Nonstationary elastic linear light scattering by spherical microparticles

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A theoretical approach is developed to solve the problem of nonstationary elastic light scattering by a dielectric spherical particle. This approach proposes optical fields of the scattering radiation to be represented as an expansion in terms of eigenfunctions of the stationary problem, in which the expansion coefficients determine the temporal behavior of the field and comply with inhomogeneous oscillation equations. Transient processes at formation of optical fields in a microparticle are studied. It is shown that nonstationary pulse scattering manifests itself, first of all, in the time shift of maximum of the internal field relative to the profile of the initial pulse and in the time delay of its trailing edge. This behavior of nonstationary fields is connected with the resonance character of the process of elastic light scattering by a particle, in which vibrational eigenmodes of the internal optical field with lifetimes comparable with or much longer than the laser pulse duration are excited.

In spite of almost century-long history, the problem of elastic linear scattering of optical radiation by dielectric spherical microparticles remains urgent by now. It is known that the basic principles of this theory formulated still in papers by Debye[1] and Mie[2] and then developed in Refs. 3–6 are concerned with diffraction of a plane monochromatic light wave at a particle under stationary conditions.

As applied to the process of elastic light scattering by a particle, the condition of stationarity means that the time of establishment of optical fields in a particle and beyond it is much shorter than the duration of the diffracting radiation pulse and, consequently, the establishment of optical fields can be considered as instantaneous. At the same time, light scattering, as any other physical process, always has nonstationary phases in its development. This circumstance is especially important in connection with promises of applying ultrashort laser pulses in aerosol optics.[7] In such time scales, the nonstationarity of the scattering process becomes comparable with the duration of a radiation pulse.

The studies of the temporal and spectral structures of the field of elastic scattering by weakly absorbing spherical particles[8] revealed the existence of free electromagnetic oscillations in dielectric spheres, whose frequencies are determined by the particle size and optical properties. If the frequency of the incident radiation coincides with the frequency of some particle eigenmode, an internal optical field is resonantly excited, and the spatiotemporal distribution of this field is completely determined by the field of the excited mode. Characteristic lifetimes τ_R of the highest-Q resonances (whispering gallery modes) in micron-sized particles usually lie in the nanosecond region.[9] Thus, if the length of the initial radiation pulse is comparable with and shorter than the time τ_R, then its scattering by a particle may have the nonstationary character.

Theoretical investigations of nonstationary light scattering are based on solutions of Maxwell's equations in their complete form with allowance for the temporal variability of the fields. A well-known approach to solution of this problem is the method of the spectral Fourier analysis.[10–13] It allows the problem on nonstationary scattering of a pulse with a spectral distribution to be reduced to scattering of a set of monochromatic Fourier harmonics. In this case, particle scattering properties are characterized by the so-called spectral response function, which is a traditional Mie series written for all frequencies of the initial pulse spectrum. The scattered and internal fields are written in the form of the convolution integral of the pulse spectrum and the spectral response function of the particle.[13] The analytical solution of this light scattering problem was obtained only for some particular cases (optically small particles[10]), when the spectral response function has a quite simple form. Some numerical solutions of this problem that describe the behavior of the internal[11,13] and external[12] fields of the scattered wave were obtained as well.

Among various numerical methods, to be noted is the finite-difference time domain method, which is, in fact, the direct numerical solution of the nonstationary Maxwell equations.[14,15] It is worth using this method in calculations of light diffraction at objects having a complex geometry, as well as inhomogeneities in their optical properties.

In our previous papers,[9,16] when studying the nonstationary problem of nonlinear light scattering in micron-sized particles, we justified the approach being under development in laser optics.[17,18] In this

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approach, it is proposed to seek the solution in the form of expansion in terms of eigenfunctions of the linear problem of stationary scattering (resonance modes of a sphere). In this case, the spatial and temporal dependences of the fields are factored so that all information about the time behavior of the scattered field is in the coefficients of the expansion series. The system of differential equations being, in essence, inhomogeneous equations of oscillations can be written for them based on the Maxwell equations. If a particular profile of the initial pulse is specified, this system can be then solved analytically or numerically.

The aim of this work is theoretical investigation of the nonstationary elastic linear scattering of light by a dielectric microparticle based on the field expansion in terms of its resonance modes. Our tasks are the detailed description of the method, derivation of equations characterizing the main features of nonstationary scattering, and numerical computations that quantitatively characterize the process under study.

The Maxwell equations describing nonstationary elastic linear scattering of light have the following form:

$$\mathrm{rot}\mathbf{E}(\mathbf{r};\, t) = -\frac{1}{c}\frac{\partial}{\partial t}\mathbf{H}(\mathbf{r};\, t);$$

$$\mathrm{rot}\mathbf{H}(\mathbf{r};\, t) = -\frac{\varepsilon_a}{c}\frac{\partial}{\partial t}\mathbf{E}(\mathbf{r};\, t) + \frac{4\pi\sigma}{c}\mathbf{E}(\mathbf{r};\, t), \qquad (1)$$

where $\mathbf{E}(\mathbf{r};\, t)$ and $\mathbf{H}(\mathbf{r};\, t)$ are the electric and magnetic field vectors, respectively; ε_a and σ are the permittivity and conductivity of the particle substance; c is the speed of light in vacuum. We consider here the complex representation of the fields.

Representation of the spherical particle as an open optical resonator allows the solution of Eqs. (1) to be sought in the form of expansion in terms of the system of eigenfunctions of such a resonator $\mathbf{E}_{np}^{\mathrm{TE,TH}}(\mathbf{r}), \mathbf{H}_{np}^{\mathrm{TE,TH}}(\mathbf{r})$ describing the spatial profile of the fields of vibrational eigenmodes of TE and TH polarizations[19,20]:

$$\mathbf{E}(\mathbf{r};\, t) = \sum_{n=1}^{\infty}\sum_{p=1}^{\infty}\left[A_{np}(t)\mathbf{E}_{np}^{\mathrm{TE}}(\mathbf{r}) - iB_{np}(t)\mathbf{E}_{np}^{\mathrm{TH}}(\mathbf{r})\right];$$

