Determination of the laser beam spatial profile by pulsed photoacoustics

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Abstract. Pulsed photoacoustics should fulfil some of the requirements to be a powerful and precise trace gas detection technique. Beside the high sensitivity and selectivity, large dynamic range and good temporal resolution, pulsed photoacoustics has the potential to measure numerous parameters of the laser beam spatial and temporal characteristics with one or no additional instruments. The different numerical methods can be distinguished for this purpose with respect to the measuring procedure and result analysis. In the following article the numerical method for the laser beam spatial profile determination will be presented. Simultaneously this method allows calculation of the molecular vibrational to translational relaxation time in different gas mixtures.

1. Introduction

It is a well known fact that the precise knowledge of the laser beam spatial profile is crucial for the molecular vibrational to translational relaxation time calculation [1]. Spatial profiles of the laser beam can be presented in the experiment using different types of the available instrumentation like: the knife-edge, slit, pinhole beam profilers, complementary metal oxide semiconductor (CMOS), and charge coupled device (CCD) camera. Avoiding the usage of such instrumentation could simplify the experimental set-up without the decreasing of the final relaxation time results quality.

Photoacoustic effects can be used in the pulsed laser beam spatial profiles determination with high spatial resolution and over a wide range of laser fluences [2]. There are different approaches and techniques concerning the laser beam spatial profile determination [2, 3], but all of them are aiming to one goal: simplification of the experimental apparatus. In our case simplification could be made using some numerical methods for the laser beam spatial profile determination instead of beam profiler usage. This method is based on the photoacoustic signal behaviour under the different experimental conditions concerning the laser beam spatial and temporal characteristics, various pressures of the investigated gas mixtures and several excitation energy decays [1,3].

Here we present a numerical method for the laser beam spatial profile determination together with the simultaneous calculation of the vibrational to translational relaxation time. It uses the temporal shape of the photoacoustic signal obtained after the multiphoton absorption in SF$_6$ – Ar mixtures explained in [3]. Like the numerical method based on the algorithm developed for photoacoustic tomography (PAT) [3,4], our proposed method gives an exact solution for the radial energy distribution in the irradiated volume. The quantity which is directly obtained in PAT is the density of
the absorbed energy. For a homogeneous distribution of the radiation and a non-homogeneous absorber, such as tissue, it corresponds to the distribution of the absorption cross-section. In our case, the absorber is homogeneous (gas mixture). One can conclude that the absorbed energy density corresponds to the radiation distribution. The spatial profile of the laser beam, representing the radiation distribution, is the quantity relevant in our study. Thus we determine that we can use any mathematical algorithm which can give us an exact radial distribution solution relevant for our studies with differences only in the time needed to compute.

2. Theoretical background

Usual procedure in the experimental photoacoustic signal (PA) analysis implies the comparison with theoretical signals obtained solving a non-homogeneous wave equation [4-8]

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

where \( \delta p(\mathbf{r}, t) \) is the pressure discrepancy from its equilibrium value, \( c \) is the speed of sound and \( S(\mathbf{r}, t) \) is the source function which can be written as [7]

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

where \( E(\mathbf{r}, t) \) is the energy density, \( H(t) \) is the Heaviside step function and \( \delta(t) \) is the Dirac delta function.

The energy density \( E(\mathbf{r}, t) \) is a quantity relevant in this study. Usually, it is factorable into spatial and temporal part, i.e. its temporal evolution is independent of the coordinates. Thus, it can be written as

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

where \( R(\mathbf{r}) \) and \( T(t) \) are the spatial and temporal parts of the energy density, respectively. The spatial part \( R(\mathbf{r}) \) is related to the distribution of the absorption cross-section in PAT, as well as to the beam profile in our study. The temporal part \( T(t) \) describes the evolution of the excitation energy through the temporal characteristics of the laser pulse and relaxation characteristics of the excited molecules.

Depending on the experimental conditions and used approximations \( T(t) \) can be presented as a Dirac delta function \( T(t) = \delta(t) \) or, for the assumed energy decay (exponential for example), as an exponential decrease which follows due to relaxation of the excited molecules:

\[
T(t) = H(t) \cdot \exp(-t / \tau_{VT}) \tag{4}
\]

where \( H(t) \) is the Heaviside step function and \( \tau_{VT} \) is the vibrational-to-translational relaxation time.

2.1. Exact solution

The exact form of the \( R(\mathbf{r}) \) can be obtained following the procedure described in [4,5]. Such solution involves summation of a series and may take much time to compute. Sometimes the simplification of this solution can be done if the detection radii are much larger than the photoacoustic signal
wavelength. This is, so called, detection radius/signal wavelength approximation [3,4]. Even though this approximation is justified (our case) it brings some errors in the \( R(r) \) calculation and has to be avoided, especially if one wants to make this \( R(r) \) calculation more general, valid for all detection radii and signal wavelengths.

