Excimer lasers

A. V. Eletskiĭ

I. V. Kurchatov Institute of Atomic Energy, Academy of Sciences of the USSR, Moscow
Usp. Fiz. Nauk **125**, 279–314 (June 1978)

A review is given of the available information on the active media of excimer lasers and specific examples of such lasers are considered. These utilize transitions between molecular terms, the lower of which is repulsive. An analysis of the elementary processes resulting in the excitation of excimer lasers is used to determine their optimal parameters.

PACS numbers: 42.55.Hq

CONTENTS

INTRODUCTION

The class of pulsed gas lasers, called excimer lasers, is a relative newcomer to quantum electronics. The first papers were published in 1970 and over 90% of the literature on excimer lasers has appeared in 1975–1977. These lasers utilize transition between two terms of a molecule, the lower of which is repulsive and is usually formed by atoms in the ground state. The upper term of a laser transition has a potential with a minimum. Molecules of this kind, existing only in an excited state, are known as excimers and hence the name of this class of lasers. In the case of a favorable relative position of the upper and lower terms (Fig. 1), population inversion may be established corresponding to electronic transitions in excimer molecules.

This mechanism of population inversion was first realized by Basov *et al.*[1,2] in liquid xenon producing transitions between excited, metastable, and ground (repulsive) terms of the Xe_2 molecule. Subsequently, stimulated emission from molecular xenon was obtained[3] in a dense gas kept at pressures of tens of atmospheres. The interest in excimer lasers increased greatly beginning from 1975 when, on the one hand, Velazco and Setser[4] showed that excited excimer molecules of inert-gas monohalides formed very readily as a result of quenching of metastable species of an inert gas by halogen-containing molecules and, on the other, the first high-power excimer lasers[5] were constructed, giving output energies up to 5 J and efficiencies[1)] of conversion of input energy amounting to 2–3%. According to the current information, excimer lasers are now capable of emitting about 100 J in 50 nsec[6] at wavelengths of 2484 or 1933 Å and conversion efficiencies in excess of 3%. The record value of the efficiency reported for an excimer laser, characterized by a lower output energy, is 23%.[7]

Such high output parameters of excimer lasers are due to a fortunate combination of a number of physical factors which favor, on the one hand, efficient conversion of the input energy into the energy of excited excimer molecules and, on the other, effective utilization of these molecules in the emission of laser radiation. Among these factors one should mention in the first place a characteristic mechanism of acquisition of the energy by the active medium. In electron-beam excitation of lasers of this type a considerable proportion of the energy entering a dense gas is expended in the formation of free electrons. The subsequent neutralization of electrons is the result of their dissociative recombination with molecular ions formed in the gas

[1)] In this case the efficiency, defined as the ratio of the laser radiation to the energy input to the active medium, represents the efficiency of conversion of the supplied energy and is not related directly to the energy parameters of the laser system as a whole.

 0038-5670/78/060502-20$01.10

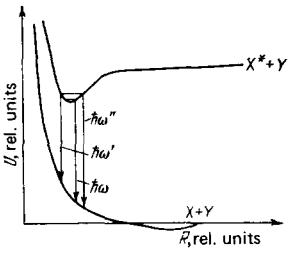

FIG. 1. Characteristic terms of an excimer molecule suitable for the operation of an excimer laser.

and this produces atoms in one or few excited states. The number of such atoms is close to the number of the electrons formed initially. The energy of these atoms is subsequently used to form excited excimer molecules. One may expect this method of formation of large amounts of excited atoms to be promising also in developing other new lasers. The mechanism of pumping ultraviolet lasers, based on the dissociative recombination of molecular ions, was first considered by Efremenkova and Smirnov.[8]

Another favorable factor ensuring high output parameters of excimer lasers is associated with the nature of the energy terms of excimer molecules (Fig. 1). The repulsive nature of the lower term is responsible for its effective emptying and for the absence of the absorption of laser radiation in the active medium, whereas the position of this lower term near the ground electronic state ensures very high (exceeding 0.9) values of the quantum efficiency of these lasers, defined as the ratio of the energy of the laser photons to the energy of the upper laser level.

The parameters of excimer lasers cited above are far better than the parameters of other lasers in the visible and ultraviolet ranges considered from the point of view of possible applications in nonlinear optics and photochemistry, as possible pump sources of tunable liquid lasers, and perhaps also in laser heating experiments. The distinguishing characteristics of excimer lasers are their short wavelengths (1700–5600 Å) and wide gain profiles, which make it possible to tune the emission wavelength by a few percent. Since the laser gain is proportional to $\lambda^2/\Delta\omega$ (λ is the wavelength of the transition and $\Delta\omega$ is the width of a spontaneous emission line), the above circumstances account for the difficulties which have been encountered in the attainment of the threshold gain in the active media of excimer lasers. For the same reasons, stimulated emission from excimer lasers is observed only at high concentrations of excited molecules, i.e., when a considerable amount of energy is introducted into the active medium in a very short time (of the order of 10^{-7} sec or less). Suitable methods of introducing energy in this way and also convenient sources have been developed relatively recently and this accounts for the "lag" in the development of excimer lasers, compared with those of other types. The present review is concerned with the processes which occur in the active media of excimer lasers. The information on these processes is used to estimate the optimal parameters of excimer lasers. An analysis is made of the experiments on excimer lasers and prospective future developments are discussed. The review covers the work published up to the middle of 1977.

1. TERMS OF EXCIMER MOLECULES

The possibility of laser action as a result of transitions in excimer molecules is based on the specific positions of the potential energy curves of the ground and excited state of a quasimolecule which is formed on close approach of atoms. It should be pointed out that our knowledge of the potential curves of quasimolecules is realtively poor: this is due to, on the one hand, insufficient amount of work done of the emission spectra of these molecules and, on the other, the extreme difficulty in the determination of the absorption spectra of such molecules. Nevertheless, a certain amount of information of this kind, obtained both experimentally and by calculation, is available in the literature and is continuously being increased. Figure 2 shows the terms of some excimer molecules which are used as the active media. Although these data are subject to some error, they illustrate quite clearly the operation principle and capabilities of excimer laser systems.

The most reliable data on the parameters of the laser term of excimer molecules are available for dimers composed of two identical atoms of inert gases. Fairly full information on these molecules can be found, for example, in Smirnov's monograph.[12] Table I gives the results of asymptotic calculations of these parameters, taken from this monograph.[12] These data are in agreement to within 10% with the results obtained by numerous other authors. It is clear from Table I that the depth of the potential well for inert gas molecules is considerably less than the thermal energy corresponding to room temperature so that these molecules do not form under laboratory conditions.

The relationship between the nature of the terms of such a quasimolecule and the principle of population inversion of excimer lasers can be followed most clearly in the case of the molecules composed of halogen and inert gas atoms. The terms of these molecules have certain general distinguishing features. The close approach of an inert gas atom to a halogen atoms in the $^2P_{3/2}$ state may result in the formation of one of two quasimolecules distinguished by the projection of the electron angular momentum on the axis joining the nuclei. This corresponds to two terms $^2\Pi_{3/2}$ and $^2\Sigma_{1/2}$, which are characterized by weak energy dependences at large internuclear distances and a strong short-range (exchange) repulsion over distances smaller than or of the order of the atomic size. In the case when a halogen atom is in a fine-structure state $^2P_{1/2}$, its approach to an inert gas atom produces a term $^2\Pi_{1/2}$, which is basically similar to the term $^2\Sigma_{1/2}$.

The question whether bound states of these molecules exist in the ground electronic state is difficult to answer because the depth of the potential well (if such a well exists) may in this situation be very small. In any case, stable dimers of this type have not been observed at room temperature although the existence of bound states of some dimers is suggested both by theoretical estimates[11] and spectroscopic observations.[13-15] According to these observations, bound states exist in the case of the molecules of XeF[13] (well depth ≈770 cm^{-1}), XeCl[13]

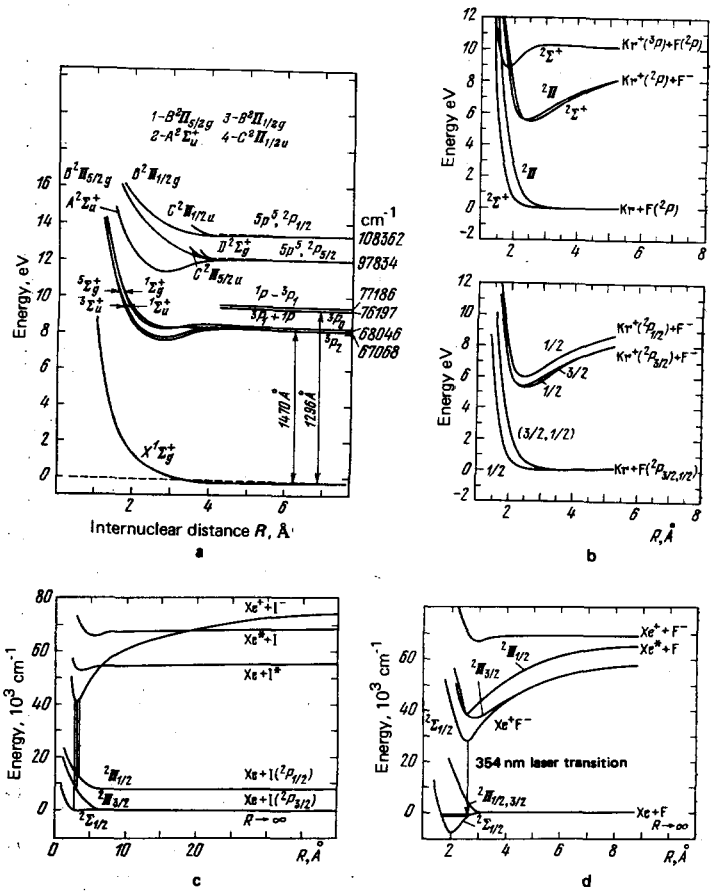

FIG. 2. Terms of some excimer molecules calculated by different authors: a) Xe_2;[9] b) KrF[10] (upper part of the figure represents calculations ignoring the spin–orbit interaction, the lower represents calculations with correction for this interaction); c) XeI;[11] d) XeF.[11]

(well depth ≈ 255 cm^{-1}), and KrF.[16]

The upper term of these laser transitions corresponds to an excited molecule with an ionic type of binding. This molecule form when an inert gas atom and a negative halogen ion come together. At large distances the interaction is the Coulomb attraction, but on closer approach this changes to the exchange repulsion of electrons and Coulomb repulsion of the nuclei. As a result, it is found that this term has a fairly deep potential well whose depth U_0 is related to the equilibrium internuclear distance R_0 by the approximate relationship

$$U_0 \approx \frac{e^2}{R_0}. \qquad (1)$$

Usually the equilibrium internuclear distance is $R_0 \sim 5$ a.u. and, therefore, the binding energy of the exicted molecule is 4–6 eV less than the energy of electrons in a negative halogen ion. This means that the minimum energy of a bound molecule is always less than the energy of R* + X and R + X* atomic systems (X and R are, respectively, halogen and inert gas atoms, and the asterisk denotes an excited atom). It follows from these considerations that an inert gas + halogen system with a large number of excited atoms provides thermodynamically favorable conditions for the formation of excited molecules with the ionic type of binding. These molecules may become radiatively deexcited, dropping down to a lower repulsive (or bound but characterized by a low binding energy) term.

TABLE I. Parameters of the interaction potential of two identical inert gas atoms calculated by an asymptotic method.[12]

Atoms	He – He	Ne – Ne	Ar – Ar	Kr – Kr	Xe – Xe
Equilibrium internuclear distance, Å	2.5	2.7	3.0	3.4	3.7
Depth of attractive potential well, °K	10.3	29	130	150	180

TABLE II. Comparison of parameters of alkali metal halides and excimer molecules of inert gas monohalides.[13*]

Molecules \ Parameters of molecules	ω_e, cm^{-1}	$\omega_e x_e$, cm^{-1}	R_e, Å	D_e, cm^{-1}
KrF	310	1.21	2.27	44 670
$^{85}Rb\,^{19}F$	373	1.80	2.27	46 750
XeF	309	1.52	2.49	42 790
$^{133}Cs\,^{19}F$	353	1.62	2.35	45 660
XeCl	195	0.54	2.94	36 540
$^{133}Cs\,^{35}Cl$	214	0.74	2.91	39 300
XeBr	120	0.23	2.96	34 700
$^{133}Cs\,^{79}Br$	150	0.36	3.07	38 000
XeI	112	0.24	3.31	32 920
$^{133}Cs\,^{127}I$	119	0.25	3.35	35 380

*The terms of the NeF excimer, whose radiation at the ~1080 Å wavelength is ≈ 30 Å wide, were calculated recently.[86]

At moderate temperatures and pressures the concentration of quasimolecules with internuclear distances corresponding to the minimum of the upper term in the normal state, is relatively low. Therefore, in a fairly wide range of parameters of the active medium and of pumping we may expect a population inversion between the upper and lower terms.

An analysis of the terms of excited molecules composed of inert gas and halogen atoms is made easier by the close analogy between such molecules and those composed of alkali metal and halogen atoms. In fact, the s valence electron in an excited inert gas atom is characterized by a binding energy close to the binding energy of a valence electron in that alkali metal atom which follows the inert gas atom in the periodic table. Since the principal features of the wave function of the weakly bound electron in an excited atom are governed by its binding energy,[12] the various parameters of these two types of molecule are very similar. The similarity is illustrated in Table II, where the parameters of alkali halide molecules are compared with the corresponding parameters deduced for excimer molecules from spectroscopic measurements.[13] Among these parameters is the energy of the vibrational quanta $\hbar\omega_e$, anharmonicity parameter $\omega_e x_e$, equilibrium distance between the nuclei R_e, and depth D_e of the potential well of a molecule formed on approach of a negative halogen ion X^- to a positive inert gas (alkali metal) ion. We can see that the principal parameters of these molecules do not differ by more than 10%.

