

Possibility of creating new class of lasers on the repulsive terms of diatomic molecules comprising the flying apart fragments

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Analysis is being carried out in the paper of a possibility of obtaining lasing effect on the transitions between the repulsive terms (with no potential wells) of diatomic molecules comprising the flying apart fragments. In our analysis we used model potential curves of the upper and lower lasing terms of such molecules. It was assumed that such a molecule can be formed due to collisions of a *B* atom in its ground state with an *A* atom excited by electrons to an upper level with the energy E_2 . Assuming the concentration of *A* atoms to be equilibrium at the electron temperature T_e , we have found the ratio T_e/E_2 and concentrations of the atoms *A* and *B* in their ground states that favor obtaining the lasing effect on the transition between the repulsive terms of molecules comprising the flying apart fragments.

Preliminary analysis of the possibility of obtaining laser generation in the gas discharge lasers that is presented in this paper has been performed for the system with the scheme of energy levels presented in Fig. 1. In the scheme considered the generation of induced radiation occurs on a transition between two repulsive terms of a diatomic molecule of flying apart fragments. The atoms of the flying apart fragments of such a molecule can be either of the same kind or of different kinds. It is just the case when atoms *A* and *B* of different kinds form the flying apart fragments of the molecule that is schematically shown in Fig. 1.

two interacting atoms are not free and form the molecule of flying apart fragments can be roughly estimated as a sum of the atomic radii of the A_1 and B_1 atoms:

$$R_M = R_A + R_B. \tag{1}$$

In the proposed scheme of creating conditions for lasing (see Fig. 1) in a pulsed or stationary electric discharge the electrons excite the atoms *A* from its ground state 1 to one of the low lying resonance levels 2 with the energy E_2 , thus yielding the excited atoms A_2 . The A_2 atoms in their turn are either transferred by the electrons into the higher energy states with the energies E_j , where $j = 3, 4, \dots, n$, and into the continuum (I_A is the ionization energy of the *A* atom) or relax due to the collisions with electrons to the ground state. Besides, the A_2 and B_1 atoms form, when colliding, the flying apart molecule A_2B_1 in its excited state with the repulsive term 2, the potential curve $U_2(R)$ of which differs from the $U_1(R)$ curve by the existence of quite an extended plateau.

Model curves $U_1(R)$ and $U_2(R)$ of the laser repulsive terms built on the base of potential curves of the $X^1\Sigma_g^+$ and $B^5\Sigma_u^+$ terms of Xe_2 molecule without a regard for quite shallow potential wells on both curves are presented in Fig. 2a. Those are just the model potential curves we have used in analysis of the possibility of obtaining the generation on the repulsive terms of a molecule comprising the flying apart fragments performed in this paper.

In accordance with Refs. 1 and 2 the concentrations of the molecules of flying apart fragments with the terms 1 (N_1) and 2 (N_2) contained in a spherical layer of the *R* radius and δR thickness may be determined as

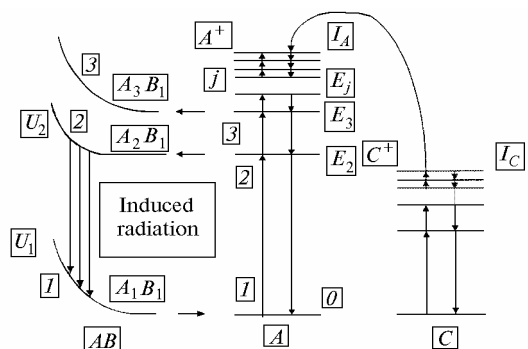


Fig. 1. Scheme of creating conditions for laser generation on repulsive terms of molecules comprising the flying apart fragments.

These atoms, denoted by A_1 and B_1 symbols, respectively, colliding with each other in their ground states, form the molecule composed of the flying apart fragments with a repulsive term 1 characterized by a potential curve $U_1(R)$, where R is the internuclear distance. Maximum internuclear distance R_M at which

$$\begin{aligned}
 N_1 &= n_1 n_B 4\pi R^2 \delta R e^{-U_1(R)/T_a}, \\
 N_2 &= n_2 n_B 4\pi R^2 \delta R e^{-[U_2(R)-E_2]/T_a},
 \end{aligned}
 \tag{2}$$

where n_B is the concentration of atoms B in the ground state; n_1, n_2 are the concentrations of atoms A in the 1 and 2 state, respectively; T_a is the temperature of atoms in energy units; $n 4\pi R^2 \delta R e^{-U(R)/T_a}$ is the probability that the B or A atom are in the spherical layer of the radius R and thickness δR surrounding the atoms A and B , respectively.