Our proposal is as follows. The general solution of the equation (1) can be written in the form [9]

\[
\delta p(r,t) = \int_0^\infty s(\omega) \left( J_0 \left( \frac{\omega}{c} r \right) - iY_0 \left( \frac{\omega}{c} r \right) \right) \exp(-i\omega t) d\omega,
\]

where \( s(\omega) \) can be denoted as a signal spectrum. One can say that

\[
\delta \overline{p}_o(\omega) \approx s(\omega) \left( J_0 \left( \frac{\omega}{c} r \right) - iY_0 \left( \frac{\omega}{c} r \right) \right),
\]

or

\[
s(\omega) \approx \frac{\text{FFT}(\delta p_o(t))}{\left( J_0 \left( \frac{\omega}{c} r \right) - iY_0 \left( \frac{\omega}{c} r \right) \right)}.
\]

Last equation means that, on the basis of a single photoacoustic signal i.e. its Fourier transform, one can calculate the signal spectrum \( s(\omega) \) and after that, using equation (5), the photoacoustic signal for an arbitrary moment in space and time. At the moment \( t = 0 \), photoacoustic signal spatial distribution is directly proportional to the spatial part of the energy density, i.e. \( \delta p(r,0) \approx R(r) \).

3. Results and discussion

All calculations used in this work are adapted for our real photoacoustic experimental set-up described in details in our previous papers [1, 3, 10]. Briefly, the TEA CO2 laser was used, tuned on 10P(16) line with the 45 ns FWHM (2 \( \mu \)s long tail) pulse, and a non-focused beam with the fluence up to 1.5 J cm\(^{-2} \). As a gas sample container the large nonresonant photoacoustic cell [10] was utilized together with a capacitive microphone as an acoustic signal detector.

A numerical example for the laser beam profile \( R(r) \) reconstruction and the vibrational-to-translational relaxation time \( \tau_{\nu-T} \) simultaneous calculation following the procedure given in section 2.1 is depicted in Figure 1. The simulated beam profile is a Gaussian, for \( \tau_{\nu-T} = 10 \mu \)s and \( T(t) \) defined by equation (4). The top-hat is the starting profile in the calculation. In Figure 1.a simulated PA signal \( \delta p(r,t) \) (grey solid curve) and calculated PA signals for the 1\(^{st} \) (black solid curve 1), 3\(^{rd} \) (dashed curve 3) and 5\(^{th} \) (black dots 5) iteration are presented. In Figure 1.b the Gaussian (grey solid curve) and laser beam profiles after the 1\(^{st} \) (black solid curve 1), 3\(^{rd} \) (dashed curve 3) and 5\(^{th} \) (black dots 5) iteration are presented, too. Here \( r \) is the distance from the center of the laser beam and \( r_L \) denotes the laser beam radius. The photoacoustic signal and laser beam profile are reconstructed with a very small discrepancy level. Finally obtained \( \tau_{\nu-T} = 9.96 \mu \)s in the fifth iteration approaches to its presetted value of \( \tau_{\nu-T} = 10 \mu \)s. Based on this result we suggest that this method can be successfully applied to different spatial laser beam profiles (Gaussian, Lorentzian, top-hat) and also to the different experimental PA signals as well. Comparing to our previous work we reduce error in \( \tau_{\nu-T} = 10 \mu \)s.
calculation from 2% to 0.4%. This error reduction also shows that non-ideal laser beam profile can significantly influence the relaxation time $\tau_{V-T}$ measurement and method presented here could help one to overcome this problem.

![Graph](image)

**Figure 1.** Simulated photoacoustic signal a) obtained for the Gaussian laser beam profile b) and their reconstruction obtained through five iterations using our numerical method explained in section 2.1, following the same analysis procedure for the method based on the PAT algorithm described in [3,4].

4. Conclusions

Presented method for simultaneous determination of the laser beam spatial profile and vibrational to translational relaxation time employs pulsed photoacoustics. Using numerical simulation it has been shown that this method accurately reproduces the spatial profile and the radius of the laser beam and, consequently, it determines the vibrational-to-translational relaxation time with good accuracy. Comparing this method to the one based on the algorithm developed for PAT, one can say that the main difference between them is the time needed to compute. In our case this time is much less than in PAT case, and comparable with the time needed in the case of detection radius/signal wavelength approximation [3,4]. Such method allows one to simplify his experimental set-up avoiding the usage of some additional instrumentation. Finally, we can conclude that this is a reliable method for the laser beam spatial profile determination (only for radially-symmetric beam profiles) and a viable technique for measuring the vibrational to translational relaxation time.

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