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High time-resolved spectroscopy of filament plasma in air

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ABSTRACT

The results of spectral–temporal studies of the filament plasma emission in air with a time resolution of 2 ps were presented. The filament was created by laser radiation with pulse duration of 60 fs and wavelength of 940 nm. It was shown that a spectral composition of the filament plasma depended on the numerical aperture (NA). At NA > 0.035, the atomic and ion lines of oxygen and nitrogen dominate in the spectrum on the background of an intense continuous continuum in the visible region. Molecular lines of the second positive system of nitrogen and the first negative system of molecular ions appear in the spectrum for NA < 0.03. In case of NA < 0.01, the spectrum consists of only the molecular and ion lines of N_2 and N_2^+. The emission intensity maxima of these lines delay relative to the laser pulse. The atomic lines begin to glow per (80–100) ps after the laser pulse. The characteristic time of the luminescent lines decay at 1/e level for ions is 18 ps, for molecules is 27 ps, and for atomic lines is 20 ns. Rapid decrease in the intensity of molecular lines emission is due to the deactivation of the particles upper states by electrons. The formation of $N_2^+(B^2\Sigma_u^+)$ excited states is most likely through their $N_2^+(X^2\Sigma_g^+)$ ground state in the process of multiphoton absorption of pumping radiation by nitrogen molecules and subsequent their excitation by electron impact.

1. Introduction

For the first time, the time dynamics of the filament plasma luminescence formed by a femtosecond radiation pulse was studied in [1]. Nd:YLF laser radiation with a wavelength of $\lambda = 1.053$ μm and a pulse duration of 500 fs was used to create the filament. The spectra were recorded using a spectrograph with temporal gating (min is 4.5 ns) and a streak camera (in the absence of spectral resolution). The authors divide three time stages in the plasma emission. During the first stage, the molecular and ion lines of N_2 and N_2^+ dominate, which appear immediately after the laser action (time t_0) with characteristic decay time of $\tau \sim 60$ ps. In the second stage, in one nanosecond from t_0, the continuum spectrum with $\tau \sim 30$ ns becomes predominant. In the third stage, atomic lines of N and O emerge from the decay of the continuum and last up to 1 μs. In [2] the luminescence duration of N_2 in the filament plasma was determined using pump-probe technology and it was ~85 ps. At the same time, in [3] only the growth front of $N_2^+$and N_2 luminescence was 150 ± 200 ps, according to measurements. It should be noted, however, that the minimum gate width was 100 ps. In the recent paper [4], measurements with a time-resolved camera (30 ps) showed that the rise of N_2^+ luminescence front was determined by a temporal resolution and $\tau = 600$ ps. The authors connect the value of τ with the lifetime of free electrons in the filament. They believe that the value of τ is determined by the electrons quenching process of $N_2^+(B^2\Sigma_u^+)$ upper state.

Application of interferometry and pump-probe technology allows measuring the electrons concentration (N_e) in filament plasma with subpicosecond time resolution [5,6]. In [6] it was shown that the electron concentration depends on the pressure and decreases within 1 ns by more than an order of magnitude (from 10^{17} to 6×10^{15} cm^{-3}) at 1 bar of nitrogen. At the same time, in [7–9] a decay time of N_e is within of 1–10 ns. Thus, the information about the luminescence temporal evolution and the filament plasma density produced in air by femtosecond pulse of radiation is contradictory.

The purpose of this paper is a further study of the temporal dynamics of the filament plasma luminescence in air with a higher temporal resolution. The luminescence of atoms, molecules, and nitrogen ions is studied when the laser beam is focused by lenses with different focal lengths.

2. Experimental setup and calibration

2.1. Experimental setup

Experiments were performed using a Ti–Sapphire laser system (Avesta Project), which generated femtosecond laser pulses of radiation with central wavelength $\lambda = 950$ nm, bandwidth of 26 nm, repetition rate of 10 Hz and transform limited pulse duration of 60 fs (FWHM). Laser pulse radiation with energy up to $E = 15$ mJ was focused into ambient air by a lens with focal length of 3–320 cm. The laser pulse

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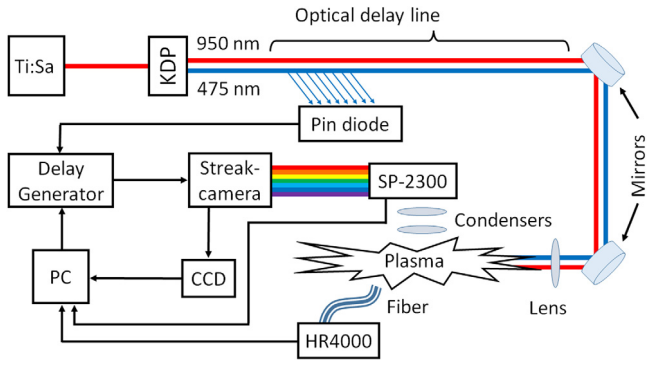

Fig. 1. Experimental setup.

energy fluctuations were ± 5%. The initial beam had diameter of 10 mm ($1/e^2$ level of intensity), quality coefficient $M^2 = 1.5$ and Gaussian-like shape of the transversal intensity profile. For part of the experiments, the radiation was partially transformed into the second harmonic (SH) in a 2-mm thick KDP crystal. SH radiation served as a reference point in the time. Schematic of the experimental setup used to measure the spectrum and filament diameter is shown in Fig. 1.