$$\mathbf{H}(\mathbf{r};\, t) = \sqrt{\varepsilon_a}\sum_{n=1}^{\infty}\sum_{p=1}^{\infty}\left[iA_{np}(t)\mathbf{H}_{np}^{\mathrm{TE}}(\mathbf{r}) + B_{np}(t)\mathbf{H}_{np}^{\mathrm{TH}}(\mathbf{r})\right], \qquad (2)$$

where still unknown coefficients $A_{np}(t)$ and $B_{np}(t)$ reflect the time behavior of the field. As is well-known, the functions $\mathbf{E}_{np}$ and $\mathbf{H}_{np}$ themselves form an orthogonal system, comply with the Maxwell equations (1):

$$\mathrm{rot}\mathbf{E}_{np} = -i\frac{\omega_{np}}{c}\mathbf{H}_{np}; \quad \mathrm{rot}\mathbf{H}_{np} = i\frac{\varepsilon_a\omega_{np}}{c}\mathbf{E}_{np} \qquad (3)$$

with the eigenfrequencies ω_{np}, and can be expressed through the vector spherical harmonics $\mathbf{M}_{np}(r, \theta, \varphi)$ and $\mathbf{N}_{np}(r, \theta, \varphi)$:

$$\mathbf{E}_{np}^{\mathrm{TE}} = c_{np}(x_{np})\mathbf{M}_{np}(r,\theta,\varphi);$$
$$\mathbf{E}_{np}^{\mathrm{TH}} = d_{np}(x_{np})\mathbf{N}_{np}(r,\theta,\varphi);$$
$$\mathbf{H}_{np}^{\mathrm{TE}} = d_{np}(x_{np})\mathbf{N}_{np}(r,\theta,\varphi);$$
$$\mathbf{H}_{np}^{\mathrm{TH}} = c_{np}(x_{np})\mathbf{M}_{np}(r,\theta,\varphi), \qquad (4)$$

where $x_{np} = \omega_{np}a_0/c$ is the resonance parameter of diffraction of the particle with the radius a_0, and the scale parameters c_{np} and d_{np} are determined from the condition of unit normalization of vector harmonics. The specific equations for the functions $\mathbf{M}_{np}(r, \theta, \varphi)$ and $\mathbf{N}_{np}(r, \theta, \varphi)$, as well as their main properties are given, for example, in Ref. 20.

Prior to further consideration, let us give some notes concerning the form and applicability of the series (2) to solution of the problem analyzed.

1. The main feature of a dielectric particle as compared to traditional metal-wall resonators is its openness. This term means that the electromagnetic field of eigenmodes is nonzero on the particle surface and exists beyond the particle as well. As a result, the resonator eigenfrequencies ω_{np} become complex: $\omega_{np} = \omega'_{np} - i\,\omega''_{np}$ with the real part determining the frequency of oscillations of eigenmode fields and the imaginary part bearing the information about the amplitude of the outgoing field.[18] In this connection, far from the particle $r \gg 1$ the harmonic part of the scattering wave $\exp\{i\,\omega_{np}\,r/c\}$ leads, apparently, to the infinite growth of its amplitude. Therefore, strictly speaking, the use of the system of eigenfunctions (2) for description of fields in open resonators is incorrect. However, since the imaginary part of the frequency ω''_{np} is determined by the parameter of the radiation Q-factor Q_{np}^R (Ref. 9): $\omega''_{np} \sim 1/Q_{np}^R$, whose characteristic values in transparent micron-sized particles are usually of the order of $Q_{np}^R \sim 10^6$–10^{15}, at $r \ll c/\omega''_{np}$ the series (2) can be thought converging within the partial zone.[21]

2. It is worth representing electromagnetic fields in the form (2) only when the functions, in terms of which they are expanded, are known. The use of spherical vector harmonics (4) as generating functions imposes certain restrictions on the spatial profile of particle permittivity, namely, we assume $\varepsilon_a = \mathrm{const}$ at $r \le a_0$. The fact that ε_a is spatially varying within the particle volume requires the problem for eigenfunctions of such a system to be solved before expansion, but this is a nontrivial problem. However, in some cases, for example, in the case of radially inhomogeneous particles consisting of concentric layers with different values of ε_a, it is possible to construct the system of eigenfunctions similar to Eqs. (4), but more cumbersome.[6,22,23]

Another obvious restriction of the considered approach to solution of the problem is a neglect of the frequency dispersion of the dielectric permittivity, because the form itself of the field expansion (2) in the presence of the dependence $\varepsilon_a(\omega)$ contradicts the

requirements of physical validity of the eigenfunctions (4). It should be noted that, although this circumstance reduces the domain of applicability of the series expansion (2), the cases, in which the allowance for ε_a dispersion is significant, are rather rare in practice, if only we do not consider strong spectral resonances of the particulate substance.

3. Natural oscillation frequencies ω_{np} are determined from solution of the Dirichlet problem for a spherical zone V_0 (see, for example, Refs. 18 and 19). In the case of an ideal sphere, they, as is known, are characterized by only two indices (besides the wave polarization TE or TH): the mode number n (angular index by the coordinate θ) and order p (radial index by the coordinate r). In the azimuth direction (by φ), the eigenfrequencies of the sphere are degenerate. Because of peculiarities of the spherical Bessel functions characterizing the radial behavior of the field of natural oscillation of a sphere, it is impossible to obtain the exact analytical equation for ω_{np}. Only approximate formulae are known.[18,24] They were obtained by approximation of the spherical Bessel functions at large values of the argument by the Airy function. Using them, we can calculate eigenfrequencies of a dielectric sphere.

4. The boundary conditions on the surface of a spherical particle consist in continuity of the tangential components of the fields $E_{\theta,\varphi}$ and $H_{\theta,\varphi}$ at transition through the surface:

$$E_{\theta,\varphi} = E^{\mathrm{s}}_{\theta,\varphi} + E^{\mathrm{i}}_{\theta,\varphi};\ H_{\theta,\varphi} = H^{\mathrm{s}}_{\theta,\varphi} + H^{\mathrm{i}}_{\theta,\varphi}, \qquad (5)$$

where the superscripts s and i stand respectively for the scattered and incident fields. But the normal components of the fields experience a jump proportional to the relative refractive index $m = m_a/m_0$, where m_a and m_0 are the complex refractive indices of the particle and the ambient medium, respectively. This circumstance prevents from the direct use of the field expansion (2) under the rot sign. Instead, the correct approach is expansion of the functions rot$\mathbf{E}(\mathbf{r}; t)$ and rot$\mathbf{H}(\mathbf{r}; t)$ in terms of Eqs. (4) and derivation of the equation for the temporal coefficients of the expansion.

