

# Laser generation of XeCl exciplex molecules in a longitudinal repetitively pulsed discharge in a Xe–CsCl mixture

A.M. Boichenko, M.S. Klenovskii

**Abstract.** By using the previously developed kinetic model, we have carried out simulations to study the possibility of laser generation of XeCl exciplex molecules in the working medium based on a mixture of Xe with CsCl vapours, excited by a longitudinal repetitively pulsed discharge. The formation mechanism of exciplex molecules in this mixture is fundamentally different from the formation mechanisms in the traditional mixtures of exciplex lasers. The conditions that make the laser generation possible are discussed. For these conditions, with allowance for available specific experimental conditions of the repetitively pulsed discharge excitation, we have obtained the calculated dependences of the power and efficiency of generation on the reflectivity of mirrors in a laser cavity.

**Keywords:** exciplex XeCl laser, discharge in a mixture of rare gas with alkali metal halide vapours, modelling of the kinetics of processes, longitudinal repetitively pulsed discharge.

## 1. Introduction

Excimer and exciplex lasers are the most high-power lasers emitting in the VUV and UV spectral range [1–9]. Among the most powerful and efficient exciplex lasers are KrF, XeCl, ArF and XeF lasers [7–9]. The mixtures of rare gases with halogen-containing molecules of HCl, F<sub>2</sub>, NF<sub>3</sub> are typically used as working mixtures in these lasers. The burn-out of the mixture at a high power is a serious problem in the repetitively pulsed regime. Its conventional solution consists in circulation of the mixture; however this turns out advantageous at low pulse repetition rates. In transition to repetition rates on the order of 1 kHz, the discharge is accompanied by the development of various instabilities [5], greatly complicating a further increase in the pulse repetition rate. The measures against the emerging characteristic instabilities are principally possible, but they hardly lead to a somewhat significant increase in the repetition rate. Nowadays, the pulse repetition rates of exciplex lasers utilising traditional mixtures are typically limited by 5 kHz.

The question arises: is it possible to significantly increase the pulse repetition rate of exciplex lasers or even make it

infinite, i.e. to pass on to a cw regime? The attempts to implement a cw exciplex laser have been undertaken in numerous works of a group of researchers from Sweden and Russia (see, e.g., [10–17]). A sharp increase in the emission yield in a narrow line  $\lambda = 146.94\text{--}147.03$  nm at cryogenic temperatures, with small amount of Xe (up to 1%) added to Kr at a pressure of hundreds of Torr, is interpreted by the authors [11, 13, 14] as a gain on the bound-free transition  $1_u\text{--}0_u^+$  of the XeKr molecule. This conclusion, as noted by the authors, is based on the studies on the calculation of the potential energy curves of this molecule [18–24] and modelling of part of the spectrum performed in [25]. However, these same authors recognise [16] that there is no reliable identification: “For reliable identification of the analysed molecular spectra, theoretical modelling of these spectra is usually performed. This procedure requires knowledge on the potential curves for the upper and lower states, on the distribution of population by the vibrational levels, and on the dipole moments of the transitions that depend on the internuclear distance. Unfortunately, we have very limited information about all these parameters, and this impedes the modelling of these spectra”. Recent calculations carried out in [26] are still unable to explain the shape of the emitted continuum. Note that the above-described drastic increase in the emission line of  $\lambda \approx 147$  nm can be explained by the radiation at optical collisions [27]. Similar spectra are identified in [28–33] as a resonance radiation of the xenon atom.

The estimates of the gain in a Kr–Xe mixture at  $\lambda \approx 147$  nm, which are obtained by researchers for 15 years (since 1991 [10] and 2006 [15, 17]) remain invariable:  $\kappa = 0.1$  cm<sup>-1</sup>. Different types of excitations such as glow or barrier discharges with different pump power levels were used in these studies. For example, in [12] the barrier discharge possesses a filamentary structure; the authors estimate the size of the filaments as not exceeding 0.1 mm and the electron density in the filaments as being no less than 10<sup>15</sup> cm<sup>-3</sup>. Excitation of the medium in such discharges occurs largely in filaments [34–37]; however, in this case the gain value presented by the authors is also the same. As a result of the studies performed, Gerasimov et al. [17] announce the implementation of continuous stimulated radiation, but the authors themselves note [15, 17] that the use of this radiation is still impossible due to many factors, including refraction, rapid decrease in transmittance of the discharge tube windows, etc., and also requires solving a set of issues relevant to withdrawal of radiation from the plasma volume.

