

Nonlinear absorption of femtosecond laser pulses (800 nm) by atmospheric air and water vapour

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Abstract. Quantitative data on the nonlinear absorption cross sections of femtosecond Ti:Sapphire laser pulses in air and water vapour have been obtained. A photoacoustic spectrometer calibrated based on the calculated value of linear absorption of laser pulses with a wavelength of 800 nm and a spectral width of 17.7 nm is used to find the nonlinear absorption cross sections of water vapour and air: $\sigma_2^w = (2.6 \pm 0.4) \times 10^{-55} \text{ cm}^4 \text{ s}$ and $\sigma_2^a = (8.7 \pm 1.0) \times 10^{-56} \text{ cm}^4 \text{ s}$, respectively. Based on measuring the absorption of femtosecond Ti:Sapphire laser pulses with a photoacoustic detector calibrated with the known linear absorption of ruby laser radiation by water vapour in air, the air nonlinear absorption cross section is found to be $(8.2 \pm 0.9) \times 10^{-56} \text{ cm}^4 \text{ s}$.

Keywords: femtosecond laser, nonlinear absorption cross section, photoacoustic technique.

1. Introduction

The results of extensive fundamental studies on the interaction of high-power femtosecond laser pulses with gases are applied in problems of remote (lidar) diagnostics of atmosphere [1] and distant objects (in particular, in the THz range) [2] and control of high-voltage electric discharges [3]. To simulate the propagation of femtosecond laser pulses in atmosphere, one needs quantitative data on the characteristics of absorption of these pulses by atmospheric air and its molecular components. Most of experiments are performed using a Ti:Sapphire laser, which generates high-intensity femtosecond pulses at the wavelength $\lambda_0 \approx 800 \text{ nm}$ with the spectral width $\Delta\lambda \sim 20 \text{ nm}$. The conventional (intensity-independent) absorption of radiation with such spectral parameters in atmospheric air is determined by the absorption at vibrational–rotational transitions in water vapor molecules. Calculations based on the HITRAN database [4] show that the water vapour absorption is low; it is characterised by the effective absorption coefficient $k_{\text{eff}} \sim 1.5 \times 10^{-8} \text{ cm}^{-1} \text{ mbar}^{-1}$. The nonlinear absorption of femtosecond pulses by the main molecular components of atmospheric air (nitrogen, oxygen, and water vapour) can result in a much larger contribution to

the attenuation of femtosecond pulses propagating in atmosphere [5]. There are no quantitative data in the literature on the characteristics of nonlinear absorption of femtosecond Ti:Sapphire laser pulses by atmospheric air.

The purpose of this study was to measure the cross sections of nonlinear absorption of femtosecond radiation with the wavelength $\lambda_0 = 800 \text{ nm}$ by atmospheric air and water vapour.

2. Experimental

A schematic of the photoacoustic spectrometer based on a Ti:Sapphire laser [6] is shown in Fig. 1. The laser generates femtosecond pulses with a width $\tau = 50 - 500 \text{ fs}$ and a repetition rate of 10 Hz. The radius of the output laser beam with a Gaussian intensity distribution is 5 mm. The spectrum of femtosecond pulses, recorded by a monochromator and a CCD linear array with a 0.04-nm resolution, has an FWHM $\Delta\lambda = 17.7 \text{ nm}$, peaking at the wavelength $\lambda_0 = 800 \text{ nm}$. The femtosecond pulse energy is changed by a polarisation attenuator and measured using a calibrated photodiode.

