A Study of the Electron Quenching of Excimers in a KrF* Laser Excited by a Coaxial Electron Beam

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Abstract. Measurements of the output energy, the optical pulse length and the build-up time of the laser pulse, obtained with a coaxially e-beam pumped KrF* laser, were performed varying the total gas fill pressure, the F₂ content and the e-beam current from 1–5 bar, 0.1–0.8% and 13.3–26.6 kA, respectively. The maximum specific extraction energy amounts to 64 J/l. The large range of measurements, especially at low F₂ concentrations, reveals the necessity to extend the kinetics of the F₂ chain in the usual computer model. With the introduction of electron quenching of KrF* and ArF* by dissociative attachment the predictions are also for low F₂ concentration in agreement with experiments.

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Because of their scalability and high efficiency, the rare-gas halogenide e-beam pumped excimer lasers, and in particular the KrF* laser, have been recognized as potential drivers for inertial confinement fusion experiments. As these lasers exhibit high gains and short excited lifetimes, they cannot be operated in an energy storage mode. As a consequence, a lot of large-scale experiments were conducted using short and high energy pumping pulses in order to achieve the high output powers required for this purpose.

Another approach would consist of developing systems with a long pump pulse at a moderate power. The resulting long laser pulse can subsequently be compressed by pulse shortening techniques.

The optical and technical problems associated with the long pulse approach are easier to solve as limitations, inferred by e-beam pinching, material fatigue and high voltage circuitry are less restrictive.

The present contribution deals with an experimental investigation of a KrF* laser, excited by a coaxial electron beam having a pulse duration of about 250 ns. The pumping current density in our laser is at

the anode in the order of 200 A/cm². This current regime is very interesting as it is the intermediate regime between the high-current e-beam pumped type of lasers with pulse lengths shorter than 100 ns and the low-current e-beam sustained discharge systems which usually have pulse lengths of 500 ns or longer. It is an extension of our earlier work done with this system. Notwithstanding the fact that we now achieved a maximum output energy of 64 J/l the aim of this paper is to report on an important conclusion we have to draw from a comparison of our experimental results and predictions obtained from a computer simulation program.

It appeared that the electron quenching of KrF*, ArF*, Kr₂F*, and Ar₂F* is not a super elastic collision process yielding F + e_s but a dissociative attachment process resulting in F⁻ ions.

Modelling of the e-beam pumped KrF* laser has been done extensively in the past decade. Cohn and Lacina [1] and Johnson and Hunter [2] described a model that is tailored to e-beam sustained and self sustained discharge KrF* lasers. In the model of

Johnson some important reactions as the quenching of KrF^* and ArF^* by fluorine and secondary electrons are not included.

Kannari et al. [3, 4] have added these quenching reactions in their model and changed the recombination rate constants according to Wadehra and Bardley [5]. The first of their two articles describes the short pulse (< 100 ns) and high excitation regime while the second one applies to longer excitation pulses.

Recently Mandl et al. [6] carried out experiments in the intermediate region and compared these values with the predictions of a computer code developed in his group and described by Klimek et al. [7]. The agreement of the predictions and the measured output values is fairly good. Klimek reported on a model in the short pulse regime. This model provides predictions of gain and absorption within an accuracy of 10–15% over a broad range of pressure, F_2 density and e-beam flux.

In this paper we used an extended version of a model [8] which was based on a set of 73 reactions involving 22 different species.

Our measurement of the output energy, the time behaviour and the build-up time of the laser pulse as a function of the total gas pressure, fluorine concentration and pumping current density are the basis of the model corrections to be presented in this paper.

1. Experimental Configuration

The use of a coaxial e-beam has advantages over a transversal pumping geometry with respect to the homogeneity of the deposited energy in the gas and the efficiency of the energy deposition [9, 10]. Technological problems however cause this coaxial technique not to be a common choice to excite excimer lasers.

Our laser apparatus has been described previously [11]. It comprises a coaxial vacuum diode directly driven by a compact home made Marx generator consisting of 5 stages each with a 80 nF, 100 kV capacitor. The diode is formed with a thin walled metal tube as anode and a cathode made out of four 50 cm long graphite edge electrodes mounted on the inside wall of a metal cylinder with 128 mm internal diameter.

