Photo-acoustic measurements of the femtosecond Ti:Sa laser radiation absorption by atmospheric air

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Received December 24, 2003

Comparative measurements of nano- and femtosecond Ti:Sa laser radiation absorption by atmospheric air were conducted using a photo-acoustic detector with spatiotemporal resolution of a signal. The dependence of the femtosecond pulse radiation absorption by atmospheric air on the pulse energy in the range ≤ 10 mJ was observed.

Introduction

In the last decade, differential absorption lidars employing tunable pulsed lasers as radiation sources are actively used for high spatial resolution monitoring of gaseous components of the atmosphere.[1,2] As a rule, such lidar systems allow detection of a very limited number of the components.

Multicomponent gas analysis of the atmosphere is carried out with spatially integrating absorption techniques, such, for example, as DOAS (differential optical absorption spectroscopy), which employ the Sun or visible and UV spectral lamps as sources of broadband radiation. An example of such devices is the SANOA open-path multiwave gas analyzer.[3] A new technology of monitoring applies the absorption spectroscopy technique with a new radiation source, namely, supercontinuum radiation, which is generated by high-power femtosecond laser pulses in a preset area of the atmosphere. It provides for multicomponent analysis of the gas composition of the atmosphere with high spatial resolution.[4]

As was shown in Ref. 4, the supercontinuum radiation is not diffuse, and it remains, as a laser beam, collimated along the propagation path. To generate supercontinuum at distances up to several hundreds of meters, femtosecond lasers with the output power of several terawatts are used. The supercontinuum radiation covers a spectral region of several hundreds of nanometers in the visible and near-IR regions.

Propagation of terawatt femtosecond pulses in gas and aerosols differs from the well-studied propagation of nanosecond pulses. Thus, when dealing with interaction of femtosecond pulses with rotational-vibrational transitions of atmospheric molecules, one should take into account the interaction nonstationarity, simultaneous excitation of a great number of molecular transitions (rotational-vibrational bands of several gases, e.g., H_2O and O_2 for the Ti:Sa laser), and very high peak power of radiation. Experimental studies of the femtosecond laser pulse absorption by molecular gases, air, and gas–aerosol media are few in number.

We report the first results of measuring the femtosecond Ti:Sa laser pulse absorption by air conducted with the help of the photoacoustic (PA) detector with the spatiotemporal signal resolution, in particular, absorption of nano- and femtosecond pulses with comparable widths of their frequency spectra.

Experiment

The experiment was conducted on a laser test bed of the Institute of Applied Physics RAS (Nizhnii Novgorod, Russia) by a standard scheme. Laser radiation was transmitted through a cell of the PA detector, which measured the laser beam energy absorbed by a gas.

The experimental laser system is described in Ref. 5. We used the pulsed radiation of the Ti:Sa laser with the pulse energy $E_p \le 10$ mJ and the pulse duration $\tau_p = 9$ ns or 80 fs. The energy of Ti:Sa laser pulses was measured by an attenuator consisting of a $\lambda/2$ phase plate and a polarizer. The radiation spectrum was recorded with a monochromator and a linear CCD array of the 0.04 nm resolution. The spectra of Ti:Sa laser pulses are shown in Fig. 1.

The intensity distribution in the beam cross section is close to the Gauss one for both nano- and femtosecond pulses. The laser beams had the same diameter equal to 8 mm at $0.135I_{max}$ level (I_{max} is the intensity at the beam axis).

Absorption of radiation of nano- and femtosecond Ti:Sa laser pulses was measured by the PA detector[6] designed at the Institute of Atmospheric Optics SB RAS (Tomsk, Russia). Thanks to the time resolution of informative and parasitic background pulsed PA signals, the detector permits measuring the radiation absorption signals in the gas under study against a zero background. Application in the PA detector of

0235-6880/04/01 83-03 $02.00

a specialized concentrator, consisting of cylindrical and spherical parabolic mirrors, to focusing acoustic signals onto the microphone improved the detector's sensitivity for the absorption coefficient up to $2 \cdot 10^{-11}$ cm$^{-1} \cdot$ J.

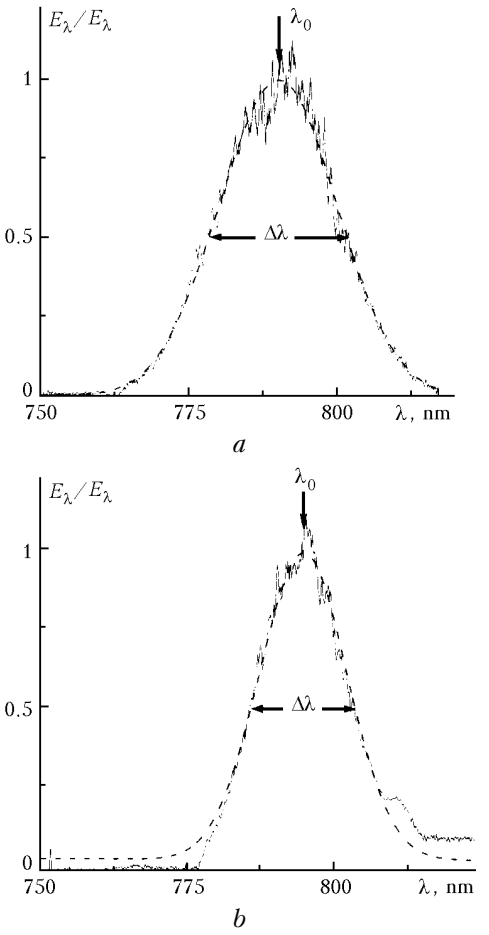

Fig. 1. Spectra of Ti:Sa laser pulses with $\tau_p = 9$ ns (*a*) and 80 fs (*b*): dashed curves are for the results of fitting by the Gauss function; $\lambda_0 = 790$ nm and $\Delta\lambda = 23.4$ nm (*a*); $\lambda_0 = 794.7$ nm, $\Delta\lambda = 17.7$ nm (*b*).

