Internal and external focusing of the optical field of a femtosecond pulse at its diffraction on a spherical particle

A.A. Zemlyanov and Yu.E. Geints

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk

Received July 17, 2003

The effect of internal and external focusing of an ultrashort laser pulse on micron-sized spherical particles is investigated numerically. Peculiarities have been found in the dynamics of formation of the spatial intensity distribution of spectrally limited light pulses scattered on particles at different pulse duration, particle size, and absorption coefficient of the particulate matter. It is shown that in the zone of the geometrical focus (behind the particle) the maximum achievable intensity of the total field may be tenfold higher than that inside the particle independent of the duration of the initial laser pulse, copying its profile. In the zone of internal focusing of the optical field at laser pulse shortening, the absolute peak of the field intensity decreases, in general, as compared to its stationary value, which is higher for larger particles. However, this decrease becomes significant only at resonance excitation of the particle internal field. The increase of the absorption coefficient of the particulate matter leads to a decrease in the maxima of the optical field intensity in the zones of its external and internal focusing, but still keeps the ratio between them.

Introduction

Scattering of laser radiation by a transparent spherical microscopic particle is accompanied by an increase in the intensity of the optical field in the internal zones near its illuminated and shady surfaces.[1] In addition, the focusing effect of the spherical surface also leads to the field intensity increase behind the particle, in the zone of its geometrical shadow, and the degree of this increase may achieve several orders of magnitude for optically "large" particles.[2] As a consequence, it becomes possible, for example, to significantly decrease the energy thresholds for manifestation of nonlinear scattering effects, such as the stimulated Raman effect, higher harmonic generation, stimulated fluorescence, as well as the effects of multiphoton absorption and tunneling ionization that lead to formation of plasma cells near the particle.[2]

In this connection, it seems important to study the dynamics of formation of the optical field intensity inside and beyond spherical micron-sized particles at nonstationary scattering of laser pulses on them, to establish the upper boundary of the achievable level of this intensity, and to determine its spatial localization and dependence on the time parameters of radiation, as well as on the particle size. This paper is devoted to solution of these problems.

Basic equations

The nonstationary problem of diffraction of a broadband radiation at a spherical microparticle was considered based on the Fourier method in combination with the linear light scattering theory, known as the Mie theory in the case of a plane monochromatic light wave incident on a particle.[3] The scattering properties of the particle in this case are characterized by the so-called spectral response function $\mathbf{E}_\delta(\mathbf{r};\omega)$, which is a traditional Mie series written for all frequencies from the spectrum of the initial pulse. Here we restrict our consideration to only a brief summary of the basic equations. The detailed description of the technique used and some details of its numerical realization are presented, for example, in Refs. 4 and 5.

In numerical calculations, we used the following representation of the electric field strength of the linearly polarized incident radiation:

$$\mathbf{E}^{\mathrm{i}}(\mathbf{r};t) = 1/2\left\{\mathbf{E}^{\mathrm{i}}(\mathbf{r};t) + [\mathbf{E}^{\mathrm{i}}(\mathbf{r};t)]^*\right\} = = 1/2 E_0 \mathbf{e}_y g(t) S(\mathbf{r}_\perp) \exp\{i\omega_0[t-(z+a_0)/c]\} + \text{c.c.}, \quad (1)$$

where $g(t)$ and $S(\mathbf{r}_\perp)$ are, respectively, the time and space profiles of the pulse; ω_0 is the carrier frequency of the pulse; E_0 is the real amplitude of the field; $\mathbf{r} = \mathbf{r}_\perp + \mathbf{e}_z z$; $\mathbf{r}_\perp = \mathbf{e}_x x + \mathbf{e}_y y$; $\mathbf{e}_x$, $\mathbf{e}_y$, $\mathbf{e}_z$ are the unit vectors along the axes x, y, and z; t is time; c is the speed of light in vacuum. It is believed that a spherical particle with the radius a_0 is located at the origin of coordinates, and the laser pulse diffracting at it propagates along the positive direction of the z-axis. The time profile of the light beam was described by the Gaussian function

$$g(t) = \exp\left\{-\frac{\left(t-(z+a_0)/c-t_0\right)^2}{t_\mathrm{p}^2}\right\} \quad (2)$$

with the duration t_p and the time lag t_0. The radiation was assumed to be a plane wave, that is, $S(\mathbf{r}_\perp) = 1$.