In this paper, to increase the pulse repetition rate of exciplex lasers, a transition to another type of the halogen-carrier, namely to alkali metal salts, is proposed. Interest in alkali metal halides as halogen donors has emerged after the studies

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[38–41], in which, using a nontraditional mixture, radiation of XeCl exciplex molecules ( $\lambda = 308$  nm) was observed: an effective luminescence was recorded during the injection of the salts of NaCl into a supersonic plasma flow. Herewith, populating of the emitting state occurs mainly due to the binary reaction of replacing sodium in the NaCl molecule by an excited xenon atom or ion. This mechanism of the formation of an excited state of the XeCl molecule is new; it differs from the previously known ion–ion and harpoon reactions that are implemented in traditional mixtures. Possible characteristics of the XeCl lamp, which may result from this mechanism in the process of excitation of the Xe–NaCl mixture by a hard ioniser (electron or ion beams, etc.) are considered in [42].

Further investigations of the exciplex sources of UV radiation by using the vapours of alkali metal salts as halogen donors are performed in [43–48], in which the mixtures of xenon and krypton with the vapours of NaCl, KCl, CsBr and CsCl are used. The vapours of alkali metal halides arise as a result of evaporation of salts from the solid state. The amount of the halogen donor molecules in the working mixture is varied by changing the temperature of the discharge tube external heater [47]. To excite the gas–vapour mixtures, a longitudinal high-voltage repetitively pulsed discharge is used. The maximal luminescence power of the exciplex molecules of XeCl is reached by the excitation of the mixture of xenon with caesium chloride [46]. The use of CsCl instead of NaCl and KCl also reduces the heating temperature of the discharge tube.

As already mentioned, the formation mechanisms of the working exciplex molecules in the mixtures of rare gases with alkali metal halides are fundamentally different from relevant mechanisms in the traditional exciplex lasers. May these mechanisms, similarly to the traditional ones, lead to laser generation? Unfortunately, there is still no experimental data on the gains in a medium based on the mixtures of rare gases with the alkali halides vapours excited by the longitudinal repetitively pulsed discharge. To answer the question on the possibility of laser generation, we resort to the numerical simulation method. Simulation of the repetitively pulsed regime of the exciplex XeCl-lamp radiation excited by the longitudinal repetitively pulsed discharge in a mixture of xenon and caesium chloride vapours has been conducted by us earlier in [49]. In this paper, we simulate the possibility of obtaining lasing in the Xe–CsCl mixture, and the attainability of the repetitively pulsed discharge characteristics needed in this connection.

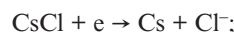
In our study on the possibility of lasing, we have been oriented on the discharge parameters being specific for operating the XeCl lamp on a Xe–CsCl mixture [46]: the relevant repetition rate reaches approximately 10 kHz, and what is more, this value is an ordinary one, i.e. there are no obstacles to its increase. Excitation of the repetitively pulsed discharge has been realised via the storage capacitor discharge by means of a thyatron switch. Being the possibility of laser generation experimentally confirmed, it would open the prospects for this type of pumping because it is well-developed and widely used in the repetitively pulsed metal–vapour lasers [50, 51]. Note also that this technique ensures the record-breaking pulse repetition rate of 750 kHz in a copper–vapour laser using the CuBr salt [52–54]. Thus, a sufficiently high pulse repetition rate of excitation can be reached experimentally. The question arises: will it be useful in terms of increasing the pulse repetition rate of exciplex lasers, and which is the limiting value of this rate? Are there any constraints on its increase and what is their nature?