2. EXCITATION MECHANISMS FOR EXCIMER LASERS

As pointed out earlier, the threshold values of the population inversion in excimer lasers are very high due to the short wavelength and the considerable line width of the relevant transitions. In fact, the expression for the gain in the case of no molecules in the lower state of the transition is[2)]

$$k=\frac{\lambda^2 a_{21}}{8\pi\Delta\nu}N_u, \tag{2}$$

where N_u is the concentration of molecules in the upper state; $\Delta\nu$ is the width of the gain profile; a_{21} is the probability of spontaneous emission as a result of the transition in question; λ is the wavelength. For excimer lasers we have $a_{21}\sim10^7-10^8$ sec^{-1} and $\lambda\approx(2-3)\times10^{-5}$ cm, so that if the threshold gain is $k\sim10^{-2}$ cm^{-1}, the threshold value of the concentration of the excited molecules is

$$N_u\approx10^{14}-10^{15}\ \text{cm}^{-3}. \tag{3}$$

This concentration of excited molecules can be produced only by means of very high pump energy densities $\xi_p\sim10^{-2}$ J/cm^3 supplied in a time of the order of $10^{-8}-10^{-7}$ sec. This high pump energy density can be achieved by passing a high-intensity beam of fast electrons or a high-power discharge pulse through a high-density gas. The mechanisms of population inversion, based on various sequences of elementary processes, are very different for these two (electron-beam and discharge) methods of supplying the pump energy. We examine these mechanisms in greater detail.

a) Electron-beam excitation of excimer lasers. Parameters of elementary processes

When an excimer laser is excited by a fast-electron beam the specific pump energy supplied to the active medium is proportional to the gas pressure so that in this case the threshold conditions are reached simply by increasing the pressure. For example, if the main mechanism by which the electron beam loses its energy in a gas is the ionization of the gas atoms, the specific energy density ξ_p supplied to the gas is[3)]

$$\xi_p\approx\frac{\pi e^4 Z}{\varepsilon}NN_b\ln\left(\frac{\varepsilon}{I}\right)c_b\tau, \tag{4}$$

where N_b is the electron density in the beam; Z is the atomic number of the elements; ε is the energy of the beam electrons; I is the binding energy of an electron in an atom; c_b is the velocity of the beam electrons, close to the velocity of light; τ is the pulse duration. For a relativistic electron beam of ~1 MeV energy, current density $j\sim10^4$ A/cm^2, and pulse duration $\sim10^{-8}$ sec, a pump energy density of ~0.1 J/cm^3 is reached at a krypton pressure of ~1 atm. Electron beam sources with these parameters are widely available[18] and their construction presents no special technical difficulties.

If the active medium consists mainly of an inert gas with a high atomic number Z, the major part of the energy lost by the electron beam in the gas is expended in the ionization of atoms. Consequently, a considerable number of free electrons is generated and their recombination produces excited atoms and molecules. We shall bear in mind the parameters of the active medium given above and analyze the following sequence of elementary processes occurring in such a weakly ionized gas:

$$\hat{e}+R\rightarrow\hat{e}+R^++e, \tag{5}$$

$$R^++2R\rightarrow R_2^++R, \tag{6}$$

$$R_2^++e\rightarrow R+R^*, \tag{7}$$

$$R^++2e\rightarrow R+e, \tag{8}$$

$$R^++X_2\rightarrow RX^*+X, \tag{9}$$

$$R^*+X_2\rightarrow R+2X, \tag{9'}$$

$$RX^*\rightarrow R+X+\hbar\omega, \tag{10}$$

[2)] This form of the expression corresponds to the limiting case when the width of a line due to a single electronic-vibrational-rotational transition is much higher than the gas temperature, so that a complete overlap of the rotational structure of the transitions is achieved (for details see below).

[3)] This expression gives the correct order of magnitude; more accurate data, which allow for the processes of multiple scattering of electrons, have been obtained as a result of cumbersome machine calculations and can be found, for example, in Ref. 17.

$$R^* + 2R \rightarrow R_2^* + R, \quad (11)$$

$$h\omega + RX^* \rightarrow R + X + 2\hbar\omega, \quad (12)$$

where R and X are, respectively, inert gas and halogen atoms; the asterisk denotes an electronically excited state; $\hat{e}$ is a fast beam electron; e is a thermal electron.

It is clear from the laser excitation mechanism that the important stages in the sequence of the processes resulting in the population inversion are the ion conversion (6) and dissociative recombination (7). These processes correspond to the main channels of conversion of the electric field energy into the energy of electronically excited atoms and molecules only under special conditions when the gas pressure and the energy of the primary electrons in the gas are sufficiently high. At low pressures, on the one hand, the conversion process (6) is very slow so that the dissociative recombination (7) competes with other recombination processes producing atoms in many electronically excited states rather than in one. This regime is unfavorable for the population inversion. On the other hand, at low electron energies the proportion of the energy expended in ionization decreases and this again lowers the selectivity of the mechanism of formation of excited atoms. In fact, if the energy of an incident electron is of the order of the binding energy of an atomic electron, an inelastic collision of such an electron with an atom is equally likely to produce atoms in a variety of excited states.

Table III gives the characteristics of the processes occurring in the active media of excimer lasers. We can see that sufficiently comprehensive and reliable information is available on the processes involving charged particles: electrons, atomic and molecular ions. On the other hand, information on the mechanisms and rate constant of the formation and destruction of excited excimer molecules is far from complete and definite. This makes it difficult to carry out systematic calculations of the gain of the active media of excimer lasers.

We shall analyze the processes which occur in a plasma formed as a result of the passage of an intense electron beam through a gas. Such an analysis will give us an idea concerning the channels along which the energy introduced into the gas is transformed. We shall compare the electron–energy loss times for elastic and inelastic electron–atom and electron–electron collisions in the specific case of an inert gas plasma with the parameters $N_e \sim 10^{15}$–10^{16} cm^{-3} and $N \leq 10^{20}$ cm^{-3}. A typical electron-energy loss time for electron–electron collisions

$$\tau_{ee} \sim \left(N_e \frac{e^4}{T_e^2} \Lambda \sqrt{\frac{T_e}{m}}\right)^{-1} \sim 10^{-11}-10^{-10} \text{ sec} \quad (13)$$

($T_e = 1-2$ eV is the electron temperature, discussed below, and Λ is the Coulomb logarithm) is much less than the time for elastic electron collisions

$$\tau_{elast} \sim \left(\frac{2m}{M} N k_{elast}\right)^{-1} \sim 10^{6} \text{ sec} \quad (14)$$

(k_{elast} is the constant for the elastic electron–atom collisions). Consequently, a Maxwellian distribution of the electron energies, characterized by an electron temperature T_e, is established in $\leq 10^{-10}$ sec in the plasma.

TABLE III. Parameters of processes occurring in electron-beam-excited excimer lasers ($T = 300$°K). *

	Process **	Constant	Reference
1	$Ar^+ + 2Ar \rightarrow Ar_2^+ + Ar$	$2.1\cdot10^{-31}$ cm^6/sec	19
2	$Kr^+ + 2Kr \rightarrow Kr_2^+ + Kr$	$2.4\cdot10^{-31}$ cm^6/sec	19
3	$Xe^+ + 2Xe \rightarrow Xe_2^+ + Xe$	$2.7\cdot10^{-31}$ cm^6/sec	19
4	$Ar_2^+ + e \rightarrow Ar^* + Ar$	$5\cdot10^{-7}\left(\frac{300°}{T_e}\right)^{1/2}$ cm^3/sec	19
5	$Kr_2^+ + e \rightarrow Kr^* + Kr$	$10^{-6}\left(\frac{300°}{T_e}\right)^{1/2}$ cm^3/sec	19
6	$Xe_2^+ + e \rightarrow Xe^* + Xe$	$2\cdot10^{-6}\left(\frac{300°}{T_e}\right)^{1/2}$ cm^3/sec	19
7	$R^+ + 2e \rightarrow R + e$	$10^{-19} N_e \left(\frac{300°}{T_e}\right)^{9/2}$ cm^6/sec	19
8	$Ar(^3P_2) + NF_2 \rightarrow ArF^+ + NF_2$	10^{-10} cm^3/sec	4, 20
9	$Kr(^3P_2) + F_2 \rightarrow KrF^* + F$	$3.6\cdot10^{-10}$ cm^3/sec	4
10	$Kr(^3P_2) + OF_2 \rightarrow KrF^* + OF$	$2.7\cdot10^{-10}$ cm^3/sec	4
11	$Kr(^3P_2) + CF_3OF \rightarrow KrF^* + CF_3O$	$1.5\cdot10^{-10}$ cm^3/sec	4
12	$Kr(^3P_2) + NF_3 \rightarrow KrF^* + NF_2$	$3.2\cdot10^{-11}$ cm^3/sec	4
13	$Kr(^3P_2) + N_2F_4 \rightarrow KrF^* + N_2F_2$	$1.1\cdot10^{-10}$ cm^3/sec	4
14	$Xe(^3P_2) + F_2 \rightarrow XeF^* + F$	$3.8\cdot10^{-10}$ cm^3/sec	4
15	$Xe(^3P_2) + OF_2 \rightarrow XeF^* + OF$	$2.7\cdot10^{-10}$ cm^3/sec	4
16	$Xe(^3P_2) + CF_3OF \rightarrow XeF^* + CF_3O$	$1.3\cdot10^{-10}$ cm^3/sec	4
17	$Xe(^3P_2) + NF_3 \rightarrow XeF^* + NF_2$	$5\cdot10^{-11}$ cm^3/sec $9\cdot10^{-11}$ cm^3/sec	4 20
18	$Xe(^3P_2) + Br_2 \rightarrow XeBr^* + Br$	$4.6\cdot10^{-10}$ cm^3/sec 10^{-9} cm^3/sec	4 21
19	$Xe(^3P_2) + N_2F_4 \rightarrow XeF^* + N_2F_2$	$1.2\cdot10^{-10}$ cm^3/sec	4
20	$Xe(^3P_2) + NOF \rightarrow XeF^* + NO$	$1.7\cdot10^{-10}$ cm^3/sec	4
21	$Kr(^3P_2) + F_2 \rightarrow Kr + 2F$	$3.6\cdot10^{-10}$ cm^3/sec	4
22	$Kr(^3P_2) + OF_2 \rightarrow Kr + OF + F$	$2.5\cdot10^{-10}$ cm^3/sec	4
23	$Kr(^3P_2) + CF_3OF \rightarrow Kr + CF_3O + F$	$2.6\cdot10^{-10}$ cm^3/sec	4
24	$Kr(^3P_2) + NF_3 \rightarrow Kr + F + NF_2$	$5.6\cdot10^{-11}$ cm^3/sec	4
25	$Kr(^3P_2) + N_2F_4 \rightarrow N_2F_3 + F + Kr$	$2.2\cdot10^{-10}$ cm^3/sec	4
26	$Xe(^3P_2) + F_2 \rightarrow Xe + 2F$	$3.8\cdot10^{-10}$ cm^3/sec	4
27	$Xe(^3P_2) + OF_2 \rightarrow Xe + OF + F$	$2.9\cdot10^{-10}$ cm^3/sec	4
28	$Xe(^3P_2) + CF_3OF \rightarrow Xe + CF_3O + F$	$3.3\cdot10^{-10}$ cm^3/sec	4
29	$Xe(^3P_2) + NF_3 \rightarrow Xe + NF_2 + F$	$4.3\cdot10^{-11}$ cm^3/sec	4
30	$Xe(^3P_2) + N_2F_4 \rightarrow Xe + F + N_2F_2$	$1.9\cdot10^{-10}$ cm^3/sec	4
31	$Xe(^3P_2) + NOF \rightarrow Xe + NO + F$	$2.2\cdot10^{-10}$ cm^3/sec	4
32	$XeBr^* + Br_2 \rightarrow Xe + 3Br$	$8\cdot10^{-10}$ cm^3/sec	22
33	$XeF^* + NF_3 \rightarrow Xe + 2F + NF_2$	10^{-9} cm^3/sec	21
34	$Xe(^3P_2) + 2Xe \rightarrow Xe_2^* + Xe$	$5\cdot10^{-32}$ cm^6/sec	21, 22
35	$Ar(^3P_2) + 2Ar \rightarrow Ar_2^* + Ar$	$1.1\cdot10^{-32}$ cm^6/sec $2.8\cdot10^{-32}$ cm^6/sec	24 25
36	$Xe(^3P_{1,2}) + Xe + He \rightarrow Xe_2^*(^{1,3}\Sigma_u^+) + He$	$1.4\cdot10^{-32}$ cm^6/sec	23
37	$Xe(^3P_{1,2}) + Xe + Ar \rightarrow Xe_2^*(^{1,3}\Sigma_u^+) + Ar$	$2.3\cdot10^{-32}$ cm^6/sec	23
38	$Ar(^3P_2) + Xe \rightarrow Ar + Xe^*$	$1.8\cdot10^{-10}$ cm^3/sec	4
39	$Ar(^3P_0) + Xe \rightarrow Ar + Xe^*$	$3\cdot10^{-10}$ cm^3/sec	4
40	$Xe_2^*(^1\Sigma_u^+) \rightarrow 2Xe + \hbar\omega$	5 nsec 5.5 nsec 2 nsec	26, 27 25 85
41	$Xe_2^*(^3\Sigma_u^+) \rightarrow 2Xe + \hbar\omega$	40 nsec 100 nsec 96.5 nsec 60 nsec	26, 27 28 25 85
42	$Ar_2^*(^1\Sigma_u^+) \rightarrow 2Ar + \hbar\omega$	3.2 nsec	25
43	$Ar_2^*(^3\Sigma_u^+) \rightarrow 2Ar + \hbar\omega$	4.2 nsec	25
44	$ArF^* \rightarrow Ar + F + \hbar\omega$	10 nsec	29
45	$KrF^* \rightarrow Kr + F + \hbar\omega$	10 – 20 nsec	30
46	$XeF^* \rightarrow Xe + F + \hbar\omega$	125 nsec 50 nsec	21 31
47	$XeBr^* \rightarrow Xe + Br + \hbar\omega$	17.5 nsec	21
48	$ArF^* + 2Ar \rightarrow Ar_2F^* + Ar$	$4\cdot10^{-31}$ cm^6/sec	88

*From the numerous data on the conversion and dissociative recombination coefficients given in the literature (for example in Smirnov's monograph[19]), we selected those which agreed with the theoretical ideas on the values of these quantities.

**In the case of quenching of metastable atoms and excimer molecules by collisions with impurity molecules, (21)–(33), the final states of the quenching molecule have not been identified and are deduced from general considerations.

The quasistationary value of this parameter is due to the exchange of energy of the plasma electrons with the beam electrons and gas atoms.[32] The quasistationary value of the electron temperature will be estimated by considering the electron energy balance in a plasma formed by a fast-electron beam.

1) *Electron energy balance.* The inelastic ionization and excitation of gas atoms by the primary and secondary beam electrons produces slow electrons of energy much lower than the first ionization potential I of a resonant atomic state. These electrons are characterized by an approximately uniform distribution of energies, so that the average energy of these electrons is $I/2$. Hence, we obtain the following expression for the rate of change of the electron energy as a result of their formation:

$$\left(\frac{\partial T_e}{\partial t}\right)_f = \left(\frac{I}{3} - T_e\right)\frac{Q_e}{N_e}, \tag{15}$$

where Q_e is the volume rate of electron generation by the electron beam.

