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COMMUNICATIONS

Water Vapor Absorption of 266-nm Nanosecond Laser Pulses with Linear and Circular Radiation Polarization

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Abstract—An increase of 50% in the amplitude of the photoacoustic signal generated when water vapor absorbs pulses of circularly polarized laser radiation at 266 nm relative to the signal for linearly polarized radiation is reported. The effect observed is associated with different dissociation efficiency of H_2O molecules as a result of one- and two-photon absorption of linearly and circularly polarized radiation.

Keywords: absorption of UV radiation, water vapor, dissociation, acoustic pulse anisotropy

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Dissociation of H_2O as a result of absorption of UV laser radiation pulses is discussed in [1], which explains the existence of the absorption band of water vapor [2] with a maximum of ~270 nm and reduced absorption coefficient at maximum $k \approx 2.3 \times 10^{-6}$ cm^{-1} mbar^{-1} [3, 4]. The authors of [1] showed that the dissociation of H_2O (photochemical reactions $O + H_2$ and $OH + H$) in the one-photon absorption of UV laser pulses with a photon energy close to but less than the dissociation energy can occur through intermediate vibrationally excited states of H_2O molecules in the ground electronic state. Photodissociation of H_2O molecules under the above absorption conditions has not been experimentally observed (see [5] and the list of cited literature).

Anisotropy of time-resolved acoustic signals generated in the chamber of an photoacoustic detector (PAD) as a result of the dissociation of Cl_2 molecules upon absorption of nanosecond (355-nm) pulses of linearly and circularly polarized laser radiation is reported in [6]. The amplitude of the PA signal is found to be maximal when the polarization plane of the linearly polarized radiation is perpendicular to the acoustic pulse propagation direction, and to be minimal when the acoustic pulse propagation direction is parallel to the radiation polarization. For circularly polarized radiation, the PA signal amplitude has a median value. The effect is observed at chlorine pressures under which the free path of the fragments (Cl atoms) is comparable to and greater than the distance from the laser beam to the microphone membrane.

The aim of this work is to study the amplitude of the PA signals generated due to absorption of nanosecond pulses of radiation at a wavelength of 266 nm by water vapor depending on the intensity of radiation with linear and circular polarizations.

The experimental setup is based on a YAG laser (LS-2134U) and a PAD with time-resolved signals (Fig. 1). The detailed description of the setup can be found in [7]. In addition to [7], the radiation polarization is controlled using a quarter-wave plate ($\lambda/4$) in this work. The polarization type is determined by the intensity of the spots for the ordinary and extraordinary radiation components on screen E1 after placing a birefringent prism in the main beam (60°). The polarization of the initial laser radiation is vertical and perpendicular to the plane of the microphone membrane. The measurement technique is the same as in [7].

Figure 2 shows the amplitudes of the PA signals U_0 recorded upon absorption by water vapor of laser pulses at a wavelength of 266 nm versus the energy E (peak intensity I_0) of radiation with circular and vertical and horizontal linear polarizations. The amplitudes of the PA signals are evidently the same in the cases of linear polarizations, while the signal amplitude is approximately one and a half times greater for circularly polarized radiation. Approximation of experimental data by the third-degree polynomial $Y = b_1X + b_2X^2 + b_3X^3$ (curves in Fig. 2) shows the contribution of the linear and two-photon radiation absorption by water vapor into the PA signal in the region $I_0 = (10^{-3} - 1.4)$ GW/cm^2; three-photon absorption is not

