Scattering of a train of femtosecond laser pulses by a spherical microparticle: the dynamics of the internal optical field

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The problem on linear scattering of a train of femtosecond laser pulses by a weakly absorbing spherical particle is solved based on the analytical solution of the Maxwell equations derived through representation of light fields as a series expansion in terms of natural electromagnetic modes of a dielectric sphere. The evolution of the optical field in the particle as it is exposed to a single pulse or a train of laser pulses is analyzed comparatively. It is found that when the particle is exposed to a series of femtosecond laser pulses, the evolution of the particle's internal field and its intensity varies depending on the gap between the pulses. This effect is shown to be connected with the excitation of natural electromagnetic modes (whispering gallery modes) in the particle with the resonance frequencies falling within the spectrum of the initial laser pulse. There exists an optimal gap between the pulses, at which the intensity of the internal optical field additionally increases in the zone of its maximum. The value of this gap is inversely proportional to the relative frequency mismatch between the natural modes excited and the central frequency of the incident radiation.

One of the features of the femtosecond-duration laser radiation is high temporal coherence in a train of pulses at the repetitively pulsed operation of a laser source. This may lead to specific effects of interaction between a train of such pulses and a nonlinear medium. In particular, this feature forms the foundation for the well known method of coherent light scattering spectroscopy.[1] Within the framework of this method, medium excitation and sensing are performed by short light pulses, and the spectroscopic information in this case is in the shape of the pulsed response from a medium. Use of this method for a microvolume of, for example, a spherical microparticle may have some peculiarities as compared to the case of a bulk medium. This is connected both with focusing of the internal optical field by the particle-to-medium interface and with the so-called resonance effect manifesting itself in excitation of long-lived electromagnetic oscillation modes (whispering gallery modes[2]) in a particle-microcavity. These effects were analyzed, in particular, in our earlier papers.[2,3]

In this connection, it seems interesting to consider linear scattering of a series of femtosecond laser pulses by transparent dielectric microspheres from the viewpoint of determining the regularities in the value and evolution of the intensity of the internal optical field at the varying gap between the pulses in a train.

To consider the problem of diffraction of a series of ultrashort laser pulses at a spherical particle, we used the theoretical approach[2,4] that is now actively developed in the laser optics. To study the evolution of optical fields, it is proposed here to search for the solution of inhomogeneous Maxwell equations as a series in terms of eigenfunctions of the linear problem of stationary scattering (resonance modes of a dielectric sphere). In this case, the spatial and temporal dependence of the optical fields is factorized so that the information about the evolution of the scattered field fully transforms to the coefficients of the series, for which the system of differential equations is written. If a particular profile of the initial pulse is specified, then this system of equations can be solved numerically or analytically.

Following this technique, let us expand the electric $\mathbf{E}(\mathbf{r}; t)$ and magnetic $\mathbf{H}(\mathbf{r}; t)$ field strength inside the particle over the system of eigenfunctions of a spherical resonator $\mathbf{E}_{np}^{\mathrm{TE,TH}}(\mathbf{r})$, $\mathbf{H}_{np}^{\mathrm{TE,TH}}(\mathbf{r})$ with the natural frequencies ω_{np} and damping coefficients Γ_{np} that describe the spatial profile of the fields of natural oscillation modes with TE- and TH-polarization[5,6]:

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

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

where ε_a is the permittivity of the particulate matter; A_{np}, B_{np} are the time-dependent coefficients. Hereinafter the complex representation of the optical fields is considered.

The corresponding system of differential equations for the expansion coefficients has the form[4]:

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}A_{np}(t) + 2\Gamma_{np}\frac{\mathrm{d}}{\mathrm{d}t}A_{np}(t) + \omega_{np}^2 A_{np}(t) = F_{np}^{i}(t), \quad (1)$$