In view of the dependence of the dissociative recombination cross section on the electron energy, this process results in a change in the average energy of electrons, which is equivalent to their heating. In fact, the rate of change of the average electron energy as a result of their dissociative recombination can be calculated from

$$\left(\frac{\partial T_e}{\partial t}\right)_{dr} = N_m \int_0^\infty \sigma_{dr}(\varepsilon)\sqrt{\frac{2\varepsilon}{m}} f(\varepsilon)\left(\frac{2}{3}\varepsilon - T_e\right) d\varepsilon, \tag{16}$$

where σ_{dr} is the cross section of the dissociative recombination of a molecular ion and an electron of energy ε; $f(\varepsilon)$ is the Maxwellian electron energy distribution function; N_m is the concentration of molecular ions. We shall represent the dissociative recombination cross section σ_{dr} by the Breit–Wigner formula[19,33]

$$\sigma_{dr} = \frac{\pi\hbar^2}{2\varepsilon m}\frac{\Gamma_{a0}\Gamma}{(\varepsilon - \varepsilon_a) + (\Gamma^2/4)} \tag{17}$$

(Γ is the width of the autoionizing level through which the process of dissociative recombination takes place, ε is the energy of this level measured from the ground state of the molecular ion, and Γ_{a0} is the inelastic part of the width of the autoionizing level, corresponding to the transition of an atom from an autoionization to a bound state). In integrating Eq. (16) we must make the simplifications which give the experimental temperature dependence of the dissociative recombination coefficient α_{dr}. This dependence is $\alpha_{dr}(T_e) \propto T_e^{1/2}$ and it is obtained using the Breit–Wigner formula for σ_{dr} on the assumption that $T_e \ll \varepsilon_a$, so that the integral in the expression for $\langle\sigma_{dr} v\rangle$ converges for low electron energies because the function $f(\varepsilon)$ is steep. We shall calculate the integral (16) using the same assumption:

$$\left(\frac{\partial T_e}{\partial t}\right)_{dr} = \frac{2}{3} N_m C_1 \sqrt{\frac{2T_e}{m\pi}}, \tag{18}$$

where N_m is the concentration of molecular ions and C_1 is the coefficient of proportionality, whose value depends on the type of molecular ion. These values, deduced by us from measurements of the dissociative recombination coefficient,[19,33] are listed in Table IV.

TABLE IV. Coefficient of proportionality in Eq. (18) calculated for molecular ions of inert gases.

Gas	Ar	Kr	Xe
C_1, 10^{-15} eV · cm^2	1	1.5	4

Comparing the expression (18) with Eq. (15) for the rate of electron heating because of their formation, we find that under quasisteady conditions corresponding to the relationships

$$Q_e = \alpha_{dr} N_e^2, \quad N_e \approx N_m, \tag{19}$$

these rates are of similar orders of magnitude.

The main electron-energy loss mechanism is the one involving inelastic electron–atom collisions. The rate of this process

$$\left(\frac{\partial T_e}{\partial t}\right)_{inelast} = \sum_n -\Delta E_{0n} k_{inelast}^n(T_e) N \tag{20}$$

will be estimated by approximating the near-threshold energy dependence of the excitation cross section of a resonant atomic state by the semiempirical formula[34]

$$\sigma_{0n}(x) = \frac{2\pi f_{0n}}{(\Delta E_{0n})^2}\frac{\ln[x - 0.9(x-1)]}{x - 0.7} \tag{21}$$

[$x = \varepsilon/\Delta E$ is the electron energy in threshold units, ΔE is the excitation energy of an atom, f_{0n} is the oscillator strength of the transition in question, N is the concentration of the inert gas atoms,

$$k_{inelast}(T_e) = \int_{\Delta E_{0n}}^\infty f(\varepsilon, T_e)\sigma_{0n}\left(\frac{\varepsilon}{\Delta E_{0n}}\right)\sqrt{\frac{2\varepsilon}{m}}\sqrt{\varepsilon}\, d\varepsilon,$$

$f(\varepsilon, T_e)$ is the Maxwellian electron energy distribution function].

The rate of cooling of the electron gas by elastic electron–atom collisions is given by

$$\left(\frac{\partial T_e}{\partial t}\right)_{elast} = -T_e\frac{2m}{M} N k_{elast} \tag{22}$$

If the duration of the electron-beam pulses exceeds the characteristic heating and cooling times of an electron gas estimated above, a quasisteady regime is established in a plasma and it corresponds to a specific electron energy T_e. This energy is found from the steady-state equation for the energy balance of electrons which, with the processes (15)–(22) having been taken into account, is

$$\left(\frac{\partial T_e}{\partial t}\right)_f + \left(\frac{\partial T_e}{\partial t}\right)_{dr} = -\left(\frac{\partial T_e}{\partial t}\right)_{inelast} - \left(\frac{\partial T_e}{\partial t}\right)_{elast}. \tag{23}$$

Since under these conditions the electron temperature T_e is much less than the threshold excitation energy of atoms E_{0n}, the first term on the right-hand side of Eq. (23) depends very strongly on T_e. Therefore, Eq. (23) regarded as describing T_e can be solved with a sufficiently high accuracy in spite of the approximate nature of the information on the constants of the elementary processes occurring in this equation.

Under quasisteady conditions described by Eq. (19) the left-hand side of Eq. (23) is proportional to the electron density and the right-hand side to the inert gas con-

centration. Therefore, a quasisteady electron temperature T_e for a given inert gas is governed by the quasisteady degree of ionization N_e/N. The results of calculations of T_e for different degrees of ionization are given in Table V.[35]

It is clear from Table V that, over a wide range of the plasma parameters, the electron temperature depends weakly on these parameters and amounts to about 1 – 2 eV. It is worth noting that the results of the calculations demonstrate a negligible influence of the elastic electron–atom collisions on the electron energy balance. The calculations apply to the case when the duration of the excitation pulses is sufficiently long compared with the characteristic time needed to establish a quasisteady energy balance. Otherwise, Eq. (23) does not contain the first term on the left-hand side. However, in view of the strong temperature dependence of the term responsible for the inelastic electron-energy losses, this results in a slight (within 10–15%) reduction in the electron temperature compared with the values listed in Table V.

2) *Charged-particle balance.* We shall now analyze the processes which generate excited inert gas atoms by dissociative recombination of molecular ions. The time dependences of these processes are found by solving a system of balance equations for electrons, atomic ions, and molecular ions:

$$\frac{dN_e}{dt}=Q_e-\alpha_{dr}\ N_eN_m, \tag{24}$$

$$\frac{dN_m}{dt}=N_aN^2k_c-\alpha_{dr}\ N_eN_m, \tag{25}$$

$$N_e=N_m+N_a,$$
$$N_e(0)=N_m(0)=N_a(0)=0, \tag{26}$$

where N_m and N_a are the concentrations of molecular and atomic enert-gas ions, respectively. We readily obtain the solution

$$N_a=\frac{Q_e}{N^2k_c}(1-e^{-N^2k_ct}), \tag{27}$$

which is valid under steady-state excitation conditions (Q_e = const). It is difficult to obtain the general analytic solution of the system (24)–(26). However, an analysis of the (24)–(26) system shows that the solution becomes stead in a time longer than the characteristic times for conversion $\tau_c\sim(N^2k_c)^{-1}\sim10^{-9}$ sec and dissociative recombination $(\alpha_{dr}Q_e)^{-1/2}\sim10^{-8}-10^{-9}$ sec. The duration of the excitation pulses in practical systems usually exceeds these values[6] so that in considering the excitation of a laser it is sufficient to have the quasisteady solution of this system, which is of the form

$$N_e=\frac{Q_e}{2N^2k_c}\left(1+\sqrt{1+\frac{4N^4k_c^2}{\alpha_{dr}\ Q_e}}\right). \tag{28}$$

TABLE V. Quasisteady electron temperatures T_e calculated from Eq. (23).

Gas / N_e/N	Ar	Kr	Xe	Gas / N_e/N	Ar	Kr	Xe
10^{-6}	1.15	1.25	1.05	10^{-4}	2.05	2.0	1.7
10^{-5}	1.61	1.65	1.4	10^{-3}	2.5	2.4	2.0

We can see that the nature of the time dependence of the electron density is largely governed by the time dependence of the dimensionless parameter $4N^2k_c^2/Q_e\alpha_{dr}$, which—in its turn—depends on the pumping rate Q_e. At a high pumping rate

$$Q_e\gg\frac{4N^4k_c^2}{\alpha_{dr}} \tag{29}$$

the process which limits the rate of formation of metastable inert gas species is the conversion of atomic ions into molecular ones. In this case we have

$$N_e\approx\frac{Q_e}{N^2k_c}+\frac{N^2k_c}{\alpha_{dr}}\approx\frac{Q_e}{N^2k_c}, \tag{30}$$

$$N_m=N_e-N_a\approx\frac{N^2k_c}{\alpha_{dr}}, \tag{31}$$

so that the volume rate of formation of metastable inert gas atoms is $\alpha_{dr}N_eN_m\approx Q_e$.

In the opposite limit of "slow" excitations, corresponding to the condition

$$Q_e\ll\frac{4N^4k_c^2}{\alpha_{dr}}, \tag{32}$$

the rate-limiting process is the dissociative recombination. It follows from Eq. (26) that

$$N_m\approx N_e\approx\sqrt{\frac{Q_e}{\alpha_{dr}}}, \tag{33}$$

and the volume rate of formation of metastable atoms is still equal to the rate of ionization of the gas Q_e by the electron beam.

A full picture of the elementary process occurring in the active media of excimer lasers can be obtained only if we have detailed information on the processes of conversion of the energy of the excited atoms formed as a result of the dissociative recombination into the energy of excited excimer molecules. Information of this kind is not sufficiently complete or reliable to draw definite conclusions on the energy conversion channels and, consequently, on the optimal conditions of energy utilization. In particular, there have been practically no detailed investigations of the process of quenching (9) of excited inert gas atoms by halogen-containing molecules, which might give information on the final state of the resultant excimer molecules, or of the rate of the competing process (9′) which results in the loss of excitation. For this reason the recently published calculations of the characteristics of excimer lasers allowing for tens of elementary processes (for example, those reported by Hart and Searles,[22] by George and Rhodes,[26] and by Werner *et al.*[27]) can hardly be compared with the experimental results because the kinetic models of laser excitation are not based on reliable values of the parameters of these processes.

3) *Optimal laser parameters.* In spite of the incomplete nature of the available information, we can use the above analysis to estimate the optimal parameters of electron-beam-excited excimer lasers. We shall first consider the lasers utilizing the dimer molecules Ar_2, Kr_2, and Xe_2, which are characterized by similar population inversion mechanisms and also by similar values of the constants of the processes governing the excitation kinetics. In lasers of this type the excimer molecules are formed by the reaction

$$R^* + 2R \rightarrow R_2^* + R, \quad (34)$$

which is characterized by a rate constant $k_{ac} \sim 10^{-32}$ cm^6/sec (Table III). Allowing for the radiation trapping, we find that the radiative lifetime of resonantly excited inert gas atoms R* is sufficiently long ($\geq 10^{-6}$ sec) so that the reaction (34) is practically the only channel for the conversion of energy of excited atoms even at inert gas concentrations $N > 10^{19}$ cm^{-3}. We shall show later that the laser in question usually operates in the near-threshold regime and, therefore, optimal conditions are obtained by accumulating the maximum possible number of excimer molecules at the upper laser (active) level. This is possible if the characteristic time of the process (34) is much less than the radiative lifetime of an excimer molecule τ_r:

$$N^2 k_{ac} \gg \tau_r^{-1}, \quad (35)$$

which gives us an upper limit to the inert gas concentration

$$N \gg (\tau_r k_{ac})^{-1/2} \sim 10^{20} \text{ cm}^{-3}. \quad (35a)$$

A similar condition imposed on the electron density is associated with the requirement that the characteristic time $\tau_{dr} \propto (\alpha_{dr} N_e)^{-1}$ of the dissociative recombination process of molecular ions is shorter than the radiative decay time of excimer molecules:

$$N_e \gg \frac{1}{\alpha_{dr} \tau_r} \sim 10^{16} \text{ cm}^{-3}. \quad (36)$$

An estimate of the threshold excitation condition in the active media of lasers of this kind is obtained from the requirement

$$k_0 l \gg 1, \quad (37)$$

where

$$k_0 = \frac{\lambda^2}{4} \frac{a_{21}}{\Delta\omega} N_u \quad (37')$$

is the gain at the center of the transition line; l is the length of the active medium; λ is the wavelength of the laser radiation; a_{21} is the probability of spontaneous emission; N_u is the concentration of the particles at the upper laser level. It should be noted that in the case of lasers utilizing inert gas dimers the line width of a transition $\Delta\omega \sim 10^{14} - 10^{15}$ sec^{-1} is much great than the width of a rotational band or even the distance between the vibrational states of an excited electron term. Therefore, all the excimer molecules contribute, irrespectively of their vibrational and rotational states, to the amplification at this frequency, which corresponds to maximum gain, i.e., N_u in Eq. (37) is the total concentration of excimer molecules.

When the energy of free electrons is converted efficiently into the excitation energy of excimer molecules, we have $N_u a_{21} \approx Q_e$. Hence, and from Eqs. (37) and (37′), we obtain the following estimate for the threshold rate of volume ionization of the gas

$$Q_e \gg \frac{4\Delta\omega}{\lambda^2 l}. \quad (38)$$

In the $l \sim 10$ cm case, which usually corresponds to practical systems, this estimate gives $Q_e \approx 10^{25}$ cm^{-3} sec^{-1}. Using the expression (38), we obtain the threshold value of the electron density

$$N_e \sim \sqrt{\frac{Q_e}{\alpha_{dr}}} \sim 3 \cdot 10^{15} \text{ cm}^{-3}.$$

The threshold gas density is governed by the excitation conditions. In fact, in the electron-beam excitation case the rate of volume ionization can be expressed, with the aid of Eq. (4), in the form

$$Q_e = \frac{\pi e^4 Z}{\varepsilon \Delta E_i} N \Lambda \frac{j_b}{e}, \quad (39)$$

where ΔE_i is the energy required to form one electron–ion pair as a result of the passage of a fast particle through the gas in question; Λ is the Coulomb logarithm; j_b is the density of the electron-beam current. Using, for example, the threshold value $Q_e \approx 10^{25}$ cm^{-3} sec^{-1}, we find that in the case of an electron beam of $\varepsilon \sim 1$ MeV energy and current density $j_b \sim 10^3$ A/cm^2, the threshold gas density is $\sim 10^{21}$ cm^{-3}. Hence, we can immediately see the technical difficulties that experimentalists have to overcome in investigating lasers with inert gas dimers as the active media. These difficulties involve trying to combine a vacuum chamber, where an electron beam is generated, with a chamber filled with an active medium kept at a pressure of tens of atmospheres.

