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Thermally nonlinear laser photoacoustic tomography

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Abstract. **The formation of a laser photoacoustic response in an inhomogeneous medium is considered taking into account the temperature dependence of the coefficient of thermal expansion. It is shown that in the one-dimensional or three-dimensional cases in the presence of individual absorbing centres (spherical particles), the shape of the photoacoustic response is the wavelet transform of the spatial distribution of heat sources. The parameters (level) of the wavelet expansion are determined by the characteristics of the laser pulse. The possibility of multiscale wavelet analysis of the medium structure is demonstrated by the example of a numerical model. The possibility of direct visualisation of individual cells in biological tissues is discussed.**

Keywords: *photoacoustics, thermal nonlinearity, tomography*.

1. Introduction

The outlook for using laser radiation in photoacoustics to excite sound is mainly determined by two reasons: the possibility to detect the ultra-low concentrations of substances upon intense laser pumping and the possibility to study narrow absorption lines requiring monochromatic radiation [1, 2]. However, there also exist other effects, which are manifested only upon laser excitation and can be important for practical photoacoustic applications.

One of these effects is the well-known temperature dependence of the coefficient of thermal expansion (the so-called thermal nonlinearity). Indeed, the coefficient of thermal expansion of water, for example, increases from $\sim 0.2 \times 10^{-3}$ up to $\sim 0.7 \times 10^{-3}$ K^{-1} (i.e., almost by a factor of four) with increasing temperature from 20 to 80 °C [3]. It is clear that this effect should be taken into account in analysis of aqueous media, in particular, biological objects. The influence of such thermal nonlinearity on a photoacoustic signal has been studied so far in homogeneous media [4, 5].

However, it is for inhomogeneous media that this effect can be most important, even when the integrated heating of the illuminated volume is small. For example, one can easily see that a particle of size ~ 10 μm with the absorption coefficient $\sim 10^3$ cm^{-1} can be heated in the laser radiation field of moderate intensity ($\sim 10^6$ W cm^{-2} for ~ 50-ns pulses) up to ~ 40 °C, resulting in doubling of the coefficient of thermal expansion. It is obvious that even in the case of lower laser intensities, the thermal nonlinearity should be taken into account in the interpretation of signals in photoacoustic experiments, especially in photoacoustic tomography.

The principle of photoacoustic tomography is rather simple [2, 6]. It is well known [4] that the front shape of an acoustic wave excited by a laser pulse is determined by the spatial distribution of heat sources. The problem of tomography is to determine the spatial structure of a medium (most often, the position and shape of some inhomogeneities in the medium, for example, a stained cell in biological tissues, etc.). The presence of inhomogeneities absorbing light leads to the appearance of a peak or inflection in the wave front of the photoacoustic response [2, 6, 7]. The detection of the position of such a peak in time allows one to determine the spatial position of this inhomogeneity. It was shown recently that the spatial resolution of such photoacoustic visualisation of heat sources can be of the order of a few tens of microns or better [7, 8].

The use of the so-called wavelet transform [9] seems promising for these purposes. The wavelet transform is a mathematical operation which can be considered as the extension of the Fourier transform to the region of non-harmonic (time-limited) basis functions [9 – 11]. In this case, the 'spectral' analysis can be performed in each relatively short time interval. At present the wavelet transform is one of the best methods for separating a signal from noise, in particular, owing to the so-called multiscale analysis [10, 11].

The aim of this paper is to find the relation between the parameters of an inhomogeneity (particle) and the shape of a photoacoustic response in the thermally nonlinear case.

2. Theory

2.1 Thermally nonlinear photoacoustic response

Consider a purely thermal mechanism of sound generation. In this case, it is assumed that the total absorbed radiation energy rapidly transforms to heat due to radiationless relaxation. The heating of the illuminated volume results in its thermal expansion and formation of a pressure pulse. The temperature field in the medium can be found from the heat conduction equation

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$$\frac{\partial T}{\partial t} = \Delta(\chi T) + \frac{kI_0}{\rho C_p} f(t)q(r), \tag{1}$$

where $T(r,t)$ is the desired temperature distribution; I_0 is the radiation intensity; ρ is the density of the medium; C_p is the heat capacity; χ is the thermal diffusivity; and k is the absorption coefficient. The normalised functions $f(t)$ and $q(r)$ describe the spatiotemporal distribution of laser radiation.