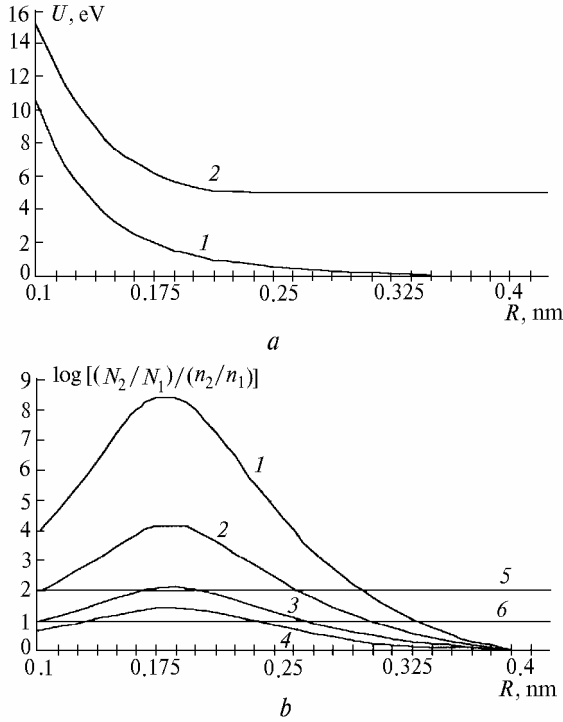


Fig. 2. Model potential curves of lower (1) and upper (2) laser repulsive terms of a molecule comprising the flying apart fragments (a); dependence of the ratio of concentration of atoms on the upper and lower laser terms normalized to the concentration ratio on the levels 2 and 1 of A atoms (b) on the internuclear distance; $T_a = 0.05$ (1); 0.1 (2); 0.2 (3); 0.3 eV (4); curves 5 and 6 – see in the text.

In Refs. 1 and 2 the thickness δR represents the size of a classically accessible area for the nuclei of a diatomic molecule. In this paper the δR parameter has different physical meaning that is based on the following reasonings. The emission on the transition between the upper and lower laser terms that occurs at the frequency ω_R corresponding to some internuclear distance R has a width $\delta\omega$ caused by different broadening mechanisms (Doppler, van-der-Waals, etc.). Therefore the molecules of flying apart fragments with the internuclear distances from the R to $R \pm \delta R/2$ interval will contribute to the emission at the ω_R frequency within the frequency interval $\delta\omega$ caused by different broadening mechanisms, where the value δR is presented in the following way

$$\delta R = \hbar \delta\omega / |dU_1(R)/dR - dU_2(R)/dR|, \tag{3}$$

To estimate the possibility of creating the inverse population of the terms with potential curves presented in Fig. 2a in a molecule comprising the flying apart fragments, assuming the equality of the statistical weights of the lower and upper terms, one can use the formula for the ratio of concentrations of such molecules on the upper and lower terms which follows from the expression (2):

$$\frac{N_2/N_1}{n_2/n_1} = e^{-[U_2(R)-E_2-U_1(R)]/T_a}. \tag{4}$$

Results of the calculation based on Eq. (4) for several values of temperature T_a of atoms are presented in Fig. 2b. The values $1/(n_2/n_1)$ for $(n_2/n_1) = 0.01$ (curve 5) and $(n_2/n_1) = 0.1$ (curve 6) are shown in the same figure. The cross points of the curves 1 to 4 with the straight lines 5 and 6 determine the limits of internuclear distances within which the condition $N_2/N_1 > 1$ holds at the above given values of the ratio n_2/n_1 and corresponding temperatures of atoms. It is seen that in order to achieve the inverse population of the laser terms at temperatures of atoms about 0.3 eV, it is necessary to provide for a high (at a level of 10% of the ground state population) population of the level 2 of the atom A , that is, it is necessary to provide high temperatures of electrons to populate the level 2 of the atom A .