0235-6880/03/10 822-04 $02.00

To calculate the distribution of the internal optical field of the particle and apply the results of the stationary Mie theory, it is necessary, first, to pass on from the time coordinates to the region of spectral frequencies, representing the initial light pulse through its Fourier transform:

$$\mathbf{E}^{\mathrm{i}}_{\omega}(\mathbf{r},\omega) = \Im[\mathbf{E}^{\mathrm{i}}_{j}(\mathbf{r},t)] =$$
$$= 1/2 E_0 \mathbf{e} S(\mathbf{r}_\perp) G(\omega-\omega_0)\exp[-ik_0(z+a_0)], \quad (3)$$

where $\Im$ is the Fourier transform operator; $G(\omega)$ is the frequency spectrum of the initial laser pulse; $k_0 = \omega_0/c$.

Equation (3) after multiplication by $e^{i\omega t}$ determines the spectral component of the initial pulse in the form of a monochromatic wave with the amplitude

$$\mathbf{A}(\omega) = E_0 \mathbf{e}_y S(\mathbf{r}_\perp) G(\omega-\omega_0). \quad (4)$$

Diffraction of such a wave at a spherical particle is described within the stationary application of the Maxwell equations:

$$\mathrm{rot}\,\mathbf{E}_{\omega}(\mathbf{r};\omega) = -ik\mathbf{H}_{\omega}(\mathbf{r};\omega);$$
$$\mathrm{rot}\,\mathbf{H}_{\omega}(\mathbf{r};\omega) = i\varepsilon_a k\mathbf{E}_{\omega}(\mathbf{r};\omega),$$

where $\mathbf{H}_{\omega}(\mathbf{r};\omega)$ is the magnetic field strength vector; ε_a is the complex permittivity of the particulate matter; $k = \omega/c$.

The boundary conditions on the surface of the spherical particle ($r = |\mathbf{r}| = a_0$) are the continuity of the tangential components of the internal field $\mathbf{E}_{\omega}$, $\mathbf{H}_{\omega}$ at transition through the surface:

$$[\mathbf{E}_{\omega} \times \mathbf{n}_r] = [(\mathbf{E}^{\mathrm{i}}_{\omega} + \mathbf{E}^{\mathrm{s}}_{\omega}) \times \mathbf{n}_r];$$
$$[\mathbf{H}_{\omega} \times \mathbf{n}_r] = [(\mathbf{H}^{\mathrm{i}}_{\omega} + \mathbf{H}^{\mathrm{s}}_{\omega}) \times \mathbf{n}_r], \quad (6)$$

where $\mathbf{n}_r$ is the vector of external normal to the particle surface, and the superscript “s” stands for the field of the scattered wave.

Solution of Eqs. (5) with regard for Eqs. (4) and (6) leads to the following representation of the total electric field:

$$\mathbf{E}^{\Sigma}_{\omega}(\mathbf{r};\omega) = \mathbf{E}_{\omega}(\mathbf{r};\omega) =$$
$$= E_0 G(\omega-\omega_0)\sum_{n=1}^{\infty} R_n \Big(c_n(m_a k a_0)\mathbf{M}^{(1)}_{n1}(kr,\theta,\varphi) -$$
$$-id_n(m_a k a_0)\mathbf{N}^{(1)}_{n1}(kr,\theta,\varphi)\Big),\ r \le a_0; \quad (7)$$

$$\mathbf{E}^{\Sigma}_{\omega}(\mathbf{r};\omega) = \mathbf{E}^{\mathrm{i}}_{\omega}(\mathbf{r};\omega) + \mathbf{E}^{\mathrm{s}}_{\omega}(\mathbf{r};\omega) =$$
$$= E_0 G(\omega-\omega_0)\sum_{n=1}^{\infty} R_n \Big[\Big(\mathbf{M}^{(1)}_{n1}(k_0 r,\theta,\varphi) - i\mathbf{N}^{(1)}_{n1}(k_0 r,\theta,\varphi)\Big) +$$
$$+\Big(a_n(k a_0)\mathbf{M}^{(3)}_{n1}(kr,\theta,\varphi) - ib_n(m_a k a_0)\mathbf{N}^{(3)}_{n1}(kr,\theta,\varphi)\Big)\Big],$$
$$r > a_0 \quad (8)$$

where

$$R_n = i^n \frac{2n+1}{n(n+1)};\ \mathbf{M}^{(1)}_{n1}, \mathbf{N}^{(1)}_{n1}, \mathbf{M}^{(3)}_{n1}, \mathbf{N}^{(3)}_{n1}$$

are the spherical vector-harmonics with the azimuth index equal to unity; m_a is the complex refractive index of the particulate matter; a_n, b_n, c_n, d_n are the Mie coefficients.[3]

In the time-space, the total electric field within the approach considered is described as a convolution integral of the spectrum of the initial laser pulse and the spectral response function of the particle:

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

Here $\mathbf{E}_{\delta}(\mathbf{r};\omega)$ stands for the series in the right-hand side of Eqs. (7) and (8).

