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Fluorescence and transient absorption spectra of $Ar/Kr/F_2$ mixtures excited by e-beam

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ABSTRACT

Time-resolved fluorescence of $Ar/Kr/F_2$ mixtures and transient absorption spectra were measured at Berdysh preamplifier module of GARPUN KrF laser facility pumped by 50 A/cm^2, 350 keV, 100 ns e-beam. The experiments pursued an aim to obtain additional data on KrF laser kinetics, in particular, on the formation of Kr_2F molecules and their influence on the absorption and extraction of KrF laser radiation in large-scale amplifiers. A possibility of femtosecond laser pulses amplification at broad-band Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) transition is discussed.

Keywords: fluorescence and transient absorption in $Ar/Kr/F_2$ gas mixtures, kinetics of e-beam-pumped KrF amplifier

1. INTRODUCTION

Emission spectra of excimer gas mixtures in the UV – visible range contains besides separate spectral lines broad bands of diatomic and triatomic molecules [see, i.e. 1, 2 and references cited therein]. For example, typical $Ar/Kr/F_2$ mixtures of KrF laser have besides KrF (B→X) laser transition band at 248-nm wavelength other emission bands associated with C→A transition of KrF molecule with a maximum around 275 nm and a broad band of $4^2\Gamma \rightarrow 1,2\ ^2\Gamma$ transition of Kr_2F triatomic molecule centered at 405 nm. As they were measured earlier in arbitrary units it is impossible to deduct relative intensities for different bands, which is of our peculiar interest for comparison with our quasi-stationary numerical code [3]. This will allow us more accurate accounting for some important kinetic reactions. In particular, of the formation of Kr_2F molecules, which, firstly, characterizes one of the main quenching processes of the upper KrF laser level in three-body collisions with Kr and Ar atoms [4], and, secondly, is an absorber of KrF laser radiation that might significantly affect on the extraction efficiency of laser radiation from the gain medium. Besides early experiments [1, 2], transient absorption in $Ar/Kr/F_2$ mixtures at 248-nm and several other excimer laser wavelengths was measured by using an effect of suppression of Kr_2F fluorescence [5]. Later this technique was extended by means of tunable dye lasers over the whole UV and visible spectral ranges [6]. Continues absorption spectra were also investigated by means of a xenon flashlamp light source in combination with a time-gated multichannal analyzer [7]. It should be noted that the absorption spectra were mostly obtained in conditions quite different of e-beam-pumped KrF laser and sometimes are contradictory in regard of the shape of absorption bands. That is why we started a new run of fluorescence and absorption measurements of $Ar/Kr/F_2$ gas mixtures under e-beam excitation [8].

Additional reason for the research of time-resolved spectra of spontaneous emission together with transient absorption spectra is the principle ability to use some broad emission bands of diatomic and triatomic molecules for simultaneous amplification of short laser pulses at other wavelengths in the gain medium of KrF laser [9]. That endows into a fast-ignition approach in the inertial confinement fusion (ICF) based on KrF laser drivers, which are capable to amplify both nanosecond laser pulses for compression of the thermonuclear fuel and picosecond pulses for igniting of collapsed pellet [10]. By today only laser oscillations were observed at trimer Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) transition in the afterglow of e-beam pumping at very high specific input power of several tens of MW/cm^3 and high working gas pressures up to 8.5 atm [11]. Lasing occurred within a broad band centered at ~430 nm. The gain volume and length were very small, ~ 0.04 cm^3 and 10 cm, respectively.

2. EXPERIMENTAL SETUP

Experiments were performed at "Berdysh" preamplifier module of GARPUN KrF laser facility [12]. A gas mixture $Ar/Kr/F_2$ = 0.3/8.9/91.8% at total pressure up to p = 1.8 atm was filling the laser chamber of 10*10 cm^2 cross-section and