If a thermal field is known, the acoustic signal can be determined from the equation for the acoustic potential φ,

$$\Delta\varphi - \frac{1}{c_s^2}\frac{\partial^2\varphi}{\partial t^2} = \frac{\partial}{\partial t}(\beta T), \tag{2}$$

where the acoustic potential is determined from the condition $p = -\rho\partial\varphi/\partial t$. Here, c_s is the sound speed; p is the acoustic pressure; and β is the coefficient of thermal expansion.

The temperature dependence of the coefficient of thermal expansion, which determines the nonlinearity of the acoustic signal, can be written in the form

$$\beta(T) = \beta_0 + \left(\frac{\partial\beta}{\partial T}\right)T = \beta_0 + \alpha T, \tag{3}$$

where $\alpha \approx 8\times10^{-3}\ \mathrm{K}^{-2}$ is the nonlinear coefficient of thermal expansion.

It is usually assumed [6] that the cooling of the volume due to heat conduction can be neglected. Indeed, taking into account that the thermal diffusivity in aqueous media is low ($\chi \sim 10^{-3}$), in the case of a nanosecond pulse of duration τ_{las}, the second term in the right-hand side of the heat conduction equation is usually greater than the second one by a few orders of magnitude. In this approximation, the temperature field can be written in the form

$$T(r,t) \approx \frac{kI_0\tau_{las}}{\rho C_p} q(r)\int f(t)\mathrm{d}t = T_0 q(r)\int f(t)\mathrm{d}t, \tag{4}$$

where T_0 is the maximum spatiotemporal value of the temperature. One can easily see that this solution is independent of the coordinate system type.

Now we can obtain the relation between the shape of the acoustic response and the spatial distribution of the absorbed energy, taking the dependence $\beta(T)$ into account. For simplicity, we consider the one-dimensional case

$$\frac{\partial^2\varphi}{\partial x^2} - \frac{1}{c_s^2}\frac{\partial^2\varphi}{\partial t^2} = \frac{\partial}{\partial t}(\beta(T)T) = \frac{\partial G(t,x)}{\partial t} \tag{5}$$

(this expression determines the parameter G). Taking (2) and (3) into account, we obtain for the nonlinear case

$$\frac{\mathrm{d}G}{\mathrm{d}t} = \beta_0\frac{\mathrm{d}T}{\mathrm{d}t} + 2\alpha T\frac{\mathrm{d}T}{\mathrm{d}t}. \tag{6}$$

One can see from (6) that the solution of Eqns (5) and (6) should be a sum of the usual linear solution [the first term in (6)] and a purely nonlinear solution [the second term in (6)]. Because the linear solutions for most of the specific cases are well known [6], we will consider below only the second term in (6) as the function $\mathrm{d}G/\mathrm{d}t$.

The general solution of Eqn (5) with certain boundary conditions is well known [6]. By applying the Fourier transform in time to (5) and then the Laplace transform in space, we obtain the expression for the acoustic pressure p at the observation point x as the convolution of the spatial and temporal forms of the function $G(t,x) = F(t)H(x)$ introduced above [6]:

$$p(x,t) = \frac{1}{2}\rho c_s\int_{-\infty}^{\infty}\frac{\partial}{\partial\tau}F\left(\tau - \frac{\xi}{c_s}\right)H^{ext}(\xi)\mathrm{d}\xi, \tag{7}$$

where $\tau = t - x/c_s$ is the time in the corresponding reference frame; $F(t)$ and $H(x)$ are the functions of the temperature distribution. It is assumed that $H(x)$ is defined only in the region $x > 0$. Depending on the boundary conditions, $H^{ext}(x)$ is the even or odd continuation of the spatial function $G(t,x)$ to the region of negative x. If there exists a rigid boundary of the medium at the point $x = 0$, the continuation should be even: $H(-x) = H(x)$. If the boundary is free, the continuation is odd: $H(-x) = -H(x)$ [6]. By using (4) and (6), we will obtain finally

$$p(x,t) = \alpha T_0^2\rho c_s\int_{-\infty}^{\infty} f\left(\tau - \frac{\xi}{c_s}\right)\int f\left(\tau - \frac{\xi}{c_s}\right)\mathrm{d}\left(\tau - \frac{\xi}{c_s}\right) \times [H^{ext}(\xi)]^2\mathrm{d}\xi, \tag{8}$$

where f is the temporal shape of the laser pulse and H^{ext} is the spatial distribution of heat sources.