For simplicity, the further consideration is conducted for waves of some particular polarization, for example, TE waves and, respectively, for the coefficients $A_{np}(t)$. It should be noted that because of symmetry of the Maxwell equations, the resulting equations for $B_{np}(t)$ are similar.

Following Ref. 17, let us multiply the first of Eqs. (1) by $\mathbf{H}^*_{np}$ from the right and integrate over the particle volume V_0, taking into account Eq. (3), as well as orthogonality and normalization of eigenfunctions:

$$\int_{V_0} (\mathrm{rot}\,\mathbf{E}\cdot\mathbf{H}^*_{np})\,\mathrm{d}\mathbf{r} = \int_{V_0} (\mathbf{E}\cdot\mathrm{rot}\,\mathbf{H}^*_{np})\,\mathrm{d}\mathbf{r} + \int_{S_0} [\mathbf{E}\times\mathbf{H}^*_{np}]\cdot\mathbf{n}_r\,\mathrm{d}\mathbf{s} = -i\frac{\varepsilon_a\omega_{np}}{c}A_{np}(t) + \int_{S_0} [\mathbf{E}\times\mathbf{H}^*_{np}]\cdot\mathbf{n}_r\,\mathrm{d}\mathbf{s} = -\frac{\varepsilon_a}{c}\frac{\mathrm{d}}{\mathrm{d}t}A_{np}(t), \qquad (6)$$

where $\mathrm{d}\mathbf{r} = r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\varphi$; $\mathrm{d}\mathbf{s} = a_0^2\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi$; $\mathbf{n}_r$ is the external normal to the particle surface; S_0 is the surface bounding the volume V_0. Then make the same operation with the second equation of Eqs. (1), but multiply it by $\mathbf{E}^*_{np}$:

$$\int_{V_0} (\mathrm{rot}\mathbf{H}\cdot\mathbf{E}^*_{np})\,\mathrm{d}\mathbf{r} = \int_{V_0} (\mathbf{H}\cdot\mathrm{rot}\mathbf{E}^*_{np})\,\mathrm{d}\mathbf{r} + \int_{S_0} [\mathbf{H}\times\mathbf{E}^*_{np}]\cdot\mathbf{n}_r\,\mathrm{d}\mathbf{s} = -\frac{\sqrt{\varepsilon_a}\,\omega_{np}}{c}A_{np}(t) + \int_{S_0} [\mathbf{H}\times\mathbf{E}^*_{np}]\cdot\mathbf{n}_r\,\mathrm{d}\mathbf{s} = \frac{\varepsilon_a}{c}\frac{\mathrm{d}}{\mathrm{d}t}A_{np}(t) + \frac{4\pi\sigma}{c}A_{np}(t). \qquad (7)$$

Differentiate Eq. (7) with respect to time and in place of the derivative $\frac{\mathrm{d}}{\mathrm{d}t}A_{np}(t)$ substitute its expression from Eq. (6). After re-grouping of terms, we obtain the following equation for the coefficients A_{np}:

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}A_{np}(t) + \frac{4\pi\sigma}{\varepsilon_a}\frac{\mathrm{d}}{\mathrm{d}t}A_{np}(t) + \omega^2_{np}A_{np}(t) = \Pi_{np}(t), \qquad (8)$$

where

$$\Pi_{np}(t) = -\frac{ic}{\varepsilon_a}\left[\int_{S_0}\left\{\omega_{np}[\mathbf{E}\times\mathbf{H}^*_{np}] - i\frac{\partial}{\partial t}[\mathbf{H}\times\mathbf{E}^*_{np}]\right\}\mathrm{d}\mathbf{s}\right]. \qquad (9)$$

In its meaning, Eq. (8) is the equation of oscillations, in which the driving force is represented by a combination of the surface integrals $\Pi_{np}(t)$. Determine their form using the boundary condition (5). Then we obtain

$$\Pi_{np}(t) = -\frac{ic}{\varepsilon_a}\left[\int_{S_0}\left\{\omega_{np}[\mathbf{E}^{\mathrm{s}}\times\mathbf{H}^*_{np}] - i\frac{\partial}{\partial t}[\mathbf{H}^{\mathrm{s}}\times\mathbf{E}^*_{np}]\right\}\mathrm{d}\mathbf{s}\right] + F^{\mathrm{i}}_{np}(t). \qquad (10)$$

Here $F^{\mathrm{i}}_{np}(t)$ has the form similar to the first term, but with the field of the incident radiation in place of the field of the scattered wave in vector products.

After simple transformations, the first term in Eq. (10) is expressed through the radiation Q-factor of the particle Q^R_{np} at the eigenmode frequency:

$$\Pi_{np}(t) = 2i\omega^2_{np}A_{np}(t)\left(1 + \frac{i}{2Q^R_{np}}\right) + F^{\mathrm{i}}_{np}(t), \qquad (11)$$

where

$$Q_{np}^R = \omega_{np} W_{np} / P_{np}^R , \quad (12)$$

and the energy stored in the mode W_{np} and the averaged power of radiation loss through the particle surface P_{np}^R have the following forms:

$$W_{np} = \frac{1}{16\pi} \int_{V_0} (\varepsilon_a \mathbf{E}_{np} \mathbf{E}_{np}^* + \mathbf{H}_{np} \mathbf{H}_{np}^*) \, \mathrm{d}\mathbf{r} ; \quad (13)$$

$$P_{np}^R = \frac{c}{8\pi} \mathrm{Re} \left\{ \int_{S_0} [\mathbf{E}_{np}^s \times (\mathbf{H}_{np}^s)^*] \mathrm{d}\mathbf{s} \right\} . \quad (14)$$