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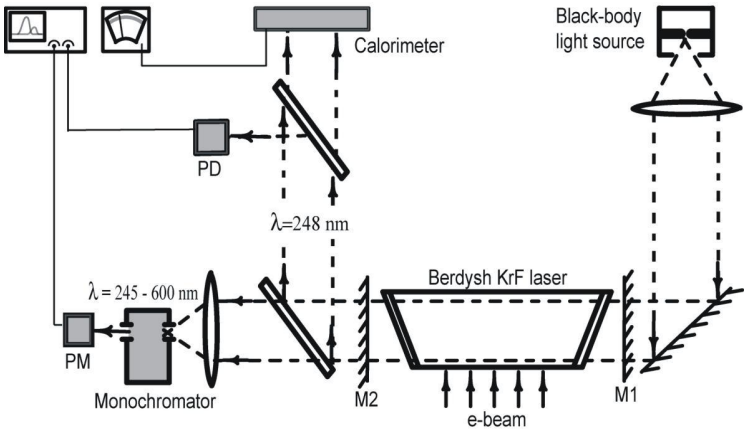
of 170-cm length. A single-side e-beam with maximum current density of 50 A/cm^2 being reached at ~80 ns delivered 900-J total energy with a pulse duration of about 100 ns at FMHM into the gain volume of 10*10*112 cm^3, thus providing a specific pumping power up to 1 MW/cm^3. Two unpumped 30-cm regions stayed at both sides of laser chamber adjacent to the windows.

The layout of fluorescence and absorption measurements is shown in Fig. 1. A pulsed capillary-discharge light source "ISI-1", which has the brightness temperature of T_b = 39±2 kK in a spectral range 220–600 nm [13], was used for absorption measurements and relative calibration of a wavelength response of the gauging equipment. The quartz lens with a 750-mm focal length transformed the image of 2-mm-diameter capillary into a parallel probe beam that passed along the laser axis and then was focused by a quartz condenser of 80-mm diameter onto the entrance slit of a grating monochromator MDR-12 with an aperture ratio 1:3. As the distance from the near boarder of e-beam-pumped region of ~ 2 m was significantly larger than the condenser focal length of 100 mm it collected the spontaneous radiation emitted by the whole 1-m pumped length. In some experiments the length of the pumped region was restricted by setting the stops for e-beam. With a grating of 1200 grooves/mm blazing at 500 nm monochromator had a dispersion of 2.4 nm/mm that corresponded to the spectral resolution of 0.2 nm for the minimal slit width ~ 0.1 mm being used. When broad emission bands were registered the slit was increased up to 0.5 mm. The second order reflection by the grating was eliminated in a visible range by means of interference filter. The wavelength reading of the monochromator was calibrated by spectral lines of a mercury lamp.



The light passing through the monochromator was measured by the 7-stage photomultiplier 14 ELU-FS (PM in Fig. 1), which has a rise time 1.2 ns, an amplification coefficient ~6*10^5 and linear current up to 2 A. A reference signal of KrF (B→X) fluorescence was monitored by a vacuum photodiode (PD). Both signals of PM and PD were registered by 200-MHz, 2 Gs/s Tektronix TDS2024 digital oscilloscope.

Fig.1. Layout of fluorescence and absorption measurements.

3. FLUORESCENCE SPECTRUM

Fig. 2 presents fluorescence signals at various wavelengths 404, 275, and 248 nm, which corresponds to $Kr_2F(4^2\Gamma\rightarrow1,2\ ^2\Gamma)$, KrF (C→A), and KrF (B→X) transitions and were measured for various lengths of e-beam-pumped region 7.5 and 100 nm. Different intensity-time histories are evident from these oscillograms. A significant shortening of the pulse-width of KrF (B→X) transition for a long pumped length is obviously due to amplification of spontaneous radiation, which is rather higher at the maximum of pumping power.

Using ISI-1 light source as the etalon of brightness with the spectrum described by Plank's formula we determined the wavelength dependence of the monochromator and PM response. Absorption by molecular fluorine when probe beam passed through unpumped gas mixture in the cavity was also taken into account in calibration procedure. This calibration was used afterwards to construct fluorescence spectra from amplitudes of fluorescence signals. They are shown in Fig. 3 and represents the main emission bands KrF (B→X), KrF (C→A), and Kr_2F ($4^2\Gamma\rightarrow1,2\ ^2\Gamma$).