The laser welded anode tubes having a diameter of 22 mm were formed with 25 μm thick Ti foil. Ti was chosen because of its high tensile strength and its low stopping power for fast electrons. In order to mount the anode in the diode assembly, flanges with a 20 mm bore made out of titanium were glued or laser welded on the ends of the fragile foil tube. The active volume of 157 cm^3 has a diameter of 20 mm, limited by the end flanges, and a length of about 50 cm. To avoid pinching on the 22 mm diameter anode a lateral current drain had to be connected to the anode foil when experi-

ments were run at a diode current above 18 kA. In this case anode tubes glued to the end flanges could be used, electrical contact between the Ti foil and end flange being unimportant. With respect to their mechanical strength the laser welded tubes are superior to the electrically welded ones used in our former experiments [11].

The laser gas volume which had a length of 80 cm was ended by two MgF_2 flats. One of these flats serves as output coupler with a reflectivity of about 11%. At the other end a 100% dielectric reflector was mounted 2 cm away from the MgF_2 window outside the gas volume. The output energy was detected by a Gen Tec ED 500 pyroelectric meter. The temporal behaviour of the laser pulse was measured with an EG&G 444 BQ photodiode. In order to measure the delay between the input current and laser pulse the R 7912 AD Tektronix digitiser was triggered by a signal derived from the Marx generator voltage.

All experiments were performed at Marx load voltages between 60 and 100 kV increased in steps of 10 kV. The total pressure of the laser gas mixtures was varied from 1 bar to 5 bar for each of the mentioned load voltages. The partial F_2 pressure ranged from 0.1% to 0.8%. The Kr content was kept fixed at 9.2%. Before each measurement a fresh gas mixture, premixed in a reservoir, was filled into the laser chamber.

At the maximum diode current of 26.6 kA, i.e. with the Marx generator charged up to 100 kV, the probability for an anode tube damage was large for fill pressures above 4 bar. Working below these critical boundary conditions the anode tubes with a wall thickness of 25 μm can withstand some hundreds of shots.

After each shot, particularly when high energy loadings are involved, a brownish red dust was to be seen in the cavity. The output decreased significantly when a second shot was done before this dust has been removed. We did not examine the origin of this dust but we suppose that it arises from some Ti_xF_y compounds which are formed on the Ti foil and vaporise under the e-beam irradiation. In order to save the optic windows from contamination we evacuated the laser cavity immediately after each shot.

2. Experimental Results

The temporal behaviour of e-beam voltage, current and laser output are shown in Fig. 1a, b, and d, respectively, for a $\text{Ar/Kr/F}_2 = 90.2/9.2/0.6$ gas mixture at 2 bar. The shown diode current (peak) of 26.6 kA and the voltage of 265 kV (averaged) were obtained with the Marx generator initially charged to 100 kV. The shape of the current- and voltage pulse did not change significantly for different initial Marx gen-

Fig. 1a–d. Oscilloscope traces of (a) the diode voltage, (b) the diode current, and (d) the output pulse for a Marx load voltage of 100 kV/stage. In (c) the current pulse as used in the model is plotted

erator load voltages. Therefore, the current pulse could be described in the computer simulation, described later on, with the pulse shape shown in Fig. 1c.

Table 1 summarises the measurements of the total laser energy and the pulse duration as a function of gas fill pressure, fluorine content and initial Marx load voltage. All the values given in Table 1 were obtained with 25 μm anode tube.

The laser pulse duration increases if the fluorine content and the excitation power are increased simultaneously, but varies only slightly with total fill pressure. The laser pulse terminates in all cases with the current pulse or earlier. The build-up time varies considerably for different gas mixtures and excitation rates. The delay between the onset of the current and laser pulse decreases from 70 to 45 ns as both pump power and gas pressure are increased. Very long build-up times, up to 130 ns, were observed in the case of fluorine rich mixtures pumped at the lowest excitation rate investigated.

Fig. 2. Output energy and specific output energy as a function of the total gas pressure for different Marx load voltages. The dashed and dotted curves are from earlier papers (see text)

In our experiment the maximum laser energy is obtained with gas mixtures containing 0.4% or more fluorine at a pressure of 5 bar excited with the near highest excitation power provided by our e-beam device.