## 2. Theoretical description

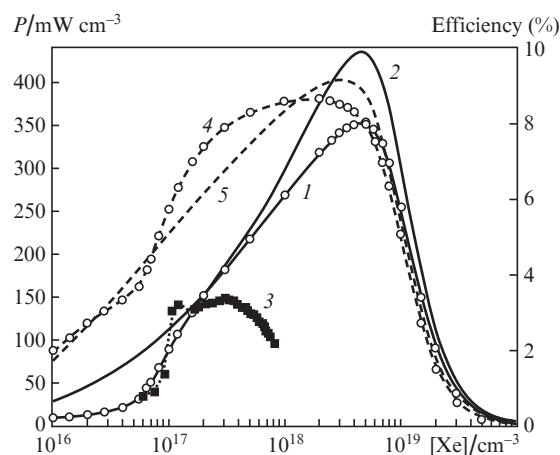
**Kinetic model.** Our simulation is based on a previously developed and tested model [49] describing the kinetics of plasma-chemical processes in the repetitively pulsed discharge in a Xe–CsCl mixture. To explore the possibility of lasing on the XeCl\* exciplex molecule that is formed in the medium under consideration, the model [49] has been supplemented with the reactions describing the process of stimulated emission and absorption of laser radiation by various plasma components [55].

In the presented model, instead of XeCl (B) and XeCl (C) states, a united state XeCl\* is used as an upper working state; therefore, the cross section of stimulated emission on the XeCl\*  $\rightarrow$  XeCl(X) transition is set equal to  $2 \times 10^{-16}$  cm<sup>2</sup>, which is less than the cross section  $4 \times 10^{-16}$  cm<sup>2</sup> for the XeCl(B)  $\rightarrow$  XeCl(X) transition (for details, see [7, 8]). The absorption cross sections of atoms and molecules at the generation wavelength for the XeCl molecule ( $\lambda = 308$  nm) are taken from our early works on the laser generation modelling in the conventional mixtures [7, 8]. The laser radiation absorption by the CsCl molecules is ignored [56].

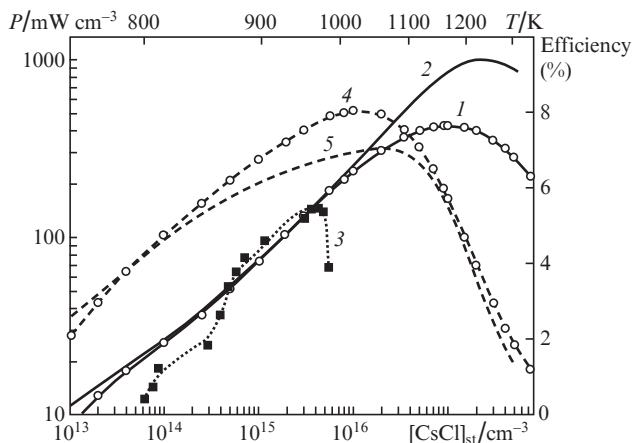
Smirnov [57] notes a negligible role of the reaction of dissociative attachment of electrons to the CsCl molecule with the formation of negatively charged ions of atomic chlorine Cl<sup>-</sup>:



therefore, this reaction has been removed from the model [49] in which the value of its rate constant is set on the basis of an estimate. The calculation results have changed, albeit not significantly (Figs 1–4). The discharge contraction starts at  $[\text{Xe}] > 3 \times 10^{17}$  cm<sup>-3</sup>, and so the experimental data in this region (Fig. 1) are illustrative. At low concentrations of Xe, after the mentioned model correction, the agreement of calculation results and experiment has improved significantly (Fig. 1). The agreement of the experiment with the calculated dependence of the radiation power on the concentration of halogen-carrier vapours in the near-wall region of the discharge tube  $[\text{CsCl}]_{\text{st}}$  has also improved (Fig. 2).

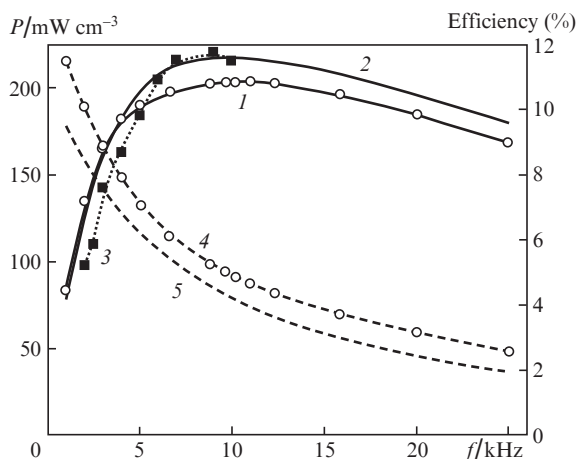


**Figure 1.** Average specific power of spontaneous radiation (1–3) and efficiency of the energy injected into the medium (4, 5) as functions of the xenon concentration: (1, 4) present paper, (2, 5) paper [49], (3) experiment [46] ( $[\text{CsCl}]_{\text{st}} = 5.6 \times 10^{15}$  cm<sup>-3</sup>,  $f = 4$  kHz,  $T = 970$  K).



