Peculiarities of formation of the transparent spherical particle optical field under irradiation by an ultrashort amplitude-modulated spatially-limited laser beam

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Peculiarities of the optical field formation inside micron-sized weakly absorbing spherical particle under illumination by a focused Gaussian laser beam with a time regime in the form of a single monopulse and a train of ultrashort laser pulses are considered. It is established that the energy transfer efficiency of incident radiation in a selected high-quality resonance mode of a particle at its illumination by a focused light beam depends greatly on the train interpulse interval and the depth of linear frequency modulation of each pulse. The influence of irradiation geometry of a particle by laser and the pulse number in the train on the time behavior of internal optical field and its peak intensity is investigated.

Introduction

A considerable body of recently published scientific investigations, devoted to the application of micron transparent particles for laser generation,[1] optical filtration of radiation,[2] aerosol spectroscopy,[3] and optical microelectronics[4] pointed to a great interest of scientists to the unique characteristics of spherical microresonators, possessing by a wide spectrum of resonance electromagnetic modes (modes of whispering gallery WGM).

The whispering gallery modes are characterized by a very high Q factor ($>10^5$), by a narrow spectral contour, great lifetimes (of the order of nanoseconds), and a high degree of localization of the optical field close to the resonator surface. Natural frequencies of WGM are determined by the microparticle size and its optical characteristics. In this case the problem of the most effective resonance optical excitation of such microresonators is actively discussed in the literature as before now.[5–7]

It is known that for the resonance excitation of the internal optical field in a particle, it is necessary to fulfill a definite condition, under which the frequency of incident light wave should coincide with the frequency of any natural mode of a particle. In this case the space-time distribution is determined by the field of an excited mode. To increase the operation efficiency of particles-microresonators, it is necessary to create optimal conditions of excitation. This is achieved mainly by the most precise tuning to the resonance. The use of laser pulses of a supershort length of the order of pico- and femtoseconds at the cost of their wider frequency range enables one to improve greatly the tuning to the resonance, and hence, to increase the excitation efficiency of the internal particle field.[8]

A question on the effective excitation of electromagnetic resonance modes in a spherical particle was considered before, both theoretically[5] and experimentally.[6,7] Thus, in Ref. 5 the resonance excitation of internal optical field of transparent spherical microparticles was simulated numerically, when irradiating them by a train of ultrashort laser pulses. It was shown that the incident radiation can be tuned optimally to a given high quality particle resonance through variation of the train interpulse interval in combination with linear frequency modulation of each pulse (chirping).

The geometry of particle irradiation plays a key role in this process. Actually, when illuminating a spherical particle with an extended (as compared with its diameter) laser beam or by a plane wave, the only condition for obtaining resonance configurations of internal optical field is the fulfillment of a definite relationship between the value of the diffraction parameter of the particle and its refractive index. Because the width of an ideal plane wave in space is infinite, probably, the spatial harmonics can be found in its composition for excitation of appropriate modes of the optical field. However, just due to its infinite extension, the plane wave is the least efficient exciting source, since the energy of the wave is infinite. In practice,[9,10] as a rule, we deal with not plane waves but with focused beams, which focal necking can be compared or is less than the particle-microcavity size. The space structure of the optical field inside the particle in this case differs from the case of its excitation by a plane wave, especially with the use of a train of ultrashort pulses.

The above-mentioned problems, to our knowledge, were not considered in the scientific literature thus far, and therefore, the goal of this paper is the theoretical study of the time dynamics of

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the optical field inside a micron spherical particle of small absorption, illuminated by a limited in space Gaussian beam with the time mode in the form of a single monopulse and a train of ultrashort laser pulses. We will use the results of the unified Mie theory with taking into account the nonstationarity of the optical field formation in the vicinity of a particle in order to study the problem of the most efficient regimes of exciting resonance electromagnetic modes of particles by the frequency-pulse radiation at variation of its time parameters and different geometry of particle irradiation.

Structure of optical fields at irradiation of a spherical microparticle by a monochromatic spatially-limited light beam

Now we consider the basic results of the generalized Mie theory,[11,12] describing the elastic scattering of the monochromatic laser radiation on a spherical microparticle, which is a limited in space focused light beam with the Gaussian transverse intensity profile and the necking size comparable with the particle diameter.

When describing the diffraction of plane electromagnetic waves on a dielectric sphere, it is well known that the classical Mie theory is used. In the case of spatially-limited light beams with an arbitrary intensity distribution in the cross section, we can also use the results of a given theory if preliminary to generalize it to a given class of beams. The central moment of the generalized Mie theory is the representation of electromagnetic field of the incident light beam on a particle in the form of expansion by partial waves (spherical harmonics), similarly to the case of the plane wave. As a result, there appear two sets of complex coefficients $(g_n^m)_{\mathrm{TE}}$ and $(g_n^m)_{\mathrm{TH}}$, describing the amplitude and the phase of each partial wave, which are called the coefficients of the beam shape (BSC) for partial waves of TE and TH polarization, respectively.[13] The value of these coefficients does not depend on space coordinates, and is determined only by a specific beam profile and the geometry of their incidence on a particle.

