Effect of accounting for spatial coordinates on the time scan of a PA signal

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A concise description of models of a PA signal formation is presented, which can be used in determination of the vibrational relaxation time. A three-dimensional model of PA signal generation that describes the gas pressure onto a microphone membrane, located at the center of a side wall of a cylindrical cell, at pulsed excitation of the gas with allowance for diffusion of molecules from the illuminated volume, is proposed.

Introduction

The technique of photoacoustic (PA) measurements is widely used in gas analysis and spectroscopic investigations for finding spectral line parameters. Several methods of determining the vibrational relaxation time from PA measurements are available.[1] In most cases, the continuous-wave modulated exciting radiation is used for this purpose.

The most common phase method consists in determination of the vibrational relaxation time for gas molecules from the dependence of the phase shift of pressure pulsations (PA signal) with respect to the modulated exciting radiation on the total gas pressure in the cell or the modulation frequency.[2–6] The amplitude method, by which the relaxation time is determined from the dependence of the PA signal amplitude on the gas pressure or the modulation frequency, is applied more rarely.[7,8] In some papers, the time of vibrational relaxation is determined from variation of acoustic resonances in the cell.[9–11]

At the pulsed exciting radiation, it is possible to determine the relaxation time from the steepness of the leading front of the recorded signal,[12,13] but this method is applicable only in the case, that the exciting pulse length is much shorter than the measured relaxation time.

The methods were also proposed to determine the relaxation time from the pressure dependence of the PA detector sensitivity,[14,15] from the phase shift of free oscillations of the microphone membrane at the gas pressure variation,[16,17] from the amplitude ratio of depression and compression pulses of the arising acoustic signal,[18,19] and from the compression pulse length of the PA signal as a function of the gas pressure in the cell at the pulsed excitation.[20] However, the results obtained are not always in a good agreement with each other and with the results obtained by other methods. This may be caused by imperfection of the PA signal formation models that are used at processing measurement results.

Formation of a PA signal under atmospheric pressure through excitation by the modulated radiation was considered in a great number of papers.[1–3,8–10,24] However, the conditions of pulsed excitation[21] and low gas pressure have some peculiarities poorly explained in the literature although just measurements under these conditions are convenient for determination, in particular, of the vibrational relaxation time.[1,3,15] This paper briefly describes the models of PA signal formation used in determination of the vibrational relaxation time and proposes a three-dimensional model of PA signal generation, which describes a response of the microphone, located at the center of a side wall of a cylindrical cell, to pulsed excitation of gas with regard for diffusion of molecules from the illuminated volume.

Models of PA signal formation at pulsed excitation

1. A simplest model describing conversion of the absorbed optical energy into the acoustic signal was proposed in Ref. 22. It takes into account only two processes following radiation absorption, namely, vibrational relaxation with the characteristic time τ_{VT} and thermal relaxation with the time τ_T. It is assumed that gas molecules, absorbing the optical radiation of the intensity $I(t)$, transit into the excited vibrational state. Then, at collisions with each other, they return into the ground state, and their vibrational energy converts into the kinetic one (the probability of radiative relaxation is usually negligibly low[23]). The increase in the kinetic energy of gas molecules results in the temperature and pressure increase in the cell, thus generating the PA signal. The excess pressure P in the cell decreases with time due to heat transfer to the cell walls and a presence of a capillary. The time of thermal relaxation is estimated as $\tau_T = \dfrac{r_0^2}{10D}$, where r_0 is the radius of the cylindrical cell; D is the gas diffusion coefficient; the coefficient 10 is found empirically.[22]

At a low gas pressure, along with the process of vibrational relaxation, the diffusion of molecules to the PA cell walls with the excess energy transfer at the rate $w_{\rm d}$ is also possible.[23] In this case, the equation for the density N^* of excited molecules includes an extra term and the equations for the excess pressure in the cell have the form[24]:

$$\frac{{\rm d}N^*}{{\rm d}t} = \frac{\kappa a^2 I(t)}{h\nu r_0^2} - (w_{\rm V} + w_{\rm d})N^*,$$

$$\frac{{\rm d}P}{{\rm d}t} = (\gamma - 1)h\nu w_{\rm V} N^* - \frac{1}{\tau_{\rm T}}P,$$

where $h\nu$ is the radiation quantum energy; a is the beam radius; κ and γ are the absorption coefficient and the specific heat ratio; $w_{\rm V} = 1/\tau_{\rm VT}$, $w_{\rm T} = 1/\tau_{\rm T}$.