The complex refractive index of the particulate matter m_a and the laser radiation wavelength λ_0 used in the numerical simulations were taken as follows: $m_a = 1.33 - i\kappa_a$; $\lambda_0 = 0.8$ μm, where κ_a is the absorption coefficient variable in the calculations. These parameters correspond, for instance, to water droplets exposed to Ti:Sapphire laser pulses. The dispersion of the particulate matter in the wavelength range chosen was neglected.[6] The influence of nonlinear optical effects, for example, multiphoton ionization and multiphoton absorption that may arise in the process of beam diffraction on the particle and, in principle, modify the spatial structure of the optical field was neglected too.

Discussion

Figure 1 shows the evolution of the spatial distribution of the relative intensity of the optical field in the plane of equatorial cross section of the particle 20 μm in radius exposed to an ultrashort laser pulse. The behavior of the field inhomogeneity factor was calculated as:

$$B(\mathbf{r};t) = (\mathbf{E}^{\Sigma}_{\omega}(\mathbf{r};t)\cdot[\mathbf{E}^{\Sigma}_{\omega}(\mathbf{r};t)]^{*})/E_0^2.$$

Figure 2 gives an idea of real values of the relative intensity of the optical field along the bigger diameter of the particle. This figure is drawn for the same stages of the process as in Fig. 1.

Figures 1 and 2 point to some regularities in the nonstationary diffraction of radiation on particles. This statement concerns the behavior of the total field intensity, namely, (a) the pulsed time dependence of the optical field intensity inside the particle; (b) the advance of formation of the external field intensity maximum as compared with the internal field maximum near the shady surface of the particle (see Figs. 1*d*–*f*). Pulsations of the internal field intensity are caused by reflections of the light wave from the internal surface of the particle and the following formation of the whispering gallery modes (WGM). On the scale of the initial laser pulse duration, the characteristic WGM lifetime can be long enough, causing the characteristic “afterglow” from the particle.[4]

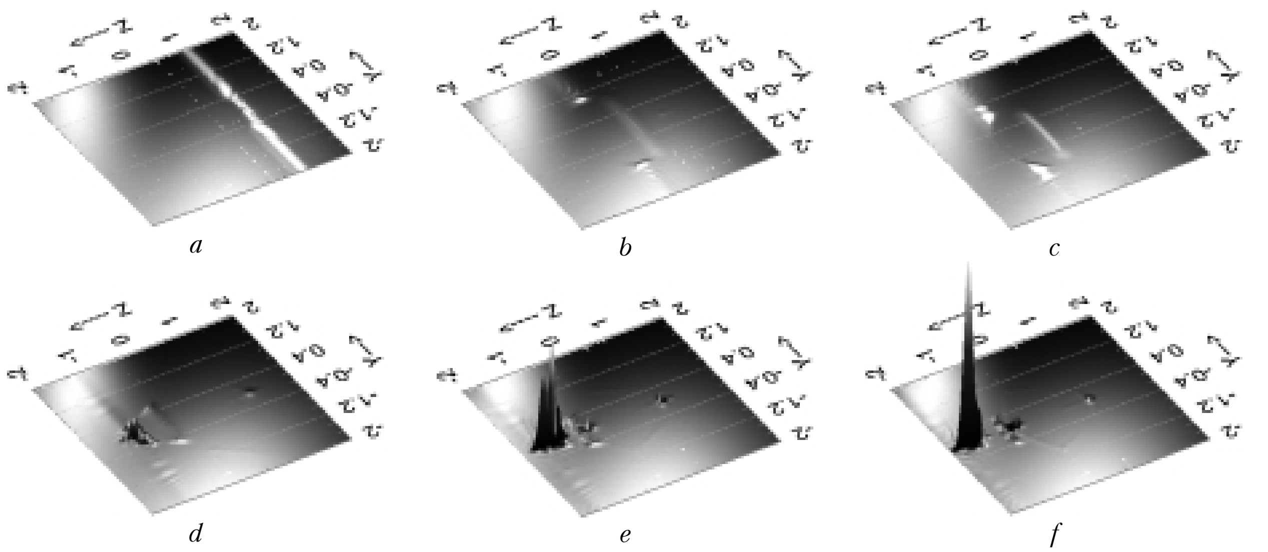

Fig. 1. Spatial distribution of the relative optical field intensity in the plane of the principal cross section of a water droplet with a_0 = 20 μm exposed to a plane wave with the following parameters: λ = 0.8 μm, t_p = 50 fs, t_0 = 100 fs at different time t = 70 (*a*), 130 (*b*), 170 (*c*), 200 (*d*), 220 (*e*), and 250 fs (*f*). The oval in white shows the particle boundary. The radiation propagates from right to left.