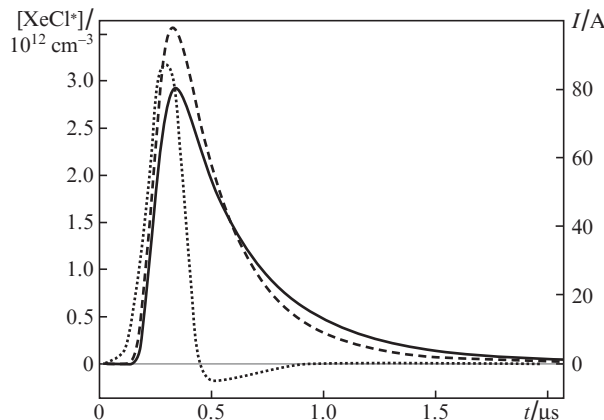
**Figure 2.** Average specific power of spontaneous radiation and efficiency of the energy injected into the medium as functions of the concentration of the  $[\text{CsCl}]_{\text{st}}$  molecules near the discharge tube wall ( $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $f = 4 \text{ kHz}$ ,  $T = 970 \text{ K}$ ). The notations are the same as in Fig. 1. The temperature corresponding to the concentration of the saturated vapours of CsCl, equal to  $[\text{CsCl}]_{\text{st}}$ , is indicated at the top.

In the region of discharge excitation frequencies exceeding 5 kHz, the calculated radiation power curve (Fig. 3) now lies below the curve obtained in [49]. In this model, at the concentration  $[\text{CsCl}]_{\text{st}} = 6.5 \times 10^{15} \text{ cm}^{-3}$  ( $T = 987 \text{ K}$ ), the experimental data turn out to be in good agreement with the calculated ones. At the concentration of CsCl molecules in the range of  $(5-7) \times 10^{15} \text{ cm}^{-3}$ , the calculated curves of the frequency dependence of the spontaneous radiation power (the lamp source power) converge in the frequency region of about 1 kHz.

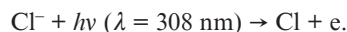


**Figure 3.** Average specific power of spontaneous radiation and efficiency of the energy injected into the medium as functions of the excitation pulse repetition rate ( $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $[\text{CsCl}]_{\text{st}} = 5.6 \times 10^{15} \text{ cm}^{-3}$ ,  $T = 970 \text{ K}$ ). The notations are the same as in Figs 1, 2.

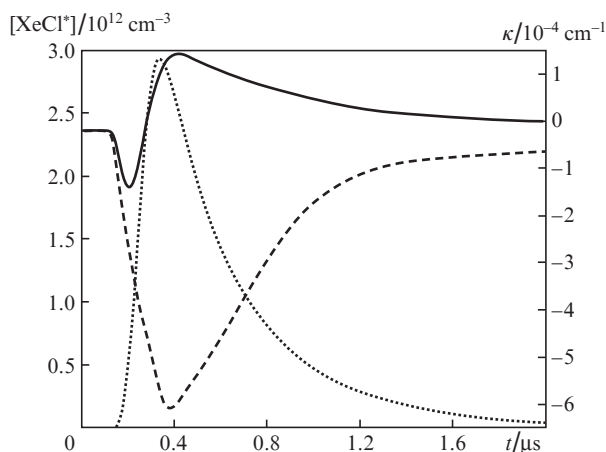
When considering the gain of the working medium, the reaction of dissociative attachment of electrons to the CsCl molecule has proved to be important from the viewpoint of the possibility of obtaining laser generation. A considerable accumulation of the negative ions of atomic chlorine in the working medium, being a result of this reaction, leads to a negative small-signal gain due to photon absorption at  $\lambda = 308 \text{ nm}$  in the photo-detachment process:



**Figure 4.** Calculated temporal dependences of the concentration of  $\text{XeCl}^*$  exciplex molecules ( $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $[\text{CsCl}]_{\text{st}} = 5.6 \times 10^{15} \text{ cm}^{-3}$ ,  $f = 4 \text{ kHz}$ ,  $T = 970 \text{ K}$ ). Solid curve stands for the present work, dashed curve – paper [49], dotted curve – discharge current pulse waveform.