The pressure optimum is due to the fact that at high pressures the laser radiation is absorbed producing excimer molecules. If this process is allowed for, the gain can be represented in the form (see, for example, the results of Ostroukhova *et al.*[36]):

$$k_0 \approx \frac{\lambda^2}{4} \frac{a_{21}}{\Delta\omega} (N_u - N^2 \cdot 4\pi R_0^2 \Delta R e^{-U(R_0)/T}), \quad (40)$$

where R_0 is the equilibrium internulcear distance in an excimer molecule; ΔR is the size of the region which is classically accessible to the nuclei of this molecule; $U(R)$ is the potential curve of the lower term of the laser transition which (in the range of the internuclear distances under consideration) is repulsive. In Eq. (40) we are ignoring the dependences of a_{21}, U, and λ on R in a small interval $\Delta R \ll R_0$. The second term in the parentheses of Eq. (40) is of the following nature: $\exp - [U(R)/T] N 4\pi R_0^2 \Delta R$ is the probability that in a spherical layer of radius R_0 and thickness ΔR, which surrounds an inert gas atom, there is a second atom of the same kind so that a laser photon may be absorbed producing an excimer molecule (this probability is less than unity for $N < 10^{23}$ cm^{-3}). When this quantity is multiplied by the concentration of inert gas atoms, it gives the number of pairs of atoms per unit volume capable of absorbing laser radiation. In view of the sensitivity of this estimate to the precision with which the curve $U(R_0)$ is known, we cannot predict reliably the inert gas pressure at which the absorption of laser radiation becomes important.

Figure 3 shows how the transparency of a xenon-filled cell to radiation of 1700 Å wavelength varies with the xenon pressure.[37] It is clear from this figure that a steep rise of absorption begins at $N > 10^{21}$ cm^{-3}. Using Eq. (40), we find that this cell is optically dense for

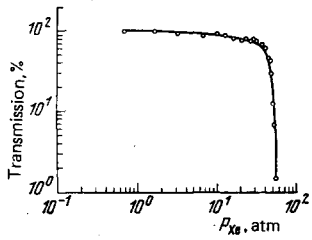

FIG. 3. Dependence of the transmission coefficient of the radiation 1700 Å wavelength on xenon pressure (in a cell 17 cm long).[37]

$N > 10^{21}$ cm^{-3} if we assume that $U(R_0 \approx 3.0$ Å$) \approx 0.26$ eV. This value is comparable with the Xe–Xe repulsive potential $U(R_0) \approx 0.42$ eV, deduced from atomic scattering experiments.[38] Figure 4a shows the dependence of the intensity of the radiation emitted by the Xe_2^* excimers, excited by an electron beam in a dense gas, on xenon pressure.[37] We can see that an optimum at $p \approx 20$ atm agrees well with the steep increase of the absorption for $p \geq 40$ atm, shown in Fig. 3.

For other inert gas dimers we can expect basically similar behavior as for the Xe_2 molecule. Thus, according to the results of atomic scattering experiments,[38] the potential of the Ar_2 molecule is $U(R_0) \leq 0.3$ eV and that of the Kr_2 molecule is $U(R_0) \leq 0.27$ eV, so that the absorption of laser radiation is important approximately in the same range of pressures as in the above case of Xe_2. This results in a similar pressure dependence of the intensity of the radiation emitted by Ar_2 (Fig. 4b).

We may conclude from this comparison that the main factor which limits the characteristics of lasers utilizing inert gas excimers is the proximity of the threshold parameters to those which are optimal from the point of view of population inversion. This suggests ways of solving this problem involving, on one hand, a reduction in the threshold pressure of the laser by increasing its length and also by increasing the Q factor of the resonator, and, on the other, by increasing the electron beam current density, which makes it possible to operate at lower working pressures when the absorption of laser radiation is slight.

We shall now estimate the optimal parameters of excimer lasers utilizing inert gas monohalides and excited by fast-electron beams. In these lasers the formation of the RX* excimer molecules is the result of the reaction (9), where the halogen-containing molecule may be either a halogen molecule X_2 or any other molecule such as NF_3, SF_6, etc. In lasers of this type the threshold conditions are much less stringent than those utilizing

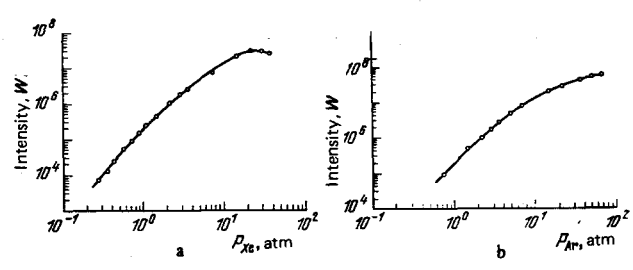

FIG. 4. Pressure dependence (in a cell of 1.3 cm^3 volume) of the intensity of spontaneous radiation emitted from Xe_2 (a) and Ar_2 (b) excimers.[35]

inert gas dimers and, therefore, the conditions (35)–(36) are no longer necessary for laser action.

The upper limit on the density of the halogen-containing substance follows from the condition that the process of quenching of the excited excimer molecules

$$RX^* + X_2 \rightarrow X + R + \frac{X_2}{2X} \tag{41}$$

should not play a significant role in the destruction of these molecules. This condition limits the concentration of the X_2 molecules to

$$N_{X_2} \leq [k_T \tau_r]^{-1} \sim 10^{17} \text{ cm}^{-3}, \tag{42}$$

if the rate constant of the process (41) is $k_T \sim 10^{-9}$ cm^3/sec (see Table III). This estimate allows us to find quite easily the upper limit on the specific output energy of an excimer laser pulse on the assumption that each molecule of the halogen-containing substance is used to form one excimer molecule:

$$\xi_{max} \leq \hbar\omega N_{X_2} \approx 5 \cdot 10^{-2} \text{ J/cm}^3, \tag{43}$$

where $\hbar\omega \approx 5 \times 10^{-19}$ J is the energy of a laser photon. The actually obtained values of $\xi \approx 3 \times 10^{-3}$ J/cm^3 [6] are over an order of magnitude smaller than the estimate given by Eq. (43). The reason for this discrepancy will be discussed in the next section.

In view of the restriction (42) imposed on the concentrationof the halogen-containing molecules, the absorption of laser radiation in this type of system does not play such an important role as in lasers utilizing inert gas dimers. In fact, it follows from the equation for the gain (40) that the absorption is unimportant if

$$\frac{NN_X \cdot 4\pi R_0^2 \Delta R \exp[-U(R_0)/T]}{N_u} \ll 1. \tag{44}$$

We shall first assume that in the range of internuclear distances of interest to us the repulsion is unimportant, i.e., $U(R_0) \leq T$. It follows from an analysis of the terms of excimer molecules (Sec. 1) that the characteristic equilibrium distances in these molecules are $R_0 \sim 4$–6 Å and $\Delta R \sim 0.1$–0.2 Å. Hence, we find that the condition (44) is satisfied if

$$\frac{CNN_X}{N_u} \ll 1, \tag{45}$$

where $C = 10^{-23} - 10^{-22}$ cm^3. If the repulsion is important for the internuclear distance R_0, i.e., if $U(R_0) > T$, the value of C is even smaller. We can see that the absorption of laser radiation is unimportant in the range of moderately high densities of the inert gas. For example, during the initial stage of the development of a laser pulse, when the number of the emitted laser photons is much less than the total number of the excited excimer molecules, we can assume that $N_u \geq N_X$, because the formation of each excimer molecule produces one halogen radical. In this case the absorption is unimportant in the range $N \ll 10^{22}$ cm^{-3}, i.e., in all cases of practical interest. In the course of the development of a laser pulse the number of the halogen radicals increases so that the ratio N_X/N_u becomes much greater than unity. However, the maximum value of N_X is of the same order of magnitude as the halogen concentration estimated above and it does not exceed 10^{17} cm^{-3}.

The minimum value of N_u is governed by the threshold conditions for laser action and, as shown below, is in any case not less than 10^{14} cm^{-3}. Hence, we obtain the upper limit on the inert gas density:

$$N \ll \frac{1}{(100-1000)\,C}. \tag{46}$$

When this condition is not obeyed, a laser pulse terminates before exhaustion of the energy of the excited excimer molecules so that the laser will not operate optimally. This has been avoided[5,6,21,30] by forming an active medium from a mixture of two inert gases, one of which is used at high pressures and serves to convert the electron-beam energy into the energy of the excited molecules. Atoms of the other inert gas participate in the excimer molecule formation so that the density of this gas satisfies the condition (46) and is much less than the total density of the buffer gas. For example, the most efficient lasers are filled with argon at a pressure of 1400 Torr and with krypton at the optimal pressure of 100 Torr.[6]

Another possible mechanism of laser radiation absorption in the active media of excimer lasers is associated with the photoionization of excited inert gas atoms. At a high concentration of these atoms the process results in undesirable absorption of the laser radiation and prevents stimulated emission. There are no published photoionization cross sections of excited inert gas atoms but, according to the recent quantum-mechanical computations,[39] typical values of the cross sections of the photoionization of resonantly excited Ar($^3P_1$) and Kr($^3P_1$) atoms by the KrF laser radiation of wavelength $\lambda = 2484$ Å are $\sim(4-5)\times10^{-18}$ cm^2. In the case of the $^3P_2$ metastable states this value is one or two orders of magnitude smaller. Since these cross sections are comparable with typical values of the stimulated emission cross sections of excimer lasers $\sigma_{st} \geqslant 10^{-18} - 10^{-17}$ cm^2,[7,40,89] the process of photoionization limits the concentration of the excited inert gas atoms in the active medium to a value lower than or of the order of the concentration of the excimer molecules. Another mechanism of the absorption of laser radiation in the active medium is associated with the presence of R_2^* molecular ions. According to calculations,[87] the cross sections for the absorption of laser radiation by the Ne_2^+, Ar_2^+, Kr_2^+, and Xe_2^+ ions are approximately 20, 12, 0.3, and 0.003 (in units of 10^{-18} cm^2) for $\lambda = 2480$ Å and 1, 9, 36, and 50 (in units of 10^{-18} cm^2) for $\lambda = 3510$ Å. This can explain the influence of buffer gases on the output parameters of excimer lasers.[41]

If the inequality (46) is obeyed, which is certainly true during the initial stage of a stimulated emission pulse, we can estimate the threshold conditions from Eq. (40) for the gain. We rewrite this equation in the form

$$k_0 = \frac{\lambda^3 a_{21}}{8\pi c \Delta\lambda/\lambda} N_u, \tag{40a}$$

where $\Delta\lambda/\lambda$ is the relative width of the gain profile of the active medium. The quantity N_u in Eq. (40a) represents the total concentration of the excimer molecules contributing to the amplification of laser radiation (compare with the discussion at the beginning of this subsection).

It is clear from Table VI that a typical value of $\Delta\lambda/\lambda$ is 10^{-2}. The value of λ is $\sim 2\times10^{-5}$ cm. If we use these parameters and assume the threshold gain $k_0 \sim 10^{-2}$ cm^{-1}, we find that Eq. (40a) gives the following estimate of the threshold concentration of the excited excimer molecules:

$$N_u \geqslant \frac{10^{22}}{a_{21}}. \tag{47}$$

A typical value of a_{21} is $\sim 10^8$ sec^{-1} and, therefore, the threshold concentration of the excited excimer molecules should be at least $10^{14} - 10^{15}$ cm^{-3}.

If we compare these results with the estimate (42), we find that the optimal concentration of the excited excimer molecules is two or three orders of magnitude lower than the density of the halogen-containing substance. Hence, we may conclude that it should be possible to generate laser radiation pulses of duration much longer than the excimer molecule lifetime. This conclusion is confirmed by the results reported by Champagne *et al.*,[41] who obtained XeF laser pulses of ~ 1 μsec duration by irradiation of the Ar:Xe:NF_3 (99.5:0.38:0.12) mixture at a pressure of 2.5 atm with a beam of electrons of 300 keV energy and a current density of 5 – 14 A/cm^2. In a tube 100 cm long and of 2.22 cm in diameter the active medium absorbed ~60 J, whereas the energy of the laser radiation pulses was 0.3 J. Estimates indicated that at the end of the laser pulse the concentration of NF_3 decreased by a factor of 2 – 3. From this it follows that an increase in the duration of the laser radiation pulse.[90]

We shall now consider the related interesting problem of the possibility of producing a cw excimer laser. In principle, such a laser can be constructed if the characteristic time for the replenishment of the halogen-containing substance can be made less than or of the order of the duration of the laser radiation pulses. In chemical recovery of the active substance the recovery time is estimated to be the time of the recombination reaction, for example,

$$2X + R \rightarrow X_2 + R, \tag{48}$$

and under the conditions in the active medium of a laser, this recombination time is

$$\tau_{rec} \sim \frac{1}{NN_X k_{rec}} \sim 10^{-5} \text{ sec}.$$

We can see that in this case we can obtain cw operation by increasing the duration of the laser radiation pulses to $\sim 10^{-5}$ sec. Turning to the cited investigation,[41] we can see that this requires a reduction of the threshold gain by another order of magnitude. The gain in the investigation of Champagne *et al.*[41] was ~0.02 cm^{-1}.

