

PACS numbers: 42.55.Lt; 42.60.Lh
DOI: 10.1070/QE2000v030n08ABEH001791

The possibility of lasing in Ne^+Ar ionic molecules pumped by a hard ioniser

A M Boichenko, S I Yakovlenko

Abstract. The kinetic model of relaxation in the Ne – Ar – Kr mixture pumped by a hard ioniser is constructed in connection with the analysis of the possibility of lasing at the $Ne^+Ar \rightarrow NeAr^+$ transition of the inert-gas ionic exciplexes. The calculations based on the typical rates of plasma-chemical reactions demonstrate that the lasing is possible but difficult to realise: One needs high pressures (greater than 16 bar) and high pumping densities (~ 1 MW cm^{-3}). In the most favourable cases, the laser efficiency lies between 0.05 and 0.25 %.

1. Introduction

An investigation of lasing at the transitions of ionic exciplexes was initiated in Ref. [1]. The large number of emission lines in inert-gas mixtures was well explained by the transitions

$$R^+R' \rightarrow RR'^+ + \hbar\omega,$$

where the molecular states R^+R' and RR'^+ asymptotically correspond to the states $R^+ + R'$ and $R + R'^+$, respectively. Here, R, R′ are inert-gas atoms, and we assume that the ionisation potential of element R is greater than that of element R′. In this work, we consider one of the types of plasma lasers [2]. The lasing in ionic exciplexes was first proposed in Refs [3, 4]. The studies of ionic exciplexes (not exclusively of inert gases) continue [5 – 9], but no lasing has been obtained so far. If ever realised, such lasers would offer a serious advantage—a non-aggressive medium.

To determine the conditions necessary for lasing, we will consider the kinetics of the active medium formation using the example of the Ne^+Ar exciplex.

2. Selection of the molecule

The spectra of the R^+R' systems lie either in the visible spectral region — He^+Ne (409.8 and 423.6 nm), Ar^+Xe (329 – 545 nm), Ar^+Kr (635.1 nm), Kr^+Xe (459.0 nm) — or in the vacuum UV (VUV) spectral region — He^+Ar (142.6 and 145.7 nm), He^+Kr (119.3 and 127.6 nm), He^+Xe (101.5 and 113.9 nm), Ne^+Ar (213 – 224 nm), $NeKr^+$ (164 – 183 nm), Ne^+Xe (133 – 156 nm).

For our investigation, we have chosen the ionic molecule Ne^+Ar. To provide favourable lasing conditions, the energy difference between the levels R^+R' and RR'^+ should be large (see Section 3 below). However, a wavelength reduction in the VUV region results in difficulties related to the manufacturing of optical elements, in particular, the cavity mirrors. For these reasons, we selected the Ne^+Ar molecule. The emission wavelengths of this molecule, 216 and to 224 nm, are the longest among the emission wavelengths of molecules of the second group of ionic exciplexes, but the absorption of the excited medium at these wavelengths is still low [10, 11].

3. Selection of the laser mixture

We will shortly discuss the difficulties in obtaining the lasing and the possible conditions that may significantly facilitate it. From the point of view of simplicity, the binary Ne – Ar mixture would be the most convenient one. However, in this case, the only reaction that could deplete the lower level of $NeAr^+$ would be the substitution reaction

$$NeAr^+ + Ar \rightarrow Ar_2^+ + Ne.$$

The maximum rates of the reactions of this type lie in the range $10^{-10} - 10^{-9}$ cm^3 s^{-1}. On the other hand, the rate of this reaction may be negligibly small as well. In this case, lasing is impossible since the rate of the reaction that populates the upper level of Ne^+Ar

$$Ne^+ + Ar + M \rightarrow Ne^+Ar + M,$$

where M = Ne, Ar, is most probably lower than that of the reaction

$$Ar^+ + Ne + M \rightarrow NeAr^+ + M.$$

which populates the lower level of $NeAr^+$. Of course, the dissociative recombination

$$NeAr^+ + e \rightarrow Ar^{**} + Ne$$

can also deplete the lower level, but it depletes the upper level as well with approximately the same efficiency. Therefore, it seems that a binary mixture does not suit our needs.

The simplest non-binary mixture is the triple mixture Ne – Ar – Kr. In this case, the lower state is depleted in the reaction

A M Boichenko, S I Yakovlenko General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 117942 Moscow, Russia