The output energy measurements obtained with the gas mixture ($\text{Ar/Kr/F}_2 = 90.4/9.2/0.4$) are visualized in Fig. 2 as a function of fill pressure and Marx load voltage. For quick reference the figure also shows earlier results obtained with a diode comprising an anode of 44 mm [11], (dashed curve) and 22 mm (dotted curve) diameter [12] made out of 50 μm thick Ti foil. These measurements were performed with the same gas mixture and an initial Marx generator charge of 100 kV. The dashed curve describing the experiments with the 44 mm diameter anode refers to the left vertical axis indicating the specific output energy. The total energy is four times higher than the value according to the right vertical axis, due to the four times larger active volume. The current density on the anode circumference is roughly doubled for the 22 mm diameter tube compared to the 44 mm anode tube. Accordingly the specific output energy is increased, too. We obtain for the laser tube of 22 mm diameter and wall thickness of 25 μm a maximum energy extraction of 64 J/l.

Comparing the results obtained with the two different 22 mm diameter anodes (Fig. 2) and keeping in mind that the initially stored energy increases with the square of the Marx load voltage, it can be seen that about twice as much input energy is needed to obtain the same output in the presence of the 50 μm compared to the 25 μm thick anode foil. A rough estimate shows that the energy loss of electrons, which is about 90 and 44 keV by traversing a Ti foil 50 and 25 μm thick, respectively, cannot explain the rather big difference of

Table 1. Summary of the output energy and pulse width measurements (FWHM) for various input parameters

% F ₂ →		0.1		0.25		0.40		0.6		0.8	
Marx Load Voltage [kV]	Total Pressure [bar]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]
60	1	0.5	100	1.6	135	1.0	138	1.3	140	1.0	150
	2	1.4	110	2.9	160	2.2	160	2.2	155	1.5	130
	3	1.8	115	2.9	165	2.7	160	2.1	130	1.2	110
	4	1.9	115	2.7	160	1.3	155	0.9	105	0.3	75
	5	1.4	115	2.0	150	1.0	95	0.1	50	0.0	
70	1	0.5	75	2.0	135	2.2	170	2.2	185	1.9	150
	2	1.7	80	4.4	140	3.7	160	4.0	180	3.2	160
	3	2.4	80	5.6	165	4.9	160	5.0	170	4.0	145
	4	2.8	80	6.0	155	5.3	165	4.3	150	3.4	135
	5	2.7	85	5.5	155	5.2	170	3.4	135	2.4	115
80	1	0.5	60	1.9	110	2.6	155	3.2	190	2.9	175
	2	1.7	70	4.3	115	5.5	170	6.0	185	5.0	170
	3	2.3	65	5.9	115	7.1	170	7.3	180	5.9	165
	4	2.7	70	7.2	110	8.3	170	7.6	165	5.3	145
	5	3.0	70	7.9	125	8.2	165	6.5	160	4.5	135
90	1	0.4	50	1.7	85	2.9	145	3.7	175	3.4	175
	2	1.3	45	4.2	95	6.3	155	6.9	175	6.5	165
	3	2.0	55	6.0	85	8.3	170	8.6	165	6.8	160
	4	2.7	60			9.7	165			8.3	150
	5					10.1	160				
100	1	0.4	40	1.7	75	2.6	100	3.8	175	3.2	180
	2	1.4	40	4.0	80	6.0	135	7.2	170	7.2	150
	3					8.3	150				
	4	2.7	50			9.7	155			10.0	150
	5					10.0	140				

input energy. Therefore it has to be concluded that a larger fraction of the e-beam current can reach the laser gas through the thinner anode foil, either due to a smaller "absorption" of electrons in the foil or due to the fact that electrons can traverse the anode and the laser gas more than once. An electron with an initial energy of 250 keV will carry enough energy after having lost averaged 90 keV on two passages through a 25 μm thick foil to penetrate the same foil at least one more time. This loss amounts to 180 keV in the presence of a 50 μm thick foil, leaving the electron with an energy no longer sufficient to travel back into the active volume. Thus, the number of electrons which are effective in the laser gas will depend critically on initial kinetic energy in combination with foil thickness.