$Kr_2F(4^2\Gamma\rightarrow1,2\ ^2\Gamma)$ (1) & KrF (B→X) (2) **KrF (C→A) (1) & KrF (B→X) (2)**

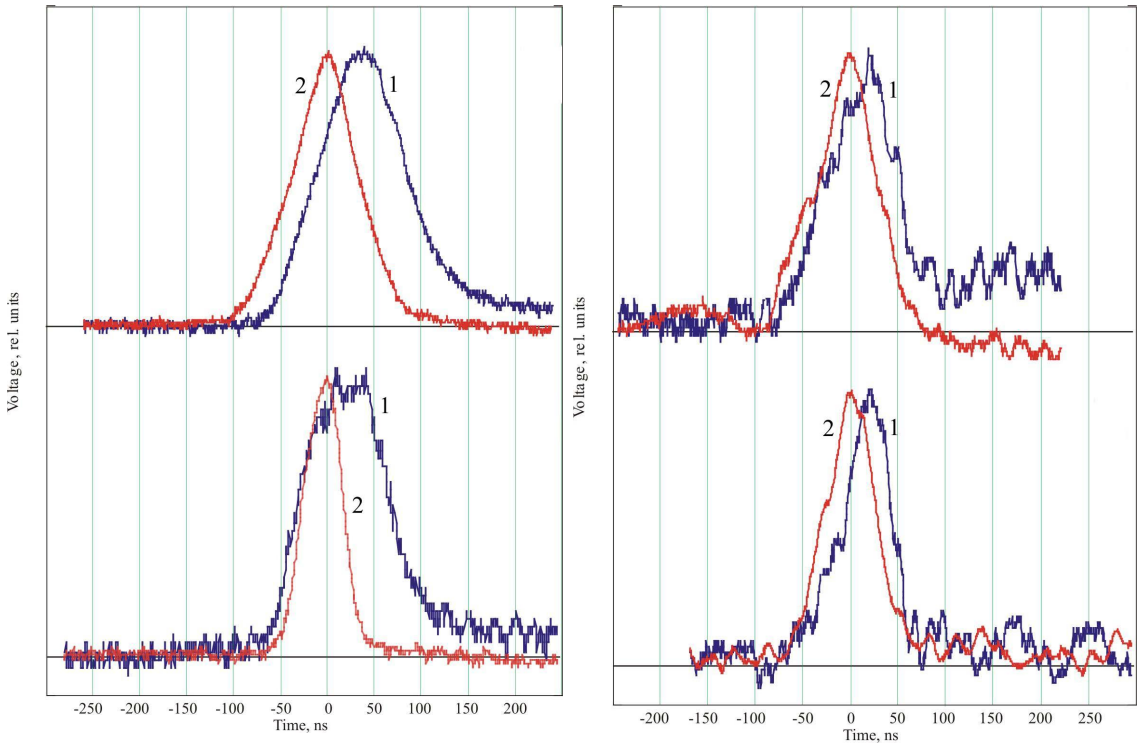


Fig.2. Fluorescence signals at λ = 404 nm (1) and 248 nm (2) (left) and λ = 275 nm (1) 248 nm (2) (right), and for various pumped lengths of L = 7.5 cm (top) and L = 110 cm (bottom).

