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Fluorescence Measurements in a Low-Pressure Kr/F_2 Medium

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Abstract. The time-resolved measurement of the sidelight fluorescence of KrF* formed by a short electron-beam pumping pulse (20 ns FWHM, 19 kA, 860 kV) was performed as a function of both the total pressure ranging from 100 Torr to 400 Torr and the partial F_2 concentration (0.1–1.0%). We have estimated the fluorescence yields (FY) for various laser-gas conditions experimentally and the results were compared with numerical predictions. This study indicates a superior amplification performance for a single ultrashort pulse in a low-pressure medium because of the higher FY for KrF*.

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Recently, there is great interest in an electron-beam (e-beam) pumped very low-pressure (<200 Torr) KrF large-aperture amplifier for ultrashort pulse (100 fs) amplification to realize a high peak-power system scalable up to the petawatt (PW) level in the ultraviolet spectral region [1,2]. The major requirement is to achieve a uniform single beam of 100 J energy with minimum wavefront distortion and very low amplified spontaneous-emission (ASE) background for the highest focussed intensity. Several considerations on amplifier physics and technology indicated a new operational mode of the e-beam-pumped Kr/F_2 medium as an alternative for this high-brightness laser amplifier. Although the high gain of the laser medium is not appropriate for subpicosecond pulse amplification for high peak-power extraction with the highest contrast ratio, the proposed lower pressure and lower excitation rate operation enable the use of thin optical windows and simultaneously decrease the ASE and the transient refractive-index change (TRIC) effects. The shorter gain window obtained by pumping with a short e-beam pulse of approximately 10 ns half-width implies higher system efficiency and a better energy contrast ratio on the target. Although there have been some investigations performed for an e-beam pumped low-pressure Kr/F_2 medium both experimentally and numerically [3–5], the characteristics of a low-pressure medium pumped by a short e-beam pulse are not yet fully understood. Moreover, low-pressure operation below 200 Torr is also essential in the recently developed microwave-excited KrF excimer lamp [6].

In this paper we report on fluorescence measurements in a short pulse e-beam pumped Kr/F_2 amplifier medium for a pressure region ranging from 100 to 400 Torr with various F_2 concentrations. The experimental results were compared with the numerical predictions using the kinetic model presented in a previous calculation [4].

1 Experiments

The experimental setup is shown in Fig. 1. The e-beam generator (Pulserad 110 A) produced 20 ns FWHM, 19 kA, 860 kV pulses. The electron beam emitted from a 5 cm di-

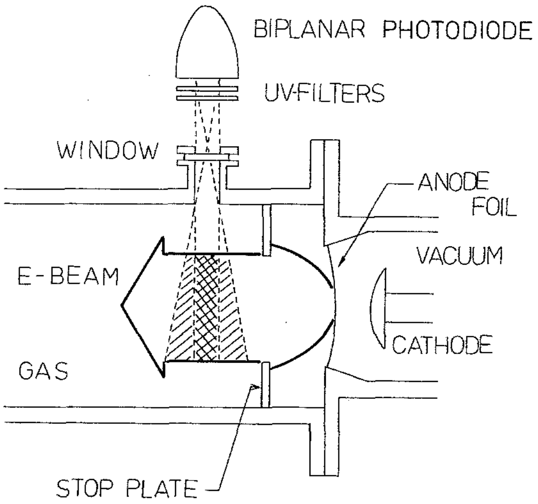

Fig. 1. Experimental setup of the measurement of the fluorescence from the electron-beam pumped low-pressure KrF medium

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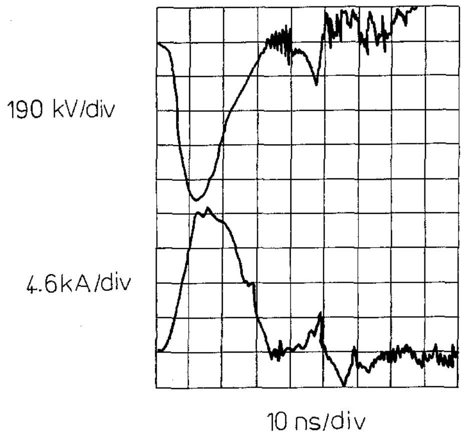