The photoacoustic detector [7] contains a cylindrical cell 20 cm in diameter and 25 cm long. 2-mm-thick MgF_2 windows are mounted on the cell sides for radiation input and output. The laser radiation is introduced into the cell using flat and spherical (with a focal length of 86.5 cm) dielectric

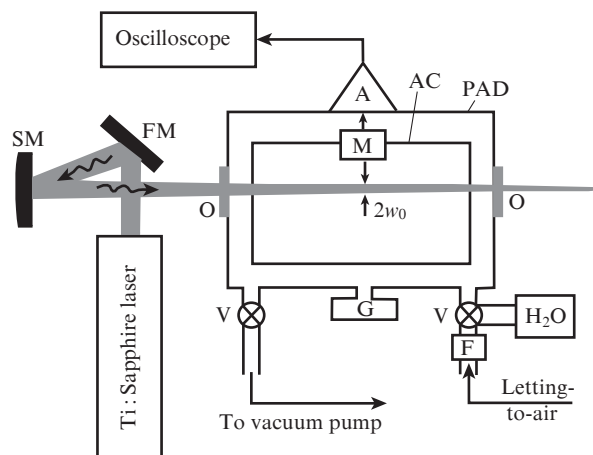


Figure 1. Schematic of the photoacoustic spectrometer: (PAD) photoacoustic detector, (AC) acoustic concentrator, (FM) flat mirror, (SM) spherical mirror, (O) optical window (MgF_2), (M) capacitor microphone MK-221, (A) broadband amplifier, (V) vacuum valve, (G) pressure gauge DVR-5, and (F) aerosol filter.

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mirrors. The cell is placed so as to make the spherical mirror focus be located behind the cell and provide the laser-beam radius $w_0 = 2.5$ mm in the acoustic-signal detection plane (Fig. 1).

An acoustic concentrator composed of two parabolic mirrors (Fig. 2) was placed in the cell to increase the detector sensitivity. The measurements are preceded by the photoacoustic-detector alignment. The laser beam axis is aligned with the focal axis of the parabolic cylinder. The acoustic wave generated in the irradiated volume of absorbing gas is directed to the paraboloid of revolution after the reflection from the parabolic cylinder surface. A capacitor microphone is placed in the focus of the paraboloid of revolution to detect the amplified pressure signal.

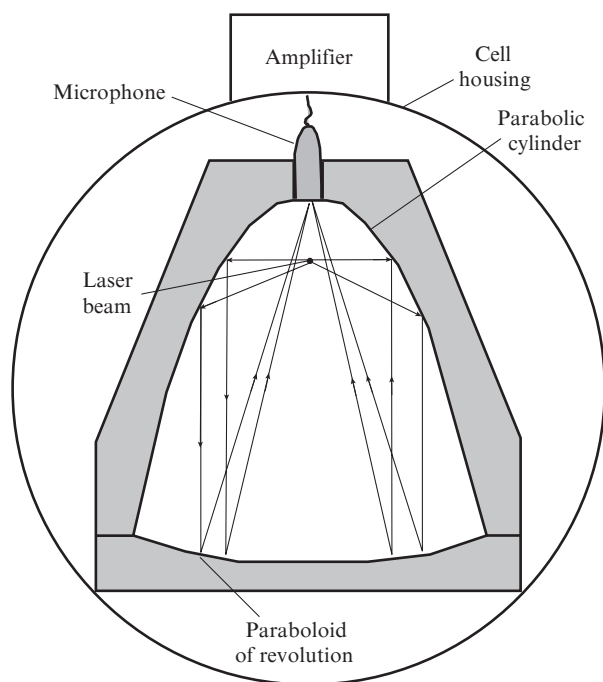


Figure 2. Photoacoustic detector with parabolic mirrors.

The microphone output signal is amplified by a broadband amplifier and recorded by a digital oscilloscope. The characteristic shape of the signal generated as a result of the partial absorption of the energy of a short laser pulse in air at atmospheric pressure is shown in Fig. 3. Three pulses are indicated by numbers 1–3 in the oscillogram. The first one corresponds to the detection of the pressure pulse propagating from the illuminated region by the microphone. The second (weaker) pulse arises with a delay $\Delta t = 550$ μ s with respect to the laser pulse; it corresponds to the detection of the pressure pulse reflected from the surface of the paraboloid of revolution by the microphone. The third pulse with an amplitude U_{PA} is delayed even more; it corresponds to the pressure pulse reflected from the parabolic cylinder surface and focused on the microphone by the paraboloid of revolution. The amplitude U_{PA} is measured in the experiment.

