KINETIC MODEL OF A NUCLEAR-PUMPED IR LASER IN A He-Ne-CCl_4 MIXTURE

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In this paper we present a detailed kinetic model of a lasing medium and the results calculated using that model for a nuclear-pumped IR laser emitting on the transitions of atomic chlorine in a high-pressure mixture of He-Ne-CCl_4. *Based on the comparison made between the calculated and experimental results we have established the mechanism of the population inversion for the transitions lasing at* 1.59 *and* 2.45 μm *wavelengths. We have also identified the optical conditions for the lasing to occur.*

Recently there appeared a report in the literature[1] on obtaining a relatively powerful lasing on the nuclear-pumped atomic chlorine transitions in the mixture of He–Ne–CCl_4. It appeared to be a characteristic feature of this laser operation that the partial pressure of the lasing component in the mixture is extremely low (~ 10 mTopp) at a sufficiently high pressure of the buffer gases (1 atm of He and 1 atm of Ne). At the same time the mechanism of lasing was unclear. The only assumption made in Ref. 1, regarding the lasing process, was that the populating of the upper lasing levels due to ion-ion recombination between the negative chlorine ions and positive ions of the noble gases could be as the most likely mechanism. In addition, it was mentioned in this paper that the excitation due to dissociation may also contribute into the lasing.

In this paper we present a theoretical investigation into the mechanisms of the lasing process in that type of laser. To do this we have constructed a detailed non-stationary kinetic model of a nuclear-pumped laser in the mixture of He–Ne–CCl_4. Using this model we have been calculating the kinetics of the plasmochemical processes in the active medium (AM), as well as the power and efficiency of lasing at 1.59 and 2.45 μm wavelengths. The input data for modeling, such as the pump pulse shape, mixture composition, and transmission of the resonator mirrors, were taken from Ref. 1. The numerical modeling has been done using the PLASER[2] software package.

1. KINETICS OF PLASMA IN THE MIXTURE OF He–Ne–CCl_4

The kinetic model of a laser in He–Ne–CCl_4 mixture considers the following plasma components: He^+, He_2^+, He_3^+, He^*, He_2^*, Ne^+, Ne_2^+, Ne_3^+, Ne^*, Ne_2^*, HeNe, $HeNe^*$, Cl^+, Cl_2^+, CCl^+, CCl_2^+, CCl_2^{+*}, CCl_3^+, CCl_4^+, C^+, C^-, Cl^-. Besides, there are eight excited energy levels of chlorine ($3d^4F_{9/2}$, $3d^4D_{5/2,7/2}$, $4p^4D_{5/2,7/2}$, $4p^4P_{5/2,3/2}$, $4s^4P_{5/2}$) and the highly excited Cl^{**} state, that efficiently allows for the levels above the $3d$ states, are involved into the modeling process. The level $4s^4P_{5/2}$ in the model represents an effective metastable state of the chlorine atom. The averaged probability of this state radiation decay equals $A = 2.4{\cdot}10^5$ s^{-1}. The probabilities of the radiation decay for all the excited states of chlorine have been calculated using the tabulated data from Ref. 3. The levels $4p$ and $3d$ in the model proposed are the lowest ones in the corresponding multiplets and have the largest statistical weights. For that reason the model assumes their population to represent the population of the entire multiplet. This assumption well agrees with the Boltzmann distribution of population over the multiplet sub-levels.

All in all the kinetic model considers about 250 plasmochemical reactions (PChR) of 34 reagents. For those reactions we have been solving the strict system of nonstationary balance equations. The number density of electrons, N_e, was determined from the condition of the plasma to be quasi-linear. Besides, we have also been solving the energy balance equation for the gas, T_g, and electron, T_e, temperatures. The working emission has been allowed for using the zero-dimensionality approach.[2]

When calculating nonsaturated gain coefficient, α_0, we have set the broadening of the lasing transition line as a sum of the collisional line width, γ_c, due to the buffer gas pressure, and the Doppler line width, γ_D. The value of the collisional line width was first determined theoretically and then corrected for using the experimentally measured value of α_0. The model used the value $\gamma_c = 10^{10}$ s$^{-1}{\cdot}$atm^{-1}. The parasitic absorption of the laser emission in the active medium and in mirrors of the resonator has been taken into account by introducing the coefficient $\varkappa^- = 1.1{\cdot}10^{-5}$ cm^{-1}.