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where the right-hand sides

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

are represented by Fourier integrals of the spectral function of the initial pulse $G(\omega-\omega_0)$ and some coefficient $K_{np}^{n}(\omega;\,\omega_{np})$ that accounts for the degree of excitation of the internal field mode (with the indices np) by the corresponding mode (n) of the external field:

$$K_{np}^{n}(\omega;\,\omega_{np}) = \frac{ic^2R_ne^{-ika_0}}{V_a\varepsilon_a kc_{np}}\left[\psi_n(ka_0)\psi_n'^{*}(n_ak_{np}a_0) - \right.$$
$$\left. -\frac{1}{n_a}\frac{\omega}{\omega_{np}}\psi_n'(ka_0)\psi_n^{*}(n_ak_{np}a_0)\right].$$

Here E_0 is the strength of the electric field of the incident wave with the central frequency ω_0; $R_n = i^n\dfrac{2n+1}{n(n+1)}$; $k=\omega/c$; $k_{np}=\omega_{np}/c$; a_0 is the particle radius; n_a is the refractive index of the particulate matter; V_a is the particle volume; c_{np} are the normalization coefficients[4]; $\psi_n(z)$ are the Riccati–Bessel spherical functions; c is the speed of light in vacuum. Further, for simplicity, we restrict our consideration to only TE-modes of the optical field.

The particular solution of the inhomogeneous equation (1), representing oscillations under the effect of the "external" force, can be written as

$$A_{np}(t) = \frac{\exp\{-\Gamma_{np}t\}}{2i\tilde{\omega}_{np}}E_0\times$$
$$\times\left[\exp\{i\tilde{\omega}_{np}t\}\int_0^t F_{np}^{i}(t')\exp\{-i(\tilde{\omega}_{np}+i\Gamma_{np})t'\}\mathrm{d}t'\right] -$$
$$-\frac{\exp\{-\Gamma_{np}t\}}{2i\tilde{\omega}_{np}}E_0\times$$
$$\times\left[\exp\{-i\tilde{\omega}_{np}t\}\int_0^t F_{np}^{i}(t')\exp\{i(\tilde{\omega}_{np}-i\Gamma_{np})t'\}\mathrm{d}t'\right], \qquad (4)$$

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

For further analysis, specify the temporal dependence of the radiation incident on the particle as a series (train) of N_p equidistant pulses with a Gaussian time profile:

$$f(t) = e^{i\omega_0 t}\sum_{j=1}^{N_\mathrm{p}} f_j(t), \qquad (5)$$

where $f_j(t) = e^{-\frac{(t-t_0-t_j)^2}{t_\mathrm{p}^2}}$; $t_j=(j-1)T$; T is the pulse repetition interval; t_0, t_p are the time parameters. The Fourier spectrum of this radiation is described by the following function:

$$G(\omega-\omega_0) = G_0(\omega-\omega_0)\sum_{j=1}^{N_\mathrm{p}} e^{-i(\omega-\omega_0)t_j}, \qquad (6)$$

where

$$G_0(\omega-\omega_0) = \frac{4\pi\sqrt{\pi}}{\Delta\omega_\mathrm{p}}\exp\left\{-4\pi^2\frac{(\omega-\omega_0)^2}{(\Delta\omega_\mathrm{p})^2} - i(\omega-\omega_0)t_0\right\}$$

describes the spectral profile of the train as a whole with the half-width $\Delta\omega_\mathrm{p} = 4\pi/t_\mathrm{p}$.

Consider the integrals (2). The calculations show that in the most cases the coefficient $K_{np}^{n}(\omega;\,\omega_{np})$ can be factored out of the integral sign at some value of the frequency ω' by applying the corresponding mean-value theorem, that is,

$$F_{np}^{i}(t) \cong \frac{E_0}{2\pi}K_{np}^{n}(\omega';\omega_{np})\int_{-\infty}^{\infty} G(\omega-\omega_0)e^{i\omega t}\mathrm{d}\omega =$$
$$= E_0K_{np}^{n}(\omega';\omega_{np})f(t). \qquad (7)$$

In the general case, this frequency ω' is determined from a particular frequency dependence of the coefficient $K_{np}^{n}(\omega;\,\omega_{np})$. However, because of the exponential limitation of the spectrum of the initial radiation the point ω' falls within the spectral profile $G_0(\omega-\omega_0)$.