The gases are prepared directly in the photoacoustic cell, which is preliminarily evacuated to a pressure of ≤ 0.1 mbar. Then the gas under study (air or H_2O vapor) is fed into the cell. Air tests are taken by the circulation method using an aerosol filter. Water vapour is introduced into the evacuated

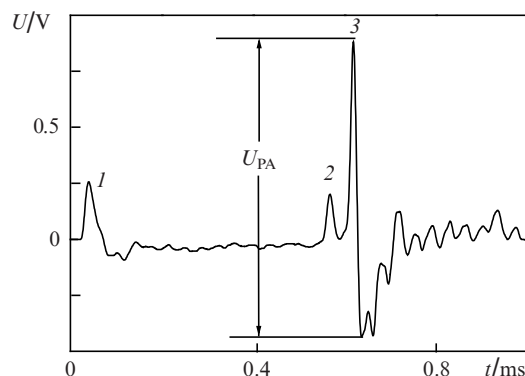


Figure 3. Oscillogram of a signal from the photoacoustic detector with an acoustic concentrator and MK-221 microphone.

cell by evaporating distilled water from a flask (Fig. 1). The gas pressure is measured by a DVR-5 pressure gauge with a VSK-5 capacitance sensor (0.1–1100 mbar).

3. Photoacoustic signal amplitude under linear and nonlinear absorption

A drawback of the photoacoustic method is that the photoacoustic spectrometer must be calibrated to obtain quantitative data on the absorption coefficients and cross sections [8]. When recording linear absorption spectra, the photoacoustic spectrometer is calibrated either by measuring the photoacoustic signal amplitude and calculating the absorption coefficient for a reference gas mixture, applying spectral line databases, or using simultaneous measurements of absorption by photoacoustic and spectrophotometric methods. The calibration of photoacoustic spectrometer for measuring the characteristics of nonlinear absorption is more difficult, because the absorption coefficient for laser beams with a non-uniform intensity distribution in the transverse beam cross section also changes over this cross section. Thus, in comparison with the case of linear absorption, where the density distribution of excited molecules in the transverse cross section and the corresponding profile of the acoustic signal source [9] coincide with the radiation intensity distribution, the profile of the acoustic signal source will be different for nonlinear absorption. The sensitivity of the photoacoustic spectrometer measuring the characteristics of nonlinear absorption will differ from the sensitivity obtained as a result of the photoacoustic-spectrometer calibration based on the known linear absorption.

According to [8], the photoacoustic signal amplitude is directly proportional to the absorbed laser pulse energy and contains information about the characteristics of linear and nonlinear gas absorption in the cell. Generally, the amplitude of an electric signal from a photoacoustic spectrometer, recorded in the case of absorption of a short Gaussian pulse in weakly absorbing gas, is determined by the expression [10]

$$U_{PA} = \alpha_M p_{PA} = \alpha_M p_{PA} E_{abs} \approx \sum_n U_n$$

$$= \alpha_M \sum_n \left[1.5926 \frac{n^{3/4}}{(2\pi\epsilon)^{3/2}} \left(\frac{v}{r}\right)^{1/2} \left(\frac{1}{C_p T}\right) \right] (\sigma_n N n^{-3/2} I_0^{n-1}) E, \quad (1)$$

