

Absorption of a femtosecond Ti:Sa-laser radiation by atmospheric air and water vapor

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We have found that in the absence of self-focusing, induced absorption of a pulsed Ti:Sa-laser radiation of femtosecond duration by atmospheric air and water vapor increases proportionally to the laser pulse energy E . In the case of Gaussian beams of 2.5-mm radius, the coefficient of induced absorption by air at atmospheric pressure changes with the pulse energy at a rate of $dk/dE = (2.48 \pm 0.45) \cdot 10^{-7} \text{ cm}^{-1} \cdot \text{mJ}^{-1}$ (pulse duration of 190 fs), and that of water vapor at the pressure of 13 mbar at a rate of $dk/dE = (1.54 \pm 0.20) \cdot 10^{-8} \text{ cm}^{-1} \cdot \text{mJ}^{-1}$ (pulse duration of 120 fs). The absorption coefficient of air measured at $E \approx 0$, linear absorption coefficient, using a femtosecond pulse well agrees with the coefficient of radiation absorption by water vapor in the atmospheric air calculated using HITRAN database.

Introduction

Propagation of high-intensity femtosecond pulses (FS) of radiation through gases and atmospheric air differs from the propagation of nanosecond-duration (NS) laser pulses. One of the most striking effects, observed and actively investigated in recent years, is the laser beam filamentation at propagation of FS laser pulses through the atmospheric air.^{1,2}

Reference 3 reports another one effect, which consists in that yet in the absence of filamentation the absorption of FS Ti:Sa-laser radiation by atmospheric air depends on the laser pulse energy. In the experiment,³ the induced absorption of 80-fs-duration radiation pulses was observed in the atmospheric air that nonlinearly grew with the increasing pulse energy being considerably stronger than absorption of NS radiation pulses (9 ns) with the comparable spectral characteristics. Quantitative data on the coefficients of induced absorption of FS-radiation by atmospheric air and its molecular components are important for estimation of the FS-radiation attenuation in the atmosphere. No such data are available in the literature.

The aim of this study was to measure absorption coefficients of atmospheric air and water vapor for FS-radiation at $\lambda_0 = 800 \text{ nm}$ and laser pulse energies, below the filamentation energy threshold.

Optoacoustic spectrometer based on a Ti:Sa-laser

Measurements have been carried out using optoacoustic (OA) method of laser spectroscopy.

Optical arrangement and block-diagram of the OA spectrometer built up around the laser complex described in Ref. 4 are shown in Fig. 1.

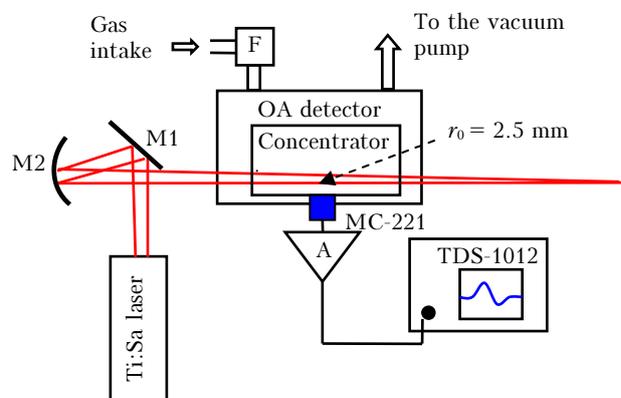


Fig. 1. Optical arrangement and block-diagram of the OA spectrometer: M1 is a plane mirror, M2 is a spherical mirror with $F = 86.5 \text{ cm}$; F is an aerosol filter; A is a wide-band amplifier with the gain coefficient of 1000; MC-221 is the half-inch microphone.