The solution of these equations has the form

$$N^* = \frac{\kappa}{h\nu}\frac{a^2}{r_0^2}\int\limits_0^t e^{-(w_{\rm V}+w_{\rm d})(t-t')} I(t'){\rm d}t',$$

$$P(t) = (\gamma - 1)\kappa \frac{a^2}{r_0^2} w_{\rm V} \int\limits_0^t e^{-w_{\rm T}(t-t')}{\rm d}t' \times$$

$$\times \int\limits_0^{t'} e^{-(w_{\rm V}+w_{\rm d})(t'-t'')} I(t''){\rm d}t''.$$

If the exciting pulse length is short as compared to the characteristic times of vibrational relaxation and diffusion of molecules to the cell walls ($\tau_{\rm p} \ll \tau_{\rm VT}$, $\tau_{\rm d}$), then

$$P(t) = (\gamma - 1)\,\kappa \frac{E}{\pi r_0^2}\frac{w_{\rm V}}{w_{\rm V} + w_{\rm d} - w_{\rm T}}\left(e^{-w_{\rm T}t} - e^{-(w_{\rm V}+w_{\rm d})t}\right).$$

As long as the pulse length meets the condition $\tau_{\rm p} \cong \tau_{\rm VT}$, $\tau_{\rm d}$ and the radiation intensity in the pulse can be thought constant, then

$$P(t) = \frac{\kappa a^2 I}{r_0^2}(\gamma - 1)\frac{w_{\rm V}}{w_{\rm V} + w_{\rm d} - w_{\rm T}} \times$$

$$\times \begin{cases} \dfrac{1 - e^{-w_{\rm T}t}}{w_{\rm T}} - \dfrac{1 - e^{-(w_{\rm V}+w_{\rm d})t}}{w_{\rm V} + w_{\rm d}}, & t \le \tau_{\rm p}, \\ \dfrac{1 - e^{-w_{\rm T}\tau_{\rm p}}}{w_{\rm T}} e^{-w_{\rm T}(t-\tau_{\rm p})} - \dfrac{1 - e^{-(w_{\rm V}+w_{\rm d})\tau_{\rm p}}}{w_{\rm V} + w_{\rm d}} \times \\ \times e^{-(w_{\rm V}+w_{\rm d})(t-\tau_{\rm p})}, & t > \tau_{\rm p}. \end{cases} \qquad (1)$$

The main advantages of this model are simple and illustrative equations, but, at the same time, there arises some uncertainty: the characteristic times of molecular thermal relaxation and deactivation on the cell walls should be determined independently, because spatial coordinates are absent in the formulation of the problem.

2. The PA signal calculation based on the gas dynamics equations for the temperature, pressure, density, velocity, and inner energy of gas molecules[25] revealed generation in the cell of signal of two types: the signal caused by thermodynamic heating of the gas and that caused by resonance acoustic properties of the medium and the cell: thermal pressure and acoustic pressure. These signals are solutions of the heat transfer equation and wave equation and can be represented in the form of the series expansions terms of trigonometric functions and Bessel functions for the cell cylindrical geometry with the corresponding boundary conditions.

The calculation of the thermal source generating the signal of both types in the cell is performed for the case of interaction between the gas incorporating particles with two energy levels and a short laser pulse ($\tau_{\rm p} \ll \tau_{\rm VT}$) having the Gauss energy distribution over the cross section. Methods for separation of the signal of only one type were proposed. It is recommended to carry out such separation during measurements, otherwise the calculation of the total signal by this model is labor-consuming and requires a large number of parameters be known.