In the absence of the reaction of dissociative electron attachment to the CsCl molecule, the gain of the medium becomes positive and reaches its maximum of  $1.4 \times 10^{-4} \text{ cm}^{-1}$  (Fig. 5).

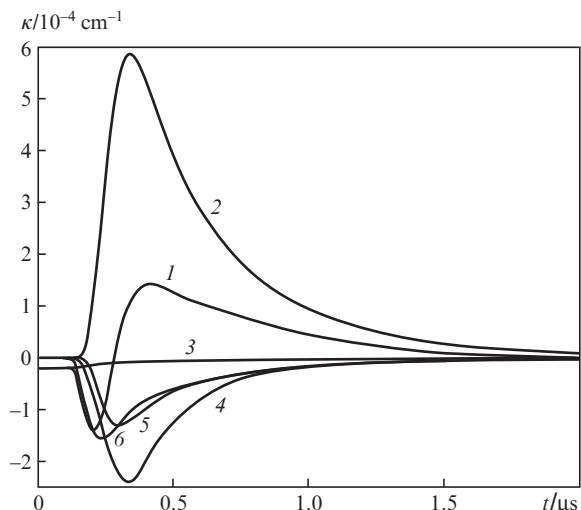


**Figure 5.** Calculated temporal dependences of the concentration of  $\text{XeCl}^*$  exciplex molecules (dot-dashed curve), of the small-signal gain with account for the process of dissociative attachment of electrons to the molecules of CsCl (dashed curve) and of the gain of medium without regard to the process of dissociative attachment of electrons to CsCl molecules (solid curve);  $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $[\text{CsCl}]_{\text{st}} = 5.6 \times 10^{15} \text{ cm}^{-3}$ ,  $f = 4 \text{ kHz}$ ,  $T = 970 \text{ K}$ .

The contribution of major components to the gain (absorption) of the medium in the small-signal regime is illustrated in Fig. 6. The greatest absorption in the working medium at the laser radiation wavelength is exhibited by the excited and, especially, highly excited atoms of xenon as a result of the photoionisation process:



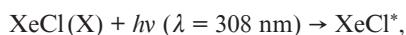
Next, in descending order of the absorption intensity, the following processes are mentioned: photoionisation of excited caesium atoms



**Figure 6.** Calculated total coefficient of the small-signal gain ( $I$ ), and the contributions to this coefficient of separate plasma components: (2)  $\text{XeCl}^*$ , (3)  $\text{Cs}$ , (4)  $\text{Xe}^{**}$ , (5)  $\text{XeCl}$ , (6)  $\text{Cs}^*$ ;  $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $[\text{CsCl}]_{\text{st}} = 5.6 \times 10^{15} \text{ cm}^{-3}$ ,  $f = 4 \text{ kHz}$ ,  $T = 970 \text{ K}$ .



absorption by exciplex molecules in the ground state on the working transition



and photoionisation of caesium atoms in the ground state



The intensity of radiation absorption by negative ions of chlorine as a result of photodetachment of electrons is by several orders of magnitude smaller than in the above processes. All other photo-processes that are taken into account in the model have virtually no effect on the gain of the medium in question.

*Laser cavity description.* Simulation of laser generation has been conducted in the zero-dimensional approximation (in the approximation of effective lifetime of a photon in the cavity) [6–9, 50, 52]. Herewith, the system of kinetic equations includes an equation for the volume-average intensity  $I$  of laser radiation:

$$\frac{dI}{dt} = (c(\kappa^+ - \kappa^-) - \gamma)I + \frac{Q}{4\pi}.$$

Here,  $c$  is the speed of light;  $\kappa^+$ ,  $\kappa^-$  are the coefficients of gain by the  $\text{XeCl}^*$  molecules and of the absorption by the medium;  $Q$  is the term responsible for the contribution of spontaneous radiation to the lasing development;

$$\gamma = \frac{c}{2l} \ln \frac{1}{R_1 R_2}$$

is a factor that represents the inverse lifetime of the photon in the laser cavity and characterises the laser radiation output from the cavity in the zero-dimensional approximation for the radiation transfer equation;  $l$  is the active region length; and  $R_1$ ,  $R_2$  are the reflection coefficients of mirrors in the laser

cavity. If the knowledge on the laser output divergence is not required (on the modelling in this case see, e.g. in [6, 58, 59]), this approach describes well the output energy dependence on the reflection coefficients of the cavity mirrors and the working medium length [6–9].