Figure 1 depicts the frequency dependence of the normalized factor

$$\bar{K}_{np}^{n} = K_{np}^{n}\frac{V_a\varepsilon_a c_{np}}{ic^2}$$

for different combinations of the mode indices at incidence of a single pulse on a water droplet. For illustration, it also shows the spectral profile $G_0(\omega-\omega_0)$. The set of natural modes in Fig. 1 is different because of different widths of the spectral profile of the initial laser radiation.

It can be seen from Fig. 1 that at t_p = 10 fs the behavior of the coefficient $\bar{K}_{np}^{n}$ is rather complicated and characterized by fast change of its value in the region of high frequencies, while for longer pulses the dependence is already smooth. However, in both of the cases, variations of the mean value of the studied function for different natural modes within the spectral profile of the radiation are small (Table), and, consequently, approximation of the integral (2) by Eq. (7) is quite correct.

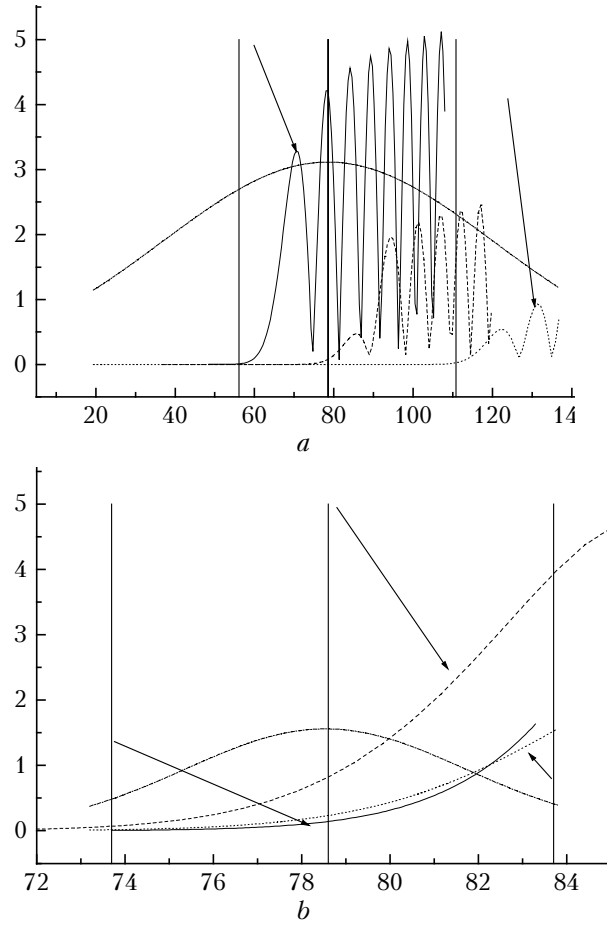

Fig. 1. Factor $\bar{K}^n_{np}(\omega; \omega_{np})$ as a function of the parameter $x = \omega a_0/c$ for the natural modes at incidence of a single pulse ($\lambda = 0.8$ μm) with the duration $t_p = 10$ (*a*) and 100 fs (*b*) on a water droplet with the radius $a_0 = 10$ μm. Vertical lines show the positions of the resonance mode frequency on the abscissa.

Table

Pulse duration, fs	Mode index	Variability range of $\bar{K}^n_{np} \cdot 10^4$, m	Mean value of $\bar{K}^n_{np} \cdot 10^4$, m	"Mean" frequency $\omega' \cdot 10^{-15}$, Hz
10	$TE_{56,1}$	$1.21 \cdot 10^{-2} - 5.12$	2.56	2.70
	$TE_{85,3}$	$1.21 \cdot 10^{-2} - 2.46$	1.23	3.15
	$TH_{120,4}$	$1.80 \cdot 10^{-3} - 0.93$	0.47	3.66
100	$TE_{90,1}$	$5.73 \cdot 10^{-3} - 0.16$	0.08	2.46
	$TE_{85,3}$	$1.21 \cdot 10^{-2} - 2.46$	1.23	2.45
	$TE_{87,4}$	$1.13 \cdot 10^{-2} - 0.16$	0.08	2.43