In the Davis terminology (see, e.g., Refs. 11 and 14), the electromagnetic field of the focused Gaussian light beam (monochromatic radiation) is described in the form of expansion along the orthogonal system of space functions in the spherical coordinate system – vector spherical harmonics. Now we introduce the Cartesian coordinates $(x'y'z')$, centre of which is located in the middle of the focal beam waist with the halfwidth w_0 (Fig. 1).

Let us assume that a linearly polarized (along x-axis) Gaussian beam is propagated along z'-axis. The second coordinate system (xyz) is commonly associated with the centre of a spherical particle and is used for expansion by partial waves. The position of the origin of the coordinate system $(x'y'z')$ relative to the coordinate centre (xyz) is characterized by a set of coordinates (x_0, y_0, z_0).

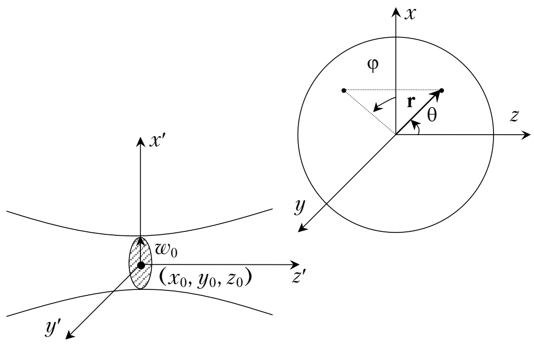

Fig. 1. Representation of disposition of coordinate systems in the problem on diffraction of a focused light beam on a spherical particle.

An expression for an internal electric field of the particle in this case represents a generalized analog of an appropriate notation for the field of a plane wave scattered on a particle[15] with taking into account the modification of the amplitude coefficients c_n, d_n by the corresponding beam shape coefficients:

$$\mathbf{E}(\mathbf{r}) = E_0 \sum_{n=0}^{\infty} \sum_{m=-n}^{n} R_n \left\{ C_{nm} \mathbf{M}_{nm}^{(1)}(\mathbf{r}) - i D_{nm} \mathbf{N}_{nm}^{(1)}(\mathbf{r}) \right\}, \quad (1)$$

where

$$R_n = i^n \frac{2n+1}{n(n+1)}, \quad \mathbf{M}_{nm}^{(1)}(\mathbf{r}), \quad \mathbf{N}_{nm}^{(1)}(\mathbf{r})$$

are spherical vector-harmonics;

$$\mathbf{r} = (\mathbf{e}_r r + \mathbf{e}_\theta \theta + \mathbf{e}_\varphi \varphi)$$

is the radius-vector in the spherical coordinate system (see Fig. 1);

$$C_{nm} = c_n (g_n^m)_{\mathrm{TH}}, \quad D_{nm} = d_n (g_n^m)_{\mathrm{TE}}$$

denotes the generalized amplitudes of partial waves connected with the Mie coefficients for a plane wave c_n, d_n (here we use designations from Ref. 15).

In its turn, the beam shape coefficients $(g_n^m)_{\mathrm{TE}}$ and $(g_n^m)_{\mathrm{TH}}$ can be found as two-dimensional integrals from radial components of electric field $E_r = (\mathbf{e}_r \cdot \mathbf{E})$ and magnetic field $H_r = (\mathbf{e}_r \cdot \mathbf{H})$ of the initial light beam:

$$(g_n^m)_{\mathrm{TE}} = -\frac{c}{4\pi E_0} (i^{n-1}) \frac{(kr)^2}{\psi_n(kr)} \frac{(n-|m|)!}{(n+|m|)!} \times$$
$$\times \int_0^{\pi} \sin\theta \mathrm{d}\theta \int_0^{2\pi} \mathrm{d}\varphi P_n^{|m|}(\cos\theta) \mathrm{e}^{-im\varphi} H_r(\theta, \varphi),$$
$$(g_n^m)_{\mathrm{TE}} = -\frac{c}{4\pi E_0} (i^{n-1}) \frac{(kr)^2}{\psi_n(kr)} \frac{(n-|m|)!}{(n+|m|)!} \times$$
$$\times \int_0^{\pi} \sin\theta \mathrm{d}\theta \int_0^{2\pi} \mathrm{d}\varphi P_n^{|m|}(\cos\theta) \mathrm{e}^{-im\varphi} E_r(\theta, \varphi), \quad (2)$$

where c is the light velocity in vacuum.

Note that the calculation of BSC for a particular type of beams presents an independent problem and is considered, e.g., in Refs. 13, 16–18. We cite the equations for BSC for the case of the incidence of a weak-focused beam of the Gaussian transverse profile on a particle, the electric field of which in the region of the focal waist is of the form:

$$\mathbf{E}(x,y,z) = E_0\mathbf{e}_x \exp\left\{-\frac{(x-x_0)^2+(y-y_0)^2}{w_0^2} - ik(z-z_0)\right\}. \tag{3}$$