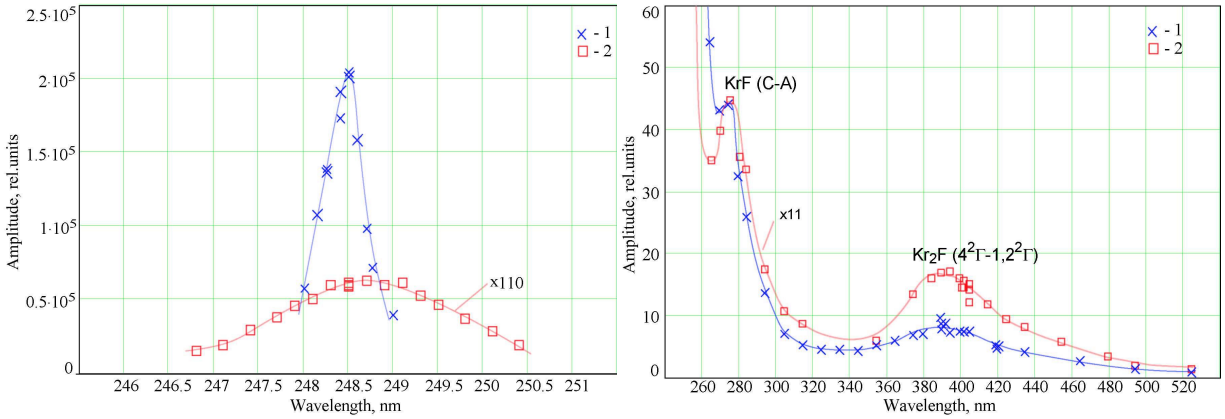


Fig.3. Fluorescence spectra of Ar/Kr/F_2 = 0.3/8.9/91.8% gas mixture at p = 1.8 atm representing KrF (B→X) (left), KrF (C→A) and Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) transitions (right) for pumped lengths of L = 110 cm (1) and L = 7.5 cm (2).

The significant growth of the maximum and narrowing of KrF (B→X) bandwidth is clearly seen for longer pumped length, and it is evidently demonstrates amplification of spontaneous emission at laser transition.

The effect of suppression of Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) fluorescence under the action of amplified spontaneous emission (ASE) at 248-nm wavelength due to preferable radiative decay of excited KrF(B) molecules, which are the donors for excited $Kr_2F(4^2\Gamma)$ molecules, was also considered. Additional plane mirrors M1 and M2 reflecting at 248-nm wavelength were set to form a plane resonator for KrF (B→X) laser (see Fig. 1). Both mirrors were transparent for visible Kr_2F radiation. Fig. 4 shows oscilloscope traces of Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) emission at λ = 404 nm acquired for resonators with different quality factor, as well as for single-pass ASE at KrF (B→X) transition (without resonator mirrors at all) or double-pass ASE (with the only rare back mirror M_1). The amplitudes of Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) fluorescence and integrals of emitted radiation over the pulse duration are shown in Fig. 5. They demonstrate approximately linear dependence on the

maximum intensity of KrF (B→X) radiation. Experimental dots designated by (1) to (5) in the graph correspond to oscillograms in Fig. 4. Intercavity intensities for (1) to (3) were determined from measurements of output energies and pulse forms with accounting for transmission of output resonator mirror. Intensities for (4) and (5) dots in the graph were obtained in direct measurements nearby output laser window.

For KrF (B→X) laser transition the saturation intensity is $I_s \sim 10^6$ W/cm^2. It means that for higher laser intensities $I > I_s$ a probability of the decay for excited KrF (B) molecules induced by radiation becomes more then all other deactivation processes including three-particle collisions, which forms Kr_2F ($4^2\Gamma$) molecules. In contrary, for $I < I_s$ radiation at 248-nm wavelength does not influence on the formation of Kr_2F ($4^2\Gamma$) molecules.

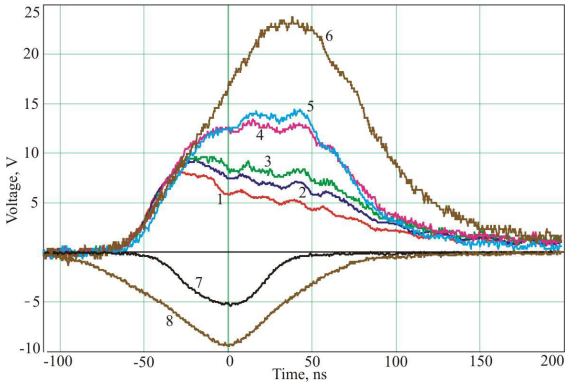


Fig.4. Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) emission at $\lambda = 404$ nm for $L = 110$ cm (1-5) and 7.5 cm (6, multiplied by 10) for different resonator performances; KrF (B→X) radiation for $L = 110$ cm (7) and 7.5 cm (8), both are out of the scale.

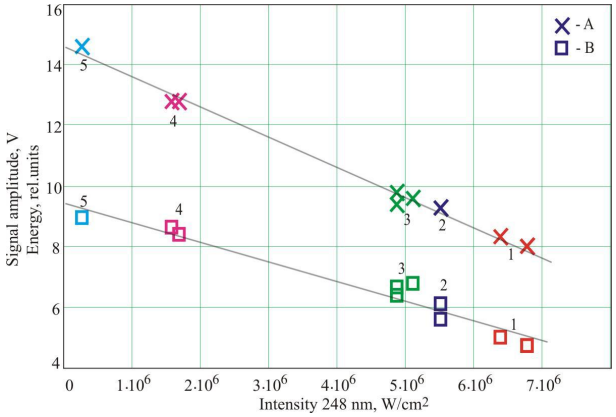


Fig.5. Amplitude (A) and energy (B) of Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) fluorescence in dependence on peak intensity of KrF (B→X) radiation. The digits near the dots correspond to the appropriate signals in Fig. 4.