The Ti:Sa laser used allows obtaining radiation pulses of FS- or NS-duration. In the absence of a FS pulse injected from the master oscillator into the feedback of the regenerative optical amplifier the laser system generates pulses of nanosecond duration ($\tau = 9 \text{ ns}$). Injection of a single FS-pulse from the master oscillator synchronously with the optical pump of the regenerative and output amplifiers provides for obtaining high-power FS pulses at a

repetition frequency of 10 Hz (for details see Ref. 4). In these experiments, to eliminate the self-focusing effect in air, the FS-pulse energy was kept below 5 mJ. Duration of the FS-pulses (FWHM) was about 190 fs. Radii of the Gaussian beams at the output from the laser system are identical in both cases being approximately equal to 5 mm. In this study, the spectrum of NS-pulsed radiation was recorded with a linear CCD array at the exit of a monochromator with the resolution of 0.04 nm. The spectrum was well-approximated by a Gaussian function centered at $\lambda_0 = 791$ nm had the width (FWHM) $\Delta\lambda = 23.4$ nm. In the case of FS-pulses, λ_0 was 800 nm and $\Delta\lambda = 17.7$ nm. Energy of the radiation pulses was changed with a polarization attenuator and measured using a calibrated photodiode.

Laser radiation was directed into a cell of the OA detector by means of two dielectric mirrors. The first mirror was plane; the second one was spherical with focal length F , equal to 86.5 cm. The cylindrical 25-cm-long OA cell of 20-cm diameter, with 2-mm thick windows made from MgF_2 , was installed in such a way that the focal spot produced by the spherical mirror was out of the cell. The radius of laser beam r_0 in the plane of the measuring microphone MC-221 was 2.5 mm. The shapes of the electric signals, generated due to the absorption of radiation pulses by air recorded at the output of a wide-band amplifier by means of a TDS-1012 oscilloscope, are presented in Fig. 2.

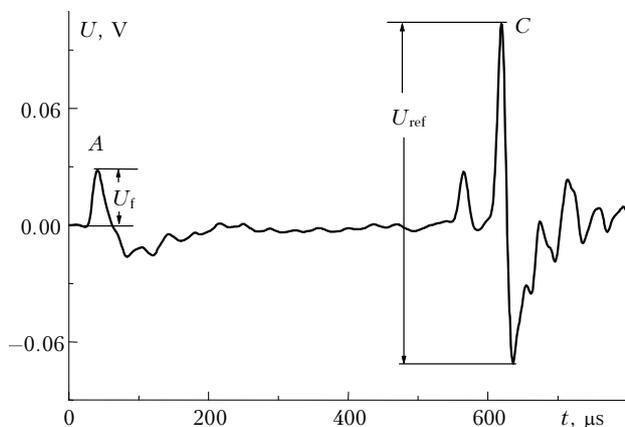


Fig. 2. Oscillogram of the OA-detector signal.

The OA cell is equipped with a sonic concentrator⁵ that consists of two parabolic reflectors and serves for increasing the detector sensitivity. The first, A, pulse with the amplitude U_f (see Fig. 2) corresponds to the action of the pressure jump at the microphone, coming directly from the illuminated area. The C pulse with the amplitude U_{ref} corresponds to the pressure pulse, focused on the microphone by means of the concentrator (for details see Ref. 5). Depending on the absorption, we measured either the amplitude U_f of the direct signal, or of the focused signal U_{ref} .

Figure 3 presents the correlation between U_{ref} and U_f . As follows from Fig. 3, the transition from measurements of U_f to U_{ref} allows one to increase the OA spectrometer sensitivity by approximately 6 times.

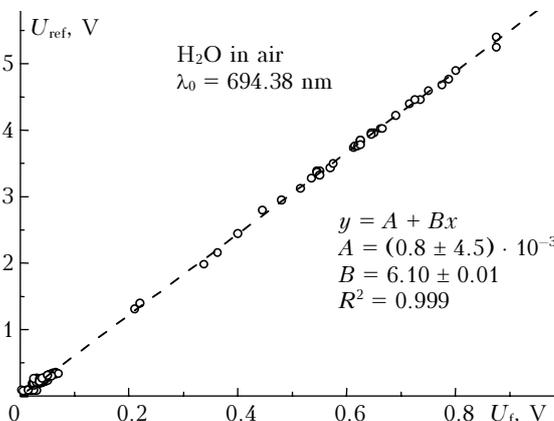


Fig. 3. Correlation between the U_f and U_{ref} amplitudes.