To analyse this result, we consider for definiteness the Gaussian pulse $f(t) = \exp(-t^2/\tau_{las}^2)$. Then, the time dependence in the integrand in (8) can be written in the form $B(t) = \exp(-t^2/\tau_{las}^2)\,\mathrm{erf}(t/\tau_{las})$. To simplify the further expressions, we will approximate this function by the first derivative of the Gaussian,

$$B^*(t) = \exp(-t^2)\,\mathrm{erf}\,(t) \approx -2t\exp(-t^2), \tag{9}$$

where $B^*(t)$ can be treated as a basis function. One can easily see that this is not a rough approximation, its error being only a few percent, whereas the typical experimental error of the approximation of a laser pulse by a Gaussian is usually greater. By substituting (9) into (8), we obtain finally

$$p(x,t) = \alpha T_0^2\rho c\int_{-\infty}^{\infty} B^*\left(\frac{\tau - \xi/c_s}{\tau_{las}}\right)q^{ext}(\xi)^2\mathrm{d}\xi. \tag{10}$$

2.2 Wavelet transform

It is known that the wavelet image (or wavelet spectrogram) of the function $f(t)$ is a function $W(a,b)$ determined by the expression [9]

$$W(a,b) = |a|^{-1/2}\int_{-\infty}^{\infty} f(t)\psi\left(\frac{t-b}{a}\right)\mathrm{d}t, \tag{11}$$

where $\psi(t)$ is the so-called basis function. Unlike the Fourier transform, the wavelet transform uses the basis functions that are noticeably nonzero only within a finite time interval [9]. This feature allows one to realise the 'spectral' analysis (in the 'frequency' $1/a$) at the given instants determined by the shift b. The wavelet decomposition (11) usually also admits the inverse procedure of reconstruction of the function $f(t)$ from the known function $W(a,b)$:

$$f(t) = C_{\psi}^{-1/2} \int_{-\infty}^{\infty} W(a,b)\psi(a,b,t)\frac{1}{a^2}\mathrm{d}a\mathrm{d}b. \quad (12)$$

There exist strict requirements on the properties of basis wavelet functions, which provide the possibility of decomposition of (11) and reconstruction of initial function (12). The most important of them is the orthogonality of the basis functions ψ. To provide the orthogonality, the basis function should be alternating, so that

$$\int_{-\infty}^{\infty} \psi(t)\mathrm{d}t = 0. \quad (13)$$

The wavelet decomposition was successfully used in many papers [see, for example, [11]) to reveal some hidden features of signals (to predict the possibility of the explosion of an airplane engine, the improvement of visualisation of the cell shape, etc.). Moreover, as was shown, the wavelet decomposition provides the so-called multiscale analysis. This means that decomposition (11) permits the study of the time dependence of the signal (the time corresponds to the shift parameter b) for different selected time resolutions [the parameter a in (11)]. In particular, this method is best suited for separating unstable signals from noise [11].

Comparison of (10) and (11) shows that a photoacoustic pulse in the case of thermal nonlinearity is the wavelet transform of the spatial distribution of heat sources [more exactly, of the square of this function, i.e., $q(x)^2$] in the basis function $B^*(t)$ determined by expression (9). One can see from (10) that the variable τ plays the role of the shift parameter b, and $1/\tau_{\rm las}$ is the wavelet frequency (or the decomposition 'level' in the multiscale analysis) $1/a$. It is also easy to verify that in the case of a Gaussian pulse, the 'basis' function in the integrand [see (9)] satisfies condition (13). Moreover, the function $B^*(t) = 2t\exp(-t^2)$ is the well-known basis function for the wavelet transform [10, 11]. As shown in [10], this function provides a stable transformation of type (11).