Received 10 April 2000
Kvantovaya Elektronika **30** (8) 681 – 686 (2000)
Translated by I V Bargatin; edited by M N Sapozhnikov

$$NeAr^+ + Kr \to Kr^+ + Ne + Ar.$$

The ultimate rates of the reactions of this type are also of the order of $10^{-10} - 10^{-9}$ cm^3 s^{-1}. The principal reaction of the exciplex KrF laser

$$Ar_2^+ + Kr \to Kr^+ + 2Ar.$$

is a well-known example; its rate is 7.8×10^{-10} cm^3 s^{-1} [12]. However, some anomalies are possible here, too. For example, the rates of the reactions

$$Ne_2^+ + Ar \to Ar^+ + 2Ne,\ Ne_2^+ + Kr \to Kr^+ + 2Ne$$

are lower, than to 5×10^{-14} and 5×10^{-13} cm^3 s^{-1}, respectively [12]. These anomalies are usually not realised in three-particle reactions; therefore, the lower level can be depleted in the reactions

$$NeAr^+ + Kr + M \to Kr^+ + Ne + Ar + M.$$

According to Refs [13–16], the rates of these reactions must be of the order of several units times 10^{-30} cm^6 s^{-1}. These rate values are widely used in the models of exciplex inert-halogen lasers. Nevertheless, they appear to be overestimated by approximately an order of magnitude [17]; therefore, we will use the value 10^{-31} cm^6 s^{-1}.

The depletion reaction is most efficient when the energy difference between the levels RR'^+ and R''^+ is minimum. This is why we selected Kr rather than, for example, Xe as R''. On the other hand, if we want to make the depletion of the upper-level in the reaction

$$R^+R' + R'' + M \to R''^+ + R + R' + M,$$

inefficient, the energy difference between the R^+R' and R'' levels must be as large as possible. Note that it is here that we make use of the above mentioned requirement of a large energy separation between the levels R^+R' and RR'^+.

Consider now the conditions under which the lasing is possible. The radiative lifetime of the upper level of Ne^+Ar is $A^{-1} \approx 10$ ns. If we wish the lower-state depletion in the reaction

$$NeAr^+ + 2Kr \to Kr^+ + Ne + Ar + Kr$$

$$(k \sim 10^{-31}\ \text{cm}^6\ \text{s}^{-1})$$

to be ten times faster than the radiative decay ($k[Kr]^2 \sim 10A$), the Kr concentration should be $\sim 10^{20}$ cm^{-3}. The upper working levels are quenched by electrons in the reaction

$$Ne^+Ar + e \to NeAr^+ + e$$

with the rate k_1, which is of the order of $\sim 10^{-7}$ cm^3 s^{-1}. Therefore, if we do not want the quenching to make a substantial contribution to the spurious depletion of the upper level, the quenching rate should not be greater than the radiative-decay rate,

$$k_1 N_e \sim A,\ \ N_e \sim 10^{15}\ \text{cm}^{-3}.$$

Using this result, we can estimate the ultimate ionisation rate (see below) that leads to this electron density:

$$\nu[Kr] \sim k_d N_e^2;\ \ \nu \sim 10^3\ \text{s}^{-1}.$$

Here, $k_d \sim 10^{-7}$ cm^3 s^{-1} is the dissociative recombination rate.

The upper state is populated through the reactions

$$Ne^+ + Ar + M \to Ne^+Ar + M,$$

where M = Ne, Ar, or Kr. Assuming that the rates of theses reactions are equal, we find that the concentrations of Ne and Ar should be no less than that of Kr. Otherwise, either the production of Ne^+ ions or the efficiency of the pumping reactions will be too low. However, we should not forget about other reactions that can also take place:

$$Ne^+ + Kr + M \to Ne^+Kr + M,$$

where M = Ne, Ar, or Kr; therefore, one should have [Ar] > [Kr]. In addition, one needs [Ne] > [Ar] for the efficient production of Ne^+ ions. Thus, the final condition is

$$[Ne] > [Ar] > [Kr] \sim 10^{20}\,\text{cm}^{-3}.$$

One can see that in this case, the total pressure of the medium exceeds 10 bar.

4. Kinetic model

Below, we will consider the case when the medium is pumped by an electron beam. In the case of hard-ionisation pumping (in particular, by an electron beam), it is convenient to characterise the specific pumping power by the ionisation rate of the medium [2, 18]

$$\nu = \xi \frac{\sigma(E) j}{e},$$

where σ is the ionisation cross section of atoms; j is the current density in the beam; e is the electron charge; and $\xi \approx 2$ is the factor that takes into account the contribution of the cascade ionisation. The excitation rate of atoms from the ground state is defined in a similar fashion. The specific input power is related to the ionisation rates by the expression

$$W = \sum_i \nu_i N_i E_{\text{pair}}^i,$$

where W is the specific power input into the medium; N_i are the concentrations of ionised medium atoms, and E_{pair}^i is the energy that a beam electron spends on the creation of electron-ion pairs from the i-th kind of atoms as it propagates in the medium until the full stop. The pumping pulse duration $\tau_{1/2}$ at half the maximum was chosen to exceed 200 ns.