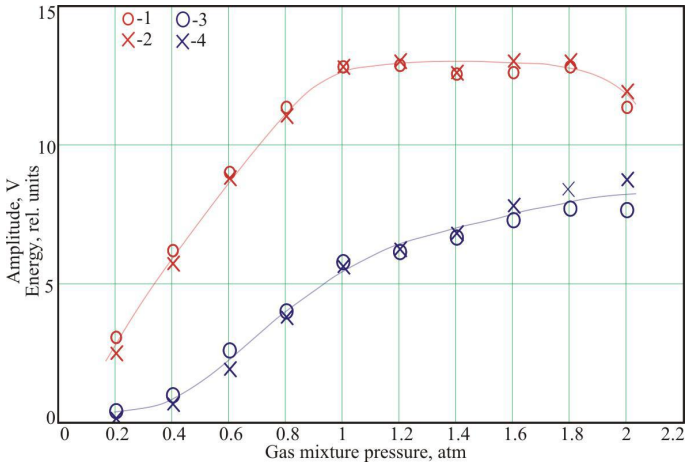


Fig.6. Amplitudes (1, 3) and energies (2, 4) of KrF (B→X) (1, 2) and Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) fluorescence (3, 4) in dependence on pressure of gas mixture.

The pressure dependence of fluorescence in 248-nm and 404-nm bands is shown in Fig. 6. KrF (B→X) radiation saturates at $p > 1$ atm, while Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) radiation continues to grow. When analyzing such behavior one should take into account that e-beam deposition falls down at low pressures while electrons range in a gas becomes comparable with a transverse size of the cavity.

4. TRANSIENT ABSORPTION SPECTRUM

Oscillogram in Fig. 7 (bottom trace) demonstrates a typical pulse of the ISI-1 light source measured at the wavelength around 430 nm. It has about 10-μs plateau with a constant brightness. The fluorescence of a gas mixture under e-bean excitation at the same wavelength is fixed at the top of the plateau. The reference pulse of KrF (B→X) fluorescence at 248 nm is shown at the upper trace. In this time scale the both look like narrow peaks. With the enlarge time resolution the signals of fluorescence are shown in Fig. 8. In comparison with the lower signal (1) when the only fluorescence at 430-nm wavelength was registered the upper signal (2) is superimposed with the probe signal of the ISI-1. By deducting from the second signal the first one we obtain the difference signal (3), which reveals the reduction in amplitude due to

transient absorption. By normalizing it to the initial value (before gas excitation) we have absorption A_λ (4) vs. time that enables us to derive an absorption coefficient $\alpha_\lambda = -\frac{1}{L}\ln A_\lambda(t)$ where L = 1.1 m is the length of e-beam-pumped region. For example, in the minimum of the curve (4) $A_\lambda = 0.75$ and $\alpha_\lambda = 0.26$ m^{-1}.

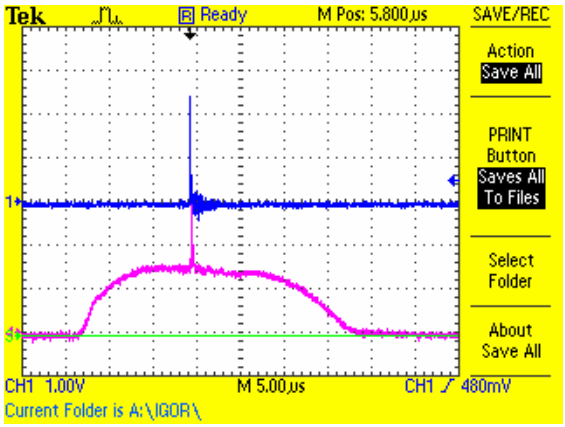


Fig.7. Pulse of the light source ISI-1 at 430-nm wavelength with a peak of gas fluorescence fixed at the plateau (bottom trace) and reference KrF ((B→X) pulse at 248 nm (upper trace).

