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ITERATIVE METHOD FOR DETERMINATION OF THE LASER BEAM PROFILE AND $\tau_{V\text{-}T}$

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Abstract. *Measuring the vibrational-to-translational relaxation time* $\tau_{V\text{-}T}$ *in gases is one of the first applications of the photoacoustic effect. The spatial profile of the laser beam is crucial in these measurements because the multiphoton excitation is investigated. The multiphoton absorption is a non-linear process. Because of this, the top hat profile is preferable. It allows one to deal with nonlinearity in a simple manner. In order to reveal the real laser beam profile, we have slightly changed the theoretical profiles in such a manner that the best matching is obtained between theoretical and experimental photoacoustic signals. Still, there was a question: Is it possible to deduce the laser beam profile directly from the photoacoustic signal, thus avoiding manual changing of the laser beam profile? According to this paper, it is possible. The appropriate method has been found in another photoacoustics application: photoacoustic tomography. Thus, the method for the simultaneous determination of the spatial profile of the laser beam and vibrational-to-translational relaxation time is presented in this paper. It employs pulsed photoacoustics and an algorithm developed for photoacoustic tomography.*

Key words: *Photoacoustic spectroscopy, laser beam profile, vibrational-to-translational relaxation time*

INTRODUCTION

The photoacoustic spectroscopy can be used for measuring vibrational-to-translational relaxation time $\tau_{V\text{-}T}$. First, continuous sources had been used [1, 2]. Afterwards, pulse sources were used [3, 4, 5]. Previously, we have used large photoacoustic cell to detect individual photoacoustic signals. From the shape of these signals we have determined vibrational-to-translation relaxation time [6, 7]. The spatial profile of the laser beam was crucial because we investigated multiphoton processes. The absorption coefficient is fluency-dependent in these processes. Because of this, the top hat laser beam profile is preferable.

Also, we have analyzed the influence of the spatial profile of the laser beam on the accuracy of vibrational-to-translational relaxation time measurements [7]. We have shown that the knowledge of the laser beam profile is crucial in these measurements. Be-

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cause of this we have tried to determine the laser beam profile from the shape of the photoacustic signal. In this paper we describe the iterative method for simultaneous determination of the laser beam profile and the vibrational-to-translational relaxation time.

THEORETICAL BACKGROUND

The photoacoustic signal shape can be calculated from the non-homogeneous wave equation [8, 9]

$$\frac{\partial^2 \delta p(\mathbf{r},t)}{\partial t^2} - c^2 \nabla^2 \delta p(\mathbf{r},t) = S(\mathbf{r},t) \tag{1}$$

where δp(r, t) is the pressure discrepancy from its equilibrium value, c is the speed of sound and S(r, t) is the source function which can be written as

$$S(\mathbf{r},t) = -\frac{\partial^2 E(\mathbf{r},t)}{\partial t^2} H(t) - \frac{\partial E(\mathbf{r},t)}{\partial t} \delta(t) \tag{2}$$

where $E(\mathbf{r}, t)$ is the energy density, $H(t)$ is the Heaviside step function and $\delta(t)$ is the Dirac delta function. Usually, the quantity $E(\mathbf{r}, t)$ is factorable into spatial and temporal part, i.e.:

$$E(\mathbf{r},t) = R(\mathbf{r}) T(t) \tag{3}$$

where R(r) and T(t) are the spatial and temporal parts of the energy density, respectively.

Previously, our goal was to solve non-homogeneous wave equation in order to obtain the theoretical photoacoustic signal. Now, we have an inverse aim. Namely, we consider $\delta p(\mathbf{r}, t)$ as a known function and our aim is to calculate $R(\mathbf{r})$. To do this, we need a back-projection method. Different back-projection methods have been developed in photoacoustic tomography [10-12]. In this study, we have used approximate algorithm developed by Xu and Wang given in [11]. If the distance between detector and source is much larger than the wavelength of the photoacoustic pulse, the $R(\mathbf{r})$ can be calculated as

$$R(\mathbf{r}) = -\mathrm{C} \cdot \iint_s \mathrm{d}s \frac{1}{t} \cdot \frac{\partial}{\partial t} (\delta p(\mathbf{r}_0, t)) \bigg|_{t=|\mathbf{r}_0 - \mathbf{r}|/c} \tag{4}$$

