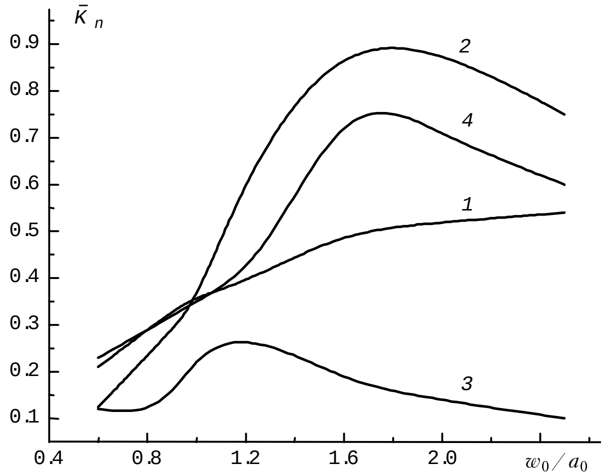


Fig. 9. Maximum possible values of the coefficient $\bar{K}_n$ for the TE^1_{100} resonance mode which can be reached for different types of beams depending on their width w_0/a_0: TEM_{00} (*1*), TEM_{01} (*2*), TEM_{11} (*3*), and TEM_{dn} (*4*).

Electromagnetic field of the ring beam TEM_{dn} is also concentrated in quite narrow zone but is uniformly distributed over the angle φ. Hence, the portion of the beam energy is not used effectively, exciting partial waves with another polarization (TH) in addition to the TE_{nm} modes. For the same reason, the low efficiency of excitation of resonances is realized for the TEM_{11} beams: $\bar{K}_n \approx 0.26$ at $w_0/a_0 = 1.24$.

Although the fundamental mode TEM_{00} of the Gaussian beam has one maximum, it is quite wide ($d_b \approx 0.84\ w_0$) that leads to the decrease of the efficiency of transforming the light energy into the internal field of the particle. Only quite wide beams can compete with the modes TEM_{01} and TEM_{dn} (see Fig. 7).

Thus, summarizing the investigations presented in this paper, we should note that for obtaining most effective excitation of electromagnetic modes in spherical particles, it is necessary to select the geometry of illuminating them according to the spatial profile of the specific beam and polarization of the excited resonance mode. In any case, the position of maxima of the spatial distribution of the pump field should be outside the particle at the distance determined by the Van de Hulst localization principle (Eq. (27)) that is unambiguously related to the value of the diffraction parameter of the particle and the number of the excited resonance mode.

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