In the considered model, we solved a self-consistent system of equations for the time dependence of the electron and gas temperatures as well as the concentrations of free electrons and the reagents Ne^+Ar, $NeAr^+$, Ne^+Kr, $NeKr^+$, Ar^+Kr, $ArKr^+$, Kr^*, Kr^{**}, Kr_2^*, Kr_2^{**}, Kr^+, Kr_2^+, Ar^+, Ar_2^+, Ar^*, Ar^{**}, Ar_2^*, Ar_2^{**}, Ne^+, Ne_2^+, Ne^*, Ne^{**}, and Ne_2^*. The asterisks here denotes the set of the lower excited electron states [10, 11, 18–21]. The constructed model is based on the kinetic reactions that we previously used to model the XeCl

(the Ne – Ar – HCl mixture) [18,19] and ArF (the Ne – Ar – F_2 mixture) [10, 11] lasers, the emission of the binary mixture Kr – Xe [20], and the exciplex KrCl lamp (the Ne – Kr – HCl mixture) [21, 22]. Therefore, we will consider here the reactions related to the kinetics of molecular ions. The system of equations ($\sim$ 110 reactions) was numerically solved using the PLAZER program package [18].

The rates of the reactions

$$Ne^+ + 2Ne \rightarrow Ne_2^+ + Ne, \quad Ar^+ + 2Ar \rightarrow Ar_2^+ + Ar,$$

$$Kr^+ + 2Kr \rightarrow Kr_2^+ + Kr$$

are known sufficiently well: They are 6×10^{-32}, 2.5×10^{-31}, and 2.5×10^{-31} cm^6 s^{-1}, respectively, and these values were used in the model. They agree well with the rates derived from the Thompson theory. The reactions of the type

$$R_1^+(R_2^+) + R_2(R_1) + R_3 \rightarrow R_1^+R_2(R_1R_2^+) + R_3,$$

where R_1, R_2, and R_3 can stand for any of Ne, Ar, Kr provided that all three elements are not identical, were assumed to have the rate 10^{-31} cm^6 s^{-1}. We do not see any reason for strong deviations of these rates from the given values. The exact calculation of these rates is probably beyond the accuracy of the used model; this is why the rates of these reactions were assumed to be equal. We used the same values of the rates for reactions

$$NeAr^+ + Kr + M \rightarrow Kr^+ + Ne + Ar + M.$$

The rate of the quenching of ionic excimers by electrons

$$Ne^+Ar + e \rightarrow NeAr^+ + e, \quad Ne^+Kr + e \rightarrow NeKr^+ + e,$$

$$Ar^+Kr + e \rightarrow ArKr^+ + e$$

was chosen to be 2×10^{-7} cm^3 s^{-1}. The rates of the dissociative recombination of the ions Ne^+Ar, $NeAr^+$, Ne^+Kr, $NeKr^+$, Ar^+Kr, and $ArKr^+$ were $T_e^{-1/2}10^{-7}$ cm^3 s^{-1}, where T_e is expressed in electron-volts.

For the absorption cross sections of different components at the wavelengths 210 – 220 nm, we used the values from our pa-pers on modelling the ArF (193 nm) [10, 11] and KrCl lasers (222 nm). The cross sections of the ions Ne_2^+, Ar_2^+, and Kr_2^+ were chosen from Ref. [23]; the cross sections of R^+R' and RR'^+ were assumed to be close to the greatest among the cross sections for R_2^+ and $R_2'^+$.

The FWHM of the emission lines of the Ne^+Ar molecule $\Delta\lambda$ is approximately 2 nm [8]. The cross section for the induced emission was calculated from the expression

$$\sigma = \frac{\lambda^2}{4\Delta\omega}A.$$

Here, λ is the emission wavelength; $\Delta\omega$ is the emission band linewidth; A is the probability of the radiative decay of the Ne^+Ar molecule. Table 1 shows the discussed kinetic reactions of the ionic molecular states, their rates, and the absorption cross sections for the components of the excited medium.

If we neglect the divergence of the laser beam, the output energy of the laser is well described by the zero-dimension approximation, in which the inverse of the lifetime of a cavity photon is given by

Table 1. Reactions involving ions used in the simulations, their rates, and the absorption cross sections for different components of the excited medium; n is the number of atoms involved in the reaction. The electron and gas temperatures in formulas should be expressed in electron-volts.