In the present work, at calibration of the OA detector and measurements of the unknown absorption coefficients of the Ti:Sa-laser radiation by water vapor and air, the U_{ref} amplitude was recorded.

Formulas for the OA-signal amplitude at absorption of the NS- and FS- radiation pulses. Calibration of the OA detector

In the case of linear absorption, amplitude of the OA-detector signal U_{ref} is related to the coefficient of radiation absorption by gas, k , laser pulse energy E , and the compression amplitude of pressure signal p_{max} by the relations from Refs. 6 and 7:

$$U_{ref} = akE = K\varepsilon p_{max}, \tag{1}$$

$$p_{max} \approx \frac{V_s^2}{2\pi C_p T r_0^{3/2} r^{1/2}} kE, \tag{2}$$

here a and ε are the OA-detector sensitivity and microphone sensitivity, respectively; K is the coupling coefficient between the amplitudes U_{ref} and U_f ; T is the gas temperature; C_p is the specific gas heat under constant pressure; V_s is the sound velocity in gas; r_0 is the radius of laser beam at the measurement point; r is the distance from the laser beam center to the microphone.

The expression (2) is valid for large size of the Gaussian beam, when the shape of the pressure signal does not depend on time of the heat release.⁷ In formula (1) for the non-monochromatic radiation, the k is determined from the equation⁶:

$$k = k(v_1) = \sum_i \int_{\Delta v} k_i(v)g(v_1 - v)dv, \tag{3}$$

where summation is carried out using the technique from Ref. 8 over the absorption lines, found into the spectral interval $(\nu_1 \pm 25) \text{ cm}^{-1}$, ν_1 is the wave number for the spectrum maximum of laser pulse radiation, related with the corresponding value of radiation wavelength in micrometers by the relation $\nu_1 = 10000/\lambda_1$. The function g characterizes radiation spectrum within the spectral interval $\Delta\nu$. According to Eq. (3), $k(\nu_1)$ is the absorption coefficient, including the contributions from i absorption lines, being in the spectral interval $(\nu_1 \pm 25) \text{ cm}^{-1}$, and distorted owing to the spread function of laser spectrometer g .

Profiles of the absorption lines are given in the form of the Voigt contour⁹:

$$k_i(\nu) = \frac{k_i^{\max} Y}{\pi} \int \frac{e^{-t^2}}{Y^2 + (X - t)^2} dt, \quad (4)$$

where for each of the i th line

$$k^{\max} = \frac{S}{\gamma_D} \left(\frac{\ln 2}{2} \right)^{1/2}; \quad Y = \frac{\gamma_L}{\gamma_D} (\ln 2)^{1/2};$$

$$X = \frac{\nu - \nu_0}{\gamma_D} (\ln 2)^{1/2}; \quad \gamma_D = \frac{\nu_0}{c} \sqrt{\frac{2RT \ln 2}{N_A m}};$$

$$\gamma_L = \Gamma_{\text{self}} P_{\text{self}} + \Gamma_{\text{br}} P_{\text{br}}.$$

Here k^{\max} and ν_0 are the absorption coefficient and wave number values for the line center; γ_D and γ_L are the Doppler and collisional line half-widths; Γ_{self} and Γ_{br} are the coefficients of line self-broadening and broadening by buffer gas; P_{self} and P_{br} are the partial pressures of absorbing and broadening gases, respectively; S is the line intensity; R is the universal gas constant; N_A is the Avogadro number; m is the mass of absorbing molecule; c is the speed of light.

Calibration of the OA detector was carried out according to known (calculated) absorption of a pulsed ruby laser radiation of 50-ns duration and the spectrum width at a half-height of $\gamma_1 = 0.04 \text{ cm}^{-1}$ by water vapor in air at atmospheric pressure. The calibration procedure is presented in detail in Ref. 10. Most accurately the constant of the OA-detector calibration a is determined from the dependence of $U_{\text{ref}} E^{-1}$ measured in the experiment at the fixed wavelength and several values of the water vapor partial pressure, on the absorption coefficient k , calculated for the same humidity values by formula (3).