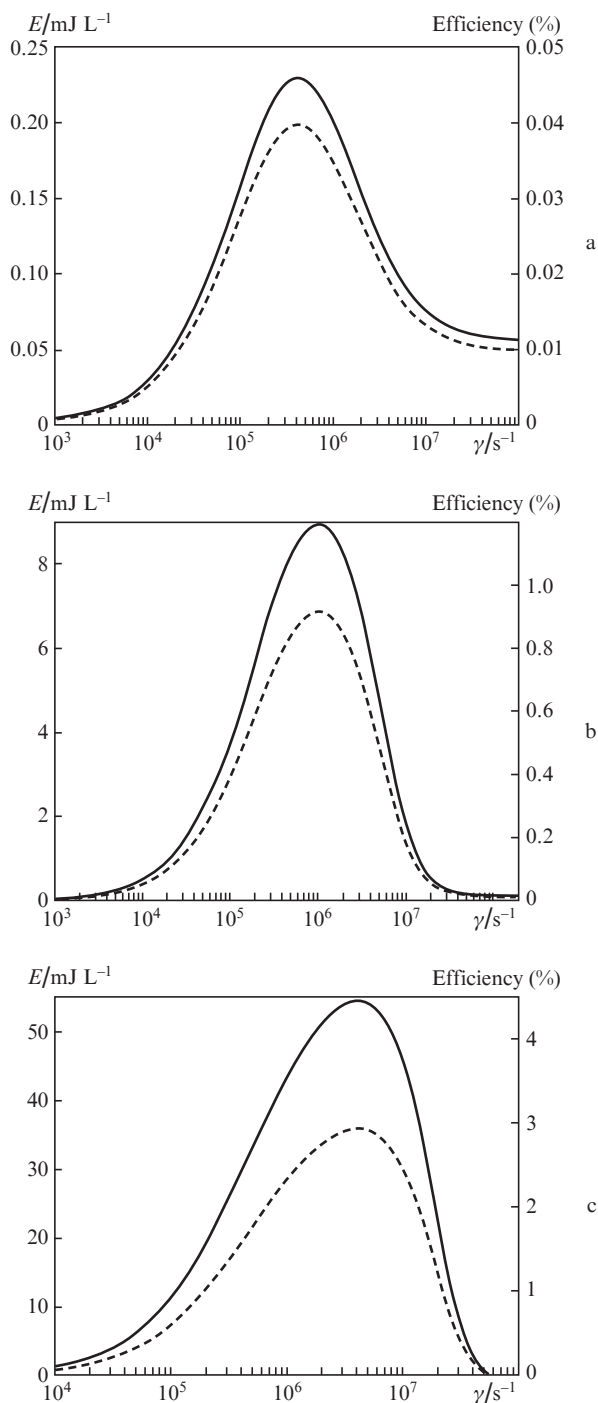
### 3. Discussion of the results

Under the experimentally defined optimal conditions [46] that ensure the maximal output power of spontaneous radiation, the calculated available lasing energy is not large. The maximum value of the laser pulse specific energy is attained at  $\gamma \approx 4 \times 10^5 \text{ s}^{-1}$ , and constitutes approximately  $0.2 \text{ mJ L}^{-1}$ , with the lasing efficiency of 0.04% with respect to the energy injected into the working medium (Fig. 7a).