To evaluate the conditions (concentrations of atoms A and B , temperatures of electrons, etc.) favorable for generation on the transition between the repulsive terms of a molecule of the flying apart fragments, it is necessary to know absolute populations of the laser terms N_1 and N_2 , wavelength λ of the stimulated emission line, and its spectral width $\delta\omega$. To estimate these parameters, let us write the amplification coefficient for the transition between terms 2 and 1 of a molecule of the flying apart fragments as a function of internuclear distance following Refs. 1 and 2:

$$\begin{aligned}
 k(R) &\approx \frac{\lambda^2 A_{21}}{4\delta\omega} \pi R^2 \delta R \times \\
 &\times (n_2 n_B e^{-[U_2(R)-E_2]/T_a} - n_1 n_B e^{-U_1(R)/T_a}),
 \end{aligned}
 \tag{5}$$

where A_{21} is the probability of spontaneous transition between the laser terms.

If one neglects, in the expression (5), the dependence of $k(R)$ on the internuclear distance R caused by the dependence of $\lambda, \delta\omega$, and δR on R , then the amplification coefficient will be proportional to the value:

$$\begin{aligned}
 k(R) \sim K(R) &= R^2 e^{-U_1(R)/T_a} \times \\
 &\times \left(\frac{n_2}{n_1} e^{-[U_2(R)-E_2-U_1(R)]/T_a} - 1 \right),
 \end{aligned}
 \tag{6}$$

that allows one to calculate the dependence of the relative amplification coefficient (i.e., the ratio of the function $k(R)$ to its maximum value)

$$k_{\text{reM}} = k(R) / k_{\text{max}}(R) = K(R) / K_{\text{max}}(R) \quad (7)$$

on the internuclear distance. The results on k_{reM} calculated using the model potential curves presented in Fig. 2a are shown in Fig. 3. It is necessary to note that the normalization of $K(R)$ dependences calculated for different conditions was performed to the same value $K_{\text{max}}(R)$ corresponding to $T_a = 0.05$ eV, and $n_2/n_1 = 0.1$ values.

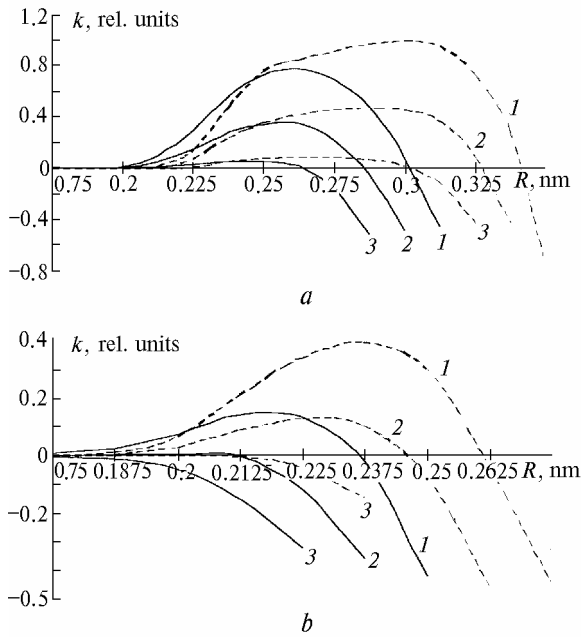


Fig. 3. Dependence of the relative amplification coefficient on the internuclear distance for $n_2/n_1 = 0.1$ (1), 0.05 (2), 0.01 (3); $T_a = 0.05$ eV (dashed curve) and 0.1 eV (solid curve) (a); $T_a = 0.2$ eV (dashed curve) and 0.3 eV (solid curve) (b).

To carry out further analysis, let us first introduce two new parameters characterizing the curves presented in Fig. 3: $\Delta R = R_{\text{max}} - R_{\text{min}}$, where R_{max} and R_{min} represent larger and smaller values of the internuclear distance at which $k_{\text{reM}} = 0.5 k_{\text{reMmax}}$, and R_0 is the parameter determined as $R_0 = (R_{\text{max}} + R_{\text{min}}) / 2$; let us also introduce the wavelength λ_0 corresponding to the internuclear distance R_0 . It is seen that both parameters ΔR and R_0 depend strongly enough on the temperature of atoms, and while relatively weakly on the concentration ratio n_2/n_1 . Second, taking into account approximate character of the estimations performed in the present paper, let us change the ratio $\delta R / \delta \omega$ in the expression (3) for the $\Delta R / \Delta \omega$ ratio with $\Delta \omega$ being determined as follows:

$$\Delta \omega = \frac{1}{\hbar} \left| [U_2(R_{\text{max}}) - U_1(R_{\text{max}})] - [U_2(R_{\text{min}}) - U_1(R_{\text{min}})] \right|, \quad (8)$$

the expression (5) is then transformed to the form:

$$k_0 \approx \frac{\lambda_0^2 A_{21}}{4 \Delta \omega} \pi R_0^2 \Delta R \times (n_2 n_B e^{-[U_2(R_0) - E_2] / T_a} - n_1 n_B e^{-U_1(R_0) / T_a}). \quad (9)$$

In accordance with the calculated results (see Fig. 3) a decrease in the n_2/n_1 is followed by a decrease in the ΔR value. However, since the decrease in ΔR is, in its turn, accompanied by a decrease in the $\delta \omega$ value, in accordance with expression (8), that compensates, to some extent, for the influence of the ΔR variation on the amplification coefficient k_0 , Eq. (9), further analysis of conditions favorable for lasing on the repulsive terms of molecules comprising the flying apart fragments was performed assuming that such parameters as R_0 , δR , and $\delta \omega$ do not depend on the n_2/n_1 ratio, being only determined by the temperature of atoms. Values of the above parameters derived for $n_2/n_1 = 0.1$ using the dependences presented in Fig. 3a, as well as $U_1(R_0)$ and $U_2(R_0) - E_2$ values, determined from the potential curves presented in Fig. 2a, are given in Table 1. Taking into account the data from Table 1, relative concentration n_2/n_1 of the atoms on the second level necessary to provide the prescribed value k_0 may be calculated using the relationship that follows from Eq. (9):

$$\frac{n_2}{n_1} = e^{-\{U_1(R_0) - [U_2(R_0) - E_2]\} / T_a} \times \left(\frac{4 k_0 \Delta \omega e^{U_1(R_0) / T_a}}{\lambda_0^2 A_{21} \pi R_0^2 \Delta R n_1 n_B} + 1 \right). \quad (10)$$

Table 1

T_a , eV	R_0 , nm	ΔR , nm	$\Delta \omega$, s ⁻¹	$U_1(R_0)$, eV	$U_2(R_0) - E_2$, eV
0.05	0.29	0.09	$7.62 \cdot 10^{13}$	0.28	0
0.1	0.26	0.06	$6.84 \cdot 10^{13}$	0.45	0
0.2	0.24	0.04	$5.53 \cdot 10^{13}$	0.71	0.02
0.3	0.22	0.03	$3.08 \cdot 10^{13}$	0.96	0.12

Then, assuming that the population of the level 2 of atoms A is in equilibrium with the temperature T_e of electrons, and that the statistical weights of the levels 1 and 2 of the atom A are equal to each other, one can calculate the value of the ratio

$$T_e / E_2 = 1 / \ln(n_1 / n_2), \quad (11)$$

which will yield the population of the level 2 of atom A necessary for achieving the lasing effect on the transition between the repulsive terms (see Fig. 2a) of the molecule AB comprising the flying apart fragments.

In further consideration of conditions favorable for achieving a continuous wave (cw) generation on the repulsive terms of a molecule comprising the flying apart fragments, we have chosen the mercury atoms ($6p(3P_1^0) \rightarrow 6s^2(1S_0)$) transition with the following set of parameters: $E_2 = 4.886$ eV, $\lambda_{21} = 253.65$ nm,

$A_{21} = 8.5 \cdot 10^6 \text{ s}^{-1}$ as a test atom A . It was supposed that the generation line λ_0 is longer than λ_{21} and equals 300 nm. It was also assumed that the probability of the spontaneous transition between the repulsive terms of the molecule comprising the flying apart fragments is equal to the reduced value of A_{21} , while the k_0 value necessary for achieving the generation was assumed to be of 10^{-2} cm^{-1} . As a test atom B we have chosen neon atom.