A rapid circulation of a gas with the aim of replenishment of the original laser mixture also can hardly help to solve the problem of constructing a cw excimer laser because the minimum transit time of a mixture through a chamber several centimeters long is of the order of $10^{-4} - 10^{-5}$ sec. Therefore, the radical approach to the solution of this problem is to use high-Q resonators and to increase the length of the active medium. This would make it possible, for a given value of the stimulated

TABLE VI. Parameters of excimer lasers

Molecule	λ, Å	Composition of active medium, Torr	Excitation method	Druation of excitation, nsec	Duration of emission	Volume of active medium, cm^3	Pump energy, J	Output energy, J	Laser efficiency, %	Gain line width Δλ, A	Gain, cm^{-1}	Reference
Xe_2	1720	Xe ($1.3\cdot10^4$)	I	100	20	—	—	10^{-3}	—	—	—	53
		Xe (10^4)	I	5	3	—	10	10^{-2}	0.1	70	—	54
		Xe ($1.2\cdot10^4$)	I	2	3	1.5	10^{-4}	—	—	50	—	55
		Xe ($1.3\cdot10^4$)	I	50	20	20	25	10^{-3}	0.004	25	0.3	56
		Xe ($1.6\cdot10^4$)	I	—	—	3.2	4	0.2	5	200	—	57
		Xe (1100)	I	1000	200	$2.5\cdot10^3$	750	0.01	0.001	160		91
Kr_2	1457	Kr ($2.4\cdot10^4$)	I	—	10	—	—	—	—	138	—	58
Ar_2	1261	Ar ($4\cdot10^4$)	I	60	15	100	600	—	—	80	—	59
ArO	5580	Ar ($3\cdot10^4$); N_2O (2)	IV	40	10^3	—	1	—	—	40	10^{-4}	60
KrO	5577	Kr (10^4); O_2 (5)	I	80	500	—	—	—	—	2,5	10^{-3}	53
		Kr ($2\cdot10^4$); O_2 (5)	I	50	80	50	2500	10^{-2}	0.0004	15	—	61
XeO	5400	Xe ($7,5\cdot10^4$); O_2 (10)	I	50	80	50	2500	10^{-2}	0.0004	250	—	61
		Xe (10^4); O_2 (10)	I	20	100	—	1	—	—	—	$2\cdot10^{-3}$	62
XeBr	2818	Xe (до 1400); Br_2 (10)	I	50	25	10	55	$4\cdot10^{-6}$	10^{-5}	1300	$3\cdot10^{-3}$	63
XeF	3511—3531	Ar (3000); Xe (10); F_2 (3)	I	100	50	15	100	$3\cdot10^{-4}$	$3\cdot10^{-4}$	—	—	64
		Ar (700); Xe (70); NF_3 (2)	I	100	100	100	260	0.08	0.03	—	—	21
		Ar (6000); Xe (35); F_2 (12)	I	15	15	3	6	$4\cdot10^{-5}$	10^{-3}	—	—	65
		He (1000); Xe (10); NF_3 (3)	V	40	20	60	10	0.065	0.6	—	—	46
		He (470); Xe (20); NF_3 (10)	II	10	10	16	20	0.01	0.5	—	—	66
		Ar (1900); Xe (7); NF_3 (2)	I	1200	1000	400	60	0.3	0.5	—	0.02	41
		Ne (3700); Xe (5); NF_3 (2)	I	1000	1000	380	55	1.0	1.8	—	0.012	41
		He (3600); Xe (4); NF_3 (1)	V	20	20	180	—	0.3	—	—	—	67
		Ar (1200); Xe (120); NF_3 (5)	I	20	10	30	40	0.005	10^{-3}	—	—	21
		He (300); Xe (4,5); NF_3 (1.5)	II	20	10	100	10	0.007	0.1	—	—	43
		He (730); Xe (9); NF_3 (9)	III	50	4	100	10	0.1	1	—	—	68
		He (400); Xe (12); NF_3 (4)	II	10	40	15	0,5	0.001	0.2	—	$2\cdot10^{-2}$	31
		He ($6\cdot10^3$); Xe (60); SF_6 (6)	II	2	2	9	to 5	0.0005	0.01	—	—	48
		Ar ($3\cdot10^3$); Xe (12); NF_3 (3)	III	500	100	80	5	0.01	0.3	15	—	20
		Ar (1500); Xe (10); SF_6 (5)	III	1000	1000	500	15	0.2	1.5	—	—	92
ArF	1933	He ($6\cdot10^3$); Ar (60); SF_6 (6)	II	2	2	9	to 5.0	0.0005	0.01	—	—	48
		Ar (1400); F_2 (4)	I	55	55	3.6	6000	92	1.6	15	—	6
		He (10^3); Ar (450); F_2 (3)	V	40	20	60	10	0.06	0.6	—	—	46
ArCl	1750	He (700); Ar (70); Cl_2 (7)	II	—	10	10^3	—	$2\cdot10^{-4}$	—	10	0.012	69
KrF	2484	He (1500); Kr (200); F_2 (4)	V	40	20	60	10	0.13	1.3	—	—	46
		He (650); Kr (40); NF_3 (1,3)	II	50	50	16	4	0.0025	0.06	—	—	66
		Ar (10^3); Kr (400); F_2 (4)	I	55	55	$3.6\cdot10^4$	6000	108	1.8	—	—	6
		He (630); Kr (70); NF_3 (1,3)	II	10	25	10	1.5	10^{-3}	0.04	—	—	70
		He (750); Kr (48); NF_3 (0,8)	III	50	25	100	10	0.03	0.03	—	—	68
		He ($6\cdot10^3$); Kr (60); SF_6 (6)	II	2	2	9	to 5	0.007	0.15	—	—	48
		Ar (740); Kr (15); F_2 (1)	III	160	90	80	2	0.004	0.2	30	—	30
		Ar ($3\cdot10^3$); Kr (150); F_2 (6)	I	50	50	—	8400	5.6	0.07	25	—	5
		Ar (10^3); Kr (400); F_2 (4)	I	55	55	$3.6\cdot10^4$	6000	108	1.8	10	—	6
		Ar ($3\cdot10^3$); Kr (30); F_2 (3)	I	100	150	15	2	0.008	0.4	40	—	64
		Ar (1500); Kr (100); NF_3 (2)	I	100	125	100	200	1.5	0.7	—	—	71
		Ar (1500); Kr (100); NF_3 (2)	I	—	125	100	—	—	23	—	—	7
		Ar (3500); Kr (100); F_2 (7)	III	70	70	240	400	1.2	0.3	—	—	93
NeF (?)	1050—1250	Ne (1000); F_2 (1,8)	I	300	—	—	$5\cdot10^3$	—	—	200	—	72
KrCl	—2220	Ar ($3,3\cdot10^3$); Kr (100); Cl_2 (5)	I	50	30	—	760	0.3	0.04	50	—	73
		He (700); Kr (70); F_2 (7)	II	—	10	10^3	—	0.0013	—	20	0.018	69
XeCl	3080	Ar ($2,7\cdot10^3$); Xe (30); Cl_2 (3)	I	100	30	15	2	$5\cdot10^{-5}$	$2\cdot10^{-3}$	25	—	64
		Ar (3700); Xe (130); CCl_4 (2)	I	25	25	70	20	0.03	0.15	—	—	92

Excitation methods: I) fast-electron beam; II) transverse electric-discharge pulses; III) discharge initiated by electron beam; IV) flash photolysis; V) discharge initiated by ultraviolet radiation.

radiation intensity inside the resonator, to use a much lower pumping rate which would increase the "burnup" time of the fluorine-containing substance to values corresponding to the recovery time (or transit time in the case of gas circulation).

b) Pulse-discharge excitation of excimer lasers. Characteristics of the excitation mechanism

The mechanism of the electric-discharge excitation of excimer lasers differs essentially from the excitation of lasers by an electron beam. The main distinguishing characteristic is the way of formation of metastable inert gas atoms. The dissociative recombination, discussed in the preceding sections, does not play a decisive role when lasers are excited by discharge pulses. The active medium now consists mainly of a buffer gas which is usually helium or neon. A typical ratio of the components of the active medium of an excimer laser is $He(Ne):R:X_2 \approx 10:1:0.1$, but the proportion of the buffer gas may be considerably greater. The total pressure in the mixture is usually about 1 atm, but in some cases much higher buffer gas pressures have been used.

Inelastic electron–atom collisions in a discharge produce a considerable number of excited inert gas atoms:

$$e + R \rightarrow R^*(^3P_1, {}^1P_1) + e. \quad (49)$$

Collisions of such atoms with electrons, accompanied by spin exchange between the incident and valence electrons, produce metastable atoms,

$$R^*(^3P_1, {}^1P_1) + e \rightarrow R(^3P_2, {}^3P_0) + e, \quad (50)$$

whose concentration is thus found to be close to the concentration of resonantly excited atoms.

A typical value of the spin exchange constant is 10^{-6} cm^3/sec.[12,42] Therefore, the process (50) may occur in a time shorter than or of the order of the characteristic laser emission time of $\sim 10^{-9}-10^{-8}$ sec when the electron

density N_e satisfies the relationship

$$N_e \geqslant 10^{14}-10^{15}\ \text{cm}^{-3}.$$

The formation of a homogeneous nanosecond discharge with this electron density in a gas at atomospheric pressure presents a serious technical problem, whose solution has been the subject of the majority of papers on electric-discharge-excited excimer lasers. In the case of inert gas dimers, when (as shown above) the optimal gas pressures are tens of atmospheres, this problem has not yet been solved so that electric discharges are used only to excite excimer lasers whose active media consist of inert gas monohalides. The experience accumulated recently in the development of high-pressure pulsed CO_2, HF, N_2, H_2, and other lasers has been found useful. In particular, excimer lasers are being excited by such thoroughly tested means as transverse discharged (TEA lasers),[43,44] traveling wave discharges or Blumlein lines,[45] and discharges with preliminary initiation by an additional ionization source such as photoionizing radiation,[46] a low-power electron beam,[47] or a preionization discharge in a small gap.[48]

The principal parameter which governs the efficiency of an electric-discharge excimer laser is the ratio of the electric field E to the buffer gas density N. For low values of E/N, the rate of excitation of inert gas atoms by electron impact, given by Eq. (49) is also low. At high values of E/N, we may expect effective simultaneous excitation of a large number of states of an inert gas atom and its ionization, which reduces greatly the efficiency of conversion of input energy into the energy of metastable inert gas atoms. The optimal values of E/N are of the order of 10^{-15} V·cm^2 at pressures of the order of atomospheric value, which corresponds to electric fields in the range $10^4 - 10^5$ V/cm. We can see that such fields can technically be produced easily only in relatively small electrode gaps amounting to a few centimeters. This leads to the necessity of using an electric field applied transversely across the laser tube.

The most effective method of overcoming the difficulties encountered in the initiation and maintenance of a homogeneous pulsed discharge in a high-pressure gas is to use an additional ionization source.[49] When such a source creates a volume-homogeneous weakly ionized plasma, an electric field is applied to the discharge gap and the amplitude of this field is no longer governed by the breakdown conditions but it is selected so as to achieve optimal excitation of the laser. Additional ionization can be provided effectively by a source of photoionizing ultraviolet radiation or by a low-power beam of fast electrons. These additional ionization sources ensure that the electron density in the active medium is $\sim 10^9-10^{12}$ cm^{-3}. Therefore, the threshold electron density $N_e \sim 10^{14} - 10^{15}$ cm^{-3} can be reached by ensuring such a value of E/N that the electron density increases by several orders of magnitude during a pulse. This allows us to estimate the optimal value of E/N. For example, let us assume that the characteristic ionization time satisfies the relationship

$$\tau_{\text{ion}} = \frac{1}{Nk_{\text{ion}}(E/N)} \sim 10^{-9}\ \text{sec}. \qquad (50')$$

Then, assuming some specific pressure, we obtain an equation for the determination of the ratio E/N. Using the experimental values of $\alpha_{\text{ion}}(E/N)$ given, for example, in Refs. 38, 50, we obtain the values of E/N needed to ensure a discharge with the required parameters in He, Ne, and Ar: these values are $E/N = 5 \times 10^{-16}$, 4×10^{-16}, and 10^{-15} V·cm^2, respectively. At lower pressures the necessary value of E/N is higher. A much higher value of E/N is required in the absence of an additional ionization source: in this case we have to ensure that τ_{ion} is 2–3 orders of magnitude less than the characteristic time for the establishment of a population inversion.

These estimates are rough and intended only by way of qualitative illustration but they allow us to understand and formulate the physical conditions which are optimal from the point of view of organization of a high-pressure pulsed discharge. First of all, high pressure (~1 atm) of a buffer gas allows us to ensure a faster development of the pulsed ionization and optimal values of the electron density $N_e \sim 10^{14} - 10^{15}$ cm^{-3}. Secondly, the use of an independent initial ionization source makes it possible to reduce greatly the applied voltage keeping it at a level which is optimal from the point of view of the population inversion. Finally, we must mention another feature of the electric-discharge excitation of excimer lasers which is that the density of the inert gas from which the excimer molecules are formed should be less than the desnity of the buffer gas. In fact, as shown above, the threshold value of the rate of formation of excited inert gas atoms

$$\frac{dN^*}{dt} \sim NN_e k_r \sim 10^{22}\ \text{cm}^{-3}\cdot\text{sec}^{-1} \qquad (51)$$

in the $N_e \geqslant 10^{14}$ cm^{-3} case is exceeded if $N \sim 10^{17}$ cm^{-3} (k_r is the rate of excitation of the resonant states of the Xe, Kr, or Ar atoms; according to calculations,[31] under optimal conditions the value of this constant is greater than or of the order of 10^{-9} cm^3/sec).

A further increase of the density N of the inert gas participating in the excimer molecule formation increases the rate of generation of excited atoms and, apparently, should help to improve the output parameters of the laser. However, the resultant accumulation of the excited atoms in the active medium alters the nature of the ionization process: instead of the direct ionization, the prime role is now played by a multistep ionization process whose efficiency rises steeply as the density of the excited atoms is increased.[51] This results in an instability accompanied by an avalanche-like increase in the electron density. In an excimer laser, when the gas contains a small amount of an electronegative impurity, this instability develops at a sufficiently high density of the excited inert gas atoms when the characteristic ionization time of the excited atoms is less than twice the electron trapping time.[51] Detailed numerical calculations[51] show that when a certain value of the excited atom density is exceeded, an instability of this kind results in a sharp drop in the proportion of the energy used to form the metastable species of the inert gas.

The efficiency of the electric-discharge excitation of

excimer lasers is approximately as high as that of the electron-beam method. This is due to the fact that under the optimal excitation conditions the proportion of the energy input which is transformed into the energy of the excited atoms is very high for a pulsed discharge in an inert gas and it may represent a few tens of percent. Consequently, the efficiency of such laser systems is frequently of the order of 1% and sometimes[7,52] ist value exceeds 10%. The specific output energy is still limited by Eq. (43). A value close to this limit, $\xi \sim 3.2\times10^{-2}$ J/cm^3, is reported by Ault *et al.*,[7] who used an electron beam as the additional ionization source.