Figure 3 shows the laser output energy and the laser pulse duration as a function of F₂ admixture at a Marx load voltage of 80 kV. For low F₂ contents the

laser output stops earlier than the e-beam current indicating that F₂ burn up is the main limitation. Laser pulse duration and laser energy increase with increasing F₂ content, each reaching a maximum at 6 and 20 mbar fluorine partial pressure for a fill pressure of 1 and 5 bar, respectively. This picture is typical as the maximum output energy is always attained with the longest output pulse. On the contrary, the maximum averaged output power is obtained in all cases for gas mixtures with 0.25% F₂ added to the total mixture, as can be seen from Fig. 4. This peak in the output flux is a consequence of a minimum in the absorption losses. This minimum with on one hand the increasing absorption due to the secondary electron density for low F₂ mixtures and, on the other hand, the increasing absorption losses caused by F₂ in the higher-doped mixtures does not shift with a rather strong variation of input power as can be seen from the figure.

Fig. 3. Output energy and pulse length as a function of the initial F_2 concentration at a Marx load voltage of 80 kV for total pressures of 1 and 5 bar

Fig. 4. Output power versus the partial F_2 pressure for different total pressures at a Marx load voltage of 70 kV

3. Computer Code

A list of all reactions comprised in our code is given in Table 2 together with their rate constants. The rate constants given in this table are in essence the same as that used by Mandl et al. [6] and Klimek et al. [7]. As can be seen from this table, Reactions 1 through 4 are the ion and metastable formation channels of Kr and Ar. Starting from the initial partial gas pressures and the electron density the whole kinetic chain is calculated yielding all species densities as a function of

Table 2. Listing of all reactions and their rate constants as used in the computer code. Dimensions of the rate constants: s^{-1} for decay reactions, $cm^3 s^{-1}$ for two-body reactions and $cm^6 s^{-1}$ for three-body reactions

