Scattering of supercontinuum radiation by spherical particles upon filamentation of laser radiation in an air medium

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We present numerically calculated scattering, absorption, and backscattering efficiency factors of water particles for the quasi-white light field (radiation of a wide spectral range) produced due to self-focusing of a high-power femtosecond pulsed radiation in the atmosphere. These integral optical characteristics were studied for a wide range of drop sizes, covering the range of fog and cloud particles. The results obtained were compared with the scattering factors for a monochromatic radiation and the broadband radiation of a femtosecond pulse.

Introduction

The possibility of using femtosecond laser radiation in sensing the parameters of gas and aerosol components of the atmosphere is intensely discussed nowadays. The promises are connected with the experimentally established fact that the supercontinuum (quasi-white) radiation is generated in the atmosphere upon the nonstationary self-focusing of a femtosecond pulse. This effect is explained by the frequency self-modulation of the laser pulse due to the Kerr effect and plasma nonlinearity, arising in the filament channel due to the multiphoton absorption of radiation by a gas. After the filamentation, the quasi-white radiation propagates as a pulse in the linear medium. Such a radiation, interacting with aerosols and gases, experiences linear scattering: Rayleigh scattering and scattering by particles. Analyzing the characteristics of this scattering in different spectral ranges within the emission spectrum of supercontinuum it is possible to acquire information on the medium structure. Just this is the idea of the inverse problem of sensing.

The initial information here is the known frequency dependence of the scattering and absorption coefficients of the aerosol medium for the quasi-white radiation. For the monochromatic radiation, this information can be obtained using Mie theory, which gives a solution of the problem on diffraction of a monochromatic wave on a spherical particle. The properties of the scattered fields are uniquely determined by the dimensionless Mie parameter $x = \omega_0 a_0/c$ (where a_0 is the particle radius, ω_0 is the radiation frequency, and c is the speed of light in vacuum) and by the values of the refractive index n_a and the absorption coefficient κ_a of the substance of a spherical particle.

What is the situation in the case the quasi-white light?

In this case, it is necessary to apply the theory of scattering of non-monochromatic (multifrequency) radiation. The principles of this theory were formulated in Ref. 1, and the first quantitative results were obtained in Ref. 2 in considering the problem on scattering of a femtosecond pulse by a spherical particle. In particular, the effect of smoothing of the frequency behavior of the integral optical characteristics (efficiency factors), arising upon the broadening of the spectrum of the radiation pulse, was noticed. In this connection, it is of undoubted interest to consider, in a more detail, the regularities of scattering of such a quasi-white light by aerosol particles, what in fact was the goal of this study.

Basic relationships

Below we briefly present basic relationships from the nonstationary Mie theory, used to calculate the scattering efficiency factors of spherical particles in the field of a spectrally limited (non-monochromatic) optical radiation.

To study the evolution of the electromagnetic field of a light wave scattered by a spherical particle, the spectral approach[3] is widely used. The initial nonstationary problem of diffraction of a broadband radiation in this case reduces to the stationary problem of scattering of a superposition of monochromatic Fourier harmonics by a spherical particle. In this case, the scattering properties of the particle are characterized by the so-called spectral response function, which is the traditional Mie series, written for all frequencies from the spectrum of the initial pulse.

Let the optical radiation incident on a spherical particle be a plane wave with the elliptical polarization, whose electric field can be presented in the following form:

$$\mathbf{E}^{\mathrm{i}}(z;t) = \frac{1}{2}\left[\mathbf{E}^{\mathrm{i}}(z;t) + \left(\mathbf{E}^{\mathrm{i}}(z;t)\right)^{*}\right] = = \frac{1}{2}E_0\mathbf{p}\hat{g}(t)\mathrm{e}^{i\omega_0(t-(z+a_0)/c)} + \mathrm{c.c.} \quad , \tag{1}$$

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where $\hat{g}(t) = g(t)\exp\{i\varphi(t)\}$ is the complex time profile of the radiation that includes also the wave phase $\varphi(t)$; ω_0 is the carrier frequency of the wave packet; E_0 is the real amplitude of the field; **p** is the vector of polarization; t is time. A dielectric spherical particle with the radius a_0 is assumed to be in the origin of coordinate system, and the radiation diffracting on it propagates along the positive direction of the z-axis.