where N is the concentration of absorbing molecules; E is the pulse energy; v is the speed of sound in the gas studied; r is the distance from the laser beam axis to the microphone; C_p is the gas specific heat at a constant pressure; T is temperature, $I_0 = 2E\pi^{-3/2}w_0^{-2}\tau^{-1}$ is the peak radiation intensity, expressed in terms of the number of photons per unit time and unit area; α_M is the acoustic sensor sensitivity; and σ_n is the absorption cross section (σ_1 is the linear absorption cross section, σ_2 is the absorption cross section with a nonlinearity factor $n = 2$, etc.). A Gaussian radiation pulse is described by the following relations [9]: $I(r = w_0, t = 0) = I_0e^{-2}$, $I(r = 0, t = \tau) = I_0e^{-1}$ и $\varepsilon = (\tau_2^2 + w_0^2/2v^2)^{1/2}$. The factor $\sigma_n N n^{-3/2} I_0^{n-1}$ is an effective absorption coefficient [10], expressed in terms of the peak intensity of Gaussian pulse. The factor in the square brackets correspond to the conversion factor for the absorbed laser pulse energy $E_{\text{abs}} = \sigma_n N n^{-3/2} I_0^{n-1} E$ into the pressure pulse amplitude p_{PA} or the photoacoustic method sensitivity α_{PA} . It follows from (1) that, when passing from linear to nonlinear absorption with a nonlinearity factor n , the sensitivity α_{PA} increases proportionally to $n^{3/4}$. The nonlinear absorption cross section σ_n is simply related to the linear absorption cross section σ_1 :

$$\sigma_n = \frac{U_n n^{3/4}}{U_1 I_0^{n-1}} \sigma_1. \quad (2)$$

It follows from (2) that, in order to determine σ_n from the known linear absorption with the cross section σ_1 , it is necessary to find experimentally the amplitudes U_n , U_1 and the peak radiation intensity I_0 . The nonlinearity factor n is determined by measuring the dependence $U_n(E)$.

4. Measurement results and discussion

Figure 4 shows the dependence of the U_{PA}/E ratio (the right coordinate axis) on E (the upper axis) for the absorption of a Gaussian pulse with a duration $\tau = 72$ fs by water vapour. The vapour pressure is $P = 13$ mbar, and the vapour concentration at the temperature $T = 295$ K is $N_w = 3.23 \times 10^{17} \text{ cm}^{-3}$. It can be seen in Fig. 4 that the energy-dependent nonlinear absorption is detected against the linear absorption background. The nonlinear absorption is directly proportional to

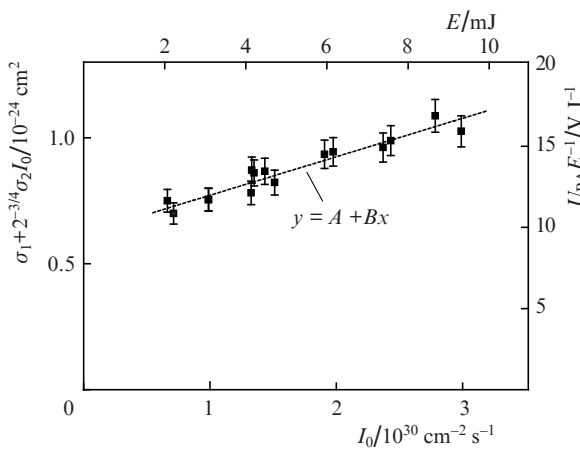


Figure 4. Dependence of the absorption of a Gaussian pulse ($\lambda_0 = 800$ nm, $\Delta\lambda = 17.7$ nm, and $\tau = 72$ fs) by water vapour at the pressure $P = 13$ mbar on the laser pulse energy (peak intensity) $E(I_0)$. The dashed line corresponds to the linear function $y = A + Bx$.

the laser pulse energy ($n = 2$) and is comparable in magnitude with the linear absorption at the pulse energy $E = 10$ mJ.

The effective (i.e., averaged over the laser radiation spectrum with a width $\Delta\lambda = 17.7$ nm) linear absorption cross section of a water vapour molecule, calculated using the HITRAN database [4], is $\sigma_1 = (6.2 \pm 0.5) \times 10^{-25} \text{ cm}^2$. This value is used to calibrate the photoacoustic spectrometer (or to pass from the right coordinate axis in Fig. 4 to the left axis). In the case of single-component gas, formula (1) yields the following relation for the U_{PA}/E ratio:

$$U_{\text{PA}}/E \sim (\sigma_1 + 2^{-3/4} \sigma_2 I_0). \quad (3)$$

Based on linear regression of the experimental data in Fig. 4 and in correspondence with relation (3), we find the nonlinear absorption cross section for water molecules: $\sigma_2^w = (2.6 \pm 0.4) \times 10^{-55} \text{ cm}^4 \text{ s}$. The rms error in determining σ_2^w contains the error in calculating the linear absorption cross section σ_1 (8%) and the rms error in determining the fitting parameter B (13%). The relative error in measuring the ratio U_{PA}/E is estimated to be at a level of 6%; it is shown by error bars in Fig. 4.