After substitution of Eq. (11) into Eq. (8), we obtain

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2} A_{np}(t) + \frac{\omega_{np}}{Q_{np}^a} \frac{\mathrm{d}}{\mathrm{d}t} A_{np}(t) + \left[\omega_{np} \left(1 + \frac{i}{Q_{np}^R} \right) \right]^2 A_{np}(t) = F_{np}^{\mathrm{i}}(t), \quad (15)$$

where, similarly to Eq. (12), the Q-factor of the resonance mode Q_{np}^a caused only by thermal loss in the particulate substance can be expressed through the thermal loss power P_{np}^a:

$$P_{np}^a = \int_{V_0} \frac{\sigma}{2} (\mathbf{E}_{np} \cdot \mathbf{E}_{np}^*) \mathrm{d}\mathbf{r} . \quad (16)$$

The final stage is determination of the specific form of the external force $F_{np}^{\mathrm{i}}(t)$ connected with the effect of the incident radiation. Assume for definiteness that the light field diffracting at the particle is a plane circularly polarized wave propagating along the axis z with the amplitude temporal profile set by the function

$$f(t) = \tilde{f}(t) \exp \{i\omega_0 t\}, \quad (17)$$

where $\tilde{f}(t)$ is the slowly varying function of time. That is, with allowance for the delay of the field at the point with the coordinate z, we have:

$$\mathbf{E}^i(\mathbf{r};t) = E_0 (\mathbf{e}_x + i\mathbf{e}_y) \tilde{f} \left(t - \frac{z + a_0}{c} \right) \exp\{i\omega_0 t - ik_0 (z + a_0)\}. \quad (18)$$

Here $\mathbf{e}_x$ and $\mathbf{e}_y$ are unit vectors; E_0 is the wave amplitude; $k_0 = \omega_0 / c$. The coordinate system is located at the center of the particle with the radius a_0.

Let us pass in Eq. (18) from the shifted time to the actual one using the Fourier transformation and use the well-known plane wave expansion in terms of spherical harmonics.[20] Then we obtain

$$\mathbf{E}^i(\mathbf{r}; t) = \frac{E_0}{2\pi} \int_{-\infty}^{\infty} G(\omega - \omega_0) \mathrm{e}^{i\omega t - ika_0} \sum_n R_n \left[\mathbf{M}_{1n}^{(1)}(kr) - i\mathbf{N}_{1n}^{(1)}(kr) \right] \mathrm{d}\omega ;$$

$$\mathbf{H}^i(\mathbf{r};t) = \sqrt{\varepsilon_a} \frac{E_0}{2\pi} \int_{-\infty}^{\infty} G(\omega - \omega_0) \mathrm{e}^{i\omega t - ika_0} \sum_n R_n \left[i\mathbf{N}_{1n}^{(1)}(kr) + \mathbf{M}_{1n}^{(1)}(kr) \right] \mathrm{d}\omega , \quad (19)$$

where $R_n = i^n (2n + 1) / [n(n + 1)]$; $k = \omega / c$; $G(\omega - \omega_0)$ is the Fourier transform of $f(t)$, and $\mathbf{M}_{1n}^{(1)}$, $\mathbf{N}_{1n}^{(1)}$ are spherical vector harmonics at the azimuth index m equal to unity. We still consider only TE modes. It should be noted that the type of polarization of the initial wave influences only the particular form of functions $\mathbf{M}_{1n}^{(1)}, \mathbf{N}_{1n}^{(1)}$ (Ref. 20).

It is obvious that in this case, because of mutual orthogonality of spherical harmonics, the sum in Eq. (19) contains only the terms with the index n equal to the index of the considered mode of the internal field, and for the external force we have the following equation:

$$F_{np}^{\mathrm{i}}(t) = \frac{E_0}{2\pi} \int_{-\infty}^{\infty} G(\omega - \omega_0) \mathrm{e}^{i\omega t} K_{np}^n(\omega; \omega_{np}) \mathrm{d}\omega, \quad (20)$$

where the coefficient $K_{np}^n(\omega; \omega_{np})$ accounts for the degree of excitation of the internal field mode by the corresponding external field mode:

$$K_{np}^n(\omega; \omega_{np}) = \frac{ic^2 R_n \mathrm{e}^{-ika_0}}{V_0 \varepsilon_a k c_{np}} \times \left[\psi_n(ka_0) \, \psi_n'^*(n_a k_{np} a_0) - \frac{1}{n_a} \frac{\omega}{\omega_{np}} \psi_n'(ka_0) \, \psi_n^*(n_a k_{np} a_0) \right].$$

For further analysis, it is convenient to modify Eq. (15), reducing it to the canonical form of the equation of forced oscillations:

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2} A_{np}(t) + 2\Gamma_{np} \frac{\mathrm{d}}{\mathrm{d}t} A_{np}(t) + \omega_{np}^2 A_{np}(t) = F_{np}^{\mathrm{i}}(t) , \quad (21)$$

which follows from Eq. (15) at the mode damping coefficient $\Gamma_{np} = \frac{\omega_{np}}{2Q_{np}}$ and $Q_{np}^R >> 1$. Here we introduce the parameter of the summarized Q-factor of the particle-resonator

$$\frac{1}{Q_{np}} = \frac{1}{Q_{np}^a} + \frac{1}{Q_{np}^R} , \quad (22)$$

that accounts for the total loss of the mode for absorption and emission of the light wave.

In the general form, the particular solution of the inhomogeneous equation (21) representing only oscillations under the action of the external force can be written as

$$A_{np}(t)=\frac{\exp\{-i\Gamma_{np}t\}}{2i\tilde{\omega}_{np}}E_0\Bigg[\exp\{i\tilde{\omega}_{np}t\}\times$$

$$\times\int_0^t F_{np}^{\mathrm{i}}(t')\exp\{-i(\tilde{\omega}_{np}+i\Gamma_{np})t'\}\mathrm{d}t'\Bigg]-$$

$$-\frac{\exp\{-i\Gamma_{np}t\}}{2i\tilde{\omega}_{np}}E_0\Bigg[\exp\{-i\tilde{\omega}_{np}t\}\times$$

$$\times\int_0^t F_{np}^{\mathrm{i}}(t')\exp\{i(\tilde{\omega}_{np}-i\Gamma_{np})\ t'\}\mathrm{d}t'\Bigg], \qquad (23)$$

where $\tilde{\omega}_{np}=\omega_{np}\sqrt{1-\Gamma_{np}^2/\omega_{np}^2}$ is the frequency of natural oscillations of the mode with allowance for loss.