1)	Ar + eFAST	$\rightarrow$ Ar+ + eFAST + eSLOW	1.26E-7
2)	Kr + eFAST	$\rightarrow$ Kr+ + eFAST + eSLOW	2.61E-7
3)	Ar + eFAST	$\rightarrow$ Ar* + eFAST	4.27E-8
4)	Kr + eFAST	$\rightarrow$ Kr* + eFAST	8.76E-8
5)	hnu	$\rightarrow$	3.64E-8
6)	Ar+ + Ar + Ar	$\rightarrow$ Ar2+ + Ar	2.50E-31
7)	Ar2+ + Kr	$\rightarrow$ Kr+ + Ar + Ar	7.50E-10
8)	Kr+ + F-	$\rightarrow$ KrF*	2.25E-6
9)	F2 + eSLOW	$\rightarrow$ F + F-	5.00E-9
10)	KrF* + Ar + Ar	$\rightarrow$ ArKrF* + Ar	8.00E-32
11)	KrF* + Ar	$\rightarrow$ Ar + Kr + F	1.80E-12
12)	KrF* + Ar + Kr	$\rightarrow$ Kr2F* + Ar	6.50E-31
13)	KrF* + F2	$\rightarrow$ Kr + F + F2	7.80E-10
14)	KrF*	$\rightarrow$ Kr + F + hnu	1.50E-6
15)	KrF* + hnu	$\rightarrow$ Kr + F + hnu + hnu	7.20E-6
16)	Ar2+ + hnu	$\rightarrow$ Ar + Ar+	3.90E-8
17)	F- + hnu	$\rightarrow$ F + eSLOW	1.50E-7
18)	F2 + hnu	$\rightarrow$ F + F	6.00E-10
19)	KrF*	$\rightarrow$ Kr + F	1.50E-8
20)	Ar+ + F-	$\rightarrow$ ArF*	2.01E-6
21)	ArF* + Kr	$\rightarrow$ KrF* + Ar	3.00E-10
22)	Ar2+ + F-	$\rightarrow$ ArF* + Ar	2.22E-6
23)	Kr+ + Ar + Ar	$\rightarrow$ ArKr+ + Ar	1.00E-31
24)	Ar+ + Kr + Ar	$\rightarrow$ ArKr+ + Ar	1.00E-31
25)	ArKr+ + Kr	$\rightarrow$ Kr2+ + Ar	3.20E-10
26)	Kr2+ + F-	$\rightarrow$ KrF* + Kr	2.07E-6
27)	Kr+ + Ar + Kr	$\rightarrow$ Kr2+ + Ar	2.50E-31
28)	Kr2+ + hnu	$\rightarrow$ Kr + Kr+	6.00E-8
29)	ArKrF* + Kr	$\rightarrow$ Kr2F* + Ar	1.00E-10
30)	ArKrF* + F2	$\rightarrow$ Ar + Kr + F + F2	1.00E-9
31)	Kr2F*	$\rightarrow$ Kr + Kr + F	6.70E-7
32)	Kr2F* + F2	$\rightarrow$ Kr + Kr + F + F2	1.00E-9
33)	Kr2F* + Kr + Ar	$\rightarrow$ Kr + Kr + Kr + Ar + F	2.50E-32
34)	Kr2F* + hnu	$\rightarrow$ Kr + Kr + F	5.00E-8
35)	F + F + Ar	$\rightarrow$ F2 + Ar	1.00E-32
36)	Ar2+ + Ar + Ar	$\rightarrow$ Ar+ + Ar + Ar + Ar	7.00E-32
37)	ArKr+ + F-	$\rightarrow$ KrF* + Ar	1.00E-6
38)	KrF* + Kr + Kr	$\rightarrow$ Kr2F* + Kr	6.70E-31
39)	Kr2F* + Kr + Kr	$\rightarrow$ Kr + Kr + Kr + Kr + F	2.90E-31
40)	KrF* + eSLOW	$\rightarrow$ Kr + F + eSLOW	2.00E-7
41)	ArF* + Ar + Ar	$\rightarrow$ Ar2F* + Ar	3.00E-31
42)	Ar* + F2	$\rightarrow$ ArF* + F	8.50E-10
43)	Ar* + Ar + Ar	$\rightarrow$ Ar2* + Ar	1.10E-32
44)	Ar* + Kr	$\rightarrow$ Kr* + Ar	6.20E-12
45)	Ar2* + Kr	$\rightarrow$ Kr* + Ar + Ar	4.00E-10
46)	Kr* + F2	$\rightarrow$ KrF* + F	8.10E-10
47)	Kr* + Ar + Ar	$\rightarrow$ ArKr* + Ar	1.00E-32
48)	Ar* + Kr + Ar	$\rightarrow$ ArKr* + Ar	1.00E-32
49)	ArKr* + F2	$\rightarrow$ KrF* + Ar + F	6.00E-10
50)	ArKr* + Kr	$\rightarrow$ Kr2* + Ar	1.10E-10
51)	Kr2* + F2	$\rightarrow$ KrF* + Kr + F	3.00E-10
52)	Kr2* + F2	$\rightarrow$ Kr2F* + F	3.00E-10
53)	Kr* + Kr + Ar	$\rightarrow$ Kr2* + Ar	1.00E-32
54)	Ar2F* + hnu	$\rightarrow$ Ar + Ar + F	3.90E-7
55)	KrF* + Kr	$\rightarrow$ Kr + Kr + F	2.00E-11
56)	Kr2F* + Ar + Ar	$\rightarrow$ Ar + Ar + Kr + Kr + F	1.00E-32
57)	ArF*	$\rightarrow$ Ar + F	2.50E-8
58)	ArF* + Ar	$\rightarrow$ Ar + Ar + F	9.00E-12
59)	ArF* + F2	$\rightarrow$ Ar + F + F2	1.90E-9
60)	ArKrF*	$\rightarrow$ Ar + Kr + F	5.00E-7
61)	Ar2F* + Kr	$\rightarrow$ ArKrF* + Ar	1.00E-10
62)	ArKr+ + hnu	$\rightarrow$ Ar + Kr+	4.50E-7
63)	ArKrF* + hnu	$\rightarrow$ Ar + Kr + F	4.50E-7
64)	ArF* + eSLOW	$\rightarrow$ Ar + F + eSLOW	2.00E-7
65)	Kr2F* + eSLOW	$\rightarrow$ Kr + Kr + F + eSLOW	1.00E-7
66)	Ar2F* + eSLOW	$\rightarrow$ Ar + Ar + F + eSLOW	1.00E-7
67)	Ar* + hnu	$\rightarrow$ Ar+ + eSLOW	1.67E-7
68)	Kr* + hnu	$\rightarrow$ Kr+ + eSLOW	1.67E-7
69)	Ar2+ + eSLOW	$\rightarrow$ Ar* + Ar	6.50E-8
70)	Kr2+ + eSLOW	$\rightarrow$ Kr* + Kr	1.90E-7
71)	Ar* + eSLOW	$\rightarrow$ Ar+ + eSLOW + eSLOW	2.80E-8
72)	Kr* + eSLOW	$\rightarrow$ Kr+ + eSLOW + eSLOW	4.80E-8
73)	Ar2F*	$\rightarrow$ Ar + Ar + F	2.00E-8