The dependence of the U_{PA}/E ratio on the energy of a laser pulse with a width $\tau = 114$ fs for wet air at a total pressure $P = 997$ mbar ($N_a = 2.44 \times 10^{19} \text{ cm}^{-3}$) and a partial water vapour pressure $P_w = 11.2$ mbar ($N_w = 2.78 \times 10^{17} \text{ cm}^{-3}$) is shown in Fig. 5. Here, the photoacoustic signal amplitude contains contributions of the linear and nonlinear absorption of laser pulses by water vapour and the nonlinear absorption by the main air components: nitrogen (80%) and oxygen (20%). Relation (3) takes the form:

$$U_{\text{PA}}/E \sim [N_w \sigma_1 + 2^{-3/4} I_0 (N_w \sigma_2^w + N_a \sigma_2^a)], \quad (4)$$

where σ_2^a is the nonlinear absorption cross section for air. As in the first case, we calibrate the photoacoustic spectrometer using the calculated value of the linear absorption coefficient for water vapour: $k_{\text{eff}} = N_w \sigma_1 = (1.7 \pm 0.2) \times 10^{-7} \text{ cm}^{-1}$. Based on the linear regression of the experimental data (Fig. 5) in correspondence with formula (4) and taking into account the

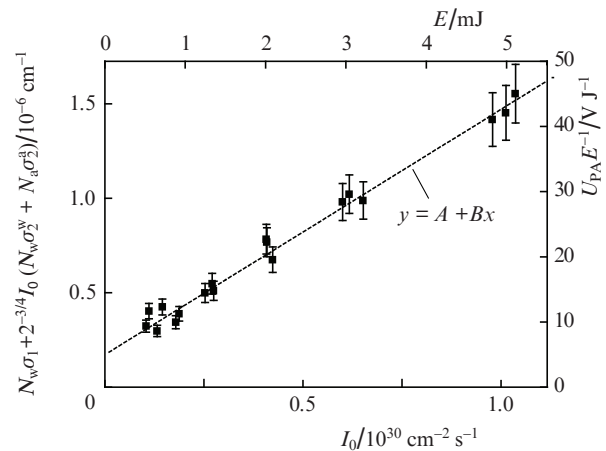


Figure 5. Dependence of the absorption of a Gaussian pulse ($\lambda_0 = 800$ nm, $\Delta\lambda = 17.7$ nm, and $\tau = 114$ fs) in air at the total pressure $P = 997$ mbar and the partial water vapour pressure $P_w = 11.2$ mbar on the laser pulse energy (peak intensity) $E(I_0)$. The dashed line corresponds to the linear function $y = A + Bx$.

nonlinear absorption of water molecules, σ_2^w , we find the nonlinear absorption cross section for air: $\sigma_2^a = (8.7 \pm 1.0) \times 10^{-56} \text{ cm}^4 \text{ s}$. The error in determining σ_2^a includes the error in calculating the linear absorption coefficient k_{eff} (10%) and the rms error in determining the fitting parameter B (5%). The relative error in measuring the U_{PA}/E ratio is estimated to be at a level of 10%; it is shown by error bars in Fig. 5.