3. SPECIFIC SYSTEMS

The existing excimer lasers have been constructed relatively recently and little experimental work has been done so far. In most cases the report of a construction of some particular excimer laser gives only a description of the apparatus and the output characteristics obtained for specific parameters of the active medium and pumping. The absence of measurement of such important properties as the gain, density of the excited and charged particles (as a function of the longitudinal and transverse coordinates of the active medium), and dependences of the laser output power on the pumping parameters and the pressure of the active medium, makes it difficult to understand the laser excitation mechanism and to established the optimal conditions for its use. In particular, the observation that the output parameters vary by several order of magnitude from one system to another, whereas the parameters of the systems themselves differ only in respect to the gas composition, has not yet been explained. Table VI compares the results obtained for different laser systems. We shall analyze these results for each type of laser listed in this table.

a) Xe_2 laser

This is one of the gas lasers emitting at the shortest wavelengths. It is distinguished by a high output power (in excess of a megawatt) in the vacuum ultraviolet range and by the ability to tune continuously the emission wavelength over a fairly wide range. Figure 5 shows the dependence of the output power of a tunable Xe_2 laser on the emission wavelength.[54]

A special feature of the xenon laser is that amplification is observed only at very high pressures $p_{Xe} \sim 10^4$ Torr. This is due to the fact that the width of the gain

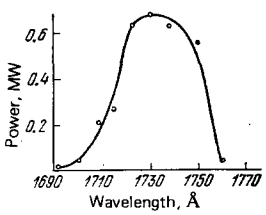

FIG. 5. Dependence of the output power of a tunable xenon laser on the emission wavelength.[54]

profile of the active medium governed by the strongly repulsive nature of the lower laser term (Fig. 2a), is very great and—as indicated by the data of Table VI—corresponds to about 5×10^{14} sec^{-1}. This high value of the intrinsic line width of the transition is independent of the gas density practically throughout the whole range of variation of the density that can be obtained under practical conditions. Therefore, in contrast to other gas lasers, an increase in the pressure in a zenon laser to several tens of atmospheres results, for fixed parameters of the pump source, in an increase in the gain.

On the other hand, for this value of the line width and with the probability of a spontaneous radiative transition $\sim 10^8$ sec^{-1} a considerable gain of ~ 0.1 cm^{-1} is achieved in accordance with Eq. (40a) when the concentration of the excited xenon excimer molecules is $N_{Xe_2^*} \sim 10^{16}$ cm^{-3}. Such high concentrations of the excited molecules are achieved only at pressures of the order of tens of atomospheres using a high-density beam of fast electrons. Thus, if we assume that the radiative lifetime of the Xe_2^* molecule is 10^{-8} sec, we find that a gain of the order of 10^{-1} cm^{-1} is achieved when the volume rate of formation of the excimer molecules is $\sim 10^{24}$ cm$^{-3}\cdot$ sec^{-1}. This imposes very stringent requirements on the parameters of the process of pumping a Xe_2 laser. Even when each electron-ion pair formed in the gas is converted effectively into a dimer molecule, the above requirement means that the specific excitation power should be of the order of 10^7 W/cm^3. In fact, the maximum efficiency of conversion of the energy of a high-power electron beam into the excitation energy of the Xe_2^* excimer molecules is about 10%,[37] which corresponds to the coefficient of conversion of the electron-ion pairs into the excimer molecules amounting to ~0.3. Thus, the above minimum excitation efficiency is underestimated severalfold and one in fact requires still higher values. This demonstrates the technical difficulties, particularly in the case of excitation of a large volume of the active medium.

It is clear from Table VI that relatively small volumes of the active medium have been excited in actual experiments. Consequently, the intensity of the emitted laser radiation has been low, so that the main mechanism of destroying the excited excimer molecules is not the stimulated but the spontaneous radiative process. In fact, the ratio of the probabilities of the stimulated and spontaneous emission from a molecule is

$$\frac{B_{21}}{A_{21}} = \frac{\rho c \sigma_{21}}{A_{21}} = \frac{P\lambda^2}{\hbar\omega S 4\Delta\omega}, \qquad (52)$$

where P is the output power of the laser; λ, ω, and $\Delta\omega$ are the wavelength, frequency, and line width of the laser radiation; σ_{21} is the stimulated emission cross section; ρ is the photon density in the resonator. If the above ratio is much less than unity, then only a very small proportion of the excited molecules participates in the stimulated emission because the majority is deexcited before there is a chance of laser action. Substituting $\Delta\omega \approx 6\times10^{14}$ sec^{-1} in Eq. (52), we find that the parameter given by this ratio is 10^{-2}, 1, and 10 for

the experimental conditions reported in three published investigations.[56,54,57] The values of the laser efficiency are approximately in the same ratio as the values of the reduced parameters. Hence, it follows that the efficiency of a xenon laser is governed by the ratio of the power of the radiation inside the resonator to the saturation parameter, given by

$$I_0=\frac{4\hbar\omega\Delta\omega}{\lambda^2}\approx 8\cdot 10^6\ \text{W/cm}^2. \tag{53}$$

Therefore, a radical way of increasing the laser efficiency to a value greater than or of the order of 1% involves an increase in the output power of the laser and this apparently can be achieved by increasing the volume of the active medium.

If we use Eq. (4) for the slowing down of fast electrons in a dense gas, we find that for electrons of energy of $\varepsilon \sim 1$ MeV the mean free path l is related to the xenon density N_{Xe} by

$$N_{Xe}l\sim\frac{\varepsilon^2}{\pi e^4 Z\Lambda}\approx 6.5\cdot 10^{22}\ \text{cm}^{-2}.$$

Bearing in mind[4)] that the xenon density N_{Xe} is limited to 10^{21} cm^{-3}, we may conclude that an increase in the output power and, consequently, in the efficiency of a xenon laser is possible when an electron beam in injected across the optic axis of the laser. This makes it possible to increase the length of the active medium by the use of ribbon-like electron beams, keeping constant the value of the input energy density and, consequently, the value of the gain.

b) Kr_2 and Ar_2 lasers

It is clear from Table VI that the information on these lasers is far too limited to make reliable conclusions on their future development and uses. The population inversion mechanism and the factors limiting the specific output power of these lasers are similar to those of the Xe_2 laser discussed above.[37] In particular, the efficiency of conversion of the electron-beam energy into the energy of the radiation emitted from the Ar_2 dimer molecules is $\approx 15\%$[37] and at pressures amounting to tens of atomspheres it is independent of the pressure. The difficulties in investigation of these dimer lasers are associated with even shorter (than in the case of Xe_2) emission wavelengths and the consequently higher values of the saturation parameter.

c) ArO, KrO, and XeO lasers

Lasers of this type emit due to transitions between the states of quasimolecules formed from a metastable oxygen atom, $O(^1S)$ or $O(^1D)$, and an inert gas atom. Radiative transitions between these states are forbidden for a free oxygen atom but the forbiddeness is lifted as a result of formation of a weakly bound quasimolecule of the inert gas oxide. If a laser is excited by a fast-electron beam, a considerable part of the energy of this beam is rapidly converted into the energy of the excited inert gas atoms (excitation energy in the range 9–11 eV) as a result of the processes (5)–(7) at inert gas pressures of the order of a few atmospheres. The subsequent reaction

$$R^* + O_2 \rightarrow R + O(^3P) + O(^1S) \tag{54}$$

produces oxygen atoms in the metastable ^1S state if the excitation energy of the inert gas atoms is at least 9.3 eV.[74,75] Formation of excimers occurs as a result of the triple association process

$$2R + O(^1S) \rightarrow RO(^1S) + R. \tag{55}$$

In this case the binding energy of the $RO(^1S)$ excimer molecules is very low, which is indicated by the fact that the wavelength of the transition of such molecules differs by not more than 5% from the wavelength of the $O(^1S)$–$O(^1D)$ transition in pure oxygen (see Table VI). This accounts for the small proportion of the bound $RO(^1S)$ molecules compared with the number of the metastable $O(^1S)$ atoms and, consequently, for the relatively poor output parameters of lasers of this type. This is confirmed by measurements of the dependences of the intensity of spontaneous and stimulated radiation on the gas temperature.[62,74] According to these measurements a reduction in the gas temperature from 300 to 240 °K increases the efficiency of conversion of the input energy into radiation energy of the $XeO(^1S)$ excimers by a factor of about 8.[74] Using this result, we can estimate the binding energy of the $XeO(^1S)$ molecule to be ~0.2 eV. Hence, we may conclude that the main limitation on the output parameters of lasers of this type is the heating of the active medium and the consequent dissociation of the $RO(^1S)$ molecules.

d) XeF, ArF, and KrF lasers

These three types of excimer laser stand out because of the exceptionally good output parameters. Their output power (in excess of 10^9 W), efficiency (above 1%), and output energy (tens of joules) represent record values among all lasers emitting in the visible and ultraviolet range. The XeF, ArF, and KrF lasers have many properties in common and they are frequently excited using the same apparatus but simply changing the gas composition. Two excitation methods have been adopted, one of which is based on the use of a fast-electron beam and the other on the use of a transverse electric discharge. It is interesting to note that the actual composition of the active medium is different for these two excitation methods. For example, in the electron-beam excitation the main component of the active medium is usually argon. In lasers excited by an electric discharge the active medium is usually based on helium. We shall now consider the characteristic features of these two methods of excitation of the fluoride lasers in question.

We shall analyze the lasers utilizing inert gas monofluorides on the basis of the result reported by Hoffman *et al.*,[6] who obtained record output parameters for the ArF and KrF lasers. We shall consider in detail the processes in the active medium of the ArF laser, be-

[4)] See Figs. 3 and 4 and the corresponding text.

cause it consists of just two components, which are Ar and F_2 (Table VI). An electron beam of ~1 MeV energy, 55 kA current, and 55 nsec pulse duration entered longitudinally a tube 15 cm in diameter and 2 m long.[6] The path traveled by the beam electrons until they were completely stopped was equal to the length of the tube. Assuming that the electron beam filled uniformly the cross section of the tube, we concluded—in accordance with Eq. (4)—that the specific pump power in the central (along the length) part of the tube was $P \approx 10^6$ W/cm^3. Consequently, the specific pump energy was $\approx 8.5\times10^{-2}$ J/cm^3. The specific laser radiation energy was $\approx 3\times10^{-3}$ J/cm^3, so that the efficiency of conversion of the input energy into laser radiation energy was close to 3%. We shall now try to analyze whether this is the maximum attainable result or whether it can be improved considerably. As established in a number of investigations (see, for example, Gerardo and Johnson[76]), the slowing down of a beam of fast electrons in high-pressure argon is characterized by about 20% efficiency of the conversion of the beam energy into the energy of the excited Ar(^3P) atoms. The energy of the laser photon emitted from the ArF molecule is half the excitation energy of Ar(^3P) and, therefore, we may conclude that under the conditions in the experiments of Hoffman *et al.*[6] the excited Ar(^3P) atoms were utilized with an efficiency of only 30%. The main source of the undesirable losses was evidently spontaneous emission, which very effectively depleted the population inversion during the initial part of the pump pulse. In fact, the duration of the pump pulses ~55 nsec was comparable with twice the time taken by photons to cross the laser tube (~15 nsec). The spontaneous deexcitation time (~10 nsec) was less than this quantity and, therefore, the spontaneous deexcitation resulted in an effective emptying of the upper laser level until the growth of the stimulated emission reached saturation. A comparison of these times indicates that a considerable proportion of the energy injected into the active medium is lost. Figure 6 compares the oscillograms of the exciting beam current and the density of the stimulated radiation emitted from the ArF laser,[6] which illustrate these conclusions.

When a small amount of krypton (tens of Torr) was added to a Ar + F_2 mixture, stimulated emission from the KrF excimer was observed whereas the intensity of the emission from ArF decreased falling to zero at the Kr pressure of about 100 Torr (Fig. 7). This was attributed to the nonresonant excitation transfer

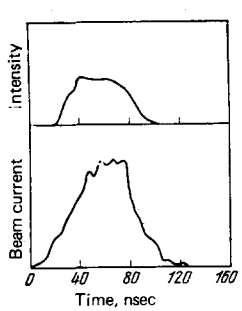

FIG. 6. Oscillograms of the electron-beam current and output radiation intensity recorded for an ArF laser.[6]

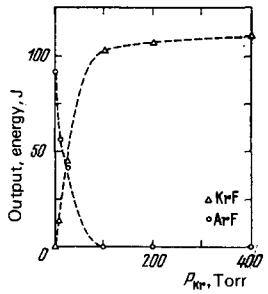

FIG. 7. Dependence of the output energy of ArF and KrF lasers on the krypton pressure[6] (p_{F_2} = 4 Torr, total pressure 1400 Torr, excitation conditions given in Table VI).

$$Ar^* + Kr \rightarrow Ar + Kr^*, \qquad (56)$$

which—at sufficiently high krypton pressures—was much faster than the reaction of excited argon with fluorine (9). The subsequent reaction

$$Kr^* + F_2 \rightarrow KrF^* + F$$

led to the formation of the KrF* excimer molecules and the transitions in these molecules were responsible for the stimulated emission. Stimulated emission was not observed from KrF* when pure krypton was mixed with fluorine. This was clearly due to the circumstance that the excited krypton atoms formed as a result of the dissociative recombination of the molecular ions, interacted less rapidly with the molecular fluorine producing the excimer. As a result of the excitation transfer process (56) the krypton atoms are formed mainly in states with higher energy than as a result of the dissociative recombination. In a state of this kind [apparently this was one of the states with the $4p^4(^2P)5p$ configuration and an excitation energy of about 12 eV] the substitution reaction, producing the KrF* excimer, was much more rapid.

The data of Fig. 7 can be used ot obtain some information on the constants of the processes involved in the conversion of the energy of the excited argon atoms formed by the dissocitative recombination process. It is specifically the fact that the output energies of the ArF and KrF lasers are similar at $P_{Kr} \approx 50$ Torr that indicates that the characteristic time of the transfer of excitation from argon to krypton is approximately equal to the corresponding time for the reaction of excited argon with fluorine:

$$N_{Kr}k_{tr} \approx N_{F_2}k_p,$$

from which it follows that the ratio of the rate constants is $k_p/k_{tr} \approx 12$. This estimate is in agreement with our rate constants $k_p \approx 10^{-9}$ cm^3/sec and $k_{tr} \approx 10^{-10}$ cm^3/sec obtained for other excited inert gas atoms and other halogens (Table III). For these values of the constants it is found that the characteristic radiative lifetime of the excited excimers ArF and KrF is ~10^{-8} sec, which again is in agreement with the ideas put forward above.

The system analyzed above is a very cumbersome and expensive apparatus, whose construction requires overcoming some serious technical difficulties. However, there are much simpler and compact systems charac-

terized by quite high output parameters. By way of example, we shall consider the system described in the paper by Champagne *et al.*,[41] which has been cited earlier. The laser cell used is shown in Fig. 8. A beam of 300 keV electrons was formed by a cold-cathode electron gun. A beam current pulse of ~10 A/cm^2 density and 1 μsec duration penetrated titanium foil 26 μ thick. The region between the resonator mirrors was a cylinder 1 m long and 2.6 cm in diameter. This region was filled with a mixture of Ar, Xe, and NF_2 in the ratio 99.5:0.38:0.12, the total pressure being about 2.5 atm. The electron-beam excitation produced stimulated emission pulses from the XeF excimer: these pulses were of 1 μsec duration and of about 0.3 J energy. Since the electron beam energy absorbed in the active medium was ~60 J, the energy conversion efficiency was ~0.5%. Long output pulses were obtained using sufficiently powerful electron-beam pulses of long duration. The possibility of a further increase of the laser pulse duration was indicated by the observation that the stimulated emission stopped at practically the same moment as the beam current.