where s is the infinite cylindrical surface, $\mathbf{r}_0$ is the position vector of the detector scanning in this surface. Quantity C is positive constant and its exact value is irrelevant for us, because we need $R(\mathbf{r})$ shape only, not an exact value. This formula is valid for delta function temporal part of the energy density. For 2D case (translational symmetry), there is a simplified formula

$$R(\mathbf{r}) = -\mathrm{C} \cdot \int_\varphi \mathrm{d}\varphi \frac{1}{t} \cdot \frac{\partial}{\partial t} (\delta p(\mathbf{r}_0, t)) \bigg|_{t=|\mathbf{r}_0 - \mathbf{r}|/c} \tag{5}$$

where φ is the polar coordinate.

RESULTS AND DISCUSSION

The back-projection method that we described can be adopted and used in photoacoustic spectroscopy of gases. Typical experimental conditions in our experiment allow further simplifications. Namely, we examine gas mixtures with low absorber concentration. Because of this, absorption coefficient is low thus we can treat our sample as an optically thin and neglect the intensity changes along propagation direction. There is a further simplification. The laser beam profile can be considered as an axial symmetric. Thus, we can conclude that there is the cylindrical symmetry. Consequently, this is 1D case thus we can reproduce the laser beam profile using only one photoacoustic signal.

The quantity $T(t)$ describes the relaxation of the excited molecules in our case. Thus, it is not delta function already it can be written as

$$T(t) = H(t) \cdot \exp(-t / \tau_{V-T}) . \tag{6}$$

We have noted that this method can be applied only when $T(t) = \delta(t)$. Because of this we have to calculate the photoacoustic signal when is $T(t) = \delta(t)$. This can be done using deconvolution. Namely, we calculate the photoacoustic signal for $T(t) = \delta(t)$ as a deconvolution of the experimental signal and $T(t)$ given by (6). According to this, we need $\tau_{V\text{-}T}$ to calculate laser beam profile $R(r)$. On the other hand, we need laser beam profile to calculate $\tau_{V\text{-}T}$ as noted above.

We can solve this problem using iterative, self-adjusted method for simultaneous determination of the laser beam profile and vibrational-to-translational relaxation time. Namely, we can assume the shape of the laser beam profile. Then, we can calculate the $\tau_{V\text{-}T}$ using this assumption. Of course, this is approximate value. Still, we can calculate the photoacoustic pulse for $T(t) = \delta(t)$ using this value. Now, we can calculate the laser beam profile. Of course, this is approximate profile, as well. Yet, we can do this repeatedly. In this way we can obtain the new laser beam profiles and relaxation times. If the starting laser beam profile was good approximation, we should obtain better approximations. Finally, the relaxation time $\tau_{V\text{-}T}$ and laser beam profile should converge to the exact values.

In order to test this method, we have performed numerical simulation. First, we have calculated photoacoustic signal for Gauss profile of the laser beam. The radius of the beam was $r_L = 1$mm and vibrational-to-translational relaxation time was $\tau_{V\text{-}T} = 10\mu$s. This photoacoustic signal simulates an experimental signal. Now we need the starting iteration (the starting laser beam profile). The simplest starting profile is top hat. Now we can start iteration procedure. First we fit simulated signal using theoretical photoacustic signal for top hat profile. We obtain relaxation time in this way. Using this relaxation time we calculate the next laser beam profile, etc. In this example we have obtained good matching in four iterations (figure 1.). Further iterations do not increase accuracy. The obtained relaxation times oscillate around the exact value. The obtained laser beam profiles are nearly the same. We have obtained $\tau_{V\text{-}T} = 9.81\mu$s. The error is less than 2% ($\tau_{V\text{-}T} = 10\mu$s). Based on these results we conclude that this method can be successfully applied to the experimental signals as well. It is worth noting that the starting laser beam profile cannot have an arbitrary shape. It has to be similar to the real profile. Otherwise the iterative procedure will not converge.