Fig. 2. Waveforms of the diode voltage (*upper*) and current (*lower*) at 2.5 cm anode–cathode gap

ameter carbon cathode was injected into the laser chamber through a 50 μm thick titanium anode foil. The diode impedance was typically 50 Ω with 2.5 cm A–K gap. The vacuum level was kept at less than 10^{-5} Torr. In order to fix the measurement volume for the estimation of the fluorescence yield (FY) at the position of viewport (8 mm diameter), we located an aluminum stop-plate with an aperture of 6 cm diameter in front of the anode foil as shown in Fig. 1. The e-beam current density measured just after the stop-plate by a Faraday cup (type FC-1, STI Optronics) was $\sim$70 A/cm^2 in the Kr gas filling conditions (100, 200, and 400 Torr) and we also confirmed the e-beam's spatial uniformity fairly well at the position of the viewpoint. We measured the temporal profiles of the sidelight fluorescence of the low pressure Kr/F$_2$ amplifier medium pumped by the e-beam for total pressures of 100, 200, and 400 Torr with a constant F$_2$ concentration of 0.5% and for the F$_2$ concentration of 0.2%, 0.5%, and 1.0% at a constant total pressure of 200 Torr. The pumped gas was replenished after each shot to avoid any contamination side-effects. In order to verify the low collisional quenching loss in the very low-pressure regime, we also estimated the FY of KrF* under the same conditions as mentioned earlier. The FY is defined as the ratio of spontaneously emitted photon density to both the rare-gas ion and metastable-atom density produced by the e-beam under non-lasing condition [7]. The spontaneously emitted KrF* photon density was estimated from the sidelight waveform detected by a biplanar phototube (type R1193U-02, Hamamatsu Photonics) located over the viewport with appropriate bandpass filters by using the values of solid angle between the biplanar phototube and the measured volume. (The measurement volume is cross-hatched in Fig. 1.) We also took into account the sensitivity of the phototube and the transmittance of both the CaF$_2$ window (4 mm thick) for the viewport and the bandpass filters for calibration.

Figure 2 shows a typical waveform of the diode voltage and the beam current when the charging voltage of the Marx bank was ±30 kV. The measurement was performed with the built-in diagnostics of the Pulserad 110 A with a storage oscilloscope (Tektronix 7834).

2 Results and Discussion

Figure 3 shows the temporal sidelight profiles of KrF* fluorescence in the low-pressure Kr/F$_2$ medium pumped by the 20 ns e-beam pulse with a current density of 70 A/cm^2 at the measuring position. The total pressure was kept at 100, 200, and 400 Torr for each (a), (b), and (c) measurement with the fixed gas-mixture ratio [Kr/F$_2$ = 99.5/0.5 (%)]. The peak intensity of (b) was 5 times larger than that of (a), that of (c) was 9 times larger. From these results we notice that the lower pressure medium of 100 Torr has a very slow risetime of over 70 ns and a longer pulse-width of 150 ns in contrast with the short risetime below 30 ns for the 400 Torr condition. At the lower pressure condition below 100 Torr, as it was expected in the previous investigation [4], the slow KrF* formation rate at the leading part of the sidelight profile can give a significant delay of the peak gain with respect to the pumping pulse peak. Recently, Datsyuk also predicted the steep decrease in the formation efficiency of the low vibrational levels at very low-pressure condition (<75 Torr) mainly due to the slowness of vibrational relaxation [5]. In spite of the termination of the pumping pulse, the temporal broadening and long tail of the gain profile at the low-pressure condition characteristic in Fig. 3a can be explained by the fact that the formation reactions were continuing after the e-beam pulse termination due to the long lifetime of