Note that the first term in the series describing the thermal pressure $P_{\rm T}$ has the form[25]:

$$P_{\rm T}(t) = P_{\rm T\,max}(e^{-t/\tau_{\rm T}} - e^{-t/\tau_{\rm VT}}),$$

that is, it corresponds to the expression obtained in the first model, but in place of the empirically determined coefficient for $\tau_{\rm T}$ the equation now includes the eigenvalues of the problem about free sonic vibrations in the cell, which are determined by the geometry and boundary conditions of the problem. The calculation results for the acoustic pressure are close to the results from Ref. 21, as well as those from Refs. 9 and 10 obtained for the modulated exciting radiation (in Ref. 10 the calculations were conducted for three-level molecules).

3. For calculation of the excess pressure in the PA cell, the wave equation similar to the equation for the acoustic pressure in the previous model was used in Refs. 19 and 26–29. The solutions were presented through the Green's function at different particular time profiles of the exciting pulse and for different distributions of the energy density over the beam cross section. A feature of these solutions is the use of the reduced time (the ratio of the current time to the time of sonic wave propagation through the beam cross section) as a parameter. In Refs. 27–29, a possible nonlinearity of the thermal source is discussed, which has a significant influence on the PA signal shape, but these works consider the PA signal generation in liquid, not in gas.

4. One more model of the PA signal formation[13] is based on the idea that the kinetic energy of a molecule increases at vibrational relaxation, and, consequently, the temperature and pressure of a small gas volume around the molecule increase as well, therefore any relaxing molecule can be considered as a point source of a divergent acoustic wave. The signal recorded by the microphone is caused by the joint effect of such sources. The model presented in Ref. 13 is two-dimensional (only cross section of the cell is considered).

Relation between the times of VT-relaxation and diffusion to the cell walls and the time of acoustic wave propagation through the beam radius

Determination of the vibrational relaxation time from PA measurements is based on the change in the ratio between the sought relaxation time and the known characteristic time of some other process at variation of the experimental conditions (for example, gas pressure in the cell). If the exciting radiation is modulated, the modulation period serves as such time scale. In Refs. 14 and 15, the time of molecular diffusion to the cell walls is used for such a comparison; in Refs. 16 and 17 – the period of free oscillations of the microphone membrane; and in Refs. 18 and 19 – the time of propagation of the acoustic wave through the beam cross section.

Figure 1 compares the characteristic times of these three processes: vibrational relaxation, diffusion of molecules to the cell walls, and sound propagation through the beam cross section, as functions of the gas pressure in the cell.