In this study, we measured $U_{\text{ref}} E^{-1}$ with a ruby laser and the calculations of k were carried out for the maximum of the H_2O absorption line at 694.380 nm of the band $\nu_1 + 3\nu_3$. In calculations of the absorption coefficients, data on the S , Γ_{self} and Γ_{br} parameters for water vapor absorption lines broadening by air were taken from HITRAN-2004.¹¹ Integration in formula (3) was carried out over the interval of $\Delta\nu = \nu_1 \pm 1.5\gamma_1$. The alternative calculations by the authors have shown that

integration over a wider interval $\Delta\nu_1$ leads to the variation of the integral value in Eq. (3) by no more than 0.01% for any wave number ν_1 .

Sensitivity of the OA detector determined from the slope of the linear regression of the $U_{\text{ref}} E^{-1} = f(k)$ experimental dependence at the radius of ruby laser radiation beam $r_0 = 2.1 \text{ mm}$ gave the value of $a = (3.97 \pm 0.44) \cdot 10^7 \text{ V} \cdot \text{cm} \cdot \text{J}^{-1}$. The calculation error of absorption coefficient was about 5%, the total calibration error was determined to be at the level of 11%. Since $a \sim r_0^{-3/2}$ (see the formulas (1) and (2)), allowing for this fact and measurement errors of laser beam dimensions for the OA-detector sensitivity in the experiment with a Ti:Sa laser, we have $a = (3.05 \pm 0.52) \cdot 10^7 \text{ V} \cdot \text{cm} \cdot \text{J}^{-1}$.

Physics of the high-power narrow light pulse absorption in molecular gases is considered in Ref. 12. The nature of the effect is that the electromagnetic field interacts with the dipole moment of molecules induced by the field of linearly polarized laser radiation. The absorbed energy of laser pulses is spent for increase of the rotational energy of molecules. Formula for the pressure signal amplitude in the case of the FS laser pulse absorption by diatomic molecules of N_2 and O_2 in the absence of self-focusing effect was obtained in Ref. 13 and has the following form

$$p_{\max} \approx \frac{V_s^2}{2\pi C_p T r_0^{3/2} r^{1/2}} \frac{2.64 G (\alpha_{\parallel} - \alpha_{\perp})^2 N E}{Q J c^2 r_0^2} E, \quad (5)$$

where $(\alpha_{\parallel} - \alpha_{\perp})$ is the difference between the polarizability components along the molecular axis and across it; N is the number of molecules in a unit volume; J is the molecular momentum; Q is the rotational partition function, which is determined as $Q = RT / BN_A$ (see Ref. 14), when $RT \gg BN_A$, $B = \hbar / (2J)$ is the rotational constant of the molecule.

According to Ref. 13, in the formula (5), G is determined by the formula

$$G = \sum_{j=0}^{\infty} e^{\left[\frac{-2B^2}{\hbar^2} (2j+3)^2 \tau^2 \right]} \frac{(j+1)(2j^3 + 4j^2 + 26j + 15)}{(2j+3)^2(2j+5)} \times$$

$$\times \left\{ e^{\left[\frac{-BN_A}{RT} j(j+1) \right]} - e^{\left[\frac{-BN_A}{RT} (j+2)(j+3) \right]} \right\}. \quad (6)$$

For the air (80% of nitrogen and 20% of oxygen) at $T = 295 \text{ K}$ and for the laser pulse duration of $\tau = 190 \text{ fs}$, $G = 0.886$ and $Q = 108$.

As follows from comparison of the formulas (2) and (5), in the case of energy absorption of a FS laser pulse by air, k is determined by the relation

$$k = \frac{2.64 G (\alpha_{\parallel} - \alpha_{\perp})^2 N E}{Q J c^2 r_0^2}, \quad (7)$$

i.e., the coefficient of induced absorption without self-focusing effect is directly proportional to the laser pulse energy.