No	Reaction	Rate/cm^{n-1} s^{-1}	σ/cm^2
Radiative decay			
1	$Ne^+Ar \rightarrow NeAr^+$	10^8	–
2	$Ne^+Kr \rightarrow NeKr^+$	10^8	–
3	$Ar^+Kr \rightarrow ArKr^+$	10^8	–
Electron quenching			
4	$Ne^+Ar + e \rightarrow NeAr^+ + e$	2×10^{-7}	–
5	$Ne^+Kr + e \rightarrow NeKr^+ + e$	2×10^{-7}	–
6	$Ar^+Kr + e \rightarrow ArKr^+ + e$	2×10^{-7}	–
Dissociative recombination			
7	$NeAr^+ + e \rightarrow Ar^{**} + Ne$	$10^{-7}T_e^{-1/2}$	–
8	$NeKr^+ + e \rightarrow Kr^{**} + Ne$	$10^{-7}T_e^{-1/2}$	–
9	$ArKr^+ + e \rightarrow Kr^{**} + Ar$	$10^{-7}T_e^{-1/2}$	–
10	$Ne^+Ar + e \rightarrow Ne^{**} + Ar$	$10^{-7}T_e^{-1/2}$	–
11	$Ne^+Kr + e \rightarrow Ne^{**} + Kr$	$10^{-7}T_e^{-1/2}$	–
12	$Ar^+Kr + e \rightarrow Ar^{**} + Kr$	$10^{-7}T_e^{-1/2}$	–
13	$Ne_2^+ + e \rightarrow Ne^* + Ne$	$3.7 \times 10^{-8}T_e^{-0.43}$	–
14	$Ar_2^+ + e \rightarrow Ar^{**} + Ar$	$6.5 \times 10^{-8}T_e^{-0.67}$	–
15	$Kr_2^+ + e \rightarrow Kr^{**} + Kr$	$1.6 \times 10^{-7}T_e^{-1/2}$	–
Creation of molecular ions in three-particle reactions			
16	$Ne^+ + 2Ne \rightarrow Ne_2^+ + Ne$	$3.9 \times 10^{-33}T_g^{-0.75}$	–
17	$Ar^+ + 2Ar \rightarrow Ar_2^+ + Ar$	$1.62 \times 10^{-32}T_g^{-0.75}$	–
18	$Kr^+ + 2Kr \rightarrow Kr_2^+ + Kr$	$1.62 \times 10^{-32}T_g^{-0.75}$	–
19	$Ne^+ + 2Ar \rightarrow Ne^+Ar + Ar$	10^{-31}	–
20	$Ne^+ + Ne + Ar \rightarrow Ne^+Ar + Ne$	10^{-31}	–
21	$Ne^+ + Ne + Ar \rightarrow Ne_2^+ + Ar$	10^{-31}	–
22	$Ne^+ + Ne + Kr \rightarrow Ne_2^+ + Kr$	10^{-31}	–
23	$Ne^+ + Kr + Ne \rightarrow Ne^+Kr + Ne$	10^{-31}	–
24	$Ne^+ + 2Kr \rightarrow Ne^+Kr + Kr$	10^{-31}	–
25	$Ne^+ + Kr + Ar \rightarrow Ne^+Kr + Ar$	10^{-31}	–
26	$Ne^+ + Kr + Ar \rightarrow Ne^+Ar + Kr$	10^{-31}	–
27	$Ar^+ + 2Ne \rightarrow NeAr^+ + Ne$	10^{-31}	–
28	$Ar^+ + Ne + Ar \rightarrow NeAr^+ + Ar$	10^{-31}	–
29	$Ar^+ + Ne + Ar \rightarrow Ar_2^+ + Ne$	10^{-31}	–
30	$Ar^+ + Ne + Kr \rightarrow NeAr^+ + Kr$	10^{-31}	–
31	$Ar^+ + Kr + Ar \rightarrow Ar_2^+ + Kr$	10^{-31}	–
32	$Ar^+ + Kr + Ar \rightarrow Ar^+Kr + Ar$	10^{-31}	–
33	$Ar^+ + 2Kr \rightarrow Ar^+Kr + Kr$	10^{-31}	–
34	$Kr^+ + 2Ne \rightarrow NeKr^+ + Ne$	10^{-31}	–
35	$Kr^+ + Ne + Ar \rightarrow NeKr^+ + Ar$	10^{-31}	–
36	$Kr^+ + Ne + Ar \rightarrow ArKr^+ + Ne$	10^{-31}	–
37	$Kr^+ + 2Ar \rightarrow ArKr^+ + Ar$	10^{-31}	–
38	$Kr^+ + Ne + Kr \rightarrow NeKr^+ + Kr$	10^{-31}	–
39	$Kr^+ + Ne + Kr \rightarrow Kr_2^+ + Ne$	10^{-31}	–
40	$Kr^+ + Kr + Ar \rightarrow ArKr^+ + Ne$	10^{-31}	–
41	$Kr^+ + Kr + Ar \rightarrow Kr_2^+ + Ar$	10^{-31}	–

(to be continued)

(continued from page 583)