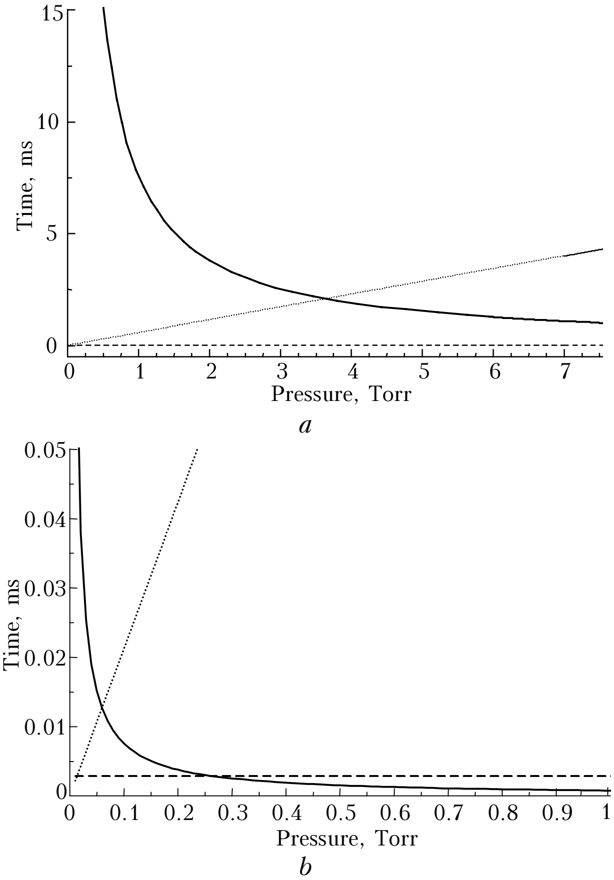


Fig. 1. Gas pressure dependence of the characteristic times of vibrational relaxation (—), diffusion of molecules to the cell walls (· · · ·), and propagation of the acoustic wave through the cross section of the exciting beam (- - -) for CO_2 (*a*) and H_2O (*b*); the cell radius is 0.5 cm and the beam radius is 0.1 cm.

The calculations were performed for the kinetic and thermophysical parameters of water vapor and carbon dioxide (Table) at the cell radius of 0.5 cm and the beam radius of 0.1 cm.

Parameter	CO_2	H_2O
Diffusion coefficient D, cm^2/s	0.1	0.27
Average velocity of molecules V_{av}, m/s	370	590
Sonic speed V_{son}, m/s	270	340

It can be seen from Fig. 1 that for the chosen geometry at the pressure of 1–10 Torr for CO_2 the times of vibrational relaxation and diffusion are quite comparable, while the time of the acoustic wave propagation through the beam cross section is too short to provide the reasonable accuracy at the comparison. For H_2O, at a rather low pressure of 0.1–0.5 Torr, all three times are comparable, but the time of the sound propagation through the beam cross section is a more convenient scale, since it is comparable with the sought time of vibrational relaxation in a wide range of gas pressures.

Three-dimensional model of PA signal generation at low gas pressures and pulsed excitation

At a low gas pressure in the cell, excited molecules can leave the illuminated volume before the vibrational relaxation occurs due to intermolecular collisions. Therefore, when constructing the model of PA signal generation, we used the equation of diffusion of excited particles[30]:

$$\frac{\partial N^*}{\partial t} - D\,\Delta N^* + N^*\, w_V = \frac{\kappa I(r,z,t) a^2}{h\nu\, r_0^2},$$

where Δ is the Laplace operator.

If

$$I(r,z,t) = I(r)\, I(t),\quad I(r) = \begin{cases} I & r \le a \\ 0 & r > a \end{cases},$$

then

$$N^* = \frac{\kappa a^2}{h\nu r_0^2} \sum_{k,n} T_{kn}(t) X_{kn}(r,z);$$

$$T_{kn} = \int_0^t I(t')\, e^{-\lambda_{kn}(t-t')} \mathrm{d}t';$$

$$\lambda_{kn} = D\omega_n^2 + D\mu_k^2 + w_V .$$

The eigenfrequencies μ_k and ω_n and eigenfunctions $X_{kn}(r, z)$ are determined by the geometry and the boundary conditions on the cell walls.

For the cylindrical cell

$$X_{kn} = \mathrm{const}_{kn}\left(\sin\omega_n z + \frac{D\omega_n}{A}\cos\omega_n z\right) J_0(\mu_k r);$$

$$\mu_k \frac{J_1(\mu_k r_0)}{J_0(\mu_k r_0)} = \frac{A}{D} \text{ and } \tan \omega_n l = \frac{2\omega_n D A}{\omega_n^2 D^2 - A^2};$$

$$A = \frac{2\varepsilon}{2-\varepsilon} \frac{V_{\mathrm{av}}}{4},$$

where $J_0(x)$ and $J_1(x)$ are zero-order and first-order Bessel functions; ε is the coefficient of accommodation on the walls; l is the cell length.