2.3 Spatial distribution of heat sources

In the general case of an arbitrary spatial distribution of heat sources, a sum of the waves incident on an acoustic sensor (receiver) from different directions should be considered. This leads to a complex multidimensional integral instead of expression (10). However, if only some limited inhomogeneities are present in the medium, whose size is much smaller than the distance to the sensor, the situation is simplified even in the three-dimensional case and becomes similar to the one-dimensional case. Solution (10) was obtained for the one-dimensional case, i.e., for the plane geometry of an absorbing inhomogeneity (for example, some plane layer). But even if the spatial distribution of heat sources is spherical (spherical particle), Eqn (5) still can be readily obtained from (2) by making the usual substitution $u = r\varphi$. (The boundary conditions for $x = 0$ change correspondingly.) In this case, the photoacoustic signal will be the wavelet transform of the function $rq(r)^2$. In the case of the cylindrical symmetry, the expression will be even more complicated.

Nevertheless, we can show that in all cases the nonlinear photoacoustic response will be the wavelet transform of a simple spatial distribution $q(r)$ of heat sources.

2.4 Arbitrary pulse shape

As mentioned above, the basis function of wavelet transform (12) is determined in this case by the pulse shape. It can be easily shown that necessary condition (13) is always fulfilled upon the nonlinear generation of sound if the laser pulse has a symmetric temporal shape, i.e., it is described by an even function of time. Then, the derivative of this pulse forms the orthogonal basis. Moreover, this fact opens up the possibility to control the parameters of the wavelet transform by selecting the shape of a radiation pulse.

Therefore, the photoacoustic response, taking the thermal nonlinearity into account, represents a natural wavelet transform of the spatial function of heat sources. This means that the shape of the generated photoacoustic signal is already the wavelet transform of this distribution and requires no mathematical processing of the signal.

However, this is not the case in the usual linear case, when the pulse shape is described by a function that does not provide the orthogonal basis.

3. Results and discussion

The possibility of the photoacoustic wavelet analysis of inhomogeneities in a medium with the help of thermal nonlinearity can be demonstrated in the three-dimensional case. According to [8], we consider a light-absorbing spherical particle in a transparent liquid. First we will find the expressions for the linear and nonlinear photoacoustic responses for a single particle.

3.1 Linear response

In the linear case, the problem can be solved exactly in the spectral form, i.e., not neglecting thermal diffusion. The acoustic pressure in a travelling wave is described by the expression

$$p(\omega,x) = -\frac{\rho\beta T_0 R}{4\pi x}\exp(\mathrm{i}\omega x)\exp\left(-\frac{\omega^2\tau_{\rm las}^2}{4}\right)$$
$$\times\exp\left\{-\left[(1+\mathrm{i})\left(\frac{|\omega|}{2\chi}\right)^{1/2}+\frac{\mathrm{i}\omega}{c_{\rm s}}\right]R\right\}$$
$$\times\left[\frac{\exp(\mathrm{i}\omega R/c_{\rm s})}{(1+\mathrm{i})(|\omega|/2\chi)^{1/2}+\mathrm{i}\omega/c_{\rm s}}-\frac{\exp(-\mathrm{i}\omega R/c_{\rm s})}{(1+\mathrm{i})(|\omega|/2\chi)^{1/2}-\mathrm{i}\omega/c_{\rm s}}\right],$$
$$p_{\rm i}(\omega,x) = -\frac{\rho\beta T_0 R}{2x}\exp(\mathrm{i}\omega x)\exp\left(-\frac{\omega^2\tau_{\rm las}^2}{4}\right)$$
$$\times\left\{\frac{2\mathrm{i}c_{\rm s}}{\omega^2}\left[\frac{\omega R}{c_{\rm s}}\cos\left(\frac{\omega R}{c_{\rm s}}\right)+\sin\left(\frac{\omega R}{c_{\rm s}}\right)\right]\right\}, \quad (14)$$

where $p_{\rm i}(\omega,x)$ and $p(\omega,x)$ are the sound generation due to the expansion of a particle itself and the heating of the environment, respectively, the total pressure being $P(\omega,x) = p(\omega,x) + p_{\rm i}(\omega,x)$.