To solve the nonstationary problem of scattering and calculate the spatiotemporal distribution of optical fields, it is necessary, first, to pass from the time coordinates to the region of spectral frequencies, representing the initial light pulse by its Fourier transform:

$$\mathbf{E}^i_\omega(z,\omega) = \Im[\mathbf{E}^i(z,t)] = \\ = \frac{1}{2}E_0\mathbf{p}G(\omega-\omega_0)e^{-ik_0(z+a_0)}, \quad (2)$$

where $\Im$ is the operator of Fourier transformation; $G(\omega) = \Im[\hat{g}(t)]$ is the frequency spectrum of the initial laser pulse; $k_0 = \omega_0/c$. Thus, if the incident radiation has the Gaussian time profile $g(t) = \exp\{-t^2/2t_\text{p}^2\}$ and the linear frequency modulation (chirping), that is $\omega(t) = \omega_0 - bt/2t_\text{p}^2$, and

$$\hat{g}(t) = \exp\left\{-\frac{t^2}{2t_\text{p}^2}(1+ib)\right\},$$

where b is the modulation parameter; t_p is the pulse duration, then the spectrum of this pulse is also a Gaussian function with the half-width $\Delta\omega = 2\pi\sqrt{1+b^2}/t_\text{p}$:

$$G(\omega) = \frac{1}{\sqrt{1+ib}}\exp\left\{-\frac{4\pi\omega^2}{2\Delta\omega^2}(1-ib)\right\}.$$

Equation (2), being multiplied by $e^{i\omega t}$, determines the spectral component of the initial radiation pulse in the form of the monochromatic wave with the amplitude

$$\mathbf{A}(\omega) = E_0\mathbf{p}G(\omega-\omega_0). \quad (3)$$

The diffraction of this wave on a spherical particle is described within the framework of the stationary approximation of the Maxwell equations:

$$\text{rot}\,\mathbf{E}_\omega(\mathbf{r};\omega) = -ik\mathbf{H}_\omega(\mathbf{r};\omega); \\ \text{rot}\,\mathbf{H}_\omega(\mathbf{r};\omega) = i\varepsilon_\text{a}k\mathbf{E}_\omega(\mathbf{r};\omega), \quad (4)$$

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

The boundary conditions on the surface of the spherical particle ($r = |\mathbf{r}| = a_0$) consist in the continuity of the tangential components of the internal fields $\mathbf{E}_\omega$ and $\mathbf{H}_\omega$ upon the passage through the surface:

$$[\mathbf{E}_\omega \times \mathbf{n}_r] = [(\mathbf{E}^\text{i}_\omega + \mathbf{E}^\text{s}_\omega)\times\mathbf{n}_r]; \\ [\mathbf{H}_\omega \times \mathbf{n}_r] = [(\mathbf{H}^\text{i}_\omega + \mathbf{H}^\text{s}_\omega)\times\mathbf{n}_r], \quad (5)$$

where $\mathbf{n}_r$ is the vector of the external normal to the particle surface; the superscript "s" corresponds to the field of the scattered wave.

The solution of Eq. (4), taking into account Eqs. (3) and (5), leads to the following representation of the electric field of, for example, a monochromatic plane wave (1) scattered by the particle:

$$\mathbf{E}^\text{s}_\omega(\mathbf{r};\omega) = E_0G(\omega-\omega_0)\times \\ \times\sum_{n=1}^{\infty}R_n[a_n(m_\text{a}ka_0)\cdot\mathbf{M}^{(3)}_{n1}(k\mathbf{r}) - ib_n(m_\text{a}ka_0)\cdot\mathbf{N}^{(3)}_{n1}(k\mathbf{r})], \quad (6)$$