No	Reaction	Rate/cm^{n-1} s^{-1}	σ/cm^2
	Recharging and capture		
42	$Ne^+ + Ar \to Ar^+ + Ne$	10^{-11}	–
43	$Ne_2^+ + Ar \to Ar^+ + 2Ne$	5×10^{-14}	–
44	$Ne^+ + Kr \to Kr^+ + Ne$	10^{-11}	–
45	$Ne_2^+ + Kr \to Kr^+ + 2Ne$	5×10^{-13}	–
46	$NeKr^+ + Kr \to Kr_2^+ + Ne$	10^{-11}	–
47	$Ne_2^+ + Ne + Kr \to Kr^+ + 3Ne$	10^{-31}	–
48	$Ne_2^+ + Ar + Kr \to Kr^+ + 2Ne + Ar$	10^{-31}	–
49	$Ne_2^+ + 2Kr \to Kr^+ + 2Ne + Kr$	10^{-31}	–
50	$Ne_2^+ + Ne + Ar \to Ar^+ + 3Ne$	10^{-31}	–
51	$Ne_2^+ + 2Ar \to Ar^+ + 2Ne + Ar$	10^{-31}	–
52	$Ar_2^+ + Ne + Kr \to Kr^+ + Ne + 2Ar$	10^{-31}	–
53	$Ar_2^+ + Ar + Kr \to Kr^+ + 3Ar$	10^{-31}	–
54	$Ar_2^+ + 2Kr \to Kr^+ + 2Ar + Kr$	10^{-31}	–
55	$ArKr^+ + Kr \to Kr_2^+ + Ar$	3.2×10^{-10}	–
56	$Ar_2^+ + Kr \to Kr^+ + 2Ar$	7.5×10^{-10}	–
57	$Ar^+ + Kr \to Kr^+ + Ar$	3×10^{-11}	–
58	$NeAr^+ + Ar \to Ar_2^+ + Ne$	10^{-11}	–
59	$NeAr^+ + Kr \to Kr^+ + Ne + Ar$	10^{-10}	–
60	$NeAr^+ + 2Kr \to Kr^+ + Ne + Ar + Kr$	10^{-31}	–
61	$NeAr^+ + Kr + Ne \to Kr^+ + 2Ne + Ar$	10^{-31}	–
62	$NeAr^+ + Kr + Ar \to Kr^+ + Ne + 2Ar$	10^{-31}	–
	Photoabsorption		
63	$Ne^+Ar + \hbar\omega \to NeAr^+ + 2\hbar\omega$	–	1.5×10^{-16}
64	$NeAr^+ + \hbar\omega \to Ne^+Ar$	–	1.5×10^{-16}
65	$Ne_2^+ + \hbar\omega \to Ne^+ + Ne$	–	0.7×10^{-17}
66	$Ne^+Ar + \hbar\omega \to Ne^+ + Ar$	–	0.5×10^{-17}
67	$Ne^+Kr + \hbar\omega \to Ne^+ + Kr$	–	3.3×10^{-18}
68	$NeAr^+ + \hbar\omega \to Ne + Ar^+$	–	0.5×10^{-17}
69	$NeKr^+ + \hbar\omega \to Ne + Kr^+$	–	3.3×10^{-18}
70	$Ar^+Kr + \hbar\omega \to Kr + Ar^+$	–	10^{-18}
71	$ArKr^+ + \hbar\omega \to Ar + Kr^+$	–	10^{-18}
72	$Ar_2^+ + \hbar\omega \to Ar + Ar^+$	–	2×10^{-18}
73	$Kr_2^+ + \hbar\omega \to Kr + Kr^+$	–	3×10^{-19}
74	$Ar^* + \hbar\omega \to Ar^+ + e$	–	10^{-19}
75	$Ar_2^* + \hbar\omega \to Ar_2^+ + e$	–	0.6×10^{-18}
76	$Ar_2^{**} + \hbar\omega \to Ar^+ + Ar + e$	–	0.4×10^{-17}
77	$Ar^{**} + \hbar\omega \to Ar^+ + e$	–	3×10^{-18}
78	$Ne_2^* + \hbar\omega \to Ne_2^+ + e$	–	10^{-18}
79	$Kr_2^* + \hbar\omega \to Kr_2^+ + e$	–	10^{-18}
80	$Kr^* + \hbar\omega \to Kr^+ + e$	–	3×10^{-19}
81	$Kr^{**} + \hbar\omega \to Kr^+ + e$	–	0.5×10^{-17}
82	$Ne^* + \hbar\omega \to Ne^+ + e$	–	10^{-19}
83	$Ne^{**} + \hbar\omega \to Ne^+ + e$	–	0.4×10^{-18}

$$\gamma = (c/2l) \ln r^{-1},$$

where c is the speed of light; l is the length of the excited medium, and r is the reflectivity of the cavity.

5. Discussion

Since the optimal output energy and the laser efficiency were always realised for the same values of the variable parameters, we will refer to them simply as optimal parameters.

5.1. Threshold characteristics

Since the lasing has not so far been realised experimentally, the calculation of the exact threshold parameters is meaningless. We used in our calculations the output specific energy of the laser radiation equal to $E = 0.1$ J litre^{-1}. The typical specific energy of the emission in exciplex inert-halogen lasers, which are currently the most powerful UV lasers, amounts to several joules per litre. For the ionisation rate $v = 10^3$ s^{-1} and the laser efficiency $\eta = 5.33 \times 10^{-2}$ %, the specific output energy $E = 0.12$ J litre^{-1} corresponds to $[Kr] = 5 \times 10^{19}$ cm^{-3}, $[Ar] = 10^{20}$ cm^{-3}, $[Ne] = 3 \times 10^{20}$ cm^{-3}, the FWHM pulse duration $\tau_{1/2} = 200$ ns, and the optimal $\gamma = 10^7 - 2 \times 10^7$ s^{-1}. If [Kr], [Ar], or $\tau_{1/2}$ are reduced by a factor of two, E drastically falls off to $\sim 10^{-2}$ J litre^{-1}. If [Ne] is reduced to 2×10^{20} cm^{-3}, E decreases to 0.05 – 0.07 J litre^{-1}; at $[Ne] = 10^{20}$ cm^{-3}, one has $E = 0.001 - 0.004$ J litre^{-1}. By optimising the cavity in the latter case, one can achieve $E \sim 0.01$ J litre^{-1}.