When the thermal diffusivity is high $\chi\to\infty$, expression (14) can be considerably simplified [8]. In this case, a very simple expression $T(x,t) = T_0R[\mathrm{erf}(t/\tau_{\rm las})+1]/x$ is obtained for temperature. In fact, this approximation was used in [8].

This approximation means that temperature increases *simultaneously* at all points around a particle (i.e., the thermal wave propagates instantly). Then, the linear acoustic response in the travelling wave is also described by a simple expression

$$P(\omega, x) \approx -\frac{i\rho\beta c_s^2 T_0 R}{2\pi x\omega^2}\exp(i\omega x)\sin\left(\frac{\omega R}{c_s}\right) \times \exp\left(-\frac{\omega^2\tau_{las}^2}{4}\right). \quad (15)$$

It is clear that such an approximation is rather rough because the velocity of a thermal wave in real media is always significantly lower than the sound speed. Because the environment of a particle is heated only by a thermal wave, it is obvious that the distribution of heat sources around the particle depends on the heat conduction wave. This distribution will differ from the approximate distribution, especially at the initial instants, when the acoustic wave is formed in fact. Therefore, the validity of this approximation should be verified.

3.2 Nonlinear response

In the case of thermally nonlinear case, it is impossible to obtain the exact solution in the spectral form. For this reason, it is convenient to use the above approximation for temperature. Then, we obtain the expression

$$P(\omega, x) \approx -\frac{\rho\alpha c_s T_0^2}{x}\exp(i\omega x)\left(1 - \frac{i\omega}{c_s}\right) \times \exp\left(-\frac{\omega^2\tau_{las}^2}{4}\right)\left\{i\pi R^2\frac{|\omega|}{\omega} + 2iR^2\mathrm{Si}\left(\frac{\omega}{c_s}R\right) - \frac{2ic_s}{\omega^2}\left[\frac{\omega R}{c_s}\cos\left(\frac{\omega R}{c_s}\right) + \sin\left(\frac{\omega R}{c_s}\right)\right]\right\} \quad (16)$$

for the total acoustic pressure. This expression, as (15), describes a travelling pressure wave.

It is convenient to verify the validity of approximate expressions with the help of a numerical model. In the numerical model, we first calculated by the method of finite differences (based on the heat conduction equation) the spatiotemporal temperature distribution. After interpolation, we used the obtained result to calculate acoustic pressure with the help of wave equation (2) also by the method of finite differences. (The steps of the division grid were selected so that a further reduction of the step would not affect the result, while the ratio of the time and spatial steps excluded the error accumulation.)

The shapes of the linear response for approximate (15) and exact (14) expressions are presented in Fig. 1. Note first of all that the results of the numerical calculation and calculation with the help of exact expression (14) were virtually coincident [curve (*1*)]. One can see that the instant heating approximation does prove to be inadequate. However, as follows from Fig. 2, the approximate shape of the acoustic response in the nonlinear case coincides much better with the numerical result (the error is noticeable only at the signal edges). This allows the use of approximate expression (16) in most cases.

At the temperature below 4 °C, when the linear coefficient of thermal expansion is close to zero and the term βT can be negative, the shape of the pressure pulse substantially

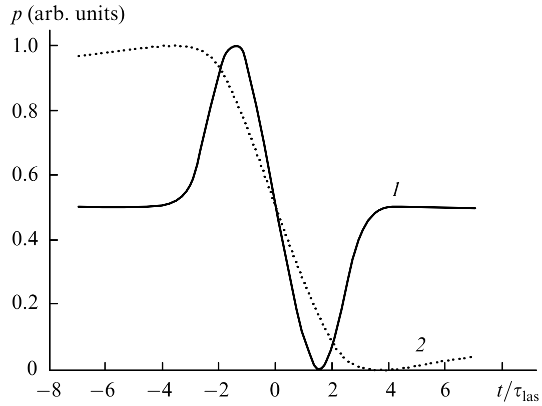

Figure 1. Linear photoacoustic response (and coinciding numerical calculation) of a spherical particle of radius $R = 30$ μm for the pulse duration $\tau_{las} = 10$ ns calculated from exact spectral solution (*1*) and in the approximation $\chi \to \infty$ (*2*).