After substituting Eq. (7) into Eq. (4) and integration, we obtain

$$A_{np}(t) ; \quad \frac{iE_0 K^n_{np}(\omega'; \omega_{np})}{2\omega_{np}} e^{i\omega_{np}t} G_0(\Delta\omega_{np} + i\Gamma_{np}) \times$$

$$\times \sum_{j=1}^{N_p} e^{-i\varphi^j_{np}} e^{-\Gamma_{np}(t-t_0-t_j)} \left[\operatorname{erf}\left(\frac{t - t_0 - t_j}{t_p} + i\frac{t_p}{2}(\Delta\omega_{np} + i\Gamma_{np}) \right) - \right.$$

$$\left. - \operatorname{erf}\left(-\frac{t_0 + t_j}{t_p} + i\frac{t_p}{2}(\Delta\omega_{np} + i\Gamma_{np}) \right) \right]. \qquad (8)$$

Here $\Delta\omega_{np} = \omega_{np} - \omega_0$; $\varphi^j_{np} = \omega_{np} t_j$; $\operatorname{erf}(z)$ is the error function. We have neglected the second integral in Eq. (4) because it is small due to the factor $G_0(\Delta\omega_{np} + 2\omega_0 + i\Gamma_{np})$. In Eq. (8) it is convenient to change the variables: $\tau = t/t_p$; $\tau_0 = t_0/t_p$; $\gamma_{np} = \Gamma_{np}/\Delta\omega_p \equiv (1/4\pi) \cdot (t_p/t_{np})$; $t_{np} = \omega_{np}/\Gamma_{np}$ is the characteristic lifetime of a natural mode; $\Delta\bar{\omega}_{np} = \Delta\omega_{np}/\Delta\omega_p$; $s_p = T/t_p$ is the pulse period-to-pulse duration ratio. Then, finally, we can write:

$$A_{np}(\tau) ; \quad \frac{iE_0 K^n_{np}(\omega'; \omega_{np})}{2\omega_{np}} e^{i(\omega_{np}t_p)\tau} G_0(\Delta\bar{\omega}_{np} + i\gamma_{np}) \times$$

$$\times \sum_{j=1}^{N_p} e^{-i\varphi^j_{np}} e^{-4\pi\gamma_{np}} \left\{ \operatorname{erf}\left[\tau - \tau_0 - \tau_j + 2\pi i(\Delta\bar{\omega}_{np} + i\gamma_{np}) \right] - \right.$$

$$\left. - \operatorname{erf}\left[-\tau_0 - \tau_j + 2\pi i(\Delta\bar{\omega}_{np} + i\gamma_{np}) \right] \right\}.$$

Analysis of the equation obtained shows that, structurally, it consists of several factors: the harmonic part with the frequency of a natural mode, the envelope of the spectral profile of the initial train of pulses that accounts for the shift of the natural mode frequency from the center of the spectrum, and the sum of contributions from individual pulses in the train. The contribution to the resulting time dependence of the coefficient A_{np} from each pulse, in its turn, has its own phase φ^j_{np}, exponentially damping part, and the function describing the initial increase of the mode amplitude. Obviously, the main difference in the repetitively pulsed excitation of the internal field mode from its excitation by a single pulse is in the phase relation between the individual pulses in the train. Consider this relation in a more detail.

Write the phase φ^j_{np} in new variables:

$$\varphi^j_{np} = 4\pi\Delta\bar{\omega}_{np}(j-1)s_p. \qquad (10)$$

At $\varphi^j_{np} = 2\pi l$, where l is the integer number, each new pulse in the train will come in phase with the field inside the particle that is generated by the previous pulses. Thus, the fields are added in-phase, and the total amplitude increases as compared to the case of a single pulse, and the smaller is the mode damping Γ_{np}, the more significant is this increase. Equation (10) imposes a condition on the gap between the pulses, at which this effect is maximum:

$$s_p = \left| 2\Delta\bar{\omega}_{np} \right|^{-1}, \qquad (11)$$

from this it follows that the larger the frequency mismatch between the natural mode and the central frequency of the incident radiation, the smaller should be the pulse ratio. Thus, for example, for efficient excitation of the mode with the frequency ω_{np} lying at the edge of the spectral profile of the initial radiation ($\Delta\bar{\omega}_{np} \approx 1$), a droplet should be exposed to a train of

pulses with the gap $s_p \approx 0.5$, that is, the partial time overlap of the pulses is needed. On the contrary, at a resonance excitation of the mode ($\Delta\bar{\omega}_{np} \approx \Gamma_{np}^2 / (\omega_0 \Delta\omega_p) \ll 1$), because of the natural damping of the mode, the effect of phasing of the pulses in the train almost vanishes.