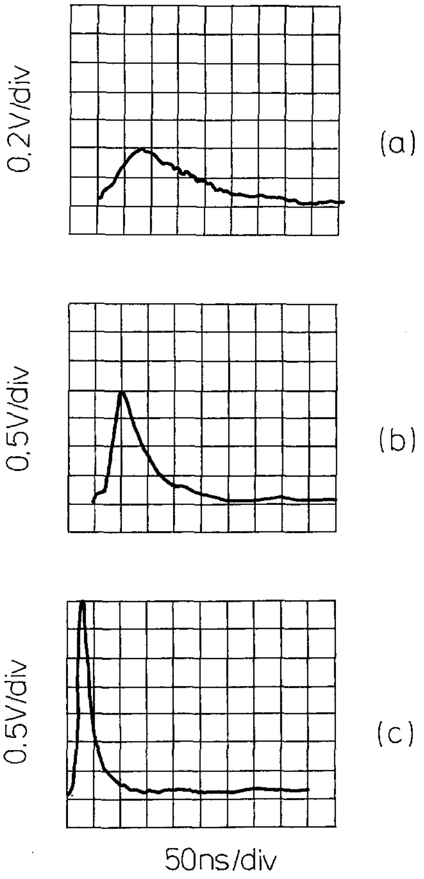

Fig. 3a–c. Temporal sidelight profiles of the KrF* fluorescence for **a** 100 Torr, **b** 200 Torr, and **c** 400 Torr total pressures. The F$_2$ concentration and current density are kept constant at 0.5% and 70 A/cm^2, respectively

some species and such as Kr^* and Kr^+, and there was no significant collisional quenching loss at the tail part of the gain profile. The lifetime of KrF^* at the tail is expected to be close to its effective lifetime of 12 ns due to the absence of main quenchers (electron, Kr, F_2 concentrations are relatively low at the tail). We notice the pulse narrowing and decrease of decay time with the increase of the total pressure.

Figure 4 shows the measured FY for KrF^* (closed circles) and calculated FY for KrF^* and Kr_2F^* (solid lines) as a function of the total pressure with constant gas ratio at an e-beam current density of 70 A/cm^2. The results of the numerical calculations fit well with the experimental results. The gradual decrease of the KrF^* FY from 0.63 at 100 Torr to 0.47 at 400 Torr and the steep increase of the Kr_2F^* FY with increasing total pressure is attributed to the three-body collisional quenching by 2 Kr. These values of FY are larger than those of the atmospheric-pressure Kr/F_2 medium a high excitation rate conditions [7]. For the low-pressure conditions below 100 Torr, the slow risetime and long temporal gain broadening are unfavorable for the energy extraction efficiency in short pulse amplification and results in larger ASE background energy ratio. Since the three-body collision is the significant quenching process at higher pressure above 400 Torr Kr/F_2 mixture, the increase of absorption is inevitably due to the steep increase of the number density of Kr_2F^* molecules as a nonsaturable absorber in the short-pulse amplification in spite of the fast risetime and higher gain due to the increase of the pumping rate in the experimental conditon. On the basis of these measurements and the numerical analysis, the total pressure range of ~200 Torr seems to be the optimal condition for the short-pulse large-aperture amplifier from these considerations on FY pulse width and absorbers.

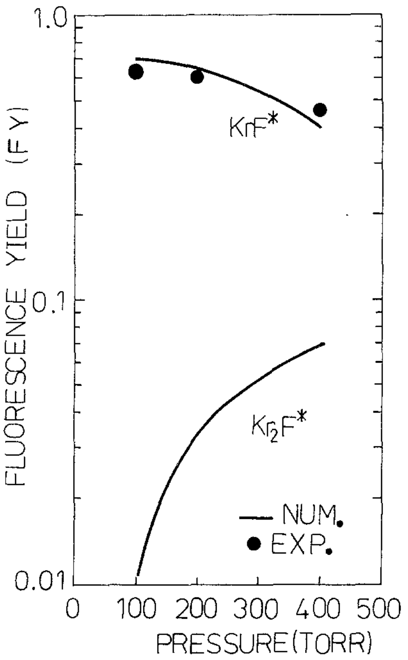

Fig. 4. Measured fluorescence yields (*closed circles*) for KrF^* and calculated fluorescence yields (*solid lines*) for KrF^* and Kr_2F^* as a function of the total pressure. The F_2 concentration and current density are kept constant at 0.5% and 70 A/cm^2, respectively