In the framework of the first order (along the parameter $s = w_0/L_D$) Davis approximation[19] derive an expression for radial component of electric field of the Gaussian beam:

$$\begin{aligned} E_r &= E_0 iQ_\zeta \exp(ikz_0)\exp(-ikr\cos\theta)\times \\ &\times\exp\left(-(skr\sin\theta)^2 iQ_\zeta\right)\exp\left(-iQ_\zeta(\xi_0^2+\eta_0^2)\right)\times \\ &\times\exp\left(2siQ_\zeta kr\sin\theta(\xi_0\cos\varphi+\eta_0\sin\varphi)\right)\times \\ &\times\left[\sin\theta\cos\varphi(1-2s^2krQ_\zeta\cos\theta)+2s\xi_0 Q_\zeta\cos\theta\right]. \end{aligned} \tag{4}$$

The equation for H_r is of a similar form with the following substitutions in the last line of the Eq. (4): $\cos\varphi \to \sin\varphi$ and $\xi_0 \to \eta_0$. Then the BSC are expressed in the form

$$\begin{aligned} (g_{nm})_{\mathrm{TH}} &= \frac{1}{2}(-is)^{|m|-1}\exp\left\{-(\xi_0^2+\eta_0^2)\right\}\times \\ &\times\frac{(\xi_0-i\eta_0)^{|m|-1}}{(m-1)!}\left[1-2iskw_0\zeta_0\left(m-\left(\xi_0^2+\eta_0^2\right)\right)\right]. \end{aligned} \tag{5}$$

Here we introduce the dimensionless variables:

$$\xi_0 = x_0/w_0,\quad \eta_0 = y_0/w_0,\quad \zeta_0 = z_0/L_D;$$

$L_D = kw_0^2$ is the beam diffraction length of the radius w_0;

$$Q_\zeta^{-1} = \left(i + 2(\zeta-\zeta_0)\right).$$

In this case the reciprocal relations are true between the coefficients[19]:

$$\left(g_{nm}(\xi_0,\eta_0)\right)_{\mathrm{TE}} = (-i)^m\left(g_{nm}(\eta_0,-\xi_0)\right)_{\mathrm{TH}},\ m \ge 0;$$

$$\left(g_{n(-m)}(\xi_0,\eta_0)\right)_{\mathrm{TE}} = -\left(g_{nm}(\xi_0,-\eta_0)\right)_{\mathrm{TE}}.$$

Correspondingly, for a plane wave (linearly polarized along x axis), since $E_r = E_x\sin\theta\cos\varphi$ and $H_r = H_y\sin\theta\sin\varphi$ all BSC are equal to zero besides two pairs:

$$(g_{n(\pm1)})_{\mathrm{TE}} = 1/2 \text{ and } (g_{n(\pm1)})_{\mathrm{TH}} = \mp(i/2).$$

As an example of the use of the generalized Mie theory, figure 2 shows the space distribution of the relative intensity of the optical field calculated by Eqs. (1)–(5):

$$B(\mathbf{r}) = \left(\mathbf{E}(\mathbf{r})\cdot\mathbf{E}^*(\mathbf{r})\right)/E_0^2$$

(factor of the field inhomogeneity) in the vicinity of a clear water droplet with radius a_0 = 10 μm, when a monochromatic Gaussian beam falls on it, having the waist radius $w_0=a_0/4$ and directed to the center $\xi_0 = 0$ and to the particle edge $\xi_0 = 1$. The water refraction coefficient n_a was equal to 1.33, which at λ_0 = 800 nm gave the value of the diffraction parameter (Mie parameter) of the particle

$$x_a = 2\pi a_0/\lambda_0 \simeq 78.5.$$

The intensity profiles are given in the plane xz passing through the centre of a droplet (equatorial plane of the particle).

It is clear from Fig. 2 that the character of the spatial distribution of the optical field varies depending on the geometry of the droplet illumination by a laser beam.

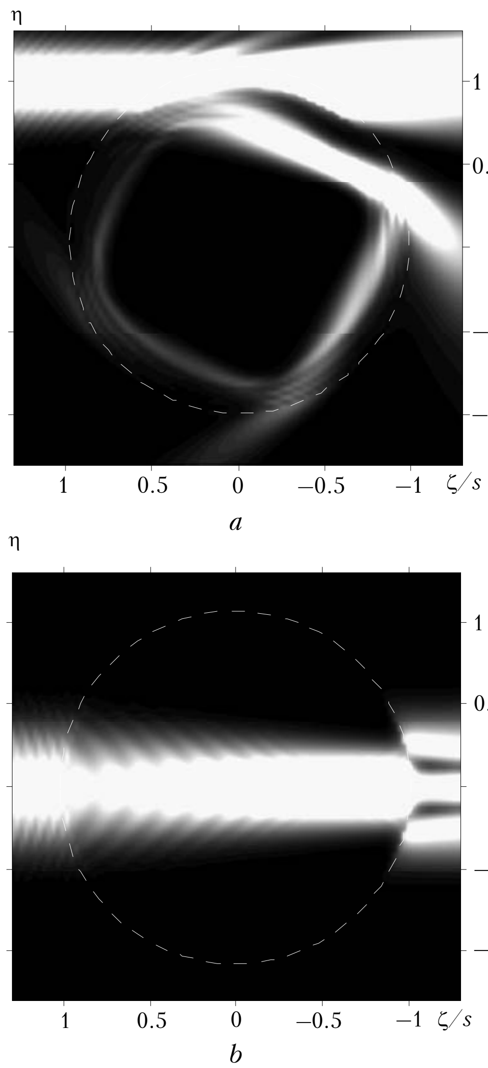

Fig. 2. Relative intensity of the optical field in the vicinity of a water droplet at the edge (*a*) and central (*b*) incidence of a narrow Gaussian light beam. The direction of radiation incidence is from the left to the right, the particle contour is shown by dash line.