where $R_n = i^n\frac{2n+1}{n(n+1)}$; $\mathbf{M}^{(3)}_{nm}$ and $\mathbf{N}^{(3)}_{nm}$ are the third-order spherical harmonics[4]; $m_\text{a} = n_\text{a} - i\kappa_\text{a}$ is the complex refractive index of the particulate matter; a_n and b_n are the partial Mie coefficients. The time-dependent electric field of the spectrally limited radiation, being scattered by a particle can, within the framework of the considered approach, be written as the convolution integral of the spectrum of the initial laser signal and the spectral response function of the particle:

$$\mathbf{E}(\mathbf{r};\, t) = E_0\Im^{-1}[G(\omega-\omega_0)\mathbf{E}_\delta(\mathbf{r};\,\omega)]. \quad (7)$$

Here $\mathbf{E}_\delta(\mathbf{r};\,\omega)$ denotes the series in the right-hand side of Eq. (6).

By definition, any of the spectral (monochromatic) efficiency factors Q_ω characterizes the fraction of the electromagnetic energy flux, converted by a particle into either absorption or scattering, with respect to the incident light energy flux through the cross section of the particle:

$$Q_\omega = \frac{1}{\pi a_0^2 I_0}\int_\Omega (\Pi_\omega)_r\, r^2 \text{d}\Omega, \quad (8)$$

where $I_0 = c/8\pi E_0^2$ is the intensity of the incident radiation; $\text{d}\Omega$ is an element of solid angle, and $(\Pi_\omega)_r = \frac{c}{4\pi}\text{Re}[\mathbf{E}_\omega,\,\mathbf{H}_\omega]_r$ is the radial component of the Poynting vector of the field of a monochromatic wave diffracted on the particle (in the case of the scattering factor) or the wave formed due to the interference of the incident and scattered waves (extinction efficiency factor).

If the optical fields (7) depend on time, to determine the stationary scattering efficiency factors, it is necessary to pass from the power parameters of radiation to its energy, which is equivalent to the measurement of the scattering characteristics by a detector with the infinite exposure time.

Passing to time integrals in Eq. (8) we have

$$Q = \frac{1}{\pi a_0^2 w_0} \int_{-\infty}^{\infty} \mathrm{d}t \int_{\Omega} \Pi_r(t) r^2 \mathrm{d}\Omega,$$

where $w_0 = \int_{-\infty}^{\infty} I_0(t)\mathrm{d}t$ is the energy density of the incident radiation; $\Pi_r(t) = \frac{c}{4\pi}\mathrm{Re}[\mathbf{E}(\mathbf{r};t), \mathbf{H}(\mathbf{r};t)]_r$, and the fields are calculated by Eq. (7), for the efficiency factors of scattering Q_s, extinction Q_{ext}, absorption Q_a, and backscattering Q_π of a spherical particle with the diffraction parameter $x = \omega_0 a_0/c$ for the broadband incident radiation, we obtain the following equations [2]:

$$Q(a_0, \omega_0) = \frac{1}{W_0} \int_{-\infty}^{\infty} \left|G(\omega - \omega_0)\right|^2 Q(a_0, \omega)\mathrm{d}\omega, \qquad (9)$$

where $W_0 = \int_{-\infty}^{\infty} \left|G(\omega - \omega_0)\right|^2 \mathrm{d}\omega$, and $Q(x)$ is any of the above spectral scattering efficiency factors[4]:

$$Q_s(x) = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2);$$

$$Q_{ext}(x) = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)\mathrm{Re}(a_n + b_n);$$

$$Q_a(x) = Q_{ext}(x) - Q_s(x);$$

$$Q_\pi(x) = \frac{1}{x^2}\left|\sum_{n=1}^{\infty}(2n+1)(-1)^n(a_n - b_n)\right|^2. \qquad (10)$$

The structure of these equations indicates that every spectral component contributes independently to the total energy of the scattered wave field and these contributions are additive and proportional to the product of the spectral density of the initial radiation by the spectral efficiency factor.

Thus, the problem of determination of these efficiency factors for a non-monochromatic radiation reduces to calculations by Eqs. (10) for all frequencies from the spectrum of the initial pulse and to the operation of convolution by Eq. (9).