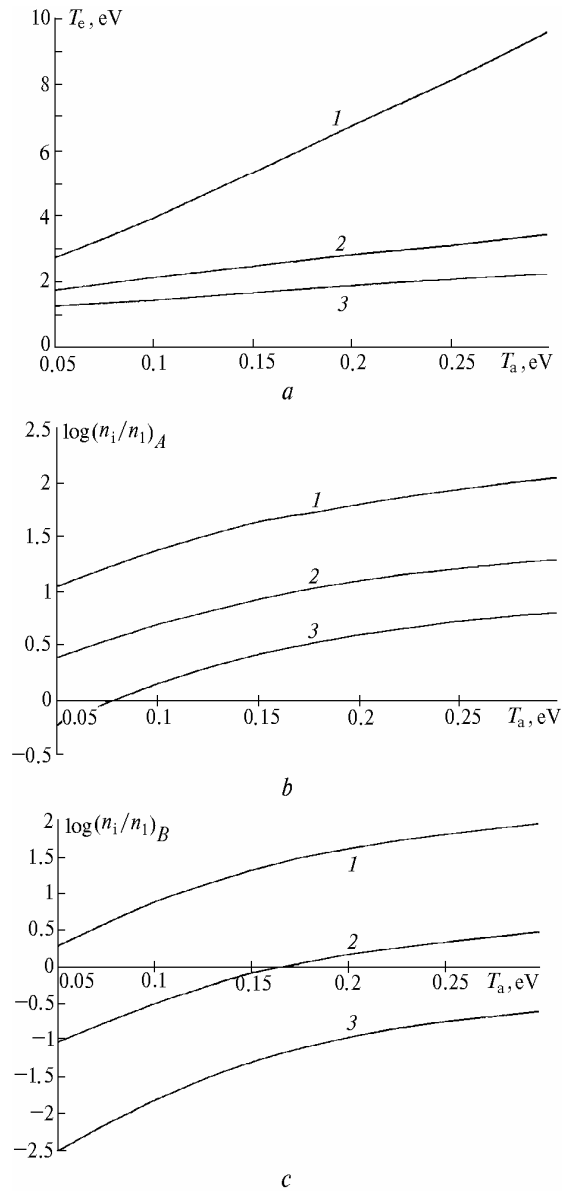


Fig. 4. Dependence of the electron temperature necessary for providing the amplification coefficient $k_0=10^{-2} \text{ cm}^{-1}$ at the center of the line of lasing on the transition between the model repulsive terms (see Fig. 2a) on T_a , when mercury atoms serve as A atoms (a). Dependences of the ratios between concentrations of ions A^+ and atoms A in the ground state (b), as well as between concentrations of the ions B^+ and atoms B in the ground state (c). The Ne atom was used as the model atom B : $n_{A1} = 10^{19} \text{ cm}^{-3}$, $n_{B1} = 10^{19} \text{ cm}^{-3}$ (1); 10^{19} cm^{-3} , $3 \cdot 10^{19} \text{ cm}^{-3}$ (2); 10^{19} cm^{-3} , 10^{20} cm^{-3} (3).

Calculated results on the temperature T_e necessary to achieve lasing on the repulsive terms of the AB molecule of flying apart fragments obtained based on expressions (10) and (11) for several sets of the atoms A and B concentrations in the ground state are presented in Fig. 4. The results calculated based on Saha formula (see, for example, Ref. 3) for the ratios of the ions A^+ and B^+ concentrations to the corresponding concentrations of atoms A and B in the ground states at the calculated temperature T_e are also shown in Fig. 4.

So, the calculated results that are presented in Fig. 4 demonstrate poor applicability of the model atoms A with the parameters chosen in the calculation for their use in the scheme proposed in this paper for achieving the lasing effect. Even the best (most easily realized) of the calculated parameters ($n_1 \approx 10^{19} \text{ cm}^{-3}$; $n_B \approx 10^{20} \text{ cm}^{-3}$; $T_a \lesssim 0.05 \text{ eV}$; $1 \text{ eV} \lesssim T_e \lesssim 1.5 \text{ eV}$) seem to be difficult to achieve. Nevertheless, the calculated results presented in Fig. 4 allow one to reveal clearly the factors hindering creation of conditions favorable for lasing in the proposed scheme, as well as to identify, at least, the ways of creating conditions for achieving a cw generation on the repulsive terms of molecules comprising the flying apart fragments.