It should be noted that, from the viewpoint of spectral analysis of signals, Eq. (11) has quite a clear interpretation. Indeed, if this condition imposed on the pulse ratio is fulfilled, then the train spectrum includes a component with the relative mismatch from the central frequency $\Delta\bar{\omega}_{np}$ (Ref. 7). This spectral component resonantly excites the particle natural mode under consideration, which leads to the increase in the intensity of the entire optical field.

As an illustration of this conclusion, Figs. 2*a*–4*a* depict the time dependence of the relative intensity

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

of the internal optical field within a water droplet in the zone of its absolute maximum B_m (shadow hemisphere) as the droplet is exposed to the train of six 100-fs pulses with the different pulse ratio.

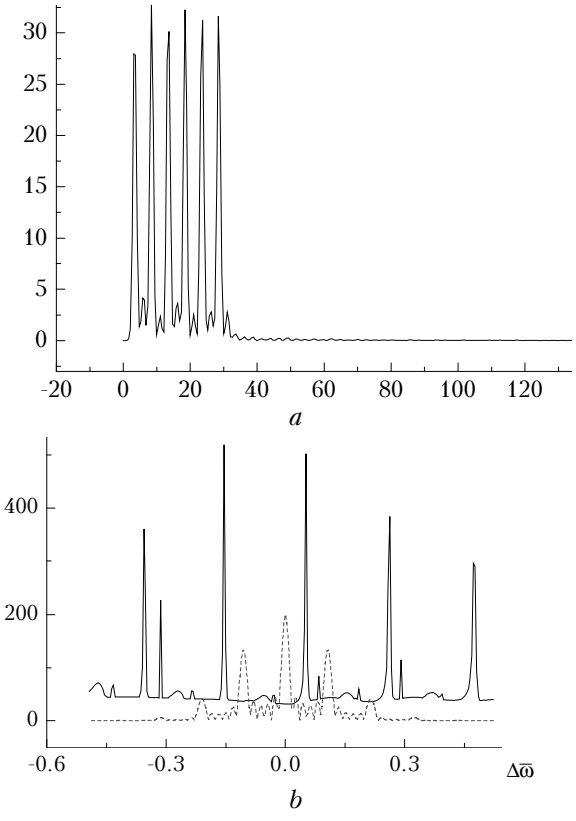

Fig. 2. Time dependence of the relative intensity B_m of the internal optical field within a water droplet (a_0 = 10 μm) exposed to a train of six pulses with λ = 0.8 μm, t_p = 100 fs, and the pulse ratio s_p = 5 (*a*); the spectral profile of the train of pulses with the parameters corresponding to Fig. 2*a* vs. relative frequency $\Delta\bar{\omega} = (\omega - \omega_0)/\omega_0$ (solid line), and the function $I_\delta(\omega)$ (dashed line) in arbitrary units (*b*).