The sensitivity is maximal, if the laser beam axis coincides with the focal axis of the cylindrical paraboloid. Under similar measurement conditions (identical thermodynamic and relaxation characteristics of the gases under study, identical space and time characteristics of radiation, etc.), the detector's sensitivity is independent of the spectral range, in which the absorption coefficients are measured.

To study absorption of the femtosecond Ti:Sa laser radiation, we used the modified PA detector without windows. Its layout is illustrated in Fig. 2.

With no windows, detector's acoustic noise from external sources (especially, low-frequency noise in the band up to 1 kHz) increased roughly 100 times. To reduce the noise level, the detector cell was equipped with buffer volumes with diaphragms, and a high-pass filter with 85% transmittance for frequencies ≥ 5 kHz was added to the electric circuit at the preamplifier output.

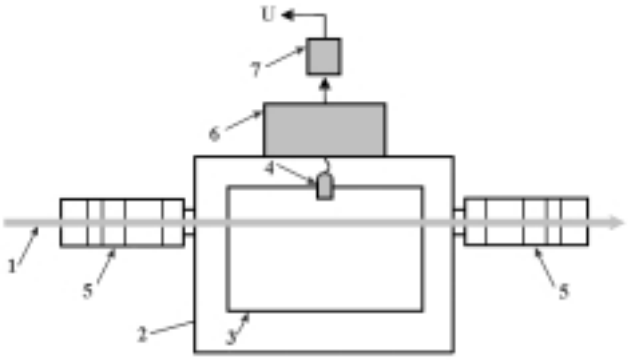

Fig. 2. PA detector: laser beam *1*, body of the PA cell *2*, concentrator *3*, microphone *4*, buffer volume *5*, preamplifier *6*, and filter *7*.

In comparative measurements of absorption of nano- and femtosecond pulses in the absence of filamentation, the radiation was focused into the cell by a lens with the focal length of 88 cm. The space between the lens and the microphone was 56 cm, and the focal plane of the lens lied behind the PA cell. In this case, the diameters of laser beams in the microphone plane were equal to 3 mm.

In the comparative measurements of absorption of femtosecond pulses under the conditions of filament generation and in its absence, the radiation was focused onto the cell by a spherical mirror with the curvature radius of 161.3 cm. The space between the mirror and the microphone was 180 cm.

Results

The comparative measurements of absorption of nano- and femtosecond pulses by air, the dependence of absorption on the pulse energy, and the difference in absorption of femtosecond pulses before and during filament generation were conducted with the indoor air at the atmospheric pressure. The water vapor partial pressure P_{H_2O} controlled by a psychrometer was 10 Torr at the air temperature of 295 K. The results obtained are shown in Figs. 3 and 4.

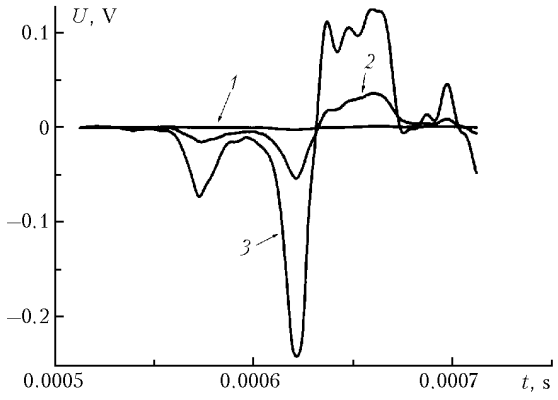

Fig. 3. PA signals generated at absorption of Ti:Sa laser pulses by indoor air.

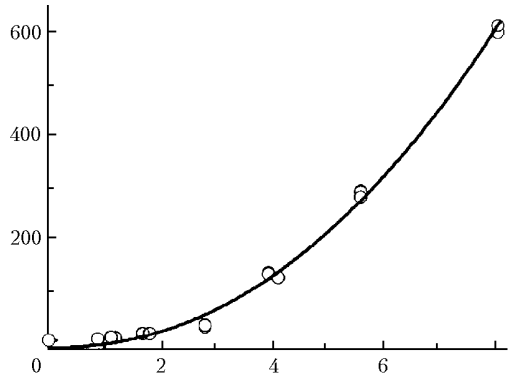

Fig. 4. Dependence of the energy of femtosecond Ti:Sa laser pulse absorbed by air on the laser pulse energy (a, b, c are fitting parameters; R^2 is the correlation coefficient).

Figure 3 illustrates a clear difference in shapes and amplitudes of the PA signal for different conditions of Ti:Sa laser pulses passing through the measurement cell. The highest PA signal amplitudes are observed in the case of the filament generation.

Figure 4 demonstrates a distinct square dependence of the femtosecond radiation absorption by air on the pulse energy. The experiments also have shown that the absorption of femtosecond radiation is much higher (by almost two orders of magnitude) than the absorption of nanosecond pulses at comparable spectral widths of radiation.

The results obtained demonstrate, for the first time, the efficiency of the PA method in investigation of absorption of ultrashort radiation pulses by atmospheric gases. They also illustrate non-trivial changes in the air absorptance under different conditions of propagation of femtosecond pulses, in particular, during filamentation. A thorough study of the discovered effects will be a subject of our further experiments.

Acknowledgments

The authors are thankful to N.N. Trifonova for the help in preparation of this paper.

This work was supported, in part, by Project 2.4 of DPhS RAS and Integration Project No. 12 of SB RAS.

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