The same results for air (see Fig. 5) can be processed based on the calibration of photoacoustic detector from the known absorption of ruby laser pulses by water vapour in air at the centre of the H_2O absorption line (694.380 nm). Relation (1) yields the U_{PA}/E ratio in the form

$$U_{\text{PA}}/E = \alpha_{\text{M}}\alpha_{\text{PA}}[N_{\text{w}}\sigma_1 + 2^{-3/4}I_0(N_{\text{w}}\sigma_2^w + N_{\text{a}}\sigma_2^a)].$$

The sensitivity of the photoacoustic spectrometer with a Ti:Sapphire laser determined from independent calibration measurements using a ruby laser is found to be $\alpha_{\text{M}}\alpha_{\text{PA}} = (3.1 \pm 0.31) \times 10^7 \text{ V cm}^{-1} \text{ J}$. We use this value to pass from the right coordinate axis to the left axis, and based on the linear regression of the experimental data, we obtain $\sigma_1 = (5.9 \pm 0.8) \times 10^{-25} \text{ cm}^2$ [which is in good agreement with the calculated linear absorption cross section $\sigma_1 = (6.2 \pm 0.5) \times 10^{-25} \text{ cm}^2$] and the nonlinear absorption cross section for air: $\sigma_2^a = (8.2 \pm 0.9) \times 10^{-56} \text{ cm}^4 \text{ s}$ (which is also in good agreement with the above value of σ_2^a). The error in determining σ_2^a includes the error in calibrating the spectrometer (10%) and the error in determining the fitting parameter B (5%). As above, we use the previously determined nonlinear absorption cross section for the water vapour molecule, σ_2^w .

5. Conclusions

We measured the cross sections of nonlinear absorption of femtosecond Ti:Sapphire laser pulses by atmospheric air and water vapour. The photoacoustic spectrometer was calibrated using the known linear absorption of Ti:Sapphire and a ruby laser pulses by water vapour. When the photoacoustic spectrometer was calibrated based on the calculated linear absorption of laser pulses with a wavelength of 800 nm and a spectral width of 17.7 nm by water vapour, the measured nonlinear absorption cross sections of this radiation by water vapour and air were, respectively, $\sigma_2^w = (2.6 \pm 0.4) \times 10^{-55} \text{ cm}^4 \text{ s}$ and $\sigma_2^a = (8.7 \pm 1.0) \times 10^{-56} \text{ cm}^4 \text{ s}$. The measurements of the absorption cross sections for femtosecond Ti:Sapphire laser pulses using a photoacoustic detector calibrated based on the known absorption of ruby laser pulses by water vapour in atmospheric air allowed us to determine the linear absorption cross section of water molecules, $\sigma_1 = (5.9 \pm 0.8) \times 10^{-25} \text{ cm}^2$ [which is in good agreement with the calculated value $\sigma_1 = (6.2 \pm 0.5) \times 10^{-25} \text{ cm}^2$], and the nonlinear absorption cross section for air: $\sigma_2^a = (8.2 \pm 0.9) \times 10^{-56} \text{ cm}^4 \text{ s}$.

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References

1. Kasparian J., Rodriguez M., Mejean G., et al. *Science*, **301**, 61 (2003).
2. Manceau J.-M., Nevin A., Fotakis C., et al. *Appl. Phys. B*, **90**, 365 (2008).

3. Tzortzakakis S., Franco M. A., Andre Y.-B., et al. *Phys. Rev. E*, **60**, R3505 (1999).
4. Rothman L.S., Jacquemart D., Barbe A., et al. *J. Quantum Spectrum Rad. Transfer.*, **96**, 139 (2005).
5. Kartashov D.V., Kirsanov A.V., Kiselev A.M., et al. *Opt. Express*, **14**, 7552 (2006).
6. Babin A.A., Kiselev A.M., Sergeev A.M., Stepanov A.N. *Kvantovaya Elektron.*, **31**, 623 (2001) [*Quantum Electron.*, **31**, 623 (2001)].
7. Tikhomirov A.B., Firsov K.M., Kozlov V.S., et al. *Opt. Eng.*, **44**, 071203-1 (2005).
8. Zharov V.P., Letokhov V.S. *Lazernaya optiko-akusticheskaya spektroskopiya* (Laser Photoacoustic Spectroscopy) (Moscow: Nauka, 1984) p. 320.
9. Heritier J.-M. *Opt. Commun.*, **44**, 267 (1983).
10. Protasevich A.E., Tikhomirov B.A. *Opt. Atmos. Okeana*, **24**, 328 (2011).