Fluorescence of the atmosphere under the exposure to fifth harmonic of Nd:YAG laser (212.8 nm)

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First results of measuring the fluorescence of the atmosphere and its basic constituents under the exposure to radiation of the fifth harmonic of a Nd:YAG laser (wavelength of 212.8 nm) are presented.

Introduction

The studies of 1984–1996 into the absorption and fluorescence of the atmosphere under the exposure to radiation of various laser sources in the region of 248–337 nm enabled revealing absorption in the region of 248–320 nm and fluorescence induced by it in the region of 285–500 nm. It was found experimentally[2] that this phenomenon is connected with water vapor.

The longwave part of the spectrum has been studied rather thoroughly ($\lambda \geq 266$ nm), whereas in the shortwave portion the fluorescence was measured only with the excitation at the wavelength of 248 nm. In this paper, we present the first results on studying the fluorescence of the atmosphere and its basic constituents under the exposure to radiation the fifth harmonic of a Nd:YAG laser ($\lambda = 212.8$ nm).

Experiment

A remote fluorescence spectrometer (Fig. 1) included a 3-m long cell with the forevacuum evacuation (minimum pressure of 0.05 mm Hg). The cell length, the angle between the directions of excitation and recording, and split entrance and exit windows were chosen and made in such a way that only radiation from the central part of the cell comes into the field of view of the receiving system (MDR-6 double monochromator and PMT). To exclude the effect of atmospheric fluorescence from the setup parts before and after the cell, the exciting radiation passed inside the screening pipes, and the inactive part of the exit window was also screened. Measuring time-resolved fluorescence spectra in several successive 50-ns long intervals enabled additional separation of the signals. Signals were recorded in the photon counting mode.

The radiation of a Nd:YAG laser with an unstable resonator and a passive Q-switch was sequentially converted into the second, fourth, and fifth harmonics in KTP, KDP, and BBO crystals, respectively. The energy per pulse at the frequency of the fifth harmonic was about 2 mJ, and the pulse repetition frequency was 5 Hz.

The fluorescence of the atmosphere, as well as gaseous nitrogen (obtained through evaporation of liquid nitrogen), technical oxygen, and water vapor (obtained through evaporation of medical water for injections) was measured; the maximum pressure of nitrogen and oxygen was 1 atm, and the water vapor pressure was 8 mm Hg. The measurements were conducted at the temperature about 18°C.

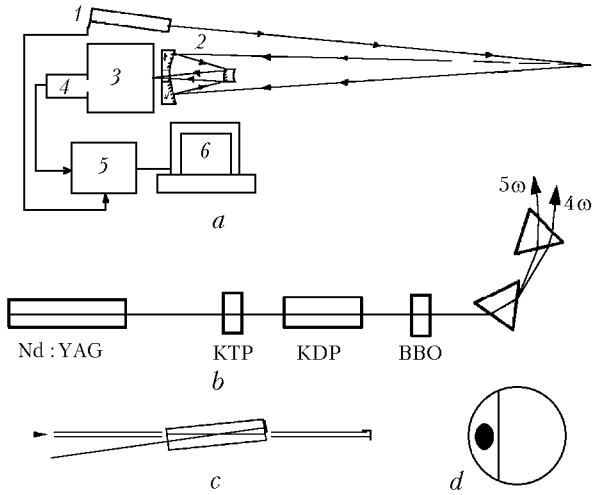

Fig. 1. Experimental setup (*a*), laser source (*b*), gas cell (*c*), and its windows (*d*): laser *1*, receiving telescope *2*, monochromator *3*, photomultiplier tube *4*, photon counter *5*, computer *6*, position of the beam exciting the fluorescence (*d*).

Results

The fluorescence of the atmosphere was measured under laboratory conditions in Moscow and Tomsk. The fluorescence signal is stably reproduced with insignificant variations; therefore, fluorescing substances are the main atmospheric gases. The fluorescence spectrum occupies the region of 260 to 420 nm and is bell-shaped with the maximum near 340 nm (Fig. 2). At the atmospheric pressure the signal is almost completely concentrated in the first 50-ns interval, the decrease of the pressure elongates the signal (at 20 mm Hg the radiation completely occupies the second time interval) with the spectral shape keeping unchanged.