If to speak about the field inside a droplet, which will be considered later on, the increase of the beam impact parameter (ξ_0 in this configuration) leads, first of all, to a glancing propagation of a part of light beam in a particle and to formation of a noticeable surface field. At the same time, the droplet center remains "dark." In contrast to this case, the central radiation incidence (when $\xi_0 = 0$) demonstrates a weaker modification of the beam intensity spatial profile inside the droplet, when varies only its amplitude, but any surface modes is not formed.

Underline that when directing a spatially-limited beam to the area, close to the particle edge, there occurs the most effective excitation of resonances of internal field with high Q factor.[12,20–22] This is a consequence of specific spatial distribution of optical field of such modes, which is localized mainly in the circle area at the particle surface. This leads to the situation, as it will be shown below, when such an edge illumination of the particle appears to be more effective for accumulation of the beam light energy inside the particle, scattering the pulse train and for obtaining higher intensity values (factor B) than, for example, at illumination of the particle centre.

Nonstationary scattering of radiation by a spherical particle. Spectral approach

The preceding consideration of light scattering by particles concerned the case of illuminating particles by a monochromatic wave, when the spatial pattern of radiation intensity distribution, diffracted by a particle, is stationary. In this section we consider a general case of this process, namely, the scattering by a spherical particle of the laser pulse, whose frequency spectrum has the finite width. This results in appearance of both spatial and temporal variabilities of optical fields inside and outside the particle, as well as to formation of dynamic character of the scattering.

To study the time evolution of electromagnetic field at scattering of a spectral-limited radiation pulse by a spherical microparticle, we have used the results of the nonstationary Mie theory[23] consisting in the combination of spectral presentation of initial radiation and linear theory of diffraction of a plane monochromatic light wave by a particle. The original nonstationary problem of diffraction of the wide-band radiation in this case is reduced to the stationary problem of the scattering by a spherical particle of a series of monochromatic Fourier harmonics. Scattering characteristics of the particle are characterized by the function of the spectral response $\mathbf{E}_\delta(\mathbf{r};\omega)$, representing the traditional Mie series written for all frequencies from the spectrum of the initial pulse.

Now we set the time profile of electric field intensity of the incident linearly polarized radiation in the harmonic form with the central frequency ω_0 and the slowly varying envelope curve $f(t)$:

$$\mathbf{E}^i(\mathbf{r};t) = \mathbf{E}^i(\mathbf{r})f(t)\mathrm{e}^{i\omega_0 t},$$

where t is the time. To calculate the distribution of the internal optical field of the particle, using the results of the stationary Mie theory, it is necessary first to go from time coordinates to spectral frequencies, presenting the initial light pulse as its Fourier transform:

$$\mathbf{E}^i_\omega(\mathbf{r},\omega) = \Im[\mathbf{E}^i(\mathbf{r},t)] = \mathbf{E}^i(\mathbf{r})G(\omega-\omega_0), \qquad (6)$$

where $\Im$ is the operator of the time Fourier transform; $G = \Im[f(t)\mathrm{e}^{i\omega_0 t}]$ is the frequency range of the initial pulse. Thus, each spectral component is formally equivalent to a monochromatic wave with the frequency ω and the amplitude, set by Eq. (6). The diffraction of such wave by a spherical particle is described in terms of the stationary Mie theory and results in the following representation, for example, of the internal electric field of the particle:

$$\mathbf{E}_\omega(\mathbf{r};\omega) = E_0 G(\omega-\omega_0)\mathbf{E}_\delta(\mathbf{r};\omega).$$

Here the function of spectral response of the particle is introduced:

$$\mathbf{E}_\delta(\mathbf{r};\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{n} R_n\Big(C_{nm}(n_a x_a)\cdot \mathbf{M}^{(1)}_{nm}(n_a kr,\theta,\varphi) - iD_{nm}(n_a x_a)\cdot \mathbf{N}^{(1)}_{nm}(n_a kr,\theta,\varphi)\Big). \qquad (7)$$

In this case the refraction coefficient $n_a(\omega)$ and the Mie particle parameter $x_a(\omega)$ should be chosen in accordance with the value of the current frequency.

Finally, the internal electric field of the particle, depending on time, is written in the form of the convolution from the spectrum of the initial laser pulse and the function of spectral response of the particle:

$$\mathbf{E}(\mathbf{r};t) = E_0\Im^{-1}\big[G(\omega-\omega_0)\mathbf{E}_\delta(\mathbf{r};\omega)\big]. \qquad (8)$$

Note that in the field of the scattered wave outside the particle is expressed in a similar way when substituting the spectral response (7) by the corresponding series for the external field.