Specify the Gaussian temporal profile of the radiation pulse

$$\tilde{f}(t)=\exp\{-(t-t_0)^2/t_{\mathrm{p}}^2\}.$$

In this case, as is known,

$$G(\omega-\omega_0)=\frac{4\pi\sqrt{\pi}}{\Delta\omega_{\mathrm{p}}}\exp\left\{-4\pi^2\frac{(\omega-\omega_0)^2}{(\Delta\omega_{\mathrm{p}})^2}-i\omega t_0\right\}, \quad (24)$$

where t_0 and t_{p} are parameters; $\Delta\omega_{\mathrm{p}}=4\pi/t_{\mathrm{p}}$ is the spectral halfwidth of the pulse. Substitute Eq. (24) into Eq. (23) and, applying the average theorem, we obtain the equation for the temporal dependence of the amplitude coefficient of the internal field mode:

$$A_{np}(\tau)\cong\frac{\sqrt{\pi}K_{np}^n(\overline{\omega};\omega_{np})E_0}{i\tilde{\omega}_{np}\Delta\omega_{\mathrm{p}}}\exp\{-i\omega_0t_0\}\times$$

$$\times\exp\left\{-4\pi^2\frac{(\delta\omega_{np}+i\Gamma_{np})^2}{(\Delta\omega_{\mathrm{p}})^2}\right\}\exp\{\tau(i\tilde{\omega}_{np}-\Gamma_{np})\}\times$$

$$\times\left[\mathrm{erf}\left(\frac{\tau+2t_0}{t_{\mathrm{p}}}-\frac{4\pi}{\Delta\omega_{\mathrm{p}}}\left[\Gamma_{np}-i\frac{\delta\omega_{np}}{2}\right]\right)+1\right]+$$

$$+O\left(\exp\left\{-\frac{\omega_{np}^2}{(\Delta\omega_{\mathrm{p}})^2}\right\}\right). \qquad (25)$$

Here $\tau=t-t_0$; $\delta\omega_{np}=\omega_0-\tilde{\omega}_{np}$; $\overline{\omega}$ is some frequency inside the spectral profile of the pulse.

Let us analyze this equation. As can be seen, the temporal dependence of the amplitude of the natural oscillation is determined by the exponential drop function with the mode damping coefficient Γ_{np} and the probability integral of a complex argument. Its imaginary part is connected with detuning of the mode frequency from the frequency of the incident radiation, and the real part gives the characteristic time, for which the amplitude reaches the maximum value τ_{m} that can be determined, based on the erf(x) properties, as follows:

$$\tau_{\mathrm{m}}\approx t_{\mathrm{p}}^2\left(\frac{1}{t_{\mathrm{p}}}+\frac{1}{\tau_{np}}\right)=t_{\mathrm{p}}(1+\gamma) \quad \text{at } \gamma\le 1,$$

$$\tau_{\mathrm{m}}\approx\tau_{np} \quad \text{at} \quad \gamma\gg 1, \qquad (26)$$

where $\tau_{np}=1/\Gamma_{np}$ is the characteristic lifetime of the mode; $\gamma=(t_{\mathrm{p}}/\tau_{np})$. The equation derived indicates that the time, for which the particle eigenmode amplitude reaches the maximum, depends on the relation between the characteristic laser pulse duration and the lifetime of the mode γ and is determined, in the general case, by the shortest of them.

As the pulse duration shortens, the value of the maximum (in time) amplitude of the natural oscillation varies by the following law:

$$A_{\max}(\gamma)\sim\gamma\exp\{-\gamma\},$$

which reflects the decrease in the fraction of the pulse spectral energy used for excitation of the selected resonance mode. In the other limit, in the region of long pulses $\gamma\gg 1$, from Eq. (23) we have the corresponding solution for the continuous-wave radiation (the delay here is not of importance):

$$A_{np}(t)\cong\frac{K_{np}^nE_0}{2\tilde{\omega}_{np}(\delta\omega_{np}-i\Gamma_{np})}\times$$

$$\times\exp\{it(\tilde{\omega}_{np}+i\Gamma_{np})\}\left[\exp\{it(\delta\omega_{np}-i\Gamma_{np})\}-1\right] \quad (27)$$

and the oscillation amplitude reaches the maximum value

$$A_{\max}(\infty)=K_{np}^nE_0/(2\tilde{\omega}_{np}\Gamma_{np}).$$

Coming back to Eq. (25), it should be also noted that the eigenmode field amplitude is also affected by its spectral position relative to the spectrum of the initial pulse. The larger is the frequency detuning of the mode $\delta\omega_{np}$, the worse is its excitation. If the pulse spectrum is rather wide, that is $\Delta\omega_{\mathrm{p}}>\delta\omega_{np}$, and the conditions are favorable for excitation of several eigenmodes, then the resulting temporal dependence of the field amplitude is determined by their superposition and has a characteristic beating shape.

Figure 1 shows the time dependence of the relative (normalized to the maximum value) intensity of the internal field $I(\tau)$ calculated with allowance for Eq. (25) at an arbitrary point inside the particle. Two model versions of the mode excitation are considered: resonance of a single mode and non-resonance excitation of three neighboring modes with different resonance characteristics (amplitudes of all modes were assumed equal to unity): $Q_1<Q_2<Q_3$; $\omega_1<\omega_2<\omega_3$. It can be seen that in the last case the dependence $I(\tau)$ at $\tau>t_{\mathrm{p}}$ has two characteristic parts. Quickly damping modes form the initial part of the drastic drop of the field intensity, while the mode with the highest Q-factor (Q_3) determines the long tail in this dependence, whose duration depends on the lifetime of this mode τ_3.

It is obvious that if the inequality $t_p < \tau_3$ is fulfilled, the optical field in the particle exists even after termination of the initial radiation pulse.