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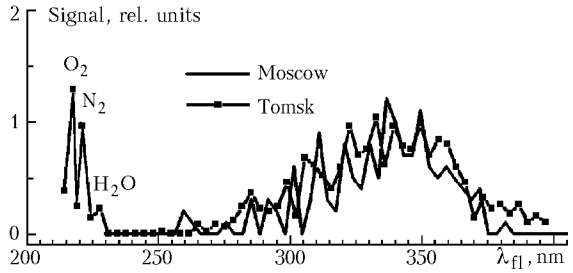

Fig. 2. Fluorescence spectrum of the atmosphere (Raman lines of three atmospheric gases are seen on the left).

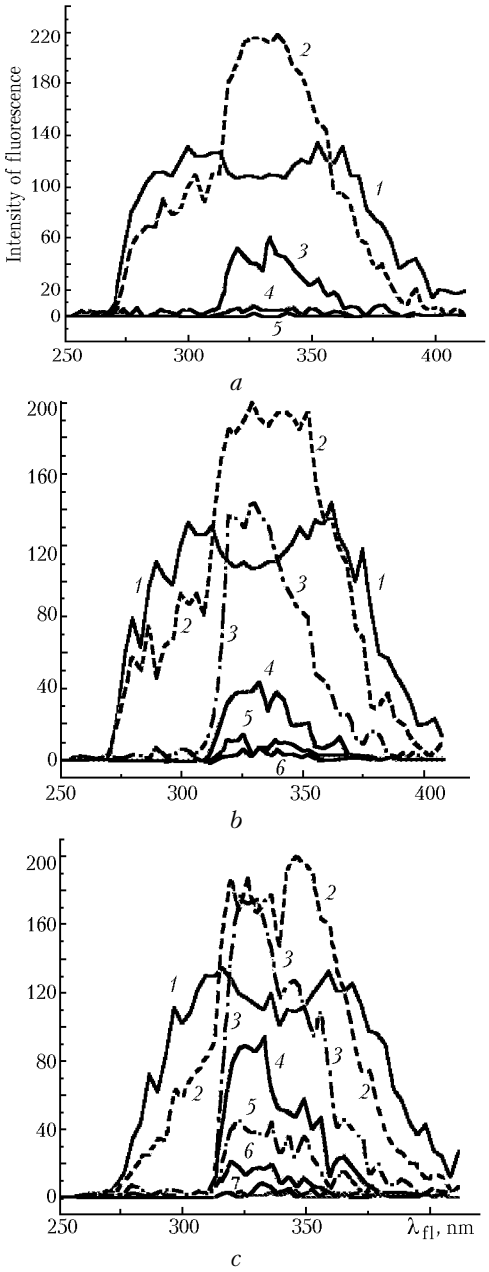

Fig. 3. Fluorescence spectrum of nitrogen at the pressure of 1 (*a*), 0.5 (*b*), and 0.2 atm (*c*). Time intervals: 0–50 (*1*), 50–100 (*2*), 100–150 (*3*), 150–200 (*4*), 200–250 (*5*), 250–300 (*6*), and 300–350 ns (*7*).

The recorded fluorescence spectrum from a cell filled with nitrogen consists of two distinct components (Fig. 3; the number of photoelectrons recorded for 100 excitation pulses is plotted along the vertical axis in this figure as well as in the figures that follow). The fluorescence signal in the first time interval (both the spectral distribution and the value of the signal) is, on the whole, similar to the signal from the atmosphere. In the second and following time intervals, a signal concentrated at 316–370 nm arises (the shortwave boundary is very sharp); the maximum value of the signal in the second interval is roughly twice as large as that in the first interval. As the pressure decreases, fluorescence drastically elongates (up to 350 ns at 0.2 atm).

The fluorescence spectrum of water vapor is similar in many aspects to the spectrum of nitrogen: the signal is also concentrated in the second and following time intervals in the region of 316–370 nm (Fig. 4). The value of this signal at the water vapor pressure of 8 mm Hg is also roughly twice as large as the signal from the atmosphere and becomes equal to that only as the pressure decreases down to 1 mm Hg. The shape of the time-integrated spectral distribution of water vapor fluorescence and the position of the maximum correspond, on the whole, to those obtained earlier at the temperature of 24°C under the exposure to the fourth harmonic of the Nd:YAG laser (λ = 266 nm) (Ref. 3).