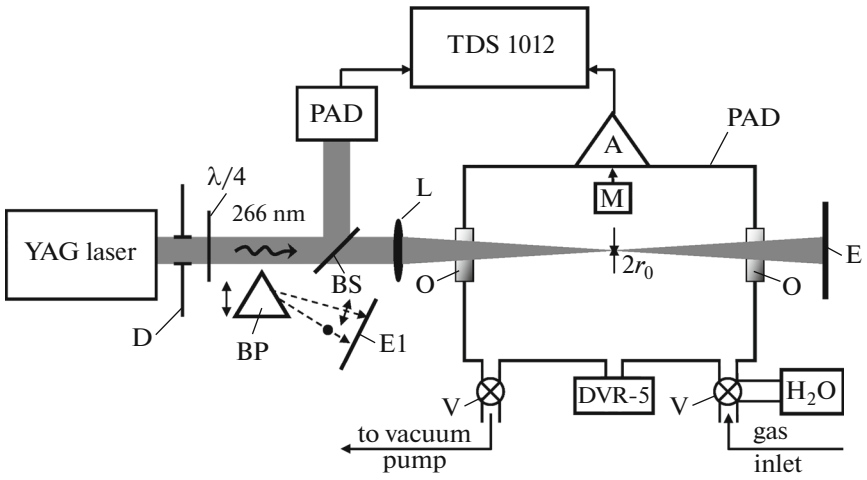

Fig. 1. Block-diagram of the setup: iris diaphragm (D), quarter-wave plate (λ/4), birefringent prism (BP), beam-splitter (BS), lens with a focal length of 100 cm (L), optical windows (O), MK-301 condenser microphone (M), screens (E and E1), vacuum valve (V), vacuum gage (DVR-5), microphone signal amplifier (A), calibrated PA receiver (PAR), and digital oscilloscope (TDS 1012).

observed in water vapor and in its mixtures with nitrogen [7]. The approximation coefficients $b_1 \approx (9 \pm 1)$ and $b_2 \approx (2.4 \pm 0.3) \times 10^3$ for linear polarizations and $b_1 = (12 \pm 1)$ and $b_2 = (4.0 \pm 0.3) \times 10^3$ for circular polarization. Coefficient $b_3 = 0$ for all radiation polarization types, i.e., the observed increase in the amplitude of the PA signal for circularly polarized radiation is associated with an increase in the contributions to the PA signal from both linear and two-photon absorption.

The physical cause of the increase in the PA signal amplitude for circularly polarized radiation is the optical orientation of atoms: when molecules interact with circularly polarized resonant radiation, photodissociation of water molecules occurs through unstable states near the dissociation threshold, as well as the transfer of angular momentum from light to the products of dissociation [8]. The effect detected is not instrumental, since measurements performed for strongly absorbing SO_2 molecules of the same symmetry C_{2V}, but a 0.6-eV higher dissociation energy [9] showed no increase in the absorption in the case of circular polarization. We did not detect the difference between the dependences $U_0(E)$ exceeding the measurement error in the PA signal amplitudes in the cases of absorption of linearly and circularly polarized radiation, since the mean free path of the dissociation products in our experiment is much shorter than the distance from the laser beam to the microphone. The decrease in the distance to 2 mm and in the water vapor pressure to 1 mbar did not maintain measurement conditions close to [6], primarily due to the significantly weaker absorption by H_2O (266 nm) as compared to Cl_2 (355 nm).

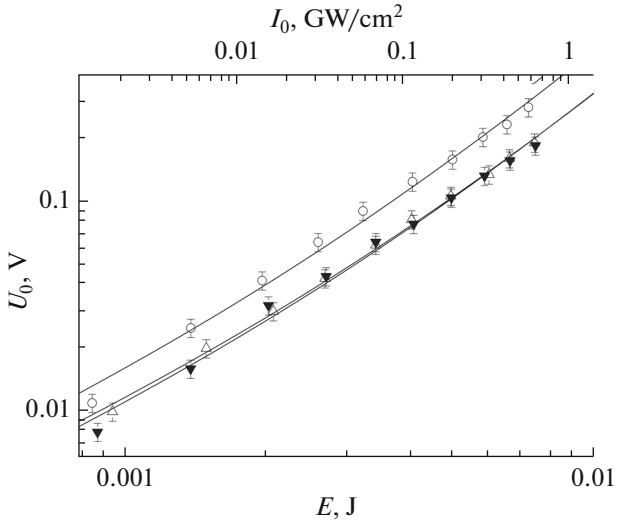

Fig. 2. PAD signal amplitudes versus energy E (peak intensity I_0) of radiation at 266 nm with circular (○), linear horizontal (▼), and linear vertical (△) polarization; the water vapor pressure is 5.5 mbar.

Thus, in this work, an increase in the amplitude of the PA signal generated in water vapor upon absorption of nanosecond pulses of laser radiation at a wavelength of 266 nm with a change in polarization from linear to circular was recorded, which is beyond the measurement errors. The contribution to the PA signal from linear and nonlinear absorption is shown in the region of variation in the peak radiation intensity $I_0 = (10^{-3} - 1.4)$ GW/cm^2, which is associated with one-photon and two-photon dissociation of H_2O molecules. Single-photon dissociation of H_2O runs through the excited state A($^1B_1$) with the formation of dissociation products H + OH (X^2P), and two-photon dissociation, through the states B($^1A_1$) with the formation of dissociation products H + OH ($A^2\Sigma^+$)

and H + OH (X^2P) [10]. Dissociation into O and H_2 is ineffective, especially two-photon, due to the existence of potential barriers.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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