Laser radiation model. Formulation of the problem

Consider the following problem. Assume that the dielectric spherical particle of the radius a_0, characterized by the refractive index n_a, is illuminated by a spatially-limited laser beam with a transverse profile of the field intensity (3) and a radius of the focal waist w_0. The time dependence of the incident radiation is set in the form of a pulse train from N_p equally spaced frequency-modulated pulses, which follow with the time interval T. The time profile of the envelope of every pulse in a train is considered to be a Gaussian with the duration t_p (by a level e^{-1} of the intensity peak). Thus, we have

$$f(t)=\sum_{j=1}^{N_p} f_j(t)=\sum_{j=1}^{N_p}\exp\left[-\frac{(t-t_j)^2}{2t_p^2}(1-ib)\right],\ \ j=1\ldots N_p. \quad (9)$$

Here $t_j = t_0 + (j-1)T$, t_0 defines the position of peak of the first pulse in a train at the time scale, and b is the parameter of depth of the linear frequency modulation (linear chirping). The instantaneous frequency of such radiation within the limits of each pulse varies linearly in time following the law: $\omega(t)=\omega_0+bt/2t_p^2$, in this case the combination of parameters $(b/2t_p^2)$ by its sense is the modulation rate. The chirping radiation is widely used in optics and spectroscopy of ultrafast fields (see, e.g., Ref. 24), because it enables one to control for the time length and spectral loading of the laser pulse at its propagation through the dispersion medium.

It should be noted that here we used the model of linearly chirped radiation, widely used in scientific literature,[24] which uses only the frequency chirping parameter b. This model considers the pulse length t_p to be stable independently of the chirping depth. Thus, an example of the chirping is a measure of broadening of only frequency pulse spectrum but not its duration. Without going into details of technical aspects for obtaining such a radiation, note that, for example, the nonlinear pulse propagation through glass fiber at simultaneous manifestation of chromatic dispersion of group velocity of light and Kerr self-focusing corresponds in practice to a similar transformation of spectral-time shape of a laser pulse. The Kerr effect results in the quasilinear frequency wave self-modulation and broadening of its spectrum.[25] The time pulse compression, accompanying this process, can be compensated partly or fully by the chromatic fiber dispersion, providing for the quasi-permanence of a frequency-modulated pulse length.

The Fourier-spectrum of the time profile (9), describing the spectral radiation contour, is given by the following function:

$$G(\omega-\omega_0)=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} f(t)e^{-i(\omega-\omega_0)t}dt = G_0(\omega-\omega_0)\sum_{j=1}^{N_p} e^{-i(\omega-\omega_0)t_j}, \quad (10)$$

where

$$G_0(\omega-\omega_0)=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} f_0(t)e^{-i(\omega-\omega_0)t}dt = \frac{2\pi\sqrt[4]{1+b^2}}{\Delta\omega_p}\exp\left[\frac{4\pi^2(\omega-\omega_0)^2(1+ib)}{2(\Delta\omega_p)^2}\right]$$

is the envelope of the spectral contour of the whole train with the halfwidth $\Delta\omega_p = 2\pi\sqrt{1+b^2}/t_p$, proportional to the chirping depth.

The expression for the spectral radiation intensity follows from Eq. (10) and is written as

$$|G(\omega-\omega_0)|^2 = |G_0(\omega-\omega_0)|^2\frac{\sin^2(N_pK/2)}{\sin^2(K/2)}, \quad (11)$$

where

$$K=(\omega-\omega_0)T=(\omega-\omega_0)s_pt_p,$$

and the parameter $s_p = T/t_p$ gives the interpulse interval. According to Eq. (11), the spectral contour of a pulse train represents a frequency-pulsed function, which has the chief maxima, equidistantly located along the frequency axis and occurring at constructive addition of exponents in Eq. (10). Coordinates of the maxima ω_m can be found from the condition $K=2\pi m$ $(m=0,\pm1,\pm2,\ldots)$, that leads to the following relation for the relative frequency tuning out of the maxima position

$$\delta\bar{\omega}_m=\frac{\omega_m-\omega_0}{\omega_0}=\frac{2\pi m}{s_pt_p\omega_0}. \quad (12)$$

Figure 3 shows the spectral intensity profile of a pulse train of ten Gaussian time pulses (t_p = 1 ps) with the phase modulation of different depths b and the different interpulse intervals s_p.

The number of pulses in the train was chosen arbitrarily, since at another value N_p the pattern will not change qualitatively. Figure 3 shows that a more dense frequency comb of the optical signal spectrum corresponds to the increase of the parameter s_p. The increase of chirping depth b results in the broadening of spectral contour throughout the pulse train (the increase of $\Delta\omega_p$) and more uniform distribution of values of spectral intensity main maxima in the frequency comb. Actually, the ratio of spectral intensity peaks at the frequency of mth side fringe and in the contour centre is determined as

$$B_\omega(\omega_m)=\frac{|G(\omega_m-\omega_0)|^2}{|G(0)|^2}=\exp\left[-\frac{4\pi^2m^2}{s_p^2(1+b^2)}\right] \quad (13)$$

and, hence, grows with the increase of modulation index b.