5.2. Dependence of E on the pump pulse duration

With an increase in the duration of the pump pulse, the specific output energy remains unsaturated until $\tau_{1/2} = 1$ μs. After the rapid increase in the region from $\tau_{1/2} > 200$ ns to $\tau_{1/2} = 1$ μs, E grows linearly (Table 2). These calculations were performed for $\gamma = 2 \times 10^7$ s^{-1}, which corresponds to the optimal energy and efficiency for $\tau_{1/2} = 200$ ns.

Table 2. Dependences of the specific emitted energy E and the laser efficiency η on the pump pulse duration for $\gamma = 2 \times 10^7$ s^{-1}, which is optimal for the FWHM pump pulse duration $\tau_{1/2} = 200$ ns; $[Ne] = 3 \times 10^{20}$ cm^{-3}; $[Ar] = 10^{20}$ cm^{-3}, and $[Kr] = 5 \times 10^{19}$ cm^{-3}.

$\tau_{1/2}$/ns	E/J l^{-1}	$\eta/10^{-2}$ %
200	0.118	5.33×10^{-2}
300	0.236	7.12×10^{-2}
400	0.358	8.11×10^{-2}
500	0.483	8.84×10^{-2}
1000	1.14	$10.5 \cdot 10^{-2}$

Table 3. Dependences of the specific emitted energy, the laser efficiency, and the optimal γ on the argon concentration for $\tau_{1/2} = 200$ ns, $[Ne] = 3 \times 10^{20}$ cm^{-3}, and $[Kr] = 5 \times 10^{19}$ cm^{-3}.

$[Ar]/10^{20}$ cm^{-3}	$\gamma/10^7$ s^{-1}	E/J litre^{-1}	$\eta/10^{-2}$%
1	2	0.118	5.33
2	2	0.349	12.2
4	5	0.479	11.5
8	2	0.224	3.32

5.3. Optimal mixtures

For $\tau_{1/2} = 200$ ns and $[Kr] = 5 \times 10^{19}$ cm^{-3}, the optimal E and η increase with increasing Ar and Ne concentrations provided that their ratio [Ar]/[Ne] lies in the range from 1/3 to unity (Tables 3 and 4). For larger values of [Kr], the optimal ratio of [Ar] to [Ne] remains the same with increasing pressure. However, if [Ar] and [Ne] are fixed, the out-put energy and the laser efficiency are lower than for $[Kr] = 5 \times \times 10^{19}$ cm^{-3}. To determine the optimal ratios $\beta = [Ar]/[Ne]$, we performed the optimisation over γ for each value of β. Table 5 shows the calculated gain.

Table 4. Dependences of the specific emitted energy, the laser efficiency, and the optimal γ for various neon concentrations, near-optimal values of $\beta = [Ar]/[Ne]$, and $\tau_{1/2} = 200$ ns.

$[Kr]/10^{20}$ cm^{-3}	$[Ne]/10^{20}$ cm^{-3}	$[Ar]/10^{20}$ cm^{-3}	E/J l^{-1}	$\eta/10^{-2}$ %	$\gamma/10^7$ s^{-1}
	3	1–4	0.118–0.479	5.33–11.5	2–5
0.5	6	2–4	0.543–1.16	13.5–21.9	5
	12	8	2.57	25.4	10
	3	1–4	$< 10^{-2}$	–	–
1.0	6	2–4	0.247–0.66	5.59–11.6	2–5
	12	8	1.66	15.6	10
	3	1–4	–	–	–
2.0	6	2–4	0.0612	0.942	2
	12	8	0.54	4.73	5

5.4. Dependence of the emission characteristics on the ionisation rate (the energy input)

For the concentrations $[Kr] = 5 \times 10^{19}$ cm^{-3}, $[Ar] = 10^{20}$ cm^{-3}, and $[Ne] = 3 \times 10^{20}$ cm^{-3} discussed above in Section 5.1 and for $\tau_{1/2} = 200$ ns, the optimal ionisation rate with respect to krypton is $\nu_{Kr} = 10^3$ s^{-1} (Table 6). When calculating these dependences, we performed optimisation over γ for each fixed value of ν_{Kr}. The existence of the optimum over ν_{Kr} is caused by the increase in the quenching rate (Section 3) of the upper laser levels by electrons at $\nu > 10^3$ s^{-1}.