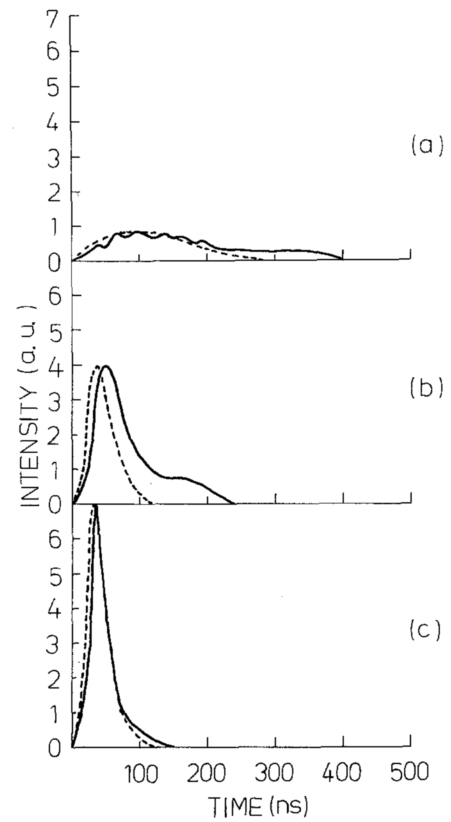

Fig. 5a–c. Temporal KrF^* fluorescence signals obtained by numerical (*broken lines*) and experimental (*solid lines*) results each for **a** 100 Torr, **b** 200 Torr, and **c** 400 Torr total-pressure conditions. The F_2 concentration and current density are kept constant at 0.5% and 70 A/cm^2, respectively

Figure 5 shows the comparison of the numerical predictions (broken lines) and the measured KrF^* fluorescence signals (solid lines) for the various total-pressure conditions. The total pressure for (a), (b), and (c) corresponds to the ones in Fig. 3. The peak values of the model calculations were normalized to the experimental results in each case. Although the pumping pulse used in the numerical simulation had a much faster risetime than in the experiment, the predicted pulse shapes well match the real KrF^* fluorescence pulse shapes.

Figure 6 shows the temporal sidelight profiles of KrF^* at various F_2 concentrations at a total pressure of 200 Torr. The F_2 concentration was kept at 0.2, 0.5, and 1.0% for each of the (a), (b), and (c) measurements. The current density of 70 A/cm^2 in the 200 Torr medium gave a 200 kW/cm^3 excitation rate. The peak intensity of (b) was 2.4 times larger than that of (a), while (c) was 6 times larger. Along with the increase of the F_2 concentration, the KrF^* fluorescence signal showed faster risetime and pulse-width narrowing. This was expected since the higher F_2 concentrations gave faster formation rates. For the case of 0.2% F_2 concentration, the risetime of the gain signal was slower than 50 ns due to the decrease of the formation rate together with the large electron quenching loss. The lower F_2 concentration results in large residual electron density due to the less effective attachment effect.

Figure 7 shows the measured FY for KrF^* and the calculated FY for KrF^* and Kr_2F^* as a function of F_2

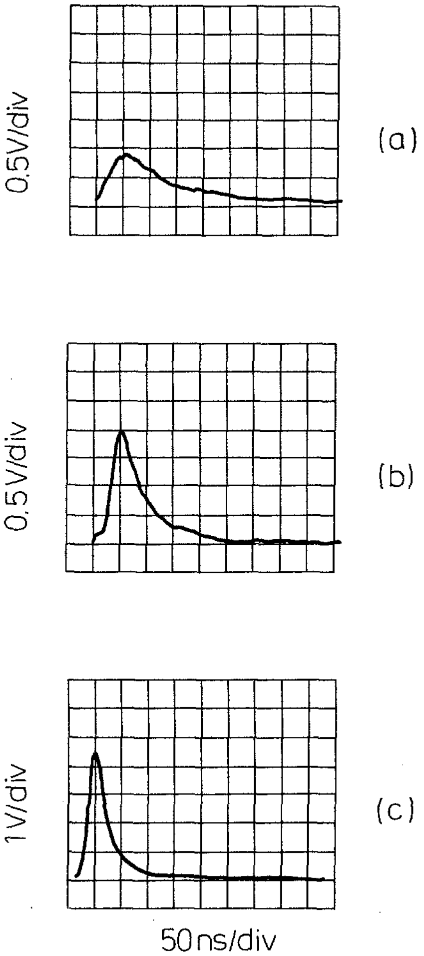

Fig. 6a–c. Temporal sidelight profiles of KrF* fluorescence for **a** 0.2%, **b** 0.5%, and **c** 1.0% F_2 concentration at 200 Torr total pressure and 70 A/cm^2 current density