Numerically calculated results

For certainty, below we shall consider the scattering of a spectrally limited optical radiation by spherical water droplets in the air. The refractive index of the air is believed to be equal to unity, whereas for water its value is taken to be $m_a = 1.33 - i\cdot 10^{-8}$ at the wavelength $\lambda_0 = 0.8$ μm (Ref. 5). As the quasi-white light, being scattered by particles, we took the supercontinuum radiation, arising upon the self-focusing of the 2-TW Ti:Sapphire-laser pulse (initial duration of 155 fs and wavelength of 0.8 μm) in the air. The spectrum of this pulse, according to the experimental data of Ref. 6, is shown in Fig. 1.

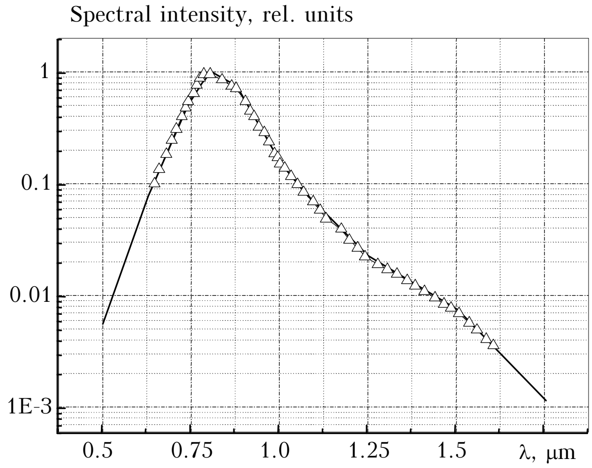


Fig. 1. Spectrum of radiation, arising upon the atmospheric self-focusing of a femtosecond laser pulse: experimental data from Ref. 6 (dots), approximated spectrum used in the calculations (solid curve).

It should be noted that similar spectrum can be described quite well by the Gaussian function with the half-width $\Delta\omega \simeq 1800$ THz, which gives the value of frequency duty factor $l_0 = \omega_0/\Delta\omega$ of the pulse about 1.25. The variation of m_a within the spectrum of the pulse was ignored.

The dependences of the scattering, absorption, and backscattering efficiency factors on the diffraction parameter of the particles for the monochromatic radiation and the quasi-white light are shown in Fig. 2.

For a comparison, Fig. 2 shows also the spectral behavior of these factors for the initial femtosecond pulse with $t_p = 155$ fs ($l_0 \simeq 60$). Analogous dependence for the extinction efficiency factor is omitted here, because the chosen level of the water absorption is very low, and it can be assumed that $Q_{ext} = Q_s$ quite accurately.

It can be seen from Fig. 2 that the wide frequency spectrum of the supercontinuum drastically changes the behavior of the efficiency factors. First, the fine spiking structure in this dependence, which is caused by the excitation of resonance eigenmodes of a transparent particle, disappears. This is especially pronounced in the plot of the dependence of the backscattering efficiency factor $Q_\pi(x)$ (Fig. 2*c*), because the spatial distribution of the field of the resonance modes is characterized by the symmetry in the forward and backward directions[7] and, consequently, the increase of the backscattering signal. This fact does not mean that resonances are no longer excited in the particle, which is evidenced by the presence of resonance spikes in the dependence $Q_a(x)$ (Fig. 2*b*), but the efficiency of this process for the broadband radiation decreases significantly.[3]