Preparation of the investigated samples and measurement of their characteristics

To remove solid fraction of atmospheric aerosol, air intake in the OA cell was carried out through an aerosol filter. Water vapor content in the air samples and temperature were recorded with a DV2TS ("Mikrofor") sensor of the relative humidity and temperature. Water vapor was bled-in by evaporating liquid water out of a flask into the preliminary evacuated cell. Air pressure or water vapor pressure in the cell was measured by means of a DVR-5 electronic manometer with the errors of 0.1 and 8%, respectively.

Results and discussion

Figure 4 presents the absorption coefficients of air as functions of the laser pulse energy for the Ti:Sa-laser radiation pulses of $\tau = 9$ ns and 190 fs duration at the partial pressure of water vapor $P_{\text{H}_2\text{O}} = 11.2$ mbar, and the total pressure of $P_{\text{total}} = 997$ mbar and temperature of 295 K.

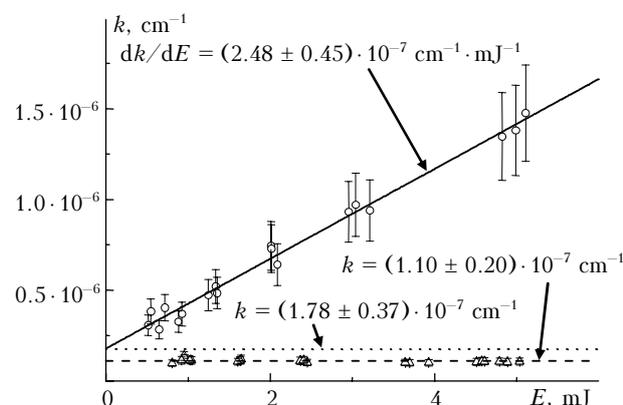


Fig. 4. Absorption coefficient of air for Ti:Sa-laser radiation depending on the laser pulse energy at $\tau = 9$ ns (Δ) and $\tau = 190$ fs (\circ).

For determination of the absorption coefficients values by formula (1) from the measurement results by the $U_{\text{ref}}E^{-1}$ relation, we have used the above-stated value of the OA-detector sensitivity $a = (3.05 \pm 0.52) \times 10^7 \text{ V} \cdot \text{cm} \cdot \text{J}^{-1}$. Figure 4 presents the total measurement errors in the absorption coefficients owing to the error of the OA-detector calibration.

As follows from Fig. 4, the absorption coefficient of NS pulses does not depend on energy and due to the above-stated characteristics for the air makes $k = (1.10 \pm 0.20) \cdot 10^{-7} \text{ cm}^{-1}$. The absorption coefficient for FS pulses is directly proportional to energy, as it should be expected according to

formula (7). The rate of the increase is $dk/dE = (2.48 \pm 0.45) \cdot 10^{-7} \text{ cm}^{-1} \cdot \text{mJ}^{-1}$. The residual absorption of FS pulses observed at $E \approx 0$ is characterized by $k = (1.78 \pm 0.37) \cdot 10^{-7} \text{ cm}^{-1}$.

Let us compare the measured values of the linear absorption coefficient with the calculated data. Figure 5 presents the absorption spectra of atmospheric air in the region from 750 to 840 nm, calculated for the experimental conditions using HITRAN-2004 database.