Table 5. Dependence of the gain on the mixture pressure for near-optimal $\beta = [Ar]/[Ne]$.

β	$[Ne]/10^{20}$ cm^{-3}	$[Ar]/10^{20}$ cm^{-3}	$\kappa/10^{-3}$ cm^{-1}
1/3	3	1	2.26
2/3	3	2	3.72
2/3	6	4	6.69
2/3	12	8	10.2

5.5. Variation in the reaction rates

If the rate of the 59th reaction (Table 1) is reduced by an order of magnitude, i.e., $k = 10^{-11}$ cm^3 s^{-1}, and the rest of the parameters are $\nu_{Kr} = 10^3$ s^{-1}, $[Kr] = 5 \times 10^{19}$ cm^{-3}, $[Ar] = 10^{20}$ cm^{-3}, and $[Ne] = 3 \times 10^{20}$ cm^{-3}, the specific energy $E \sim 0.1$ J litre^{-1} was achieved only for $\tau_{1/2} = 500$ ns ($E = 7.3 \times 10^{-2}$ J litre^{-1}, $\eta = 1.32 \times 10^{-2}$%). In this case, γ must be 5×10^6 s^{-1}.

Table 6. Dependences of the specific emitted energy, the laser efficiency, and the optimal γ on the ionisation rate ν (with respect to krypton) for $\tau_{1/2} = 200$ ns, $[Ar] = 10^{20}$ cm^{-3}, $[Ne] = 3 \times 10^{20}$ cm^{-3}, and $[Kr] = 5 \times 10^{19}$ cm^{-3} ($\nu = 10^3$ corresponds to the energy input $W = 1.1$ MW cm^{-3}).

$\nu/10^2$ s^{-1}	$E/10^{-2}$ J litre^{-1}	η (%)	$\gamma/10^7$ s^{-1}
1	< 1	–	–
3	7.11	$1.07 \cdot 10^{-1}$	0.5
10	11.8	$5.33 \cdot 10^{-2}$	2.0
30	1.21	$1.82 \cdot 10^{-3}$	0.5
100	< 1	–	–

So far, we did not discuss the quenching reactions of the type

$$Ne^+Ar + M \rightarrow \text{products}$$

where M = Ne, Ar, or Kr. In the presence of such quenching, the concentration of the [Kr] + [Ar] + [Ne] misture is bound from above by the condition

$$\sum_i K_{R_i}[R_i] \leqslant A, \tag{1}$$

where $R_1 = Kr$, $R_2 = Ar$, $R_3 = Ne$, $[Kr] \sim 5 \times 10^{19}$ cm^{-3} $< [Ar] = (1/3 - 1)[Ne]$. Condition (1) does not contradict to the threshold reagent concentrations derived in Section 5.1 if the rates k are much lower than 10^{-12} cm^3 s^{-1}.

5.6. Recommendations

It follows from the above results that, in order to achieve the specific output power $E \sim 0.1$ J litre^{-1}, the concentrations have to be rather high: $[Kr] = 5 \times 10^{19}$ cm^{-3}, $[Ar] = 10^{20}$ cm^{-3}, and $[Ne] = 3 \times 10^{20}$ cm^{-3}. In this case, the total pressure will be at least 16.6 bar, the duration of the pump pulse $\tau_{1/2}$ must be no less than 200 ns (preferably, $\tau_{1/2} \sim 1$ μs), and γ must be in the range $5 \times 10^6 - 2 \times 10^7$ s^{-1}. Given that $\nu_{Ne} \approx 0.32\nu_{Kr}$ and $\nu_{Ar} \approx 0.76\nu_{Kr}$, the optimal ionisation rate $\nu_{Kr} = 10^3$ s^{-1} will correspond to the specific power input to the medium

$$W = \sum_i E_{R_i} \nu_{R_i}[R_i] = 1.1 \text{ MW cm}^{-3}.$$

For example, if the electrons of the pump beam have the energy 200 keV, the current density of the electron beam should be

$$j \approx e\nu_{Ar}/2\sigma_{Ar} = 61 \text{ A cm}^{-2}.$$

The transit of such electrons in the neon at a pressure of 10 bar is $R_{Ne} \simeq 6.5$ cm, and in the argon at a pressure of 3.7 bar, $R_{Ar} = 10$ cm [24]. Under conditions, considered, the electron transit is

$$R \sim \left(\frac{1}{R_{Ne}} + \frac{1}{R_{Ar}}\right)^{-1} = 3.9 \text{ cm}.$$

Because this transit occurs along the electron paths, the width of the excited region will be 2–3 cm; i. e., only the transverse pumping of the medium can be realised.