For more effective optical excitation of the microresonator it is necessary to fulfill the resonance condition for a wavelength (frequency) of the incident radiation and the particle radius. This is not easy to realize in practice, because the spectral width of the resonator excited mode can be much less than the width of the lasing line. Besides, the use of ultrashort pulses with the spectral contour width roughly equal to the carrier frequency for pumping a microparticle results in multiple excitation of a wide range of eigenmodes, frequencies of which fall in the radiation contour and neighbour with the required mode, that is undesirable. Figure 3 shows that the pulse train has a quasilinear spectrum, that enables us to concentrate the radiation energy in the given spectral intervals.

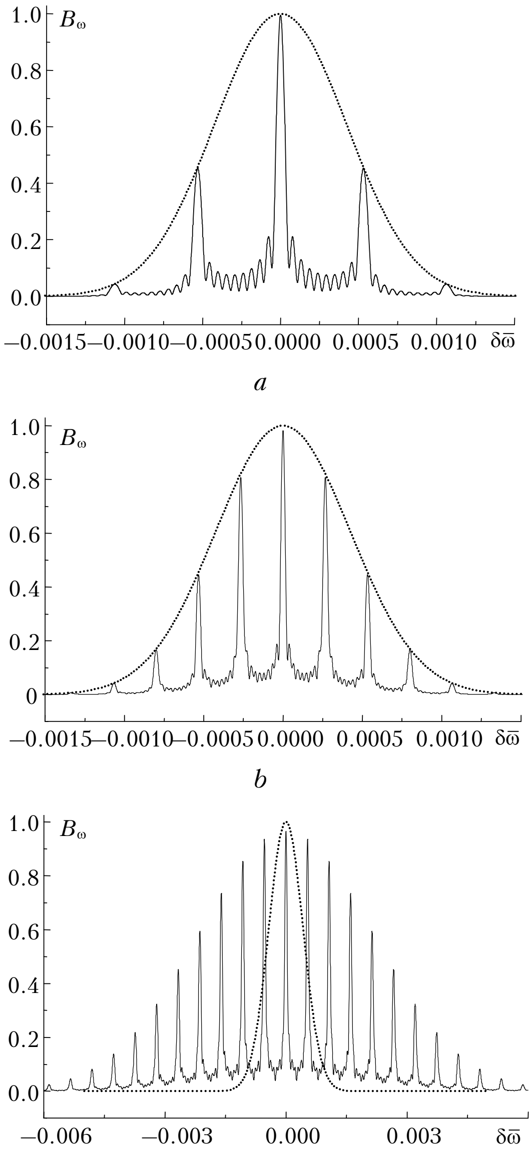

Fig. 3. Spectral radiation intensity, normalized to the maximum value, depending on the relative frequency detuning at different time regimes: unchirped monopulse (dash lines); pulse train of 10 unchirped pulses at $s_p = 5(a)$ and 10 (b); pulse train of 10 chirped pulses at $s_p = 5(a)$ and $b = 6$ (c).

Thus, the pulse train, when varying the spacing of their sequence, presents great potentialities for variation of the spectral composition of an optical signal as compared with a single pulse. Besides, the radiation chirping allows the control for the loading of a given frequency spectral interval. These arguments have determined the choice of the radiation, affecting the particle, in the form (10).

Discussion of results

Now we consider the results of numerical solution of the problem on the linear nonstationary scattering by a spherical microparticle of a train of modulated ultrashort laser pulses, presenting in a space a narrow light beam. Let, for definiteness, a droplet has $a_0 = 10$ μm at the focal beam waist $w_0 = a_0/2$, and the carrier wave of laser radiation $\lambda_0 = 800$ nm. Under such conditions the water refractive index $n_a = 1.33$ and, in fact, the water does not absorb the radiation.[26] The every pulse duration in a train was equal to $t_p = 200$ fs $(t_0 = 4\, t_p)$. This corresponds to spectral contour width of unchirped radiation of about 10 nm, which increased by one order of magnitude, when varying b from 0 to 10. In calculations, we did not take into account the frequency dispersion of the water refractive index within the limits of the spectral interval under study. In the numerical experiments, a light beam moved only along the axis x $(\xi_0 \geq 0,\ \eta_0 = \zeta_0 = 0)$ and was directed to different zones of a water droplet.

First, we illustrate the conclusion, drawn before, (see Fig. 2) on the effect of the reciprocal position of the laser beam and the particle on the character of the optical field, formed inside it. Figure 4 shows the function of spectral transmission of a water droplet

$$K(\mathbf{r};\omega) = \left(\mathbf{E}_\delta(\mathbf{r};\omega)\cdot\mathbf{E}_\delta^*(\mathbf{r};\omega)\right)$$

at three impact parameters of the incidence of a light beam.