In the case of pulsed exciting radiation

$$T_{kn} = \begin{cases} [1 - e^{-(\lambda_{kn} t)}]/\lambda_{kn} & t \le \tau_{\mathrm{p}} \\ [e^{-(\lambda_{kn}(t-\tau_{\mathrm{p}}))} - e^{-(\lambda_{kn} t)}]/\lambda_{kn} & t > \tau_{\mathrm{p}} \end{cases}.$$

Since excited particles colliding in the cell volume convert the energy of vibrational excitation into the thermal one with the time constant w_{V}, that results in generation of the PA signal, each relaxing particle can be considered as a source of a spherical acoustic wave. The resulting PA signal at the point with coordinates **R** can be represented as a superposition of waves generated due to relaxation of particles at the points **r** [Ref. 31]:

$$P(t,\mathbf{R}) = \frac{\gamma - 1}{4\pi V_{\mathrm{son}}^2} \int_V \frac{1}{|\mathbf{R} - \mathbf{r}|} H\left(t - \frac{|\mathbf{R} - \mathbf{r}|}{V_{\mathrm{son}}}, \mathbf{r}\right) \mathrm{d}V,$$

where $H(t, \mathbf{r})$ is the function of the thermal source at the point $\mathbf{r}$; V is the PA cell volume.

Thus, the pressure on the membrane of a microphone located at the point $\mathbf{R}_{\mathrm{m}}$ can be described by the following equation:

$$P(t) = (\gamma - 1) w_{\mathrm{V}} \frac{\kappa a^2}{r_0^2} \times$$

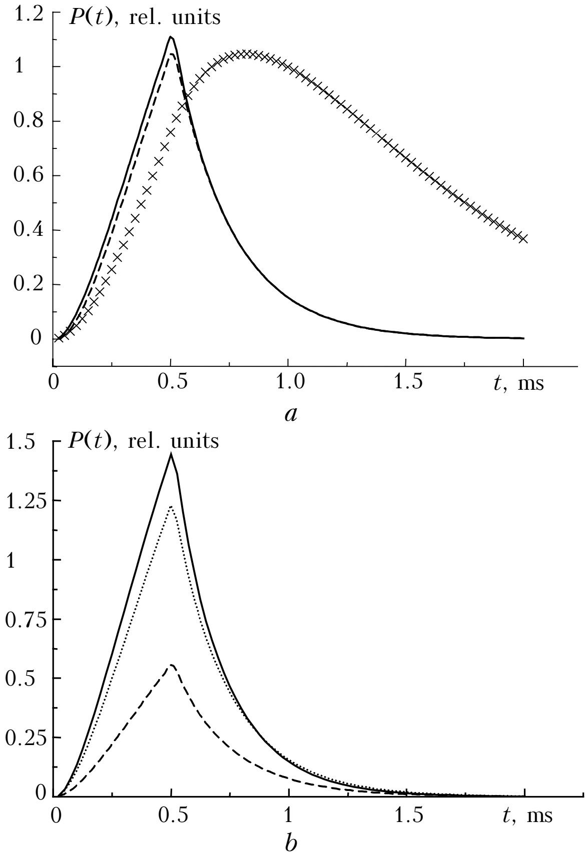
$$\times \int_V \frac{1}{|\mathbf{R}_{\mathrm{m}} - \mathbf{r}|} \sum_{kn} X_{kn}(\mathbf{r}) \mathrm{d}V \int_0^{t - \frac{|\mathbf{R}_{\mathrm{m}} - \mathbf{r}|}{V_{\mathrm{son}}}} I(t') e^{-\lambda_{kn}\left(t - t' - \frac{|\mathbf{R}_{\mathrm{m}} - \mathbf{r}|}{V_{\mathrm{son}}}\right)} \mathrm{d}t. \quad (2)$$

Equation (2), as opposite to the model (1), does not require the knowledge of additional parameters of the gas, cell, or experimental conditions (except for the cell length), but it allows the diffusion of excited molecules in the cell volume and the related change in the shape of the thermal source to be taken into account already at formulation of the problem.

Calculated results

For comparison of the proposed (2) and the traditional (1) models, we have calculated the time scan shape of the PA signal acting on the microphone membrane in the cell filled with CO_2 and H_2O. The radiation pulse was assumed rectangular. The radiation pulse lengths in the calculation (50 ns and 0.5 ms) were close to experimental.[32] The beam radius was taken equal to 1 mm, and the cell radius and length were 0.5 and 10 cm, respectively. The vibrational relaxation times used in the calculations (at the pressure of 1 atm) were 10^{-5} s for CO_2 and 10^{-9} s for H_2O [Refs. 2 and 20].