1. Among the calculated results shown in Fig. 4 most important seem to be the high temperature of electrons and high concentrations of atoms A and B . Both these factors are, first of all, caused by the difficulty of achieving the prescribed, in the calculations, amplification coefficient ($k = 10^{-2} \text{ cm}^{-1}$) at the parameters used in the calculations: $\lambda_0 = 300 \text{ nm}$ ($\lambda_{21} = 253.65 \text{ nm}$), $A_{21} = 8.5 \cdot 10^6 \text{ s}^{-1}$, and $E_2 = 4.886 \text{ eV}$. Use of atoms with $\lambda_{21} > 253.65 \text{ nm}$ ($E_2 < 4.886 \text{ eV}$), and $A_{21} > 10^8 \text{ s}^{-1}$, providing the possibility of achieving the generation at $k_0 < 10^{-2} \text{ cm}^{-1}$ as an A atom will allow one to move into the region of both lower electron temperatures and lower concentrations of A and B atoms.

2. Influence of triple collisions on the radiative properties of a laser transition is quite probable at large concentrations of the B atoms. Assuming the atoms A and B to be elastic balls of the same radius $r_a = R_M/2$, the lifetime of the molecule of flying apart fragments can be estimated as $\tau_M \approx R_M/v_a$, where v_a is the thermal speed of atoms. Assuming also that each collision of a such a molecule with an atom results in changing the radiative properties of the lasing transition, and estimating the cross section σ_M of the molecule collision with the third atom as a sum of the collision cross sections of two atoms $\sigma_M \approx 2\pi R_M^2$, while the rate constant q_M of the collision of the molecules comprising the flying apart fragments with the atoms as $q_M \approx \sigma_M v_a$, one can obtain a constraint on the concentration of atoms under which a lifetime of the molecule comprising the flying apart fragments is less than the interval of time between its successive collisions with atoms:

$$n < n_{\max} = 1/2\pi R_M^3. \quad (12)$$

Assuming that $R_M \approx 0.4$ nm, one obtains using Eq. (12) that $n_{\max} \approx 2.5 \cdot 10^{21}$ cm⁻³. This means that probable influence of triple collisions on the radiative properties of the lasing transition is to be taken into account at concentrations $n_B \approx 2.5 \cdot 10^{21}$ cm⁻³.

3. In accordance with the calculations presented in Fig. 4, the degree of ionization of atoms A is about 50% even under most favorable conditions that corresponds to the concentration n_e of electrons at the level of 10^{19} cm⁻³. At such values of n_e an intense mixing of the electronic laser terms of the molecule of flying apart fragments may be expected, what excludes a possibility of achieving the inversion of their populations. Assuming the constant q_e of the quenching rate of the upper laser term by electrons to be of the order of 10^{-8} cm⁻³·s⁻¹, while the probability A_{21} of a spontaneous decay of the molecule comprising the flying apart fragments due to the lasing transition to be of the order of $3 \cdot 10^8$ s⁻¹, one can estimate the concentration of electrons at which the quenching rate of the upper laser term by electrons will be less than the frequency of its spontaneous decay, i.e., $n_e < A_{21}/q_e \approx 3 \cdot 10^{16}$ cm⁻³ that is significantly less than the concentration of electrons corresponding to the calculated results presented in Fig. 4.

4. At the concentration n_e of the order of 10^{16} – 10^{17} cm⁻³ in the nonequilibrium plasma the evolution of strong inhomogeneities is quite probable. To avoid this and to decrease the concentration of ions A , the atoms C (vapor) of alkali metals with a low ionization potential may be added as a third component, as it was proposed in Ref. 4, into a two-component active medium consisting of atoms A and B . In this case the recombination flow of ions A^+ will increase, while their concentration decrease. With the help of Saha formula one can easily obtain a relation determining the decrease of the concentration n'_i of ions A^+ relative to the equilibrium concentration n_i of ions, at a given electron temperature, by adding easily ionized component with the concentration n_C , all atoms of which are ionized:

$$\begin{aligned} n'_i/n_i &= (\sqrt{J^2 + 4} - J)/2 \\ J &= n_C/n_i. \end{aligned} \quad (13)$$

The presence of alkali metal atoms in the active medium changes the proposed scheme of generation to its new shape shown in Fig. 1.