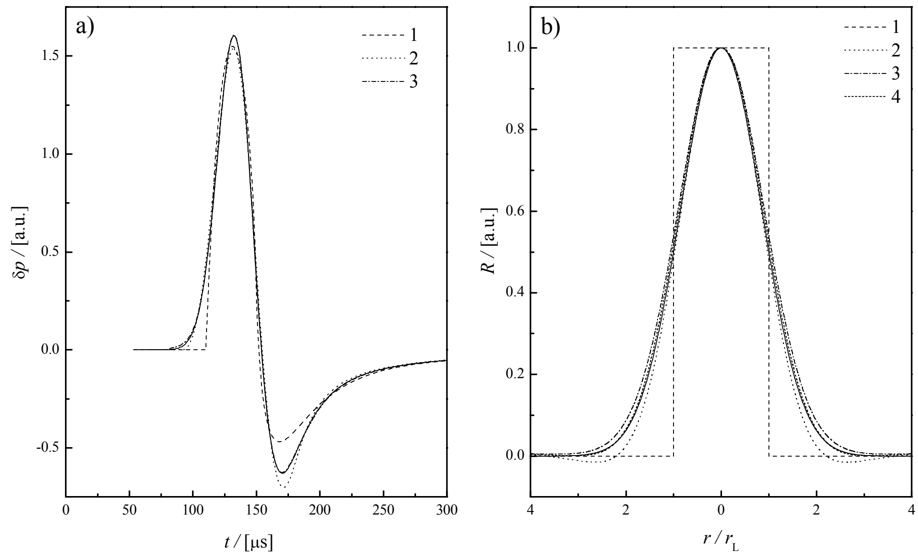

Fig. 1. A numerical simulation of the method for simultaneous determination of the laser beam profile and $\tau_{V\text{-}T}$. The simulated profile is Gaussian. The starting profile in simulation is a top-hat. a) Photoacoustic signals. b) The laser beam profiles. The solid lines depict real profile and photoacoustic signal.

We have tested our procedure in an experimental example. The experimental set-up is described previously [6]. We have tested our procedure using experimental photoacoustic signal obtained in SF_6-Ar gas mixtures. A small amount of the absorbing species was (p_{SF6} = 0.47mbar) diluted in the buffer gas (p_{total} = 100mbar). The obtained results are displayed in figure 2. In figure 2.a) is displayed experimental photoacoustic signal as well as three iterations. The other iterations are not displayed because of clarity. In figure 2.b) are depicted appropriate laser beam profiles. The eight iterations increase matching between theoretical and experimental signal. It is apparent that we need more iterations in this experimental example than in numerical simulation. Experimental errors and limitations are reason for this. The obtained laser beam profile (8^{th} iteration) possesses a different shape and width in comparison to the starting profile (top hat).

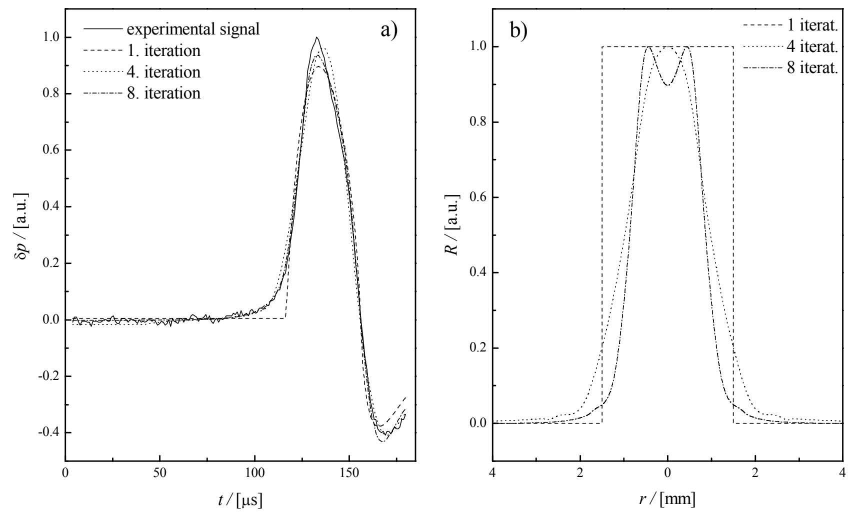

Fig. 2. An experimental example for determination of the laser beam profile and τ_{V-T}. The experimental signal is obtained in SF_6-Ar gas mixture. The starting profile in calculation is top-hat. a) The experimental photoacoustic signal (solid line) and obtained theoretical signals. b) Obtained laser beam profiles.