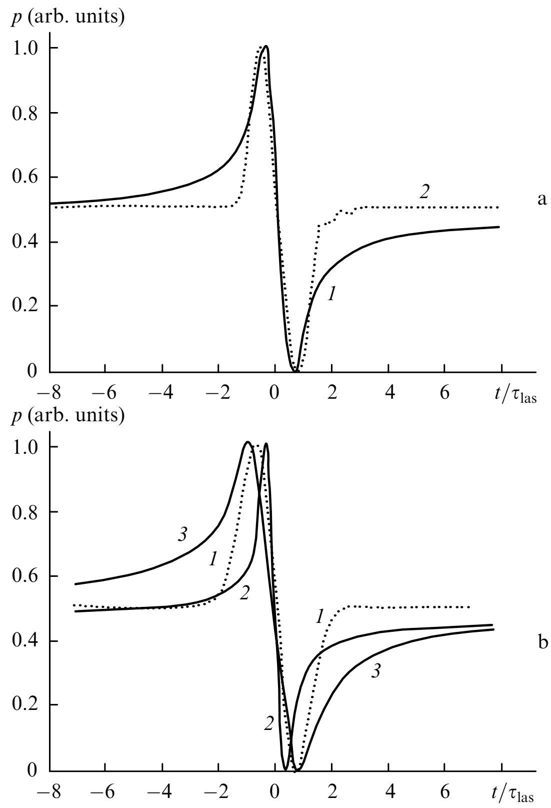

Figure 2. Thermally nonlinear photoacoustic response of a spherical particle of radius $R = 30$ μm for $\tau_{las} = 10$ ns calculate from the spectral solution in the approximation $\chi \to \infty$ (*1*) and numerically (*2*) (a), as well as the linear (*1*) and nonlinear (*2*, *3*) photoacoustic responses of a spherical particle with $R = 1$ (*1*, *2*) and 30 μm (b).

changes (Fig. 3). Such a signal is obtained from expression (16) when the tabulated values of β and α are used. This result can explain, in particular, the appearance of a 'three-polar' pulse observed in experiments [12].

3.3 Multiscale analysis

Consider the model representing a structure consisting of two spherical particles with radii $R = 30$ and 1 μm separated by a distance of 50 μm. Note that the thermal fields of the particles in this case are not overlapped during

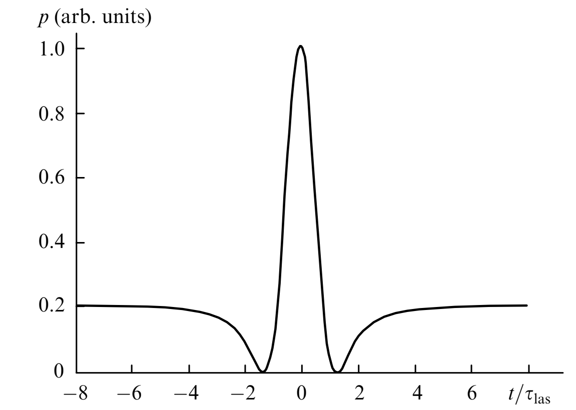

Figure 3. Thermally nonlinear photoacoustic response of a spherical particle of radius $R = 30$ μm at temperature no more than 4 °C and $\tau_{las} = 10$ ns.

the time of the signal formation. If particles are quite small, the acoustic effects (scattering of sound, etc.) can be neglected. The number of calculation points was selected in accordance with the time resolution of the acoustic signal digitising achieved at present ($\sim$0.2 ns per point).

To demonstrate the possibility of the wavelet analysis of such a structure under real conditions, the white noise with the amplitude equal to the signal amplitude was added to the signal. Two different durations (10 and 100 ns) of the laser pulse were used as two wavelet decomposition levels.

The results are presented in Fig. 4. One can see that it is difficult to make any conclusion on the inhomogeneity structure from the shape of the linear response even in the case of a short pulse, whereas in the nonlinear case the position of the inhomogeneity can be accurately determined even for long pulses. (It would seem that this conclusion can be made in the linear case as well; however, due to the presence of a negative half-wave, this is difficult to implement under real conditions, when the waves from different inhomogeneities can interfere.)