time. Their rate constants are calculated according to Johnson and Hunter [2]. Included in the model are pressure dependent rate constants for the recombination reactions like $X^+ + F^-$ where $X = Ar, Ar_2, Kr$, and Kr_2 after Flannery and Yang [13, 14]. The rare-gas dimer ion-electron recombination rate coefficients were calculated for a mean secondary electron temperature of 1 eV.

The effect of the distribution of KrF^* molecules in the vibrational state of the B level is not included in our code because of the fact that too little information is available with respect to the instantaneous distribution in these levels. Moreover the relaxation rates and stimulated emission cross-sections are unknown.

The input parameters of the code are a time dependent current density, the electron energy, the initial gas pressures, geometrical factors as gain length, cavity length and electron path length as well as outcoupling area and percentage.

Except the current density, for which only the time dependence but not the absolute value is known, all these input parameters are measurable quantities.

As a consequence we take in our model calculations the time dependence of the input current, as shown in Fig. 1c, while the absolute value which is the only parameter in our model, is fitted according to the experimental results.

If the model is correct this fitted value should be in agreement with the experimental peak current density.

Compared to the transverse e-beam geometry our coaxial configuration is more complicated with respect to the real current estimation. Only the total anode current can be measured easily.

Assuming the total e-beam current divided in n current beams, each irradiating an anode tube area (length $\times$ diameter) from another side, the average current density due to all this n beams can be estimated as

$$n(I_{\text{tot}}/n)/l \cdot D, \quad (1)$$

where l is the active pumping length and D is the tube diameter. At a Marx generator charging of 60 kV the diode current (peak) and voltage (averaged) were measured to be 13.3 kA and 170 kV, respectively. Using the formula above an average current density of 133 A/cm² can be estimated. In order to fit our experiments at the Marx load voltage of 60 kV, an effective peak current of 150 A/cm² has to be inserted in the model. This value agreed very well with the value of 133 A/cm². For other measured diode current values the code input current was scaled accordingly. So we used a current density of 300 A/cm² in the code describing the experiments done at a total diode current of 26.6 kA (100 kV load voltage). According (1) the measured peak current density in the tube was 266 A/cm².

The temporal behaviour of the current pulse was simulated by multiplying the peak current density with the function depicted in Fig. 1c, corresponding to a pulse having a duration of 250 ns FWHM and a risetime of about 100 ns.

The input voltage used in the code is kept constant at the measured diode voltage minus 44 kV over the

whole pulse length. Corrections at the beginning and at the end of the pulse would contribute only little to the total production of ion-electron pairs as the e-beam current is rather low in these time intervals.

Other than the fitted current density all input parameters are measurable quantities as gain length (50 cm) resonator length (91 cm), mirror reflection (0.11) and outcoupling area (3.14 cm²).

The code is runned on a DEC 2060 computer with a cpu time varying from about 20 s for the medium and high F_2 mixtures up to 90 s for the bean F_2 mixtures.

4. Model Calculations

We covered a broad area with our experimental parameters. Moreover we measured for each of these parameters the optical pulse length, the delay time between the onset of the current pulse and the optical pulse and the total energy. With all these results it is possible to check every model very thoroughly as it has to predict all these values over the whole range. As we shall describe later on our model calculations showed that we had to modify some reactions, for the rest without changing the rate constants, in order to fit the experimental data at the lowest F_2 concentrations. From our calculations it appears that the contributions to the various formation and quenching channels may show large changes with respect to the input parameters. We will discuss them with the help of the results of one particular calculation out of the whole matrix we had to carry out. For this purpose we took a gas mixture of 16 mbar F_2 , 380 mbar Kr and pressurized up to 4 bar with Ar as buffer gas. The mixture was excited by an e-beam with a current density of 300 A/cm² at a voltage of 265 kV yielding an output energy of 9.2 J.