An increase in the lasing power can be expected in conditions when a greater concentration of  $\text{XeCl}^*$  exciplex molecules is attained, which, according to the dependences shown in Figs 1, 2, requires an increase in the concentrations of xenon and caesium chloride. The calculations show that the maximal value of the gain is achieved at  $[\text{Xe}] \approx 2.5 \times 10^{18} \text{ cm}^{-3}$ . However, an increase in the xenon concentration leads to the discharge contraction [43–48], this is why we present below the data for  $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ , which, according to [46], are optimal for a lamp source. At this Xe concentration, the calculated maximal gain of the medium is attained at  $[\text{CsCl}]_{\text{st}} = 1 \times 10^{17} \text{ cm}^{-3}$  and constitutes  $\kappa \approx 2.3 \times 10^{-3} \text{ cm}^{-1}$ ; therefore, we also present the simulation results for the larger halogen-carrier concentrations (Figs 7b, 7c).

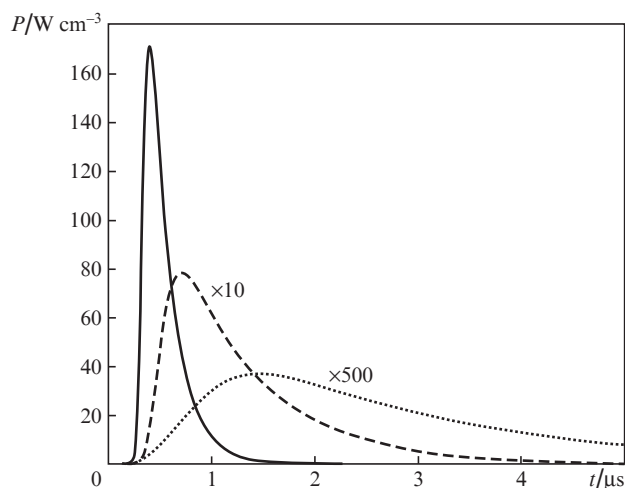
At  $[\text{CsCl}]_{\text{st}} = 2 \times 10^{16} \text{ cm}^{-3}$ , the maximal value of the laser output pulse specific energy is attained at  $\gamma \approx 1 \times 10^6 \text{ s}^{-1}$  and constitutes about  $9 \text{ mJ L}^{-1}$ , with the lasing efficiency of 0.9% relative to the energy introduced into the medium (Fig. 7b). The required temperature to ensure the saturated vapour concentration  $[\text{CsCl}]_{\text{st}} = 2 \times 10^{16} \text{ cm}^{-3}$  is approximately 1055 K. With increasing  $[\text{CsCl}]_{\text{st}}$  up to  $1 \times 10^{17} \text{ cm}^{-3}$ , the maximal value of the laser output pulse specific energy is achieved at  $\gamma \approx 4 \times 10^6 \text{ s}^{-1}$  and exceeds  $50 \text{ mJ L}^{-1}$  (Fig. 7c). Herewith, the lasing efficiency relative to the energy introduced into the medium is  $\sim 3\%$ . To ensure  $[\text{CsCl}]_{\text{st}} = 1 \times 10^{17} \text{ cm}^{-3}$ , heating of caesium chloride up to the temperatures of about 1170 K is required. With a gain increase, the optimum value of the product of the reflectivities of the mirrors is reduced, which leads to the reduction in duration and increase in the power of the laser pulse generation (Fig. 8).

The spectroscopic and kinetic aspects of the principal difference between the traditional formation mechanisms of exciplex molecules and the substitution reactions that occur simultaneously through the neutral and ion channels are considered in [38, 42, 48]. The predominance of the substitution reactions compared to the traditional channels of populating the working states of the exciplex molecules must be also manifested in the active medium of lasers on such mixtures in the case of their possible implementation. Indeed, the estimated contribution of the neutral and ion channels to the formation of exciplex molecules constitutes 66% and 30%, respectively (for the conditions  $f = 4 \text{ kHz}$ ,  $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $[\text{CsCl}]_{\text{st}} = 1 \times 10^{17} \text{ cm}^{-3}$ ,  $T = 1170 \text{ K}$ ,  $\gamma = 4 \times 10^6 \text{ s}^{-1}$ ). The traditional channels of the exciplex molecule formation (reaction of ion–ion recombination, harpoon reaction) only account for about 1%. A low contribution of the harpoon reaction ( $2 \times 10^{-4}\%$ ) is caused by a small accumulation of the chlorine dimers, which is due to more efficient binding of atomic chlorine with caesium atoms, i.e. the reactions of regeneration of