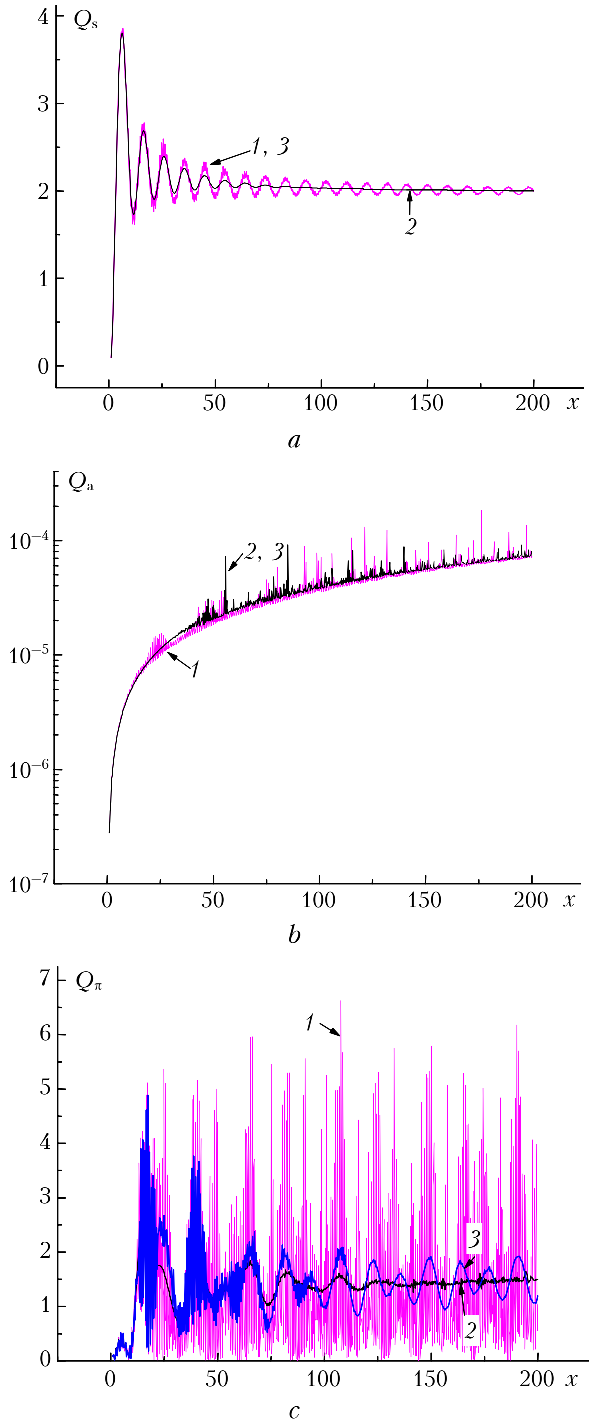


Fig. 2. Efficiency factors for scattering $Q_s(x)$ (*a*), absorption $Q_a(x)$ (*b*), and backscattering $Q_\pi(x)$ (*c*) as functions of the diffraction parameter of the particles x for the monochromatic radiation (*1*), supercontinuum radiation (*2*), having the spectrum as in Fig. 1, and 155-fs laser pulse (*3*). Curves *1*, *3* in Fig. 2*a*, as well as curves *2*, *3* in Fig. 2*b* are very close to each other.

In Figs. 2*a* and *b*, one can see the effect (noticed in Ref. 2) of smoothing of large-scale pulsations of the efficiency factors Q_s, Q_π in the region of large x values at broadening the spectrum of a light pulse. These pulsations are of the interference origin and are characteristic of the monochromatic radiation.[4] The frequency range of the quasi-white light is rather wide, and, as a result, the pulsations of the efficiency factors are damped and approach a constant level.

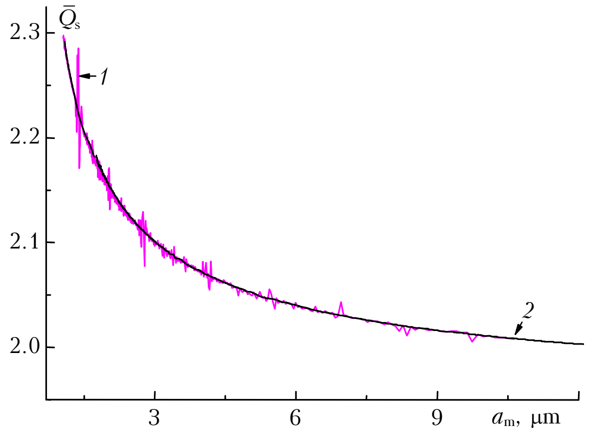
At the same time, for the model 155-fs pulse, marked differences from the case of the monochromatic radiation are seen only in the Q_π and Q_a factors while the spectral behavior of the scattering efficiency factor almost fully copies its monochromatic analog.