In the nonlinear case at a shorter pulse duration (Fig. 4d), not only two particles can be distinctly distinguished, but their size and the distance between them can be estimated. Therefore, by comparing signals at different durations of the laser pulse (Figs 4c, d), we can reconstruct the parameters of the structure in Fig. 4a by the methods used in the wavelet analysis.

3.4 Conditions for nonlinear photoacoustic generation

Thus, we have shown that the nonlinear thermal mechanism of photoacoustic generation, due to the properties of the wavelet transform, can substantially improve the spatial resolution of photoacoustic tomography. In this case, the interpretation of the signal is simplified. However, the question remains: Under what conditions the nonlinear thermal mechanism of sound generation will be more efficient than the usual linear mechanism? It follows from (3) that the nonlinear mechanism dominates when $2\alpha T > \beta$. In aqueous solutions and biological media, α and β are usually $\sim 8 \times 10^{-6}$ K^{-2} and $\sim 10^{-3}$ K^{-1}, respectively [3]. This results in the temperature jump $T > 60$ K. This temperature is close to the physiological temperature. In addition, upon irradiation by a short laser pulse, the heating by radiation exists only during the physiologically short time ($\sim$1 ms). This means that the described method of photoacoustic tomography of biological media is non-invasive. Note also that the mean absorption coefficient of biological tissues is usually small ($k \sim 1$ cm^{-1}), and for the radiation intensities of $10^6 - 10^7$ W cm^{-2}, the total heating is $0.01 - 0.1$ K. Nevertheless, as pointed out above, the nonlinear thermal mechanism can be efficient.

Therefore, this method provides a new possibility of photoacoustic visualisation, for example, individual cells *in vivo* in biological tissues. According to our estimates for the characteristic size of a cell of the order of a few tens of micron, an acoustic sensor with the $\sim$100-MHz pass band will be required, which is available at present [7]. To perform the multiscale wavelet analysis, a semiconductor laser with the variable pulse duration can be used. Therefore, this method can be comparatively simply realised at present.

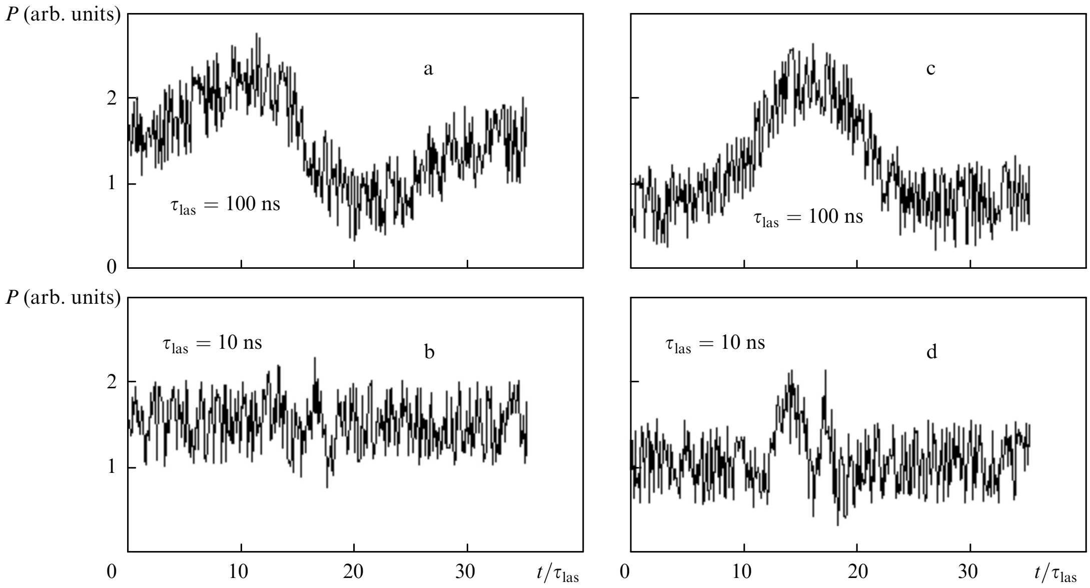

Figure 4. Example of the multiscale wavelet analysis. The model photoacoustic linear (a, b) and nonlinear (c, d) responses of the system of two spherical particles with $R = 30$ and 1 μm separated by a distance of 50 μm for $\tau_{las} = 10$ and 100 ns.

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