**Figure 7.** Specific pulse energy of laser radiation (solid curve) and laser radiation efficiency relative to the energy injected into the medium (dashed curve) as functions of the inverse lifetime of laser radiation in the laser cavity for  $[\text{CsCl}]_{\text{st}} = 5.6 \times 10^{15} \text{ cm}^{-3}$ ,  $T = 970 \text{ K}$  (a),  $[\text{CsCl}]_{\text{st}} = 2 \times 10^{16} \text{ cm}^{-3}$ ,  $T = 1055 \text{ K}$  (b) and  $[\text{CsCl}]_{\text{st}} = 1 \times 10^{17} \text{ cm}^{-3}$ ,  $T = 1170 \text{ K}$  (c);  $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $f = 4 \text{ kHz}$ .

the molecules of halogen-carrier CsCl. The contributions of the remaining reactions of populating the states of the XeCl molecule are virtually offset by the contributions of reverse reactions [e.g., reactions of excitation and de-excitation of the states of  $\text{XeCl}^*$  and  $\text{XeCl}(\text{X})$  by means of electron impact], and represent no interest from the viewpoint of pumping the working laser levels.



**Figure 8.** Temporal dependences of the specific generation power:  $\gamma = 4 \times 10^6 \text{ s}^{-1}$ ,  $[\text{CsCl}]_{\text{st}} = 1 \times 10^{17} \text{ cm}^{-3}$ ,  $T = 1170 \text{ K}$  (solid curve),  $\gamma = 1 \times 10^6 \text{ s}^{-1}$ ,  $[\text{CsCl}]_{\text{st}} = 2 \times 10^{16} \text{ cm}^{-3}$ ,  $T = 1055 \text{ K}$  (dashed curve),  $\gamma = 4 \times 10^5 \text{ s}^{-1}$ ,  $[\text{CsCl}]_{\text{st}} = 5.6 \times 10^{15} \text{ cm}^{-3}$ ,  $T = 970 \text{ K}$  (dotted curve);  $[\text{Xe}] = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $f = 4 \text{ kHz}$ .

As we mentioned above, the possibility of lasing depends essentially on the reaction of dissociative attachment of electrons to the CsCl molecule. At this stage of research, it is important to conduct an experimental verification on the presence of lasing in the conditions discussed.

The required temperatures (about 1200 K) are attainable for the discharge tube designs used in [43–48]. We should note that, for example, in the case of metal vapour lasers, the characteristic values of operating temperatures may be even higher. Thus, from the engineering viewpoint, an increase in heating temperature of halogen salts up to 1200 K should not cause any significant problems.

## 4. Conclusion

Thus, using an example of the Xe–CsCl mixture, we have considered a possibility of employing nontraditional halogen-containing mixtures of alkali metal halides with rare gases to obtain laser generation on exciplex molecules.

The use of a nontraditional mixture leads to a new mechanism of pumping the working levels – a binary reaction of replacing the atom or ion of an alkali metal in the molecule of alkali halide by the excited atom or ion of a rare gas, respectively. In this case, the total contribution of the traditional mechanisms to populating the exciplex molecules is negligibly small. The alternative pumping mechanism may expand the operational possibilities of the exciplex lasers.

A possibility of lasing is demonstrated in the Xe–CsCl mixture excited by means of a longitudinal repetitively pulsed discharge. The calculated values of the specific lasing energy in the pulse can attain  $0.05 \text{ J L}^{-1}$ , with the efficiency of 3% relative to the energy injected into the medium.

A possible reason for a failure in lasing or its significant suppression in a laser on the nontraditional mixture may be the reaction of dissociative attachment of electrons to the molecule of an alkali metal halide. However, according to the available data, this process is of no importance in the case of CsCl molecules.

The typical pressures in the exciplex lasers on the traditional mixtures constitute the values of about 1 atm or higher. This leads to intensification of the quenching processes, to the need for pre-ionisation and also to a rapid development of instabilities with increasing the excitation pulse repetition rates. According to the results obtained, the use of the non-traditional mixture ensures lasing at significantly lower pressures – it is quite sufficient to have a pressure of a few dozen Torr, which, in turn, may result in a substantial increase in the pulse repetition rate of generation in the exciplex lasers.

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