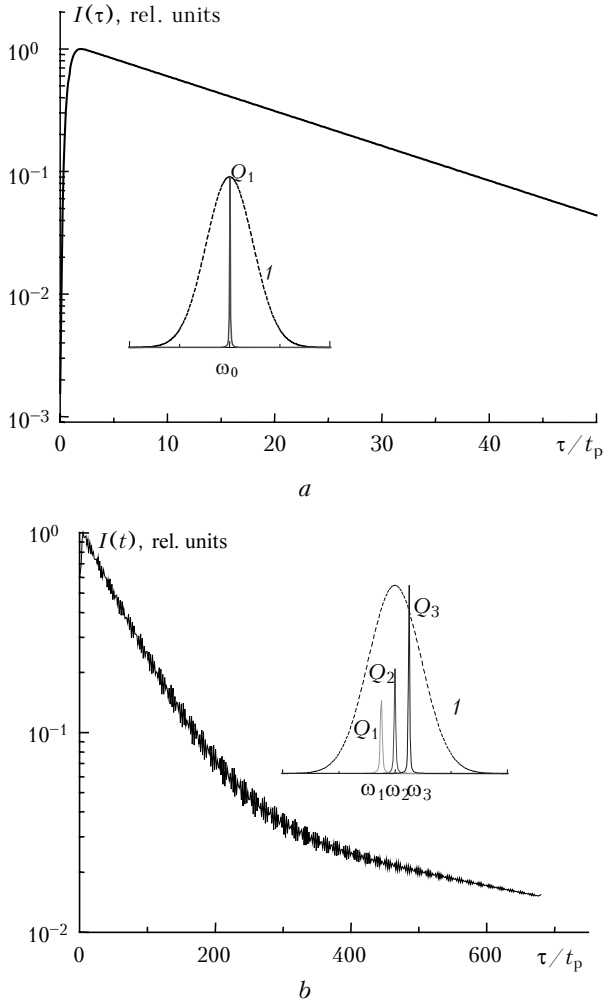

Fig. 1. Model dependence of the internal field of the spherical particle at excitation of a single resonance mode (*a*) and three modes (*b*). The spectral position of the mode is shown schematically in small fragments; the pulse spectrum is plotted as curve *1*.

The last statement directly follows from the solution obtained for the coefficients of the optical field expansion (25). At the same time, it is somewhat unusual for the linear theory of light diffraction at a spherical particle that traditionally considers the scattering process under stationary conditions: $t_p >> \tau_{np}$. This allows us to speak that nonstationary elastic scattering of light by a particle has a resonance character, and the shorter the excitation pulse, the more pronounced this resonance character. Since characteristic lifetimes of eigenmodes in weakly absorbing particles can reach $\sim 10^{-8}$–10^{-9} s (Ref. 9), scattering of pico- and femto-second laser pulses can be supposed resonance.

In the case of diffraction of long pulses $t_p >> \tau_{np}$ at a particle, the t_0 field of the TE mode inside the particle with allowance for Eq. (27) can be written as

$$\mathbf{E}(\mathbf{r};t) = E_0 \exp\{i\omega_0 t\} \sum_{n,p} \frac{K_{np}^n c_{np}}{2\hat{\omega}_{np}(\delta\omega_{np} - i\Gamma_{np})} \mathbf{M}_{np}(\mathbf{r}) . \quad (28)$$

However, as is known, the Mie theory in this case gives

$$\mathbf{E}(\mathbf{r};t) = E_0 \exp\{i\omega_0 t\} \sum_n c_n^{\text{Mie}}(x_a) \mathbf{M}_n(\mathbf{r}) . \quad (29)$$

Here c_n^{Mie} are the Mie coefficients.[20] Comparing Eqs. (28) and (29), we obtain

$$c_n^{\text{Mie}}(x_a) = \sum_p c_n^{\text{Mie}}(x_{np}) \frac{\Gamma_{np}}{(\delta\omega_{np} - i\Gamma_{np})} , \quad (30)$$

where $c_n^{\text{Mie}}(x_{np}) = K_{np}^n c_{np} / (2\Gamma_{np}\hat{\omega}_{np})$ is the resonance value of the Mie coefficient at $\omega_0 = \hat{\omega}_{np}$.

Equation (30) demonstrates the relation between the above expansion of the electromagnetic field in terms of the particle eigenmodes (the so-called Fourier–Bessel series[19]) and the traditional representation of the field as a superposition of partial waves in the Debye theory. It follows from this equation that the information about natural resonances of the particle is directly contained in the Mie coefficients in the form of the infinite sum of functions of resonance profiles. This fact is illustrated in Fig. 2, which depicts the absolute value of one of the Mie coefficients at the varying particle diffraction parameter x_a. Numerical calculation was based on Eq. (30).

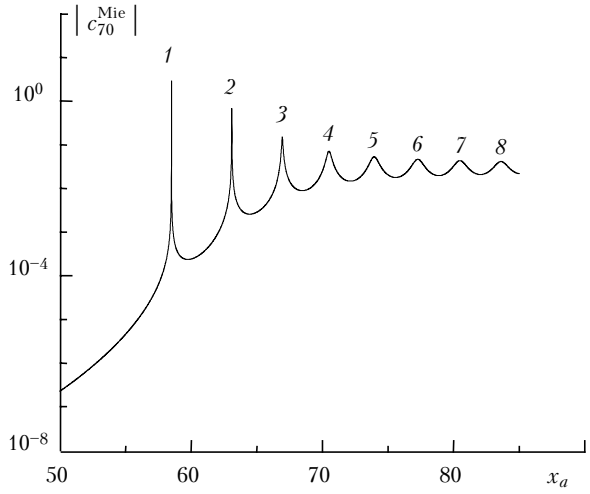

Fig. 2. Absolute value of the Mie coefficient $|c_{70}^{\text{Mie}}|$ as a function of the particle diffraction parameter $x_a = 2\pi a_0/\lambda$. Digits enumerate the orders of resonance modes.

The above consideration of regularities in transition processes of light wave diffraction at a particle would be incomplete without discussing the evolution of the electromagnetic field of the scattered wave. The direct use of the same technique for

expansion of the scattered field in terms of eigenfunctions of the particle-resonator in the form (2) as for the internal field is apparently incorrect, because the domain of definition of resonance modes is bounded by a surface close to the particle surface (see the above notes). Consequently, it is beyond reason to describe the field at an arbitrary separation from the particle by the functions (4).

This problem can be solved by passing from consideration of wave diffraction at a spherical object to wave emission by a spherical object with a preset (known) distribution of the electromagnetic field. Following Refs. 4 and 9, we consider briefly the main stages of solution of this problem with the so-called method of integro-differential equation.