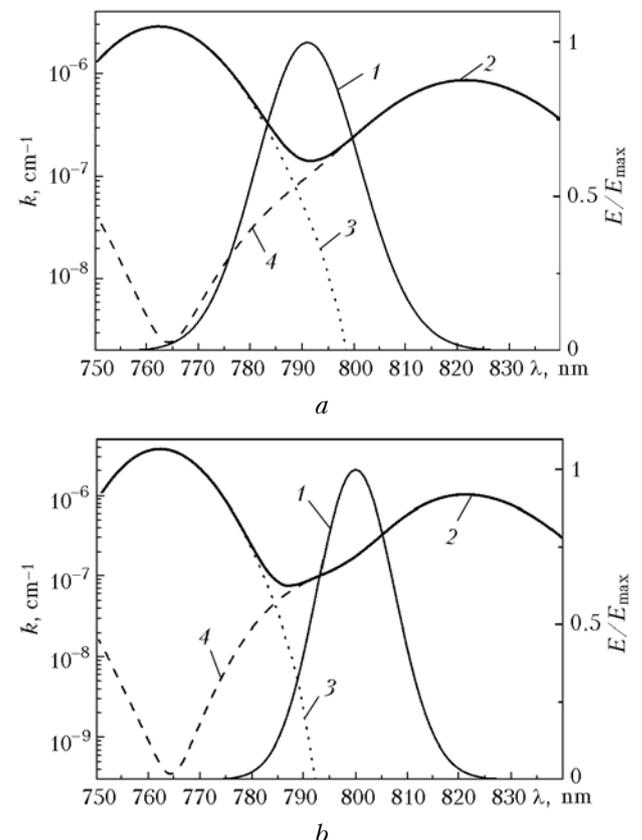


Fig. 5. Model spectra of NS (a) and FS (b) laser pulses (curve 1) and calculated spectra (curve 2) of air absorption corresponding to the experimental conditions. Contributions to the air absorption coefficient from oxygen (content of O_2 of 20%) (curve 3) and from water vapor (content of H_2O of 11.2 mbar) (curve 4).

The calculations were made by formula (3) at $P_{\text{H}_2\text{O}} = 11.2$ mbar, $P_{\text{O}_2} = 197$ mbar, $P_{\text{total}} = 997$ mbar and $T = 295$ K allowing for the spectrometer instrumental function (Gaussian width $\Delta\lambda = 23.4$ nm for the NS pulses (Fig. 5a) and $\Delta\lambda = 17.7$ nm for the FS pulses (Fig. 5b)). According to estimates,¹⁵ the errors of such calculations do not exceed 8%.

As follows from Fig. 5, for the FS radiation at $\lambda_0 = 800$ nm, the absorption by air of the low-intensity radiation is caused only by water vapor. Contribution from the absorption due to rovibrational water vapor transitions to the air absorption coefficient of the Ti:Sa-laser NS-pulses makes about 70%, the rest being caused by the

electron excitation of the oxygen molecules into the $b^1\Sigma_g^+$ state.

The calculated and measured values of the absorption coefficients are given in the Table.

Measured and calculated values of the linear absorption coefficients of the Ti:Sa-laser FS- and NS-radiation pulses in air at $P_{\text{H}_2\text{O}} = 11.2$ mbar, $P_{\text{O}_2} = 197$ mbar, $P_{\text{total}} = 997$ mbar and $T = 295$ K

Spectral radiation characteristics	Absorption coefficient, 10^{-7} cm^{-1}			
	H ₂ O	O ₂	H ₂ O + O ₂	
	Calculation			Measurement
FS				
$\lambda_0 = 800$ nm,	1.74±0.14	0	1.74±0.14	1.78±0.37
$\Delta\lambda = 17.7$ nm				
NS				
$\lambda_0 = 791$ nm,	0.98±0.08	0.43±0.03	1.41±0.11	1.10±0.20
$\Delta\lambda = 23.4$ nm				

The calculated absorption of the FS pulses almost coincides with the measured residual absorption. The measured absorption coefficient of air for NS pulses is somewhat smaller than the calculated value and it best agrees with calculation for water vapor when oxygen absorption is ignored. The obtained relation between the calculated and measured values of the absorption coefficients for NS pulses can be explained if one takes into account that collisional relaxation of the excited oxygen molecules is considerably slower and the contribution into the OA signal is less effective owing to this process than contribution from the excited H₂O molecules. In particular, time of vibrational-translational relaxation of the O₂ molecules due to collisions with each other at $P = 1$ mbar, makes about 14 s (see Ref. 16). At the same time water vapor molecules in air relax, at the same pressure, due to the H₂O–O₂ and H₂O–N₂ collisions for the time of about 10^{-4} s, but owing to the H₂O–H₂O collisions during about 10^{-6} s (Ref. 17). Investigation of the generation efficiency of the OA signal at the Ti:Sa-laser NS-pulse absorption by oxygen and water vapor can be the object of a special experiment that is beyond the scope of the present study.