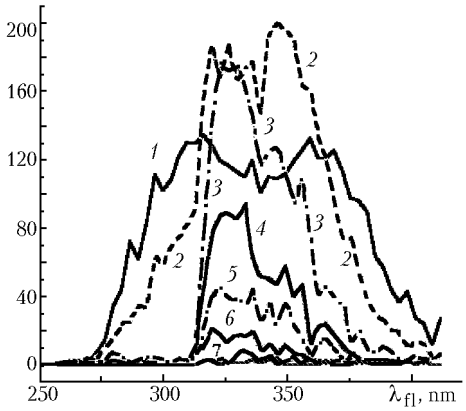

Fig. 4. Time behavior of water vapor fluorescence spectrum at the pressure of 8 mm Hg (designations are the same as in Fig. 3).

Table. Parameters of water vapor fluorescence

Excitation wavelength, nm	Fluorescence cross section, 10^{-24} cm^2	Quenching cross section, 10^{-15} cm^2
212.8	2	by water vapor –270
248.5, Refs. 1 and 2	1.1	by nitrogen – 10^{-2} by oxygen– 10 by air – 0.5
266, Ref. 3	0.56 at 24°C 6.9 at 72°C	by water vapor – 1.5
270, Ref. 2	2.5	
308, Ref. 2	0.1	

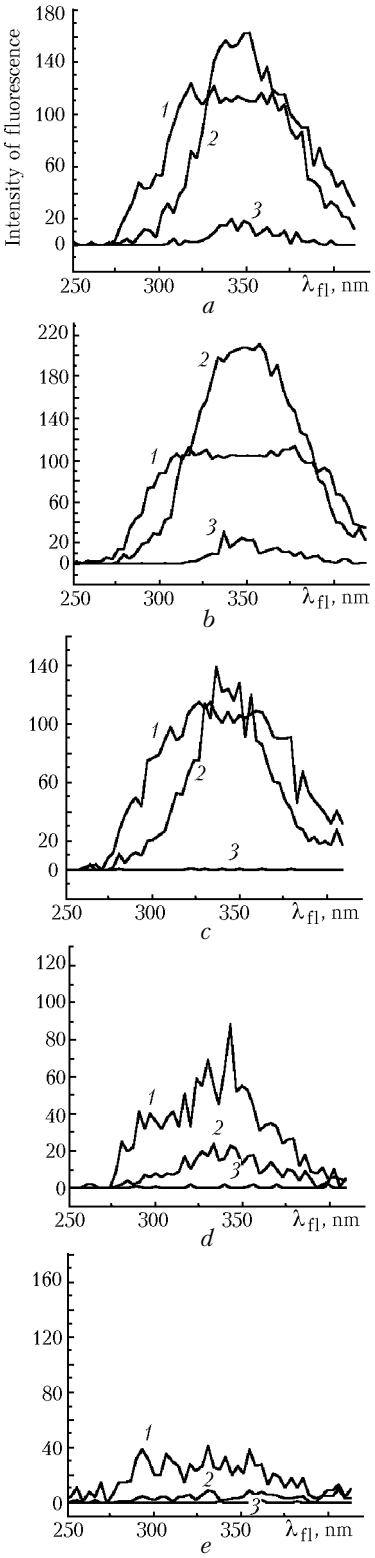

Fig. 5. Oxygen fluorescence spectrum at the pressure of 0.1 (*a*), 1 (*b*), 8 mm Hg (*c*), 0.1 (*d*), and 1 atm (*e*). Time intervals: 0–50 (*1*), 50–100 (*2*), and 100–150 ns (*3*).

The Table gives the cross sections of water vapor fluorescence and its quenching as measured earlier and in this work. It can be seen that the fluorescence cross section varies non-monotonically with the wavelength.

The shape of the oxygen fluorescence spectrum coincides, on the whole, with that of the atmospheric fluorescence spectrum (Fig. 5). At the oxygen pressure of 0.5–4 mm Hg the maximum value of the signal in the second time interval exceeds by 1.5 to 2 times the signal in the first interval, at the pressure of 8 mm Hg they become equal, and then the signal in the second channel rapidly decreases. Starting from 30 mm Hg the total fluorescence signal decreases, and the decrease reaches 2.5 to 3 times at the atmospheric pressure of oxygen. This decrease is obviously the consequence of stronger quenching.

Conclusions

Fluorescence of the atmosphere under the exposure to radiation at the wavelength of 212.8 nm is mostly caused by oxygen and nitrogen.

Water vapor also fluoresces under these conditions, but contributes insignificantly to the fluorescence of air at the atmospheric pressure. The delay in appearance of the fluorescence signal is likely connected with a complex step-wise mechanism of the fluorescence excitation.

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