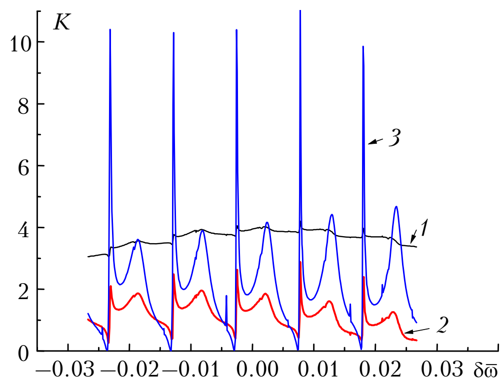

Fig. 4. The transmission function of water droplet K depending on the relative frequency difference $\delta\omega$;⁻ at different vertical shifts of the light beam: $\xi_0 = 0$ (*1*); 0.5 (*2*), and 1 (*3*).

The values of the function of spectral response (7) here and further were calculated at a definite point inside the particle $\mathbf{r}_m$, corresponding to the space coordinate of the absolute (in time) intensity peak of the optical field. For a water droplet of 10 μm radius the calculations of the internal field profile have shown that the point $\mathbf{r}_m$ is on the principal diameter in the particle shadow hemisphere and has the following spherical coordinates: $\mathbf{r}_m = (r/a_0,\ \theta,\ \varphi)|_m = (0.845;\ 0.0;\ 0.0)$.

It follows from Fig. 4 that the central geometry of the particle illumination is characterized by a weak, practically neutral spectral dependence of the transmission function. In this case, the excitation of the field resonances does not appear. The transfer of the laser beam in the direction of the droplet edge leads to a gradual development of the resonance structure of the spectral response, which becomes more pronounced at $\xi_0 \simeq 1$. The sign "$\simeq$" here denotes the dependence of a precise value of the impact parameter ξ_0, at which the maximal value of K is realized, on the morphology of the particle excited resonance mode. As it was shown in Refs. 13 and 27, for the optimal tuning to the resonance with the increasing number of excited WGMs of the internal field, ξ_0 must grow. In this paper we do not pursue the goal to analyze in detail the behavior of the particle transmission function from the geometry of its illumination, and, therefore, further all presented results refer to the case of the accurate edge radiation incidence $\xi_0 = 1$.

The time profile of the optical field intensity $B(t)$ (inhomogeneity factor) at a chosen point $\mathbf{r}_m$ inside a drop in the train field of 15 femtosecond laser pulses is given in Fig. 5*a*. Figure 5*b* shows in relative units the spectral behavior of the transmission function of a droplet and the initial radiation for this case.

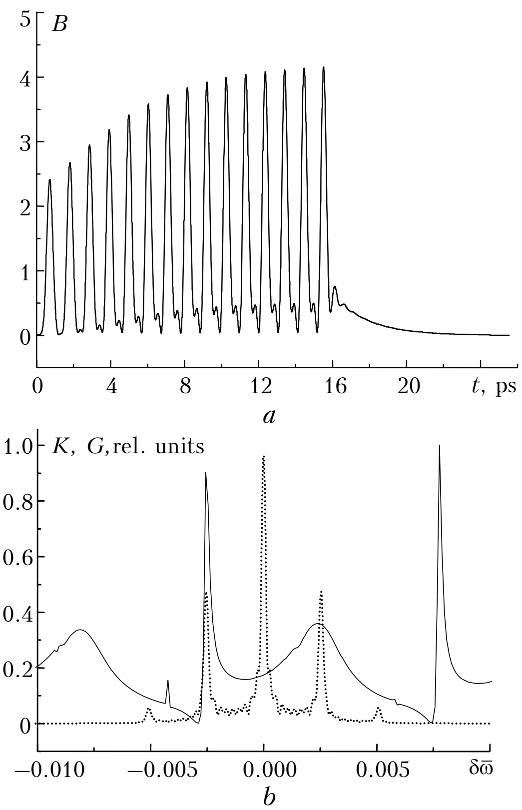

Fig. 5. The time dependence of the relative intensity of internal optical field $B(r_m)$ of water drop (*a*) and its spectral transmission function K (*b*) at illumination by a train of femtosecond pulses with $s_p = 5.25$ and $b = 0$. The profile of spectral radiation intensity is shown by a dash line.

The pulse repetition frequency in the train is specially selected [according to the condition (12)] for the resonance excitation by the first main lateral fringe of the radiation spectrum with the tuning $\delta\omega_{-1} = 2.5\cdot10^{-3}$ of the eigenmode TE^3_{85}, which $Q_R \sim 7\cdot10^3$.

It follows from Fig. 5 that the pulse train provides for the resonance character of the radiation scattering by particle. This is expressed through the increase of the intensity maximum of the internal particle field from pulse to pulse by a factor of two and the corresponding radiation energy accumulation in an excited resonator mode. After completion of the train the particle de-excited the stored energy during the characteristic lifetime of mode $\tau_R = Q_R/\omega_1 \sim 3$ ps and the particle field intensity decreased exponentially. Besides, figure 5*a* shows the intensity variations, which are connected with the excitation one more eigenmode of the droplet (see Fig. 5*b*), namely, TE^4_{81}, with much lower Q_R (~10^2).

The number of pulses in a train, necessary for providing for maximal level of internal field intensity $B_m = \max\limits_t\{B(\mathbf{r}_m, t)\}$ during their scattering by the particle, is determined by resonance characteristics of the excited whispering gallery mode. Figure 6 shows the results of investigation of B_m depending on the number of pulses, which show that the field intensity maximum is at $N_p = 10 \div 15$ and after that it is not changed with the pulse train elongation.