The time measurement of plasma spectral lines was made transverse to the filament. For this purpose, the plasma radiation was collected and focused on the spectrometer slit by two condensers. After spectrometer, the radiation was applied to the entrance slit of the streak camera, at the output of which the image expanded in time and over the spectrum was recorded by a CCD camera. Then the information was transferred and processed through the interface to the computer. The spectrometer (Acton Spectra Pro SP-2300) had 3 interchangeable gratings: 50, 300 and 1200 grooves/mm. The limiting spectral resolution at the minimum input slit was 0.27 nm. The streak camera (Hamamatsu, Universal Streak Camera C10910) had two time sweeps: fast (minimum scan was 100 ps) and slow (minimum scan was 1 ns). The maximum time resolution of the streak camera was 0.644 ps. In pulse accumulation mode, the temporal resolution for 10^4 pulses was 4.3 ps due to the presence of jitter in streak camera operation. The camera was triggered by a scattered pump radiation detected by a pin-diode installed near the spectrometer to reduce the jitter. The resulting signal from the streak camera was averaged over 300 pulses. The laser radiation passed about 14 m and only then it was focused to compensate the time delay of the streak camera scanner.

Measurement of the diameter of filament plasma column in time was carried out in a scheme in which the radiation was focused along the slit of the spectrometer. At the same time, it was switched to "0" mode, in which there was no spectral resolution. To estimate the plasma electron concentration the dynamics of the hydrogen line H_α broadening under the action of Stark effect was also studied. In this case, the laser radiation was focused into a cell filled with a mixture of nitrogen and hydrogen in a ratio of 15/1 at a total pressure of 1.5 atm. The lens focal length was 15 cm and the radiation energy $E = 15$ mJ. The time-integrated emission spectra of the filament plasma were recorded by the HR-4000 spectrometer (Ocean Optics, 200–1100 nm, 0.75 nm spectral resolution).

2.2. Calibration

When the spectrometer is combined with the streak camera, a temporal error occurs in the registration of different wavelengths. It is due to the fact, that because of the diffraction grating dispersion the different wavelengths pass different distances to the streak camera photocathode. This error becomes significant when the sweep is less than 1 ns. We performed temporal calibration of the instrument to find the time difference for different wavelengths. Highly directional supercontinuum (SC) was used as a light source in the spectral range from 350 to 1000 nm [10]. The measurements of SC duration with the help of interferometry have shown that it is no more than 1 ps, so this radiation can be used for calibration. With such a wide SC spectrum and a rather large path of its transportation, the effect of air dispersion on the calibration accuracy is possible. However, our estimates have shown that when the radiation at $\lambda = 350$ nm ($n_{350} = 1.00028973$) and $\lambda = 1000$ nm ($n_{1000} = 1.00027505$) passes 14 m way the time difference is $\Delta t = \Delta n l/c = 0.685$ ps, where Δn is the difference between the refraction index of air n at these wavelengths, l is the path length, and c is the speed of light in vacuum. Thus, the effect of air dispersion is at the level of device temporal resolution. The measurements by streak camera showed that there is a delay of short-wave components of SC. So the delay of the radiation at $\lambda = 375$ nm from $\lambda = 600$ nm is 5 ps. The time difference found for different wavelengths was taken into account to register the temporal behavior of luminescent spectra of filament plasma.

Before the experiments, the spectral instrument was calibrated by the position of the spectral lines and their intensity using the Hg–Ar lamp and the blackbody source. To determine the limiting temporal resolution of the streak camera, 50 fs SH radiation was directed on the spectrometer input slit in the absence of condensers. The maximum resolution in this case for a chronograph slit of 5 μm and averaged by 300 pulses was 2 ps.

3. Experimental results

The shape and luminescence of the filament plasma essentially depended on the lenses focal length (F). With a tight focusing ($F = 3$–13 cm), the luminescence was close to the optical breakdown observed in the nanosecond pulse duration range of the radiation. At the same time, an extremely heterogeneous and conical SC was observed behind the plasma. Its luminescence changed from pulse to pulse. Longer plasma channel with variable intensity appeared in case of $F = 15$–50 cm. In this case a conical SC appeared too in the form of a bright yellow ring, whose directivity depended on the pump energy. The plasma channel length for $F = 100$–300 cm was 10–100 cm. An axial SC of a white color appeared behind the filament at a certain pump energy level.

An investigation of temporal dynamics of the plasma emission was carried out for the first two regimes, since the intensity of plasma glow in third mode was insufficient for recording. In Fig. 2(a) the time integral spectrum in the range of 200–1100 nm for $F = 3$ cm is presented.

As can be seen from the figure, only atomic and ion lines of nitrogen, hydrogen, and oxygen are observed in the spectrum. The continuous spectrum is due to the bright white plasma glow. Its maximum is in the region of 500 nm and, according to the Planck formula for absolutely black body, indicates a gas temperature ~5000 °K. The development dynamics of the atoms luminescence is shown in Fig. 2(b). The inset shows that the beginning of their luminescence is delayed relative to the beginning of the laser pulse by 87 ps. An analogous delay was also observed in [3].

The maximum intensity was achieved after ~0.2 ns from the luminescence beginning. The decay time of the luminescence intensity by 1/e level is $\tau = 20$ ns.

The intensity of the continuous spectrum, atomic and ionic lines sharply decreased with increasing of lens focal length. The lines of the second positive system (2^+) of N_2 molecular nitrogen and the first negative system (1^-) of N_2^+ ions began to manifest themselves clearly in the short-wavelength part of the spectrum.

With increasing of the focal length ($F > 15$ cm) N_2^+ and N_2 lines began to dominate over other lines and the atomic lines practically disappeared. An overview of the spectrum lines development in time in the range of 250–530 nm (a) and time-resolved lines of N_2^+ and N_2 (b) are given in Fig. 3. Under these conditions ($F = 32$ cm and $E = 13$ mJ) an extended filament of sufficient intensity was formed.