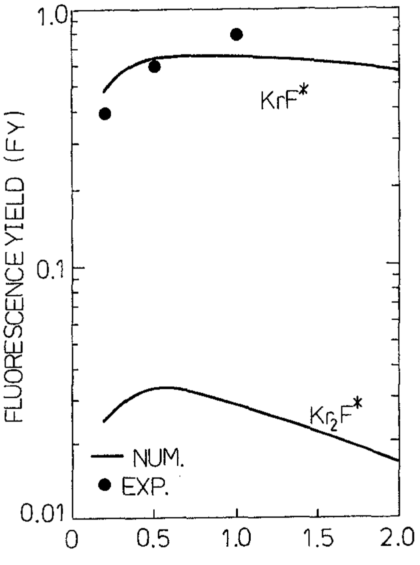

Fig. 7. Measured fluorescence yields (*closed circles*) for KrF* and calculated fluorescence yields (*solid lines*) for KrF* and Kr_2F* as a function of the F_2 concentration at 200 Torr total pressure and 70 A/cm^2 electron-beam current density

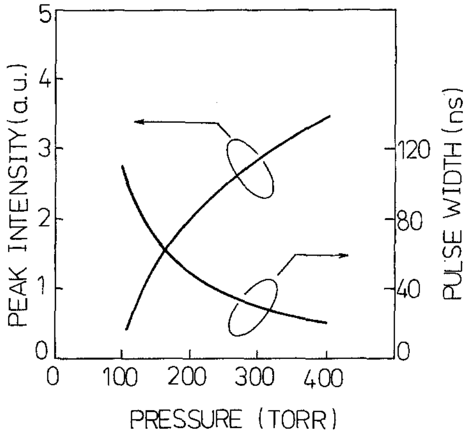

Fig. 8. Numerical results of the fluorescence peak intensity and pulse width as a function of the total pressure. The F_2 concentration and current density are kept constant at 0.5% and 70 A/cm^2, respectively

concentration at a constant total pressure of 200 Torr. From the calculated curve for Kr_2F*, it is indicated that the effect of three-body collisional quenching by the 2 Kr is decreased with increasing F_2 concentration in excess of 0.5%. In spite of the decrease of quenching by 2 Kr, the calculated FYs of KrF* have almost constant values with the F_2 concentration over 0.5%. According to our numerical analysis, the main quenching species are electrons and 2 Kr at 0.5% F_2 condition, but the main quencher at 1.0% F_2 condition is F_2. For the larger F_2 concentration (1.0% F_2), the measured FY value of 0.8 for KrF* is larger than that of the model prediction. It is possible that the rate constants used in the calculation are only valid for normal mixtures (normally <0.5%) of F_2, and hence the discrepancy at larger concentrations. For example, the rate constants for dissociative attachment to F_2 are still uncertain [8].

For a short-pulse KrF amplifier application, the Kr/F_2 medium with a larger F_2 concentration may lead to unfavorable results such as larger absorption and TRIC in the amplifier. The F_2 concentration around 0.5% seems to be the best for the dedicated application.

Finally, Fig. 8 summarizes the dependence of the fluorescence peak intensity and pulse width from the experimentally confirmed numerical results. The F_2 concentration is 0.5% and the current density is 70 A/cm^2.

3 Conclusion

We have measured the time-resolved sidelight fluorescence of KrF* in a very low-pressure Kr/F_2 medium pumped by a 20 ns e-beam pulse to study the validity of our numerical code and characterize the low-pressure gain behavior. Although the higher total pressure and higher F_2 concentration lead to a faster risetime, a shorter gain window, and a larger peak gain, the total pressure of 200 Torr and a F_2 concentration of 0.5% seem to be the best choice for the short-pulse large-aperture amplifier considering the technical points of view such as the use of a thin optical window, ASE and TRIC effects, extraction efficiency, and better gain/absorption ratio. From the estimation of the FY, we confirmed that Kr/F_2 media with pressures below 200 Torr reduce the three-body collisional quenching process and absorption significantly, which in turn gives a large value of the gain/absorption ratio, high system efficiency, and lower distortion in a petawatt-level amplifier.

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