Different time needed for the formation of the intensity maxima of the external and internal fields is explained by different mechanisms of their formation. The intensity maximum of the external field is located just behind the particle, and it is formed by the light wave having passed through the particle without reflections (Fig. 1*d*). Therefore, it arises first compared to the "rear" maximum of the internal field, which is formed by the rays reflected from the internal surface of the particle (Fig. 1*e*). The further increase in the intensity of the external maximum (Fig. 1*f*) is associated with the arrival of the central part of the pulse.

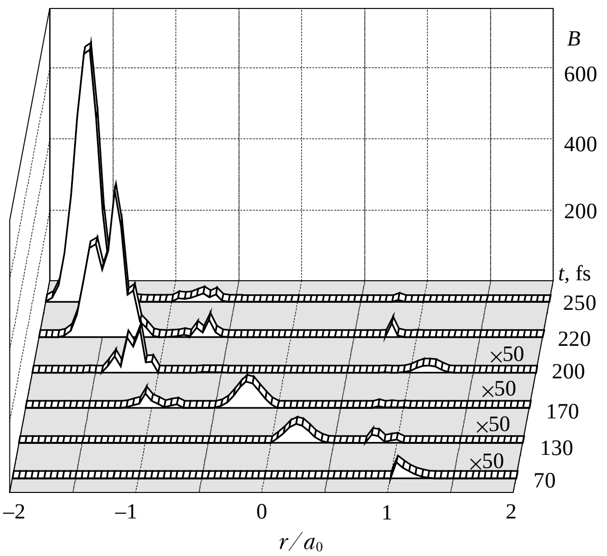

Fig. 2. Distribution of the relative intensity of the optical field along the principal diameter of a water droplet with a_0 = 20 μm exposed to a plane wave. The radiation parameters are the same as in Fig. 1. The intensity at the first four curves is increased by 50 times.

The particle-size dependence of the intensity maxima of the internal and external fields for different values of the refractive index of the particulate matter κ_a is depicted in Fig. 3, where B_m is the maximum achievable value of the optical field inhomogeneity factor for the entire period of consideration of the scattering process (absolute intensity maximum).

Analysis of data presented in Fig. 3 shows the general growth of the intensity maximum in the considered zones with the increase of the particle radius. In addition, at any level of radiation absorption by the particle the increase of the external field intensity in the focus is always, by at least an order of magnitude, higher than that inside the particle. The increase of the absorption coefficient leads to stabilization of these dependences at some level independent of the particle size.

The calculated results shown in Fig. 3 were obtained at a widely varying duration of the incident pulse, and they demonstrate that in the zone of the geometrical focus (behind the particle) the time dependence of the factor B copies the time profile of the initial pulse. At the same time, the value of the maximum achievable intensity B_m in this zone is independent of t_p.

A somewhat different situation is observed for a similar dependence of B_m in the zone of the rear intensity maximum of the internal field. The data for this case are summarized in Fig. 4, which shows that, as the laser pulse becomes shorter, the absolute maximum of the field, generally, decreases from its stationary level achieved at $t_\text{p} > 10^{-10}$ s for the considered range of the particle size, and the larger the particle, the higher this level is.

This decrease is rather small and does not exceed 15%, except for the two particle radii used in the calculations: a_0 = 11 and 20 μm, for which the decrease in the intensity is quite significant (see Fig. 4).