Monohalide excimer lasers excited by electric discharge pulses are even more compact and simpler. Usually these systems are based on the designs developed earlier for the excitation of high-pressure CO_2, N_2, CO, and other pulsed lasers, and therefore the corresponding devices are, as a rule, sufficiently highly developed and convenient for laboratory applications. One of such systems[40] was based on a commercial CO_2 laser system with transverse excitation (Tachisto TAC-11) and preliminary ionization by ultraviolet radiation. The discharge chamber dimensions were 60×2×0.5 cm. The energy stored in a capacitor was 10 J. Preliminary ionization of the active medium was provided by a spark discharger located at a distance 4 cm from the resonator axis. A ~25 kV voltage pulse of the main discharge was applied 0.1–25 μsec after the preionization pulse and lasted 40 nsec. Depending on the composition of the mixture, the system could generate stimulated emission from XeF, KrF, or ArF, Helium was used as the buffer gas. The output energies were determined for various lasers as a function of the percentage of the main inert gas (Fig. 9). The total pressure in the mixture was 1000 Torr and the pressure of F_2 was 3 Torr. In all cases the maximum output energy was about 0.1 J per pulse, which corresponded to an overall laser efficiency of the order of 1%. The optimal composition of the mixture varied from laser to laser apparently because the discharges behaved in different ways in different gas mixtures for a given voltage across the gap. Clearly an efficiency of ~1% is

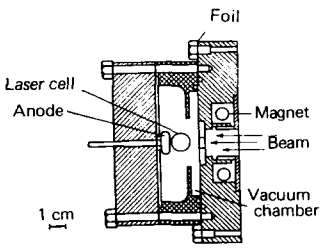

FIG. 8. Laser cell used by Champagne *et al.*[41]

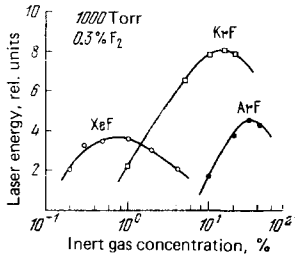

FIG. 9. Dependence of the output energy of laser radiation on the inert gas concentration.[40]

achieved in the case of electric-discharge excitation more easily than in the electron-beam excitation case.

Another important feature of the electric-discharge excitation method is the feasibility of pulse-periodic operation. This extends greatly the range of possible applications of these lasers. The value of the pulse repetition frequency is limited by the requirement that the characteristic time for the chemical recovery of the halogen-containing substance should not exceed the time during which a considerable proportion of this substance, participating in a chemical reaction, is used up. On the basis of this requirement we can estimate the maximum average (per period) specific power:

$$P_{av} \approx \frac{N_{F_2}}{\tau_{chem}} \hbar\omega \approx \frac{N_{0F_2}^2 k_{rec}}{4} N_R \hbar\omega \approx 10^3 - 10^4 \text{ W/cm}^3;$$

here, $\tau_{chem}^{-1} = N_F N_R k_{rec}$ is the characteristic recombination time of the halogen-containing molecule (the third particle is the inert gas of density N_R); N_{0F_2} is the initial density of fluorine. In obtaining the above estimate we have assumed that only a small proportion of the moelcules react during each pulse because the average time between the pulses is much shorter than the time needed for the chemical recovery of the gas. The average power attained in practice is considerably less than that given by the above estimate. The operation of a laser usually results in irreversible chemical changes in the active medium and, therefore, pulse-periodic operation requires circulation of the gas through the discharge region, though the rate of circulation may be slow. It is then possible to obtain stimulated emission at a repetition frequency of ~100–200 Hz. For example, pulse-periodic operation of a KrF laser of simple construction and with a double transverse discharge was recently reported.[77] A chamber of 40×2×0.3 cm dimensions was filled with a He:Kr:SF_6 mixture and excited with 5–7 nsec pulses of about 8 J energy. A slow circulation of the mixture at a rate of 0.1 liter/min made it possible to generate output pulses at a repetition frequency of 100 Hz and an average laser power of about 200 mW.

An even higher pulse repetition frequency—about 200 Hz—was achieved by Christensen[78]: in this case the gas was driven at a rate of 10 liter/min through the system and the average power output was 60 mW. An interesting feature of the system was the closed circulation of the active mixture through a ballast chamber.

e) XeBr, XeCl, KrCl, ArCl, and NeF lasers

These lasers excited by an electron beam have been investigated relatively recently. The poor output pa-

rameters of the XeBr laser[63] could be due to two factors. On the one hand, the considerable width of the gain profile of the active medium ($\Delta\lambda \sim 1300$ Å) made it very difficult to achieve saturation of the active transition in this laser, so that the excited XeBr* excimer molecules were deexcited mainly by spontaneous emission. On the other hand, a reduction in the output paramaters of the laser could be due to the quenching of the excited xenon atoms

$$Xe^* + Br_2 \rightarrow Xe + Br + Br,$$

which was a process with a constant that could be several times greater than the rate of formation of the XeBr excimer molecules. The output parameters of the XeCl and KrCl lasers seem to be more promising but final conclusions will not be possible until more detailed experiments have been carried out.

CONCLUSIONS

We shall conclude the review by drawing attention to the constantly widening range of variation of the parameters of excimer lasers and the associated expansion of their potential application. For example, the most attractive feature of excimer lasers is apparently related to the possibility of continuous tuning of the wavelength over quite wide ranges of visible and ultraviolet radiation. This is illustrated in Fig. 10, where the dashed line surrounds the parts of the spectrum within the gain band of excimer lasers. In spite of the very approximate nature of such a diagram, it follows from this diagram that—in principle—excimer lasers may cover about 20% of the ultraviolet range. This can be extended even further by the use of excimer lasers to pump tunable liquid lasers. Progress depends on the success in the search for dyes emitting ultraviolet wavelengths. The first experiment of this kind[75] was carried out using a KrF laser to pump a solution of *p*-terphenyl in *p*-dioxane. Stimulated emission was found to be continuously tunable over the wavelength range 335–346 μ with a peak power output of 3 kW and a pulse duration of ~20 nsec. Another way of extending the spectral range of the excimer laser radiation involves the use of Raman scattering. The high energy

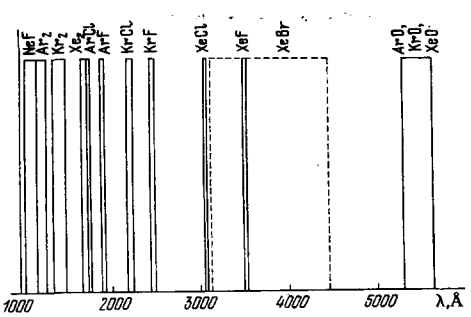

FIG. 10. Parts of the optical spectrum corresponding to potential tuning range of excimer lasers. An anomalously wide gain profile of the XeBr molecule is reported in just one paper [39] and it is unlikely to represent the possible width of the tuning range of the XeBr laser.

density of the excimer laser radiation permits the attainment of very high values of the efficiency of the conversion process which then reduces the output frequency by an amount which is a multiple of the energy of a vibrational quantum of the illuminated gas. In one such experiment,[79] gaseous H_2, D_2, and CH_4 compressed to about 50 atm were used to convert the output frequencies of the KrF and ArF excimer lasers. When the power density of the laser radiation was $\sim 10^9 - 10^{10}$ W/cm^2 and the pulse duration was $\sim 10^{-8}$ sec, a number of lines of coherent emission was observed in the Stokes and anti-Stokes regions and the conversion efficiency reached 50%. Moreover, the conversion efficiency of the XeF laser radiation into radiation of the 5850 Å wavelength, achieved by the Raman scattering of this radiation by Ba vapor, was close to unity.[80]

The most fruitful applications of excimer lasers with continuously tunable ultraviolet wavelengths are clearly those in isotopically selective laser photochemistry. In view of this, it is interesting to note the recent report[81] of the use of the ArF laser radiation ($\lambda \approx 1930$ Å) for the enrichment of oxygen isotopes. The laser was excited by a double electron discharge and emitted pulses of 25 nsec duration, 0.05 J energy, and 1 Hz repetition frequency. The width of the stimulated emission line was of the order of 10 Å. The laser radiation wavelength corresponded to the Schumann–Runge absorption band of molecular oxygen, whose upper state $B^3\Sigma_u^-$ was of the predissociation type. The radiation passed through natural air, which filtered off the part of the spectrum absorbed by the $^{16}O_2$ molecules. The radiation filtered in this way was directed to a cell containing molecular oxygen of normal isotopic composition at a pressure of 10 atm and it was absorbed only by the molecules which contained the ^{17}O and ^{18}O atoms. The oxygen atoms formed as a result of predissociation were characterized by an increase in the heavy-isotope content and were rapidly bound by the simple reaction

$$O + O_2 + M \rightarrow O_3 + M$$

(*M* was a third particle), which was easily detected by means of silica gel crystals present in the cell which absorbed the molecular ozone. After irradiation of the mixture with 500 laser pulses, the enrichment coefficient was close to 100%. This demonstartion experiment was carried out under conditions far from optimal from the point of view of enrichment yield and efficiency but provided a clear idea of the possibilities of one of the potential applications of excimer lasers.

Another range of potential applications of excimer lasers involves the interaction of high-power ultraviolet radiation with matter, particularly, laser heating and compression of matter with the aim of bringing about thermonulcear reactions. In this case the use of excimer lasers meets with two serious problems, one of which is the need to generate high-power pulses of short duration (≤ 1 nsec) and the other is the need to reduce the angular divergence of the laser radiation, which is a prerequisite of satisfactory focusing. Recent investigations report encouraging results along the path of overcoming these problems. For example, Christensen *et al.*[82] generated XeF laser pulses of about

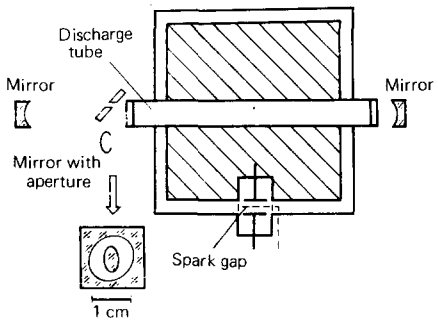

FIG. 11. Configuration of the unstable resonator used by McKee *et al.*[84]

2 nsec duration by the mode-locking technique. Even shorter pulses were obtained from the same laser by Tomov *et al.*[83] who used the active medium of the XeF laser to amplify the third harmonic of the radiation emitted from a neodymium glass laser. This harmonic was of wavelength close to the emission wavelength of the XeF excimer molecule and was applied in the form of a train of pulses of about 0.2 nsec each (such a train was obtained by active mode locking). One of the pulses was amplified as a result of its passage through the active medium of the SeF laser, which increased the intensity by a factor of 6000 to ~50 kW. The active medium of the laser was the $He:Xe:NF_3 = 250:2.5:1$ mixture at a total pressure of 2 atm, filling a volume of 85 cm^3. The excitation was provided by a transverse electric discharge after preliminary ionization and the excitation energy was about 0.5 . Although the output energy per pulse was relatively low, this experiment demonstrated one of the ways of solving the problem of reducing the laser pulse duration.

Encouraging results were also obtained by McKee *et al.*[84] in efforts to reduce the angular divergence of the radiation emitted from excimer lasers. They used for the first time an unstable resonator in an excimer laser, which immediately resulted in over a tenfold reduction in the angular divergence of the laser radiation. An XeF or KrF exximer laser was excited by a pulse discharge in a gap 1.8 cm wide between electrodes 87 cm long; the voltage across the electrodes was 30 kV which was applied in pulses of ~10 nsec duration and that carried an energy of ~5 J stored in a Blumlein line. The composition of the mixture in the case of the XeF laser was $He:Xe:F_2 = 96.5:1.5:2$ at a total pressure of 450 Torr; in the caes of the KrF laser the composition was $He:Kr:F_2 = 93:6:1$ at a total pressure of 600 Torr. McKee *et al.* used the unstable resonator configuration shown in Fig. 11; this figure also gives the characteristic shape of the output spot. The use of this resonator reduced the angular divergence of the laser radiation from 3–5 to ≤ 0.2 mrad without a significant change in the energy characteristics: the output energy of the laser was $(5-7)\times10^{-2}$ J, corresponding to an efficiency in excess of 1%. The diameter of the exit aperture was ~0.5 cm and the reduced angular divergence was then close to the diffraction limit of $\sim 10^{-4}$ rad.

These briefly reviewed directions for the development of the physics and technology of excimer lasers should result in a considerable extension of the capabilities of these lasers. Other equally important problems are still to be solved, including continuous frequency tuning of the radiation emitted from excimer lasers, as well as the construction of cw or quasi-cw systems. There is no double that research directed to the construction, improvement and application of excimer lasers, will lead to further progress in quantum electronics.