5. The above analysis of the possibility of creating conditions favorable for lasing on the repulsive terms of molecules comprising the flying apart fragments was performed assuming that the population of the level 2 of A atoms was in equilibrium at the electron temperature T_e . However, to produce the efficient generation on the transition between the

repulsive terms of such a molecule, it is necessary that the population of the level 2 of A atoms caused by the radiative collisions with atoms B be significantly decreased compared with the equilibrium population at T_e . Only in this case the locking of the main flow of excitation of the A atoms within the path $A_1 \rightarrow A_2 \rightarrow A_2B_1 \rightarrow A_1B_1 \rightarrow A_1$ is possible thus providing for high efficiency of the transformation of electric energy introduced into a discharge into the energy of laser radiation.

Thus, analysis performed in this paper, has shown that there exists the possibility of creating lasers on the repulsive terms though one faces a number of problems on this way and in the case of a success one can hope to create highly efficient lasers operating in a wide range of wavelengths.

Chemical elements, whose atoms can be used, as the A atoms in the proposed scheme of achieving the lasing seem to be promising are given in Table 2, where the comparison is performed of the pressure of saturated vapor of these elements at the temperature of 1000 K (see Ref. 5) as well as of some characteristics of the corresponding atoms, taken from Ref. 6, with the analogous characteristics of the mercury atom used as an atom A in the above described calculations (see Fig. 4). The values of the parameter $\alpha = (\lambda_{21}^2 A_{21})_{\text{Hg}} / (\lambda_{21}^2 A_{21})_A$ characterizing to some extent, in accordance with the Eq. (9) the decrease in population of the level 2 of atoms presented in the Table 2 as compared to the population of level 2 of the mercury atoms assuming the same amplification coefficient k_0 are also given in Table 2. The value of the parameter α at the level of 0.02 for these atoms means that their usage as an atom A may essentially decrease the concentration of atoms A , the electron temperature, and the degree of ionization of atoms compared to the values obtained in the case when mercury atoms are used as A atoms in the above discussed scheme.

Further analysis of the possibility of achieving the lasing effect on the repulsive terms of molecules comprising the flying apart fragments should be carried out both for some particular pairs of atoms and for actual potential curves of the repulsive terms of such molecules. The atoms of Li, Ca, Mg, Sr, Tl, Yb, and Eu may be included into the consideration if the temperature T_a is increased up to 1500 K; these atoms appear to be suitable for creating lasers on the repulsive terms of molecules comprising the flying apart fragments can be expanded more if the temperature T_e is increased up to 2000 K.

In conclusion it should be noted that alkali metals and inert gases (see, for example, Refs. 7–10) are the atoms that can form molecules of flying apart fragments with the potential curves of repulsive terms, which appear to be promising for achieving the lasing effect on the repulsive terms.

Table 2

Element	p , kPa	E_2 , eV	λ_{21} , nm	A_{21} , s ⁻¹	I , eV	E_2/I	$\lambda_{21}^2 A_{21}$, nm ² ·s ⁻¹	α
Hg	$\approx 1.3 \cdot 10^3$	4.886	253.65	$8.5 \cdot 10^6$	10.434	0.468	$5.5 \cdot 10^{11}$	1
Cd	53.2	5.417	228.80	$6 \cdot 10^8$	8.994	0.602	$3.1 \cdot 10^{13}$	0.018
Zn	10.6	5.796	213.86	$7.4 \cdot 10^8$	9.394	0.617	$3.4 \cdot 10^{13}$	0.016
Na	33.2	2.102	589.592	$6.1 \cdot 10^7$	5.139	0.409	$2.1 \cdot 10^{13}$	0.026
Na	33.2	2.104	588.995	$6.1 \cdot 10^7$	5.139	0.409	$2.1 \cdot 10^{13}$	0.026
K	79.8	1.610	769.90	$3.9 \cdot 10^7$	4.341	0.371	$2.3 \cdot 10^{13}$	0.024
K	79.8	1.617	766.49	$4.0 \cdot 10^7$	4.341	0.372	$2.4 \cdot 10^{13}$	0.023
Rb	133	1.560	794.76	$3.4 \cdot 10^7$	4.177	0.373	$2.0 \cdot 10^{13}$	0.028
Rb	133	1.589	780.03	$3.7 \cdot 10^7$	4.177	0.380	$2.3 \cdot 10^{13}$	0.024
Cs	133	1.386	894.35	$3.3 \cdot 10^7$	3.894	0.356	$2.6 \cdot 10^{13}$	0.021
Cs	133	1.455	852.11	$3.7 \cdot 10^7$	3.894	0.374	$2.7 \cdot 10^{13}$	0.020

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