In Fig. 5 the contribution of the formation channels during the pulse evolution is plotted.

The formation channels are in sequence of their importance, the recombination reactions of Kr^+ , Kr_2^+ , and ArKr^+ with F^- , the replacement reaction $\text{ArF}^* + \text{Kr}$ and the reaction of Kr^* and Kr_2^* metastables with F_2 .

With the exception of the bean F_2 mixtures and besides the beginning and the end of the pulse where the contributions change considerably due to the F_2 burn-up, in general the variations during the pulse are relatively small. However larger deviations occur when the input parameters are changed.

For high partial pressures of F_2 the contribution of the Kr_2^+ channel is 20–30% lower than at a low F_2 density. This decreased contribution in the presence of higher F_2 densities is at a total gas pressure of 2 bar almost completely compensated by the contribution

Fig. 5. Contributions in percent to the formation of KrF^* during the evolution of the pulse for a Marx load voltage of 100 kV and for a 0.4/9.2/90.4 ($\text{F}_2/\text{Kr}/\text{Ar}$) gas mixture at 4 bar

increment of the $\text{ArF}^* + \text{Kr}$ channel while at a pressure of 4 bar the contribution of both the $\text{ArF}^* + \text{Kr}$ and the $\text{Kr}^+ + \text{F}^-$ channel are enhanced at the expense of the Kr_2^+ channel. The changes in the other channels are less pronounced with respect to both the time dependency and the F_2 density.

In Fig. 6 the KrF^* losses are plotted as a function of time. Besides the stimulated emission which is responsible for the main losses of KrF^* molecules, three body quenching of KrF^* by 2Ar and by $\text{Ar} + \text{Kr}$ is an important loss mechanism as can be seen from the figure.

Our calculations show that at a total pressure of 4 bar the contribution to the relaxation of KrF^* by 2Ar and by $\text{Ar} + \text{Kr}$ are roughly two times higher than at a pressure of 2 bar. At the same time the contribution of the spontaneous emission to the total relaxation decreases more than two times.

The photon losses are given in Fig. 7. The outcoupling of stimulated radiation delivers the largest contribution as can be seen from the figure. The second important photon loss channel is the absorption by F_2 . The contribution of both mechanisms vary strongly depending on input parameters.

Except for the lowest investigated F_2 concentration, where all F_2 is burned up quickly, 10% up to 30% of the photons are lost through absorption by F_2 in low power pumped gas mixtures containing 0.25%

Fig. 6. Percentual contribution of KrF^* loss channels during the pulse under the conditions as mentioned in Fig. 6

Fig. 7. Percentual losses of photons via different channels as a function of time under the conditions as mentioned in Fig. 6

to 0.8% F_2 , respectively. In the high power pumping regime the F_2 absorption is less dominant because of the quickly lowered F_2 concentration due to the reaction $\text{F}_2 + \text{e}_s \rightarrow \text{F} + \text{F}^-$.

5. Discussion

Table 3 summarizes the calculated and measured laser output energies and laser pulse durations for the experiments run at 60, 80, and 100 kV initial charging voltage. In general good agreement is found, especially, in the case of gas mixtures with medium or high F_2 densities. The comparison of the measured optical pulse delays and the computed values also shows a good agreement even for the low current and high F_2 density region where, as we mentioned above, the optical delay can be as large as 130 ns.

However for mixtures with a F_2 content of only 0.1% a large deviation of the predictions with measurements is obtained. Tracing the contributions of single reactions to the whole kinetics it can be seen that the density of secondary electrons play a crucial role in the case of F_2 lean mixtures. In Fig. 8 the uninterrupted curve present the calculated densities of F_2 , e_s , F^- , and KrF^* during the pulse for a 4 bar gas mixture containing 0.1% F_2 and excited by an e-beam current density of 300 A/cm². The calculations show that, as the F_2 burns up, the density of secondary electrons increases rapidly due to the decreased rate of electron attach-

ment by F_2 . At the same time the production of F^- ions decreases and KrF^* is mainly quenched by secondary electrons resulting in a poor laser performance. Yet, the experimentally obtained values for output energy and pulse duration are higher and longer, respectively, than those calculated.