[1]N. G. Basov, E. M. Balashov, O. V. Bogdankevich (Bogdankevitch), V. A. Danilychev, G. N. Kashnikov, N. P. Lantzov, and D. D. Khodkevich (Khodkevitch), J. Lumin. **1**, 834 (1970).
[2]N. G. Basov, V. A. Danilychev, Yu. M. Popov, and D. D. Khodkevich, Pis'ma Zh. Eksp. Teor. Fiz. **12**, 473 (1970) [JETP Lett. **12**, 329 (1970)].
[3]H. A. Koehler, L. J. Ferderber, D. L. Redhead, and P. J. Ebert, Appl. Phys. Lett. **21**, 198 (1972).
[4]J. E. Velazco and D. W. Setser, J. Chem. Phys. **62**, 1990 (1975); IEEE J. Quantum Electron. **QE-11**, 708 (1975); L. G. Piper, J. E. Velazco, and D. W. Setser, J. Chem. Phys. **59**, 3323 (1973); J. E. Velzco, J. H. Kolts, and D. W. Setser, J. Chem. Phys. **65**, 3468 (1976).
[5]G. C. Tisone, A. K. Hays, and J. M. Hoffman, Opt. Commun. **15**, 188 (1975).
[6]J. M. Hoffman, A. K. Hays, and G. C. Tisone, Appl. Phys. Lett. **28**, 538 (1976).
[7]E. R. Ault, M. L. Bhaumik, and R. S. Bradford Jr., Bull. Am. Phys. Soc. **21**, 169 (1976); R. S. Bradford Jr., E. R. Ault, and M. L. Bhaumik, Opt. Commun. **18**, 116 (1976); R. S. Bradford Jr., W. B. Lacina, E. R. Ault, and M. L. Bhaumik, Opt. Commun. **18**, 210 (1976).
[8]B. M. Smirnov, Dokl. Akad. Nauk SSSR **183**, 554 (1968 [Sov. Phys. Dokl. **13**, 1148 (1969)]; L. Ya. Efremenkova and B. M. Smirnov, Dokl. Akad. Nauk SSSR **203**, 779 (1972) [Sov. Phys. Dokl. **17**, 336 (1972)].
[9]R. S. Mulliken, J. Chem. Phys. **52**, 5170 (1970).
[10]T. H. Dunning Jr. and P. J. Hay, Appl. Phys. Lett. **28**, 649 (1976); P. J. Hay and T. H. Dunning Jr., J. Chem. Phys. **66**, 1306 (1977).
[11]C. A. Brau and J. J. Ewing, J. Chem. Phys. **63**, 4640 (1975).
[12]B. M. Smirnov, Asimptoticheskie metody v teorii atomnykh stolknovenii (Asymptotic Methods in the Theory of Atomic Collisions), Atomizdat, M., 1973.
[13]J. Tellinghuisen, J. M. Hoffman, G. C. Tisone, and A. K. Hays, J. Chem. Phys. **64**, 2484 (1976); J. Tellinghuisen, A. K. Hays, J. M. Hoffman, and G. C. Tisone, J. Chem. Phys. **65**, 4473 (1976); J. Tellinghuisen, G. C. Tisone, J. M. Hoffman, and A. K. Hays, J. Chem. Phys. **64**, 4796 (1976).
[14]F. J. Adrian and V. A. Bowers, J. Chem. Phys. **65**, 4316 (1976).
[15]J. Goodman and L. E. Brus, J. Chem. Phys. **65**, 3808 (1976).
[16]B. S. Ault and L. Andrews, J. Chem. Phys. **65**, 4192 (1976).
[17]U. Fano, Annu. Rev. Nucl. Sci. **13**, 1 (1963).
[18]G. Wallis, K. Sauer, D. Sunder, S. E. Rosinskiĭ, A. A. Rukhadze, and V. G. Rukhlin, Usp. Fiz. Nauk **113**, 435 (1974) [Sov. Phys. Usp. **17**, 492 (1975)].
[19]B. M. Smirnov, Iony i vozbuzhdennye atomy v plazme (Ions and Excited Atoms in Plasma), Atomizdat, M., 1974.
[20]J. A. Mangano, J. H. Jacob, and J. B. Dodge, Appl. Phys. Lett. **29**, 426 (1976).
[21]E. R. Ault, R. S. Bradford Jr., and M. L. Bhaumik, Appl. Phys. Lett. **27**, 413 (1975).
[22]G. A. Hart and S. K. Searles, J. Appl. Phys. **47**, 2033 (1976).
[23]J. K. Rice and A. W. Johnson, J. Chem. Phys. **63**, 5235 (1975).
[24]E. Ellis and N. D. Twiddy, J. Phys. B **2**, 1366 (1969).
[25]J. W. Keto, R. E. Gleason Jr., and G. K. Walters, Phys. Rev.

Lett. **33**, 1365 (1974).
[26]E. V. George and C. K. Rhodes, Appl. Phys. Lett. **23**, 139 (1973).
[27]C. W. Werner, E. V. George, P. W. Hoff, and C. K. Rhodes, Appl. Phys. Lett. **25**, 235 (1974).
[28]E. E. Huber Jr., L. R. Jones, E. V. George, and R. M. Lerner, IEEE J. Quantum Electron. **QE-12**, 353 (1976).
[29]M. Bourene, O. Dutuit, and J. Le Calve, J. Chem. Phys. **63**, 1668 (1975).
[30]J. A. Mangano and J. H. Jacob, Appl. Phys. Lett. **27**, 495 (1975).
[31]C. P. Wang, H. Mirels, D. G. Sutton, and S. N. Suchard, Appl. Phys. Lett. **28**, 326 (1976).
[32]C. A. Brau, Appl. Phys. Lett. **29**, 7 (1976).
[33]A. V. Eletskiĭ and B. M. Smirnov, in Modelirovanie i metody rascheta fiziko-khimicheskikh protsessov v plazme, pod. red. L. S. Polaka (in: Simulation and Methods of Calculation for Physicochemical Processes in Plasma, ed. by L. S. Polak), Nauka, M., 1974.
[34]A. V. Eletskiĭ and B. M. Smirnov, Zh. Tekh. Fiz. **38**, 3 (1968) [Sov. Phys. Tech. Phys. **13**, 1 (1968)].
[35]A. V. Eletskiĭ and V. D. Kulagin, Fiz. Plazmy (in press) [Sov. J. Plasma Phys. (in press)].
[36]I. I. Ostroukhova, B. M. Smirnov, and G. V. Shlyapnikov, Zh. Eksp. Teor. Fiz. **73**, 166 (1977) [Sov. Phys. JETP **46**, (1977)].
[37]H. A. Koehler, L. J. Berderber, D. L. Redhead, and P. J. Ebert, Phys. Rev. A **9**, 768 (1974); **12**, 968 (1975).
[38]A. V. Eletskiĭ, L. A. Palkina, and B. M. Smirnov, Yavleniya perenosa v slaboionizovannoĭ plazme (Transport Phenomena in Weakly Ionized Plasma), Atomizdat, M., 1975.
[39]H. A. Hyman, Appl. Phys. Lett. **31**, 14 (1977); K. J. McCann and M. R. Flannery, Appl. Phys. Lett. **31**, 599 (1977).
[40]R. Burnham, F. X. Powell, and N. Djeu, Appl. Phys. Lett. **29**, 30 (1976).
[41]L. F. Champagne, J. G. Eden, N. W. Harris, N. Djeu, and S. K. Searles, Appl. Phys. Lett. **30**, 160 (1977); L. F. Champagne and N. W. Harris, Appl. Phys. Lett. **31**, 513 (1977).
[42]A. V. Eletskiĭ and B. M. Smirnov, Gazovye lazery (Gas Lasers), Atomizdat, M., 1971.
[43]R. Burnham, N. W. Harris, and N. Djeu, Appl. Phys. Lett. **28**, 86 (1976).
[44]C. P. Eang, Rev. Sci. Instrum. **47**, 92 (1976).
[45]R. Burnham and N. Djeu, Appl. Phys. Lett. **29**, 707 (1976).
[46]R. Burnham, F. X. Powell, and N. Djeu, Appl. Phys. Lett. **29**, 30 (1976).
[47]J. A. Mangano and J. H. Jacob, Appl. Phys. Lett. **27**, 495 (1975).
[48]V. N. Ishchenko, V. N. Lisitsyn, and A. M. Razhev, Appl. Phys. **12**, 55 (1977); Pis'ma Zh. Tekh. Fiz. **2**, 839 (1976) [Sov. Tech. Phys. Lett. **2**, 327 (1976)].
[49]A. V. Eletskiĭ and B. M. Smirnov, Dokl. Akad. Nauk SSSR **190**, 809 (1970) [Sov. Phys. Dokl. **15**, 109 (1970)]; A. V. Eletskiĭ, Avtoreferat dokt. dissertatsii (Author's Abstract of Doctoral Thesis), Kurchatov Institute of Atomic Energy, Moscow, 1976.
[50]S. C. Brown, Basic Data of Plasma Physics, MIT Press, Cambridge, Mass., 1959 (Russ. Transl., Atomizdat. M., 1961).
[51]J. D. Daugherty, J. A. Mangano, and J. H. Jacob, Appl. Pys. Phys. Lett. **28**, 581 (1976).
[52]C. A. Brau and J. J. Ewing, Opt. Commun. **18**, 115 (1976).
[53]A. W. Johnson, J. B. Gerardo, E. L. Patterson, R. A. Gerber, J. K. Rice, and F. W. Bingham, Kvantovaya Elektron. (Moscow) **3**, 916 (1976) [Sov. J. Quantum Electron. **6**, 495 (1976)].
[54]D. J. Bradley, D. R. Hull, M. H. R. Hutchinson, and M. W. McGeoch, Opt. Commun. **11**, 335 (1974); **14**, 1 (1975).
[55]S. C. Wallace and R. W. Dreyfus, Appl. Phys. Lett. **25**, 498 (1974).
[56]A. W. Johnson and J. B. Gerardo, J. Appl. Phys. **45**, 867 (1974); J. B. Gerardo and A. W. Johnson, Appl. Phys. Lett. **26**, 582 (1975).
[57]V. A. Danilychev, O. M. Kerimov, and I. B. Kovsh, Tr. Fiz. Inst. Akad. Nauk SSSR **85**, 49 (1976).
[58]P. W. Hoff, J. C. Swingle, and C. K. Rhodes, Appl. Phys. Lett. **23**, 245 (1973).
[59]W. M. Hughes, J. Shannon, and R. Hunter, Appl. Phys. Lett. **24**, 488 (1974).
[60]W. M. Hughes, N. T. Olson, and R. Hunter, Appl. Phys. Lett. **28**, 81 (1976).
[61]H. T. Powell, J. R. Murray, and C. K. Rhodes, Appl. Phys. Lett. **25**, 730 (1974).
[62]N. G. Basov, A. N. Brunin, V. A. Danilychev, A. G. Degtyarev, V. A. Dolgikh, O. M. Kerimov, and A. N. Dobanov, Pis'ma Zh. Tekh. Fiz. **2**, 337 (1976) [Sov. Tech. Phys. Lett. **2**, 129 (1976)].
[63]S. K. Searles and G. A. Hart, Appl. Phys. Lett. **27**, 243 (1975); S. K. Searles, Appl. Phys. Lett. **28**, 602 (1976).
[64]J. J. Ewing and C. A. Brau, Appl. Phys. Lett. **27**, 350 (1975); C. A. Brau and J. J. Ewing, Appl. Phys. Lett. **27**, 435 (1975).
[65]N. G. Basov, A. N. Brunin, V. A. Danilychev, V. A. Dolgikh, A. G. Degtyarev, and O. M. Kerimov, Pis'ma Zh. Tekh. Fiz. **2**, 1057 (1976) [Sov. Tech. Phys. Lett. **2**, 417 (1976)].
[66]C. P. Wang, Appl. Phys. Lett. **29**, 103 (1976).
[67]W. J. Sarjeant, A. J. Alcock, and K. E. Leopold, Appl. Phys. Lett. **30**, 635 (1977).
[68]R. Burnham, F. X. Powell, and N. Djeu, Appl. Phys. Lett. **29**, 30 (1976).
[69]R. W. Waynant, Appl. Phys. Lett. **30**, 234 (1977).
[70]D. G. Sutton, S. N. Suchard, O. L. Gibb, and C. P. Wang, Appl. Phys. Lett. **28**, 522 (1976).
[71]M. L. Bhaumik, R. S. Bradford Jr., and E. R. Ault, Appl. Phys. Lett. **28**, 23 (1976).
[72]J. K. Rice, A. K. Hays, and J. R. Woodworth, Appl. Phys. Lett. **31**, 31 (1977).
[73]J. R. Murray and H. T. Powell, Appl. Phys. Lett. **29**, 252 (1976).
[74]V. Ya. Aleksandrov, V. Yu. Vinogradov, V. V. Lugovskiĭ, and I. V. Podmoshenskiĭ, Opt. Spektrosk. **41**, 390 (1976) [Opt. Spectrosc. (USSR) **41**, 224 (1976)].
[75]C. D. Cooper, G. C. Cobb, and E. L. Tolnas, J. Mol. Spectrosc. **7**, 223 (1961).
[76]J. B. Gerardo and A. W. Johnson, IEEE J. Quantum Electron. **QE-9**, 647 (1973).
[77]V. N. Ishchenko, V. N. Lisitsyn, and A. M. Razhev, Pis'ma Zh. Tekh. Fiz. **3**, 690 (1977) [Sov. Tech. Phys. Lett. **3**, 281 (1977)].
[78]C. P. Christensen, Appl. Phys. Lett. **30**, 483 (1977).
[79]T. R. Loree, R. C. Sze, and D. L. Barker, Appl. Phys. Lett. **31**, 37 (1977).
[80]N. Djeu and R. Burnham, Appl. Phys. Lett. **30**, 473 (1977).
[81]R. K. Sander, T. R. Loree, S. D. Rockwood, and S. M. F Freund, Appl. Phys. Lett. **30**, 150 (1977).
[82]C. P. Christensen, L. W. Braverman, W. H. Steier, and C. Wittig, Appl. Phys. Lett. **29**, 424 (1976).
[83]I. V. Tomov, R. Fedosejevs, M. C. Richardson, W. J. Sarjeant, A. J. Alcock, and K. E. Leopold, Appl. Phys. Lett. **30**, 146 (1977).
[84]T. J. McKee, B. P. Stoicheff, and S. C. Wallace, Appl. Phys. Lett. **30**, 278 (1977).
[85]M. Ghelfenstein, H. Szwarc, and R. Lopez-Delgado, Chem. Phys. Lett. **52**, 236 (1977).
[86]N. W. Winter, C. F. Bender, and T. N. Rescigno, J. Chem. Phys. **67**, 3122 (1977).
[87]W. R. Wadt, D. C. Cartwright, and J. S. Cohen, Appl. Phys. Lett. **31**, 672 (1977).
[88]M. Rokni, J. H. Jacob, J. A. Mangano, and R. Brochu, Appl. Phys. Lett. **31**, 79 (1977).
[89]A. M. Hawryluk, J. A. Mangano, and J. H. Jacob, Appl. Phys. Lett. **31**, 164 (1977).
[90]C. H. Fisher and R. E. Center, Appl. Phys. Lett. **31**, 106

(1977).
[91]C. E. Turner Jr., Appl. Phys. Lett. **31**, 659 (1977).
[92]Yu. I. Bychkov, N. V. Karlov, I. N. Konovalov, G. A. Mesyats, A. M. Prokhorov, and V. F. Tarasenko, Pis'ma Zh. Tekh. Fiz. **3**, 1041, 1233 (1977) [Sov. Tech. Phys. Lett. **3**, 427, 509 (1977); Yu. I. Bychkov, N. V. Karlov, V. F. Losev, G. A. Mesyats, A. M. Prokhorov, and V. F. Tarasenko, Pis'ma Zh. Tekh. Fiz. **4**, 83 (1978) [Sov. Tech. Phys. Lett. **4**, 34 (1978)].
[93]N. G. Basov *et al.*, Pis'ma Zh. Tekh. Fiz. **3**, 1297 (1977) [Sov. Tech. Phys. Lett. **3**, 534 (1977)].

Translated by A. Tybulewicz