The diffraction pattern and wings are clearly visible in obtained profile. Also, the new relaxation time is as much as 2.16 times longer than the starting one. Values of relaxation time oscillate randomly around 3.80µs after 8^{th} iteration. Based on these oscillations, we have estimated the error to be lower than 10%.

CONCLUSION

The iterative method for the simultaneous determination of the spatial profile of the laser beam and vibrational-to-translational relaxation time is presented in this paper. It is related to the pulse photoacoustic spectroscopy of gases. It uses back-projection method developed for photoacoustic tomography. The method has been tested using numerical simulation and experimental signals. This method increase accuracy of the translation-to-vibration relaxation time measurement. This is important because different methods give very different values for this time.

An exact back-projection method can further increase accuracy of the relaxation time measuring. Because of this, the goal of further studies will be the development of a simple and exact back-projection method.

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REFERENCES

1. G. S. Gorelik, *Dokl. Akad. Nauk. SSSR* **54**, 779 (1946)
2. P. V. Slobodskaya, *Izv. Akad. Nauk. SSSR* **12**, 656 (1948)
3. J. Schuurman, G. H.Wegdam, *Chem. Phys. Lett.* **73**, 429 (1980)
4. K. M. Beck, A. Ringwelski and R. J. Gordon, *Chem. Phys. Letters* **121**, 529-534 (1985).
5. K. M. Beck and R. J. Gordon, *J. Chem. Phys.* **87**, 5681-5686 (1987).
6. M. D. Rabasović, D. D. Markushev and J. Jovanović-Kurepa, *Meas. Sci. Technol* **17**, 1826-1837 (2006).
7. M. D. Rabasović, J. D. Nikolić and D. D. Markushev, *Meas. Sci. Technol* **17**, 2938-2944 (2006).
8. P. M. Morse and K. Uno Ingard, *Theoretical Acoustics*, Princeton: Princeton University Press, 1987
9. R. T. Bailey, F. R. Cruickshank, R. Guthrie, D. Pugh and I. J. M. Weir, *Mol. Phys.* **48**, 81-95 (1983).
10. C. G. A. Hoelen, F. F. M. de Mul, R. Pongers and A. Dekker, Opt. Lett. **23**, 648-650 (1998).
11. M. Xu and L. V. Wang, *IEEE T. Med. Imaging* **21**, 814-822 (2002).
12. X. Wang, Y. Pang, G. Ku, X. Xie, G. Stoica and L. V. Wang, *Nat. Biotechnol.* **21**, 803-806 (2003).

ITERATIVNI METOD ZA ODREĐIVANJE PROFILA LASERSKOG ZRAKA I $\tau_{V\text{-}T}$

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Merenje vibraciono-translacionog relaksacionog vremena $\tau_{V\text{-}T}$ *u gasovima je jedna od prvih primena fotoakustičkog efekta. Prstorni profil laserskog zraka je veoma značajan u ovim merenjima jer je ispitivana višefotonska pobuda. Višefotonska apsorpcija je nelinearan proces. Zbog ovoga je poželjno koristiti laserski zrak sa homogenim prostornim profilom. Ovakav profil omogućava da se uticaj nelinearnosti eliminiše na jednostavan način. Da bismo odredili stvarni prostorni profil zraka, neznatno smo menjali teorijski profil zraka dok nismo dobili najbolje slaganje između teorijskog i eksperimentalnog fotoakustičkog signala. Ipak, postavlja se pitanje da li je moguće direktno iz fotoakustičkog signala odrediti prostorni profil laserskog zraka i na taj način izbeći ručnu promenu teorijskog profila laserskog zraka. U ovom radu je pokazano da je to moguće. Odgovarajući metod za ovo je pronađen u drugoj primeni fotoakustike, fotoakustičkoj tomografiji. u ovom radu je prezentovan metod za istovremeno određivanje prostornog profila laserskog zraka i vibraciono-translacionog relaksacionog vremena. Ovaj metod koristi impulsnu fotoakustiku i algoritam razvijen za potrebe fotoakustičke tomografije.*