Under study is the Helmholtz equation for the Hertz vector $\mathbf{\Pi}_e(\mathbf{r}; t)$ (the similar equation for the vector potential of the field can be considered as well[9])

$$\Delta\mathbf{\Pi}_e(\mathbf{r}; t)-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{\Pi}_e(\mathbf{r}; t)=-4\pi\mathbf{P}(\mathbf{r}; t)\,, \qquad (31)$$

which follows from the Maxwell equations (1), and the vector $\mathbf{\Pi}_e(\mathbf{r}; t)$ is determined through electromagnetic field vectors as

$$\mathbf{E}(\mathbf{r}; t)=-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{\Pi}_e(\mathbf{r}; t)+\nabla\mathrm{div}\,\mathbf{\Pi}_e(\mathbf{r}; t);$$

$$\mathbf{H}(\mathbf{r}; t)=-\frac{1}{c}\frac{\partial}{\partial t}\mathrm{rot}\mathbf{\Pi}_e(\mathbf{r}; t)\,. \qquad (32)$$

Here

$$\mathbf{P}(\mathbf{r}; t)=\left[\frac{\varepsilon_a-1}{4\pi}-i\frac{\sigma}{\omega}\right]\mathbf{E}(\mathbf{r}; t)$$

is polarization of a substance. The solution of Eq. (31) is known and can be written in the form of a delay integral at the spatial point with the radius vector $\mathbf{r}$:

$$\mathbf{\Pi}_e(\mathbf{r}; t)=\int_{V_0}\frac{\mathbf{P}(\mathbf{r}'; t)}{R}\exp\{-ikR\}\,d\mathbf{r}'\,, \qquad (33)$$

where $R=|\mathbf{r}-\mathbf{r}'|$ is the distance between an observation point and an elementary source. The integral is taken over the whole volume occupied by sources of the scattered wave (particle volume V_0).

For the total field beyond the particle $\mathbf{E}(\mathbf{r}; t)$, it follows from Eqs. (33) and (32) that

$$\mathbf{E}(\mathbf{r}; t)=\mathbf{E}^i(\mathbf{r}; t)+\mathbf{E}^s(\mathbf{r}; t)=$$

$$=\mathbf{E}^i(\mathbf{r}; t)+\mathrm{rot}\,\mathrm{rot}\int_{V_0}\frac{(\varepsilon_a-1)\mathbf{E}(\mathbf{r}'; t)}{4\pi R}\exp\{-ik_aR\}\,d\mathbf{r}'\,, \qquad (34)$$

where $k_a=m_ak$, m_a is the complex refractive index of the particulate substance. In the far zone ($k_a\mathbf{r}\gg1$; $\mathbf{r}\gg\mathbf{r}'$), taking into account that

$$R=\sqrt{r^2-(r')^2-2rr'\cos\vartheta}\cong r-r'\cos\vartheta\,,$$

where ϑ is the angle between the vectors $\mathbf{r}$ and $\mathbf{r}'$, we can simplify Eq. (34) by permutation of the operations of integration and differentiation. The resulting field of the scattered radiation $\mathbf{E}^s(\mathbf{r}; t)$ can be represented as a wave formed by superposition of the fields generated by polarized elements of the particle volume[9]:

$$\mathbf{E}^s(\mathbf{r}; t)\cong\frac{k^2(\varepsilon_a-1)}{4\pi r}\exp\{ikr\}\times$$

$$\times\int_{V_0}\mathbf{E}(\mathbf{r}'; t-r/c)\exp\{ik_ar'\cos\vartheta\}\,d\mathbf{r}'\,. \qquad (35)$$

As can be seen, the key point in this approach is the knowledge of the structure of the field inside the particle $\mathbf{E}(\mathbf{r}'; t)$, which is just described by the series (2). Substitute Eq. (2) into Eq. (35) and write the final equation for the electric field far from the particle (TE mode):

$$\mathbf{E}^s(\mathbf{r}; t)\cong\frac{k^2(\varepsilon_a-1)}{4\pi r}\exp\{ikr\}\sum_n\sum_p A_{np}(t-r/c)I_{np}^{\mathrm{TE}}(\theta, \varphi), \qquad (36)$$

where the integrals

$$I_{np}^{\mathrm{TE}}(\theta, \varphi)=\int_{V_0}\mathbf{E}_{np}^{\mathrm{TE}}(\mathbf{r}')\exp\{ik_ar'\cos\vartheta\}d\mathbf{r}'$$

give the angular behavior of the scattered field. It follows from this equation that the time dependence of the internal and scattered fields within one mode is the same. However, since every mode enters into the sum (36) with its own coefficient I_{np}^{TE}, the time dependence of the resulting (total) scattered field, generally speaking, may differ from the time behavior of the field inside the particle. And only when all modal coefficients A_{np} have the same time dependence, for example, in the case of long pulses (27), the temporal part can be factorized out of the summation sign.

Figure 3 depicts the calculated dependence of the relative intensity of the optical fields inside and outside a water droplet $I(\tau)$ (in the far zone) at diffraction of pulsed radiation with different pulse duration at it. The numerical calculations were conducted by the technique described in Ref. 13. The intensity of the internal field was calculated at the point of the absolute field maximum at the following values of spherical coordinates: $r = 0.92\,a_0$, $\theta = \pi$, $\varphi = 0$. The field intensities were normalized to the corresponding temporally maximum level. It was assumed that the pulses have the Gaussian temporal profile and the wavelength $\lambda = 0.81$ μm typical of femtosecond Ti:Sapphire lasers.[25] For comparison, Fig. 4 shows the same dependences, but in the case of a long square pulse, the Mie theory deals with.