The results of calculation by the model (2) have shown that although the main contribution to the signal is due to the first radial and longitudinal modes, accounting for several terms of the series somehow changes the value and shape of the PA signal. Therefore, Figs. 2–5 show the calculation results with regard for only radial (k = 1, 20) and longitudinal (n = 1, 10, 20) cell modes. Higher-order modes do not affect the results. In calculations by the both models, it was believed that the diffusion coefficient of excited molecules coincides with the diffusion coefficient of molecules in the ground state and the coefficient of accommodation on the wall surface is equal to unity.



a

b

Fig. 2. Evolution of PA signal: CO_2, p = 1 Torr, τ_{p} = 0.5 ms: (*a*) calculation by the model (2): sum of radial modes up to k = 20 (——), first radial mode (– – –); calculation by the model (1) (×); (*b*) summation over the longitudinal modes up to n = 1 (– – –), 10 (· · ·), 20 (——).

Figures 2 and 3 depict $P(t)$ calculated for CO_2. The radiation pulse length is 0.5 ms in Fig. 2 and 50 ns in Fig. 3. The contribution from the radial harmonics with $k > 1$ decreases with increasing k, therefore the shape of $P(t)$ and the peak value are largely determined by the first radial harmonic (Fig. 2*a*). The contributions from the longitudinal harmonics with $n > 1$ (Fig. 2*b*), unlike those of the radial components, significantly increase the peak value of $P(t)$, although their time behaviors differ insignificantly (Fig. 3).

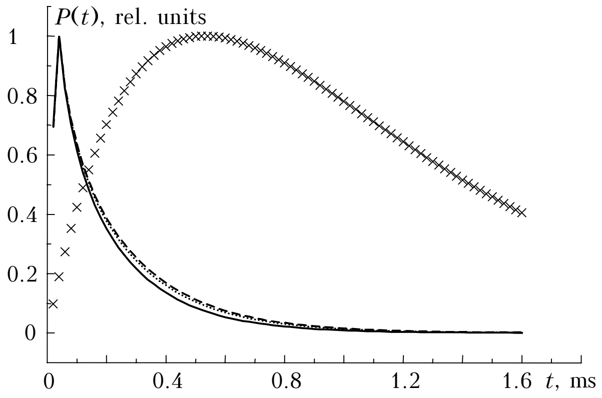


Fig. 3. Evolution of PA signal: CO_2, $p = 1$ Torr, $\tau_p = 50$ ns. Summation over the longitudinal modes of the model (2) up to $n = 1$ (– – –), 10 (···), 20 (——); calculation by the model (1) (×). All signals are normalized.

The results of calculations for H_2O are shown in Figs. 4 and 5. Due to the fast vibrational relaxation in H_2O, the shape of $P(t)$ calculated for the pressure of 1 Torr in the cell at the radiation pulse length of 0.5 ms (Fig. 4) is close to the shape of the exciting pulse. Therefore, for better illustration, further calculations were performed for $p = 0.1$ Torr and the pulse lengths of 50 ns and 50 μs.

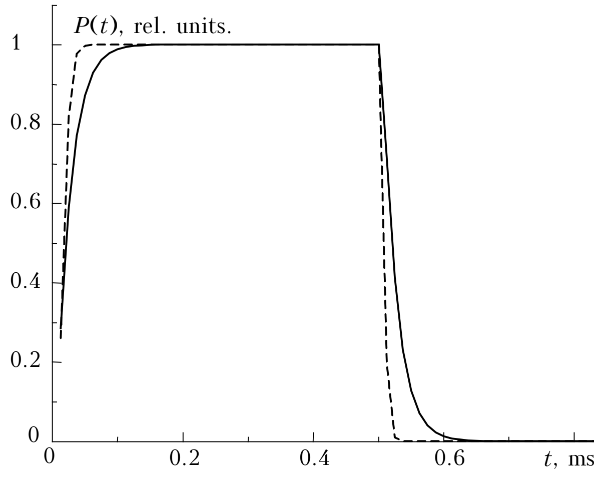


Fig. 4. PA signal calculated by model (2) for H_2O at the pressure of 1 Torr (— — —) and 0.1 Torr (——), $\tau_p = 0.5$ ms.