Assuming that there is no doubt about the rates at which slow electrons are produced and captured by F_2 or deactivate KrF^* , ArF^* , Kr_2F^* , and Ar_2F^* [15] it must be concluded, that the electron quenching of these species should be a dissociative attachment process yielding F^- rather than a super-elastic collision process resulting in $F + e_s$. This dissociative attachment would be responsible for an increased production rate of F^- and a decreased density of secondary electrons, both having a beneficial effect on the net production of KrF^* . The simulation of this effect is done in our computer model by changing the reactions numbered 40, 64, 65, and 66 (Table 2) accordingly, for the rest using the same rate constants. The dashed curves in Fig. 8 show the densities of the same species under the same input conditions after the code has been changed. The results of the calculations have been included in Table 3 indicated with an

Table 3. Comparison of experimental and computed output energies and pulse widths. The values denoted with an asterisk are computed after a slight modification of some reactions in the code (see text)

% F ₂		0.1		0.25		0.4		0.6		0.8		
Marx load voltage [kV]	Total pressure [bar]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]	Out-put [J]	Pulse length [ns]	
60	2	exp	1.4	110	2.9	160	2.2	160	2.2	155	1.5	130
		code	0.4	67	2.5	182	2.4	181	1.9	170	1.4	153
		*	1.1	122					1.9	169		
	4	exp	1.9	115	2.7	160	1.3	155	0.9	105	0.3	75
		code	0.3	45	2.8	166	2.4	146	1.2	120	0.5	88
		*	0.9	80								
80	2	exp	1.7	70	4.3	115	5.5	170	6.0	185	5.0	170
		code	0.5	55	3.1	147	5.1	200	4.6	196	3.9	190
		*	1.4	95	4.4	180	5.1	200	4.6	196		
	4	exp	2.7	70	7.2	110	8.3	170	7.6	165	5.3	145
		code	0.5	40	4.3	139	6.9	183	5.8	173	4.4	165
		*	1.5	72	6.2	163	6.9	183	5.8	173		
100	2	exp	1.4	40	4.0	85	6.0	135	7.2	170	7.2	150
		code	0.5	48	3.3	125	6.0	195	6.2	205	5.2	200
		*	1.4	84	5.1	170						
	4	exp	2.7	50			9.7	155			10.0	150
		code	0.6	37	4.8	116	9.1	189	7.9	189	6.0	178
		*	1.7	68	6.8	150	9.2	192

Fig. 8. Calculated temporal behaviour of the densities of KrF^* , F^- , F_2 , and e_s assuming the quenching of KrF^* , ArF^* , Kr_2F^* , and Ar_2F^* by e_s to be a dissociative attachment process (dashed curve). The solid curve shows the same densities treating these quenching process as a super elastic collision process

asterisk. Actually the results approach the measured ones much better.

Moreover the simulations for F_2 rich mixtures are hardly changed, as expected, because in these cases the density of secondary electrons does not attain such a value to dominate the KrF^* quenching.

With this modification of the computer model it was assumed that in all collisions of KrF^* , ArF^* , Kr_2F^* , and Ar_2F^* molecules with a slow electron a F^- ion is released. Thus, an even better model fit can possibly be obtained if for each of these species the correct branching ratio between dissociative attachment of electrons and super-elastic electron collision would be known.

In our model the temperature dependency as given by Shui [16] is not included. As a consequence the quenching rate constants of KrF^* by 2 Ar and by $\text{Ar} + \text{Kr}$ is probably too large. This on its turn may be the reason why the computer code predicts values for

the output energy which are somewhat too low for the experiments in the high energy density and high pressure regime.

6. Conclusion

We investigated a KrF^* excimer laser excited by a coaxial e-beam and compared the results with the predictions of a computer model. The experiments cover a large area of input parameters. Substantial agreement between this large range of experiments and the model is only obtained if electron quenching by dissociative attachment of the excimer molecules is also included in the kinetic chain.

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