The efficiency factors considered above are the main quantitative characteristics of the radiation extinction by isolated spherical particles. The actual atmospheric aerosol formations (fog, mist, clouds) are usually polydisperse and characterized by some particle size distribution function $f(a)$. For such aerosol ensembles, the light scattering theory uses the so-called averaged scattering efficiency factors $\bar{Q}_i$, defined as statistical averages of the corresponding optical characteristic of a single particle Q_i using the density function $\pi a^2 f(a)$ (see Ref. 5):

$$\bar{Q}_i = \frac{\int_0^\infty \pi a^2 Q_i(a) f(a)\mathrm{d}a}{\int_0^\infty \pi a^2 f(a)\mathrm{d}a}. \tag{11}$$

In Eq. (11), the subscript "*i*" denotes any of the factors (10). By its meaning, Eq. (11) gives the ratio of the optical cross section of particles in a unit volume to their geometrical cross section.

Figure 3 compares the averaged efficiency factors $\bar{Q}_s$, $\bar{Q}_\pi$ of the water aerosol with the particle size characteristic of clouds, calculated by Eq. (11) for the monochromatic radiation of $\lambda_0 = 0.8$ μm and the supercontinuum radiation with the frequency spectrum shown in Fig. 1. As the particle size distribution function, we took the gamma distribution with the variable modal radius a_m and the half-width parameter $\mu = 2$. It follows from Fig. 3 that the averaging of the efficiency factors over the ensemble of particles produces the smoothing effect on both the monochromatic radiation and the quasi-white light. However, in the latter case, the degree of manifestation of this effect is much higher, because the statistical averaging is combined with the spectral one, and, in particular, the values of the backscattering efficiency factor almost do not change upon the variation of the cloud microstructure, remaining at the level $\bar{Q}_\pi \simeq 1.5$.



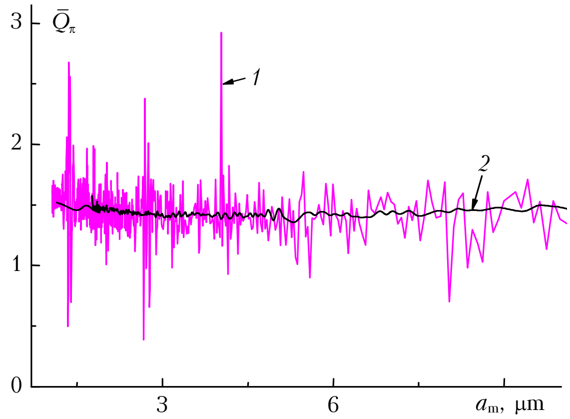


Fig. 3. Averaged efficiency factors of scattering $\bar{Q}_s(x)$ (*a*) and backscattering $\bar{Q}_\pi(x)$ (*b*) for a water cloud with the gamma size distribution of particles ($\mu = 2$) as functions of the modal radius of droplets for the monochromatic radiation (*1*) and a supercontinuum radiation (*2*).

Conclusions

Thus, the presented results on the integral optical characteristics of scattering of a broadband light signal, modeling the actually observed supercontinuum radiation from a femtosecond laser pulse after the interaction with the atmosphere by weakly absorbing spherical particles, have shown that, in the field of this radiation, the most pronounced effect is the smoothing of optical characteristics of light scattering upon the change of the particle radius. One can believe that the factors Q_s, Q_π have weak dependence on the particle size starting roughly from $x = 100$, which, for $\lambda_0 = 0.8$ μm, corresponds to the particle radius $a \simeq 13$ μm.

In a polydisperse ensemble of particles with the size distribution function modeling atmospheric clouds, the averaged scattering efficiency factor $\bar{Q}_s$ decreases smoothly with the increase of the modal radius of particles and tends asymptotically to $\bar{Q}_s = 2$, while the averaged backscattering efficiency factor $\bar{Q}_\pi$ varies slightly near the constant level $\bar{Q}_\pi = 1.5$.

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