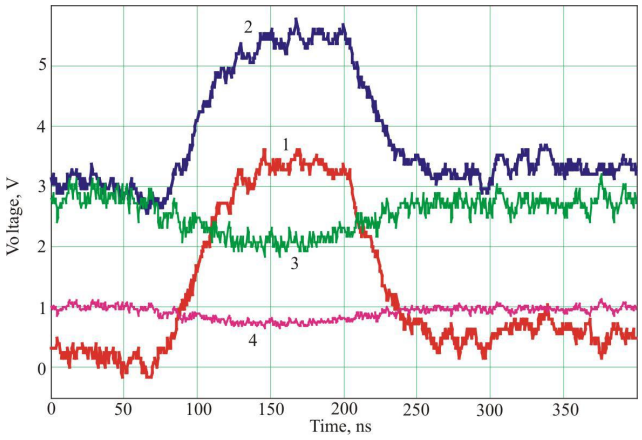


Fig.8. Oscilloscope signals of gas fluorescence at 430-nm wavelength (1), its superimposition with the probe ISI-1 radiation (2), the difference signal (3), and normalized transient absorption (4).

The absorption spectrum for the maximal absorption in the pulse was constructed from described measurements by today in a restricted wavelength range of 320−460 nm and it is presented in Fig. 9. It is extended in shorter wavelengths UV region by the spectrum taken from the paper [6]. A group of absorption lines associated with bond-bond transitions of transient species is distinguished in the range of 420-460 nm. Better spectral resolution will be implemented in further experiments to identify these lines. A broad continues absorption band, which begins from 420 nm and lasts to UV, originates due to transition of Kr_2F ($9^2\Gamma \leftarrow 4^2\Gamma$) molecule resulted in a photodissociation of Kr_2F molecule.

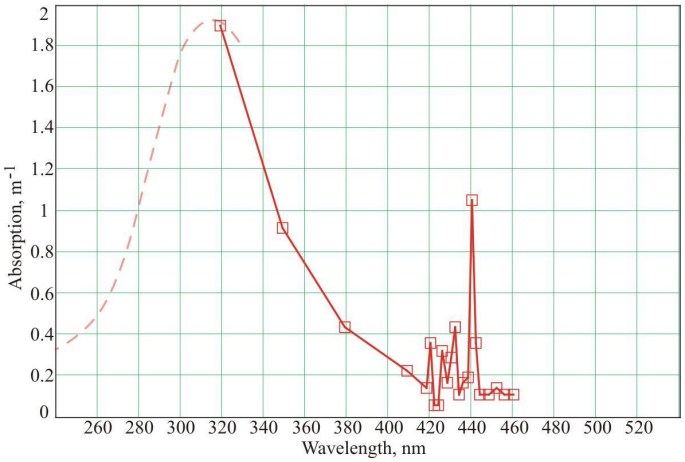


Fig.9. Absorption spectra of Ar/Kr/F_2 = 0.3/8.9/91.8% gas mixture at p = 1.8 atm measured at the maximum of absorption pulse. Dashed line is a part of the spectra from paper [6].

5. CONCLUSIONS

The experimental data were obtained on time-resolved spontaneous emission in various spectral bands of KrF (B→X), KrF (C→A), and Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) transitions of Ar/Kr/$F_2$ gas mixtures under moderate-rate e-beam pumping (~ 1 MW/cm^3), the temporal radiation dynamics and suppression of Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) fluorescence by stimulated KrF (B→X) radiation. This will allow us to verify the numerical code and to account for more accurately in simulations, which are going on, important kinetic processes and radiation transfer in the gain medium of large-scale KrF laser amplifiers [14]. The comparison of fluorescence and transient absorption spectra demonstrates that there is a wide enough spectral range of about 400-440 nm where amplification of femtosecond laser pulses at broad-band Kr_2F ($4^2\Gamma \rightarrow 1,2\ ^2\Gamma$) transition is expected to be higher than absorption. Such experiments will be further performed aiming to verify the fast-ignition ICF concept [9].

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