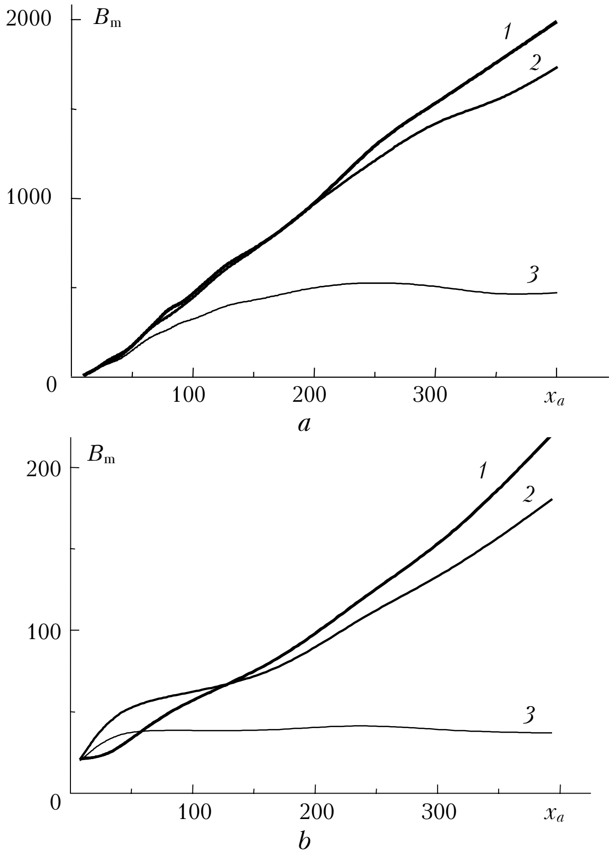

Fig. 3. Dependence of the maximum values of the optical field inhomogeneity factor B_m in the zone of geometrical focus behind the particle (*a*) and at the rear maximum inside the particle (*b*) on the diffraction parameter $x_a = 2\pi a_0/\lambda_0$ at different level of absorption: $\kappa_a = 10^{-8}$ (*1*), 10^{-4} (*2*), 10^{-3} (*3*).

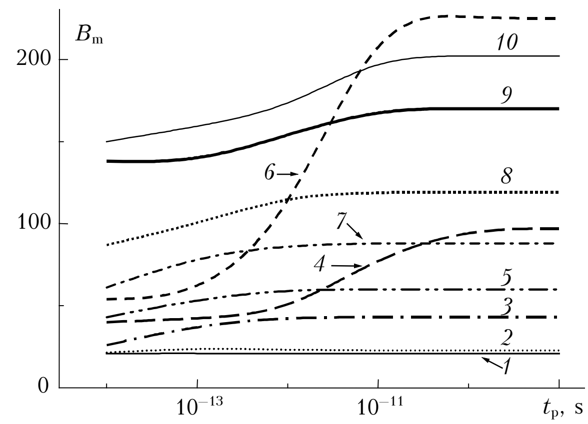

Fig. 4. Dependence of B_m in the zone of internal focus of the optical field on the laser pulse duration t_p for different particle radius a_0 = 1 (*1*), 5 (*2*), 10 (*3*), 11 (*4*), 15 (*5*), 20 (*6*), 22 (*7*), 30 (*8*), 40 (*9*), and 50 μm (*10*).

This is explained by the fact that for these particle sizes the excitation of the internal field is close to resonance, that is, the central frequency of the pulse spectrum falls (though not exactly) at one of the particle natural resonances. This leads both to the general increase in the maximum of the internal field of the stationary intensity distribution and to its stronger dependence on the spectral width of the radiation.[4]

Conclusion

Thus, the results obtained point to the following regularities in the manifestation of the effect of external and internal focusing of the optical field by a spherical microparticle for the case of the particle exposed to laser pulses of femtosecond duration. In the zone of the geometrical focus of the incident radiation (behind the particle) the maximum achievable intensity of the total field may exceed by an order of magnitude the intensity inside the particle and does not depend on the duration of the initial pulse, copying its profile. In the zone of internal focusing of the optical field, as the laser pulse shortens, the absolute maximum of the field intensity decreases from its stationary value, which is higher for larger particles. This decrease of the intensity is rather small and becomes significant only at resonance excitation of the internal field. The increase of the absorption coefficient of the particulate matter leads to a decrease in the maximum levels of the optical field intensity in the external and internal focal zones, but keeps the intensity ratio between them.

Acknowledgments

This work was partly supported by the Project 2.9 of the Program of Department of Physical Sciences of RAS.

References

1. A.P. Prishivalko, *Optical and Thermal Fields inside Light Scattering Particles* (Nauka i Tekhnika, Minsk, 1983), 190 pp.
2. P. Chylek, M.A. Jarzembski, V. Srivastava, R.G. Pinnick, J.D. Pendleton, and J.P. Cruncleton, Appl. Opt. **26**, No. 5, 760–762 (1987).
3. C.F. Bohren and D.R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1983).
4. A.A. Zemlyanov and Yu.E. Geints, Atmos. Oceanic Opt. **14**, No. 5, 316–325 (2001).
5. A.A. Zemlyanov and Yu.E. Geints, Atmos. Oceanic Opt. **15**, No. 8, 619–627 (2002).
6. K.S. Shifrin and I.G. Zolotov, Appl. Opt. **34**, No. 3, 552–558 (1995).