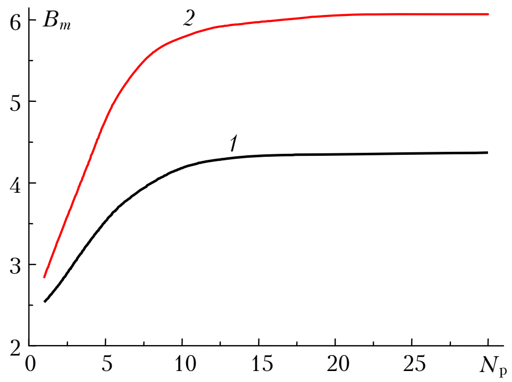

Fig. 6. The dependence of maximal value of the function of droplet inhomogeneity B_m in the field of laser pulse train on their number N_p at $s_p = 5.25$ and $b = 0$ (*1*); 3 (*2*).

As it follows from Eq. (11), the characteristic width of any of principal maxima of the train spectrum is inversely proportional to N_p, the existence of optimal number of pulses is connected with attaining equality between the spectral lobe width and the WGM width. The finer is the spectral resonance, excited in a particle, the longer should be the pulse train under other equal conditions for to transmit most effectively the train energy to the particle.

One more problem is connected with the influence of the radiation frequency modulation on the magnitude of the attainable level of the optical field intensity. The train chirping, as it was noted above, widens the spectral contour of the entire pulse train $\Delta\omega_p \sim b$, which at constant frequency position of the spectral maxima ω_m increases their relative value as compared with the absolute intensity maximum at the centre [see Eq. (13)]. More effective excitation of the field resonance by the chirping radiation is shown in Fig. 6 (curve *2*), where at $b = 3$ the field intensity maximum increases by a factor of 1.5 as compared with maximum of the unmodulated pulse train.

However, the radiation frequency modulation has also a negative effect on the efficiency of the field resonance excitation, because at the total spectral broadening simultaneously the absolute maximum of the lobe intensity decreases. Actually, it follows from Eq. (10) that for the first lobe

$$\left|G(\omega_1)\right|^2 \sim \frac{1}{\sqrt{1+b^2}} \exp\left[-\frac{t_p^2(\omega_1-\omega_0)^2}{(1+b^2)}\right].$$

This function has its maximum at $b_1 = \sqrt{\left|2(\omega_1-\omega_0)^2 t_p^2 - 1\right|}$, that gives the value of optimal chirping: $b_1 \simeq 2.3$ at experimentally selected pulse train parameters.

The results of calculations of the function $B_m(b)$ are shown in Fig. 7.

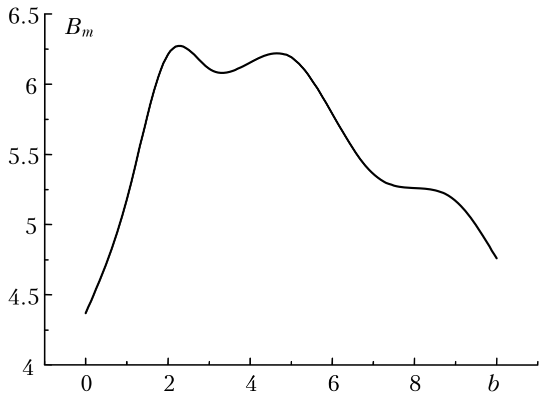

Fig. 7. The intensity maximum of the droplet internal field B_m irradiated by the laser pulse train at $N_p = 15$ and $s_p = 5.25$, depending on the linear chirping depth b.

It is seen that this dependence has a maximum at $b = b_1$. The second, lower maximum is observed at $b \simeq 5$ due to the excitation of the mode TE_{85}^3, as well as a series of WGM with a high Q-factor by a wide spectral contour of the chirping radiation.

Conclusion

Thus, the investigations of nonstationary scattering by a transparent spherical particle of a train of frequency-modulated femtosecond laser pulses, having a spatial configuration in the form of a limited narrow beam of the Gaussian transverse intensity profile, as well as time dynamics of the formation and characteristics of resonance excitation of internal optical field of the particle by such radiation have made it possible to draw the following conclusions:

1. The efficiency of incident radiation energy transfer to intrinsic resonance of a particle with a given high Q-factor (WGM), illuminated by a spatially-limited focused light beam, depends greatly on the combination of such parameters as relative pulse length in the trainand the depth of linear frequency modulation of each pulse.

2. The geometry of particle irradiation makes an essential effect on the values of relative intensity of the inner optical field. The excitation of a weakly absorbing spherical particle by a narrow laser beam is the most efficient at its focusing to a particle edge, because only in this case the excitation of high-quality WGM becomes possible as opposed to the case of the light beam direction to the droplet centre.

3. A comparative analysis of time dynamics of formation of the inner optical field of the particle irradiated by a single pulse or by a laser pulse train has shown that the increase of pulses in the train results in the variation of time behavior of the inner particle field and its peak intensity. At the same time, there exist an optimal number of pulses in the train, some excess of which does not lead to further increase of the intensity peak of the particle inner optical field.

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