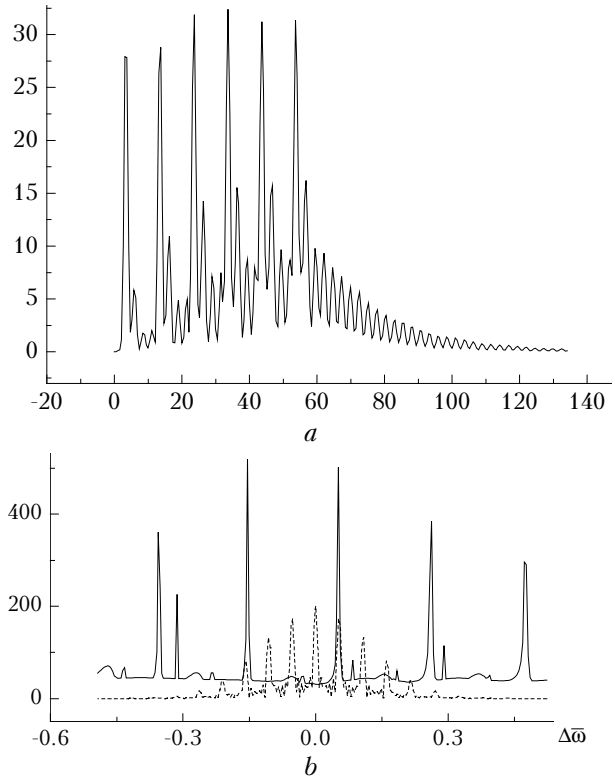

Fig. 3. The same as in Fig. 2*a* but at s_p = 10.

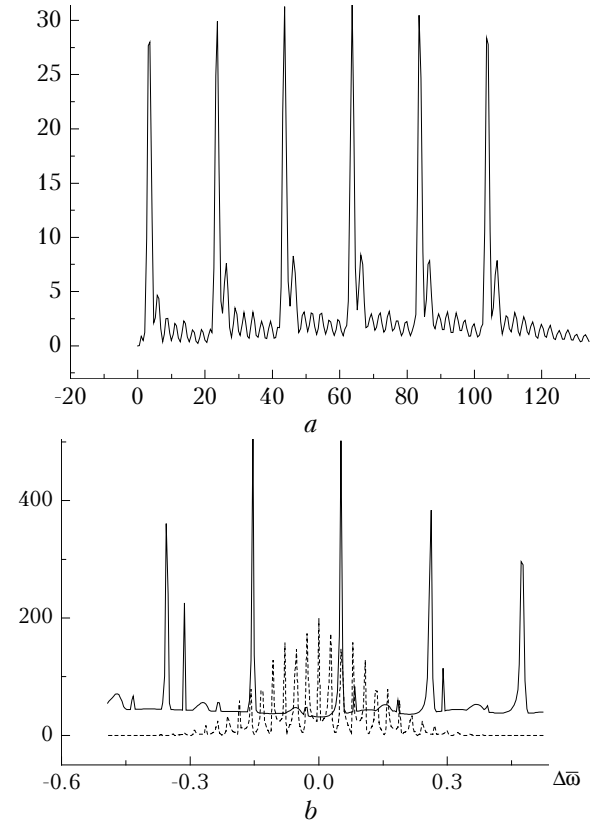

Fig. 4. The same as in Fig. 2*a* but at s_p = 20.

Figures 2*b*–4*b* show, in arbitrary units, the spectral profile of the train at these values of the pulse ratio

and the function $I_\delta(\omega) = \left(\mathbf{E}_\delta(\omega;\mathbf{r}) \cdot \mathbf{E}^*_\delta(\omega;\mathbf{r})\right)$, where $\mathbf{E}_\delta(\omega;\mathbf{r})$ is the so-called spectral response of the droplet.[3] These calculations were carried out by the technique described in Refs. 4 and 8 and involving the use of the Fourier method in combination with the theory of linear light scattering.

It is clearly seen from Figs. 2–4 that in some cases the fields from individual pulses add in-phase in the particle. This is especially true, when the local spectral maxima in the train spectrum coincide with the strong resonance modes of the droplet. Thus, for example, at $s_p = 10$ and 20, when the condition (11) is fulfilled, we can see an increase in the field intensity in the gap between pulses due to excitation of the natural mode lying just near the central frequency $TE_{85,3}$. At the same time, at $s_p = 5$ this mode is excited inefficiently.

Thus, the main difference of the repetitive scattering of pulses by a spherical microparticle from scattering by a single pulse consists in the possibility of some phasing of individual pulses in the train, when the whispering gallery modes falling within the spectral profile of the train are excited resonantly. In this case, the larger is the frequency mismatch between the natural mode and the central frequency of the incident radiation, the shorter should be the gap between the pulses.

Acknowledgments

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