6. Conclusions

We have theoretically analysed the necessary conditions required for lasing in ionic exciplexes of inert gases. The $Ne^+Ar \rightarrow NeAr^+$ transition was selected as the most promising for lasing. We found the conditions that are most favourable for lasing. The kinetic model was constructed by using the reaction rates that were typical for the reactions involved. However, the reaction rates can obviously be greater or lower than these typical values. For example, the rates of reactions 19, 20, and 26 (Table 1), which result in the production of a Ne^+Ar molecule, can be lower than 10^{-31} cm^3 s^{-1}, while the rates of reactions 27, 28, and 30, which result in the production of a $NeAr^+$ molecule, can be greater than 10^{-31} cm^3 s^{-1}. This may be unfavourable for lasing. Moreover, we assumed that the quenching reactions of the upper laser level

$$Ne^+Ar + Kr + M \rightarrow Kr^+ + Ne + Ar + M,$$

$$Ne^{+}Ar + M \rightarrow \text{products}$$

(M = Ne, Ar, or Kr) as well as some other reactions are not important for the considered kinetics.

It may happen that the reaction rates will deviate in the direction unfavourable for lasing and (or) the above quenching reactions of the upper laser level will be significant. In this case, lasing may be completely infeasible. On the other hand, even under the most favourable conditions, an appreciable lasing can be realised only at pressures exceeding 16.6 bar. In this case, the laser efficiency η corresponding to $E = 0.1 - 2.5$ J litre^{-1} is as small as $0.05 - 0.25\%$.

References

1. Tanaka Y, Yoshino K, Freeman D E *J. Chem. Phys.* **62** 4484 (1975)
2. Gudzenko L I, Yakovlenko S I *Plasma Lasers* (Moscow: Atomizdat, 1978)
3. Basov N G, Voitik M G, Zuev V S, Kutakhov V P *Kvantovaya Electron. (Moscow)* **12** 2204 (1985) [*Sov. J. Quantum Electron.* **15** 1455 (1985)]
4. Basov N G, Voitik M G, Zuev V S, Kutakhov V P *Kvantovaya Electron. (Moscow)* **12** 2213 (1985) [*Sov. J. Quantum Electron.* **15** 1461 (1985)]
5. Basov N G, Voitik M G, Zuev V S, Klementov A D, Kutakhov V P, Pendyur S A *Kvantovaya Electron. (Moscow)* **14** 185 (1987) [*Sov. J. Quantum Electron.* **17** 106 (1985)]
6. Scumann M, Langhoff H *J. Chem. Phys.* **101** 4769 (1994)
7. Tishler H, Delaporte P, Fontaine B, Sentis M L *IEEE J. Sel. Topics in Quantum Electron.* **1** 877 (1995)
8. Hure L, Robert E, Cachoncinlle C, Pouvesle J M, Wieser J, Ulrich A *Proc. VIII Int. Symp. Sci. Technol. Light Sources* (Greitswald: INP, 1998)
9. Xing D, Wang Q, Tan S, Ueda K *Appl. Phys. Lett.* **71** 2584 (1997)
10. Boichenko A M, Derzhiev V I, Yakovlenko S I *Laser Phys.* **2** 210 (1992)
11. Boichenko A M, Derzhiev V I, Zhidkov A G, Yakovlenko S I *Kvantovaya Electron. (Moscow)* **19** 486 (1992) [*Sov. J. Quantum Electron.* **22** 444 (1992)]
12. Rhodes C K (Ed.) *Excimer Lasers* (Berlin: Springer-Verlag, 1984)
13. Lee F W, Collins C B, Waller R A *J. Chem. Phys.* **65** 1605 (1976)
14. Collins C B, Lee F W *J. Chem. Phys.* **68** 1391 (1978)
15. Collins C B, Lee F W *J. Chem. Phys.* **71** 184 (1979)
16. Collins C B, Lee F W *J. Chem. Phys.* **72** 5381 (1980)
17. Boichenko A M, Karelin A V, Yakovlenko S I *Laser Phys.* **5** 80 (1995)
18. Yakovlenko S I (Ed.) *Trans. Gen. Phys. Inst. RAS* **21** 44 (1989) (in Russian)
19. Boichenko A M, Derzhiev V I, Zhidkov A G, Yakovlenko S I *Kvantovaya Electron. (Moscow)* **16** 278 (1989) [*Sov. J. Quantum Electron.* **19** 182 (1989)]
20. Boichenko A M, Yakovlenko S I *Kvantovaya Electron. (Moscow)* **19** 1172 (1992) [*Sov. J. Quantum Electron.* **22** 1094 (1992)]
21. Boichenko A M, Skakun V S, Sosnin E A, Tarasenko V F, Yakovlenko S I *Kvantovaya Electron. (Moscow)* **23** 344 (1996) [*Quantum Electron.* **26** 336 (1996)]
22. Boichenko A M, Skakun V S, Sosnin E A, Tarasenko V F, Yakovlenko S I *Laser Phys.* **10** 540 (2000)
23. Wadt W R *J. Chem. Phys.* **73** 3915 (1980)
24. Stopping powers for electrons and positrons *Int. Commission on Radiation Units and Measurements (ICRU), rep. 37.7910* (Woodmont Avenue, Bethesda, MD, 20814 USA, 1984)