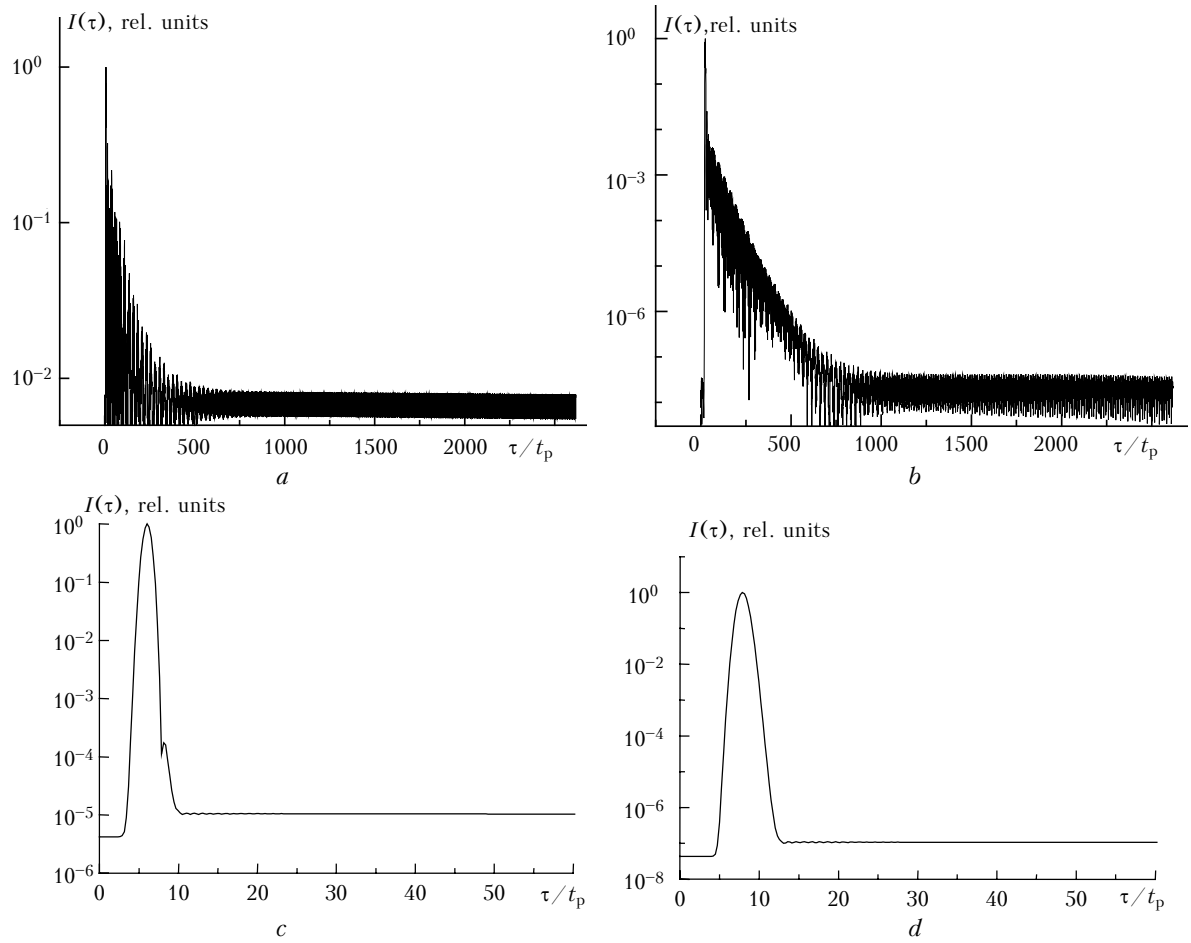

Fig. 3. Relative (normalized to the maximum value) intensity of the internal optical field (*a*, *c*) and the backscattered wave field (*b*, *d*) as a function of dimensionless time τ/t_p at scattering of pulsed radiation (λ = 0.81 μm, t_p = 100 fs (*a*, *b*) and 1 ps (*c*, *d*)) by a water droplet with a_0 = 10 μm.

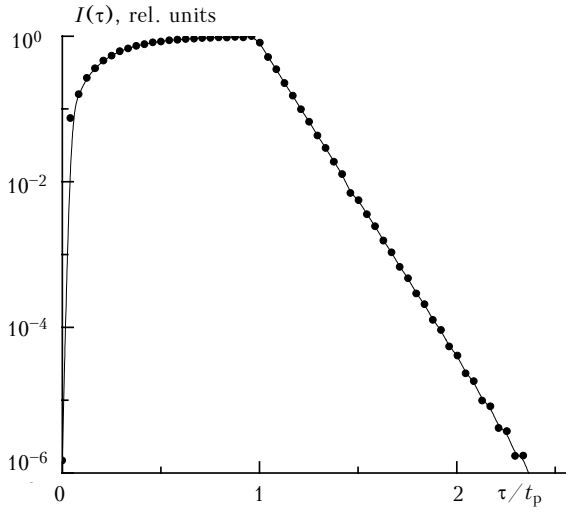

Fig. 4. The same as in Fig. 3, but at resonance scattering of a square pulse ($TE_{100,1}$ mode) with t_p = 1 μs; internal (solid line) and scattered (dots) fields.

One can easily see the difference in the time behavior of the fields at droplet illumination by ultrashort pulses and, consequently, significantly resonance character of light scattering. At the same time, the time behaviors of the internal and scattered fields shown in Fig. 4 are the same.

Thus, we can draw the following conclusions:

1. The theoretical approach to the problem of nonstationary linear elastic light scattering by dielectric spherical particles has been developed. This approach proposes optical fields of the scattered radiation to be represented in the form of expansion in terms of eigenfunctions of the stationary problem, in which the expansion coefficients determine the time behavior of the field and satisfy inhomogeneous equations of oscillations.

2. The study of transient processes at formation of optical fields in a transparent spherical particle carried out based on this approach has shown that the nonstationarity of pulse scattering manifests itself, first of all, in the time shift of the internal field maximum with respect to the profile of the initial pulse and in elongation of the pulse trailing front. This behavior of nonstationary fields is connected with the resonance character of the process of light wave scattering by a particle, when resonance oscillation modes of the internal optical field are excited, and the lifetimes of these modes

may be comparable with or much longer than the laser pulse duration.

3. The main parameters affecting the transient stage in formation of the internal optical field of the particle are the laser pulse duration and characteristic lifetimes of the resonance modes. The time of increase of the internal field depends on the relation between these parameters and is determined by the shortest of them. The characteristic time of the intensity drop is determined by the lifetime of the excited resonance mode with the highest Q-factor. Scattering nonstationarity is inherent in the pulses, whose duration does not exceed the characteristic mode lifetime inside a microparticle resonator. For picosecond pulses such scattering occurs in rare cases, but for femtosecond pulses it always exists for optically "large" particles.

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