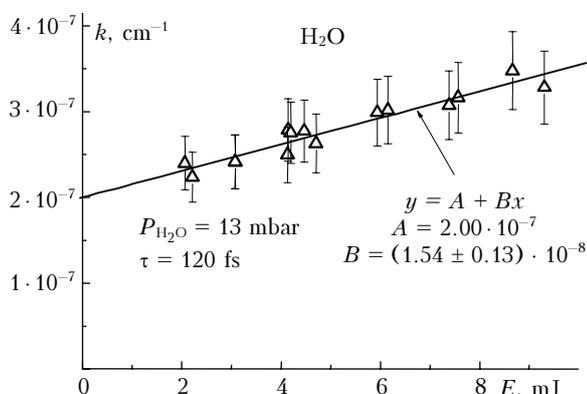


Fig. 6. The dependence of absorption coefficient of water vapor for Ti:Sa-laser FS-pulse ($\tau = 120$ fs) on the pulse energy; $dk/dE = (1.54 \pm 0.20) \times 10^{-8} \text{ cm}^{-1} \cdot \text{mJ}^{-1}$.

Figure 6 presents the dependence of the absorption coefficient of water vapor for the FS radiation at $P_{\text{H}_2\text{O}} = 13$ mbar on the laser pulse energy at $\tau = 120$ fs. As follows from Fig. 6, for the FS radiation an induced absorption of radiation occurs, in addition to the absorption due to rovibrational transitions in H₂O. At $P_{\text{H}_2\text{O}} = 13$ mbar and laser pulse duration of $\tau = 120$ fs, the coefficient of induced absorption increases at a rate of $dk/dE = (1.54 \pm 0.20) \cdot 10^{-8} \text{ cm}^{-1} \cdot \text{mJ}^{-1}$. It should be noted that the OA-detector sensitivity obtained from the calibration measurements using a ruby laser can not be applied to the measurement results on the $U_{\text{ref}}E^{-1} = f(E)$ dependence for water vapor, and formulas (5)–(7) for the H₂O molecule will have another form. In this case, the spectrometer calibration was carried out according to the linear (residual) FS-pulse absorption of the Ti:Sa-laser radiation by water vapor at self-broadening, for which the calculated value of the absorption coefficient is $k = (2.00 \pm 0.16) \cdot 10^{-7} \text{ cm}^{-1}$.

Conclusion

In this study, the coefficients of induced absorption of FS-radiation pulses by air and water vapor have been measured for the first time by means of the OA spectrometer, built up around a Ti:Sa laser. In the absence of self-focusing effect, the coefficient of induced absorption increases proportionally to E . For the Gaussian beams of 2.5 mm radius, at $\lambda_0 = 800$ nm and spectral width $\Delta\lambda = 17.7$ nm, the coefficient of induced absorption by air at atmospheric pressure increases at a rate of $dk/dE = (2.48 \pm 0.45) \cdot 10^{-7} \text{ cm}^{-1} \cdot \text{mJ}^{-1}$ (pulse duration of 190 fs). In water vapor at the partial pressure of 13 mbar, the increase rate is $dk/dE = (1.54 \pm 0.20) \times 10^{-8} \text{ cm}^{-1} \cdot \text{mJ}^{-1}$ (pulse duration of 120 fs).

The coefficient of linear (residual) absorption of the Ti:Sa-laser FS-radiation pulses by air determined in measurements, well agrees with the absorption coefficient of laser pulses at vibrational-rotational transitions of water vapor molecules, obtained from the calculations using the HITRAN database.

Acknowledgments

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