Conclusions

The calculated results clearly demonstrate that the proposed models of $P(t)$ differ in the steepness of the signal growth, peak position in time, and the rate of decrease for both slowly relaxing CO_2 and fast relaxing water vapor. This difference is the least at the comparable values of the laser pulse length and the time of vibrational relaxation of a gas.

It should be noted that, in practice, the microphone membrane is affected not only by the signal caused by propagation of the pressure wave from a relaxing particle as a source, but also by the waves reflected from the cell walls. Unfortunately, under conditions of pulsed excitation the problem cannot be considered as a stationary one. Therefore, we do not use a standing wave formed in the cell under boundary conditions and given distribution of excited particles (the time of sound propagation along the axis of the 10-cm cell is about 0.3 ms, while several reflections are needed for formation of the standing wave). The closest, in time, reflected wave, formed at reflection from the side surface of the cell opposite to the microphone, can be expected in 0.03 ms (for the cell of 1 cm in diameter). This time is close to the time of the initial increase of the pressure due to the direct wave; therefore, the resulting signal shape is distorted, but the amplitude of such reflected wave is far lower than the amplitude of the direct wave, because it propagates to the threefold longer distance to reach the microphone and, in addition, there are inevitable losses at reflection. Refinement of the proposed model with regard for reflections from the side walls of the cell will be the subject of our further analysis, but already the presented results explain quite adequately the difference in the times of vibrational relaxation[15–17,20] at interpretation of the data of PA measurements with the help of different models of signal generation.

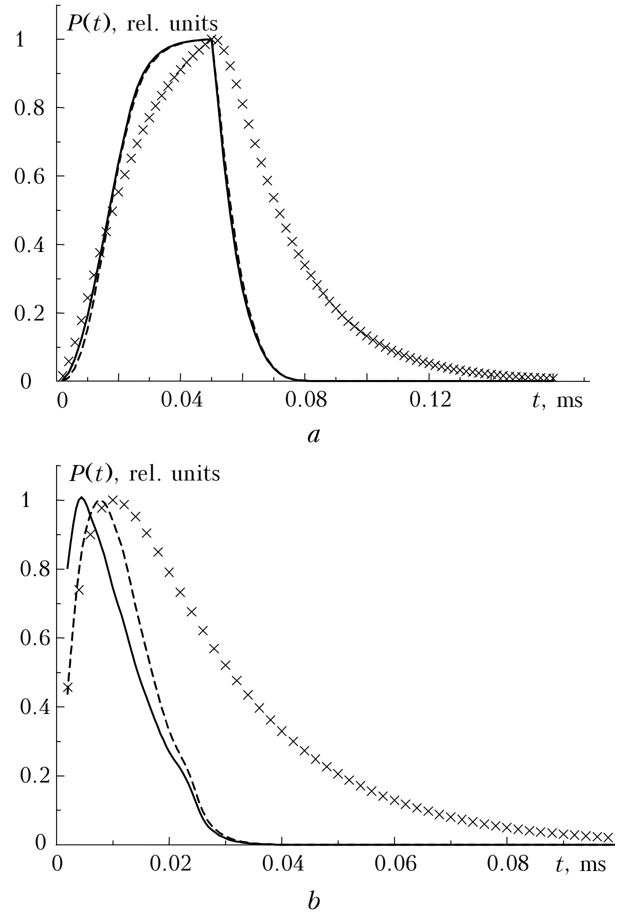


Fig. 5. Evolution of PA signal; H_2O, $p = 0.1$ Torr; calculation by the model (2), sum of radial modes up to $k = 20$ (——), first radial mode (— — —); calculation by the model (1) (×). $\tau_p = 50$ μs (*a*) and 50 ns (*b*). All signals are normalized.

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