The plasmochemical reactions involving He and Ne have been borrowed from our kinetic model of a laser in the mixture of He–Ne–Ar–H_2 presented in Ref. 4 that

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dealt with a detailed description of the He–Ne plasma relaxation.

Under the action of hard ionization the relaxation flux of helium and neon components yields the ions of Ne and He and these atoms in the excited states. In the mixture He:Ne = 1:1 the main components yielded are ions and neon components in the excited states. Most abundant among them is the molecular ion Ne_2^+ whose number density exceeds that of other components by more than an order of magnitude.

It is a characteristic feature of mixtures containing CCl_4 that there is a high concentration of Cl^- ions that strongly affect the plasma kinetics. The efficient production of these ions occurs due to the dissociative capture of electrons

$$CCl_n + e = CCl_{n-1} + Cl^-, \quad (1)$$

$$Cl_2 + e = Cl + Cl^-, \quad (2)$$

where n varies from 1 to 4.

The rate constants for reactions (1) and (2) have been set by applying spline to data from Refs. 5 and 6. Since the temperature T_e used in calculations made in Ref. 6 was taken below 1 eV no processes, competing with the reaction (2) in excitation and ionization of Cl_2, were taken into account.

The ions Cl^- take part in the triple ion–ion recombination with the atomic and molecular ions of helium and neon that yields the intermediate complexes of $NeCl^*$ and $HeCl^*$. The excitation energy of these complexes is much higher than the ionization potential of the atomic chlorine. Decomposition of the above complexes is the primary source of Cl^+ ions. The other, though weaker, source of Cl^+ ions is the capture of electrons by the atomic chlorine from the ions of the buffer gases, Ne_2^+, He_2^+, Ne^+, and He^+. Then in the reactions of charge exchange and three-particle conversion there are produced the molecular Cl_2^+ ions

$$Cl^+ + Cl_2 = Cl_2^+ + Cl, \quad (3)$$

$$Cl^+ + Cl + M = Cl_2^+ + M, \;\; M = \text{He, Ne}. \quad (4)$$

From the condition of best fit between the calculated and experimental values we have obtained the rate constants for these reactions to be $1.2\cdot10^{-10}$ cm^3/s and $5\cdot10^{-31}$ cm^6/s, respectively.

2. MECHANISMS OF CREATING INVERSE POPULATION OF THE WORKING ENERGY LEVELS

The two working transitions under study, $3d^4F_{9/2} - 4p^4D_{7/2}$ and $3d^4D_{7/2} - 4p^4D_{7/2}$, lasing at the wavelengths of 1.59 and 2.45 μm have common lower level so the maximum lasing power is achieved at one and the same CCl_4 partial pressure of 30 mTorr.[1] For this reason we assume, in the model proposed, that the inverse population in these transitions has the same nature.

The results of modeling the nuclear-pumped laser on IR transitions in chlorine atoms have shown that the main channels of the upper lasing levels pump are the triple recombination of atomic Cl^+ ions with electrons, the dissociative recombination of the excited CCl_2^{+*} ions with the negative Cl^- ion

$$Cl^+ + e + M = Cl^* + M, \quad (5)$$

$$Cl^+ + e + e = Cl^* + e, \quad (6)$$

$$Cl_2^+ + e = Cl^* + Cl, \quad (7)$$

$$CCl_2^+ + e = Cl^* + CCl, \quad (8)$$

$$CCl_2^{+*} + Cl^- = Cl^* + CCl_2. \quad (9)$$

The final rate constants of the reactions (5) to (9) are $2\cdot10^{-31}T_gT_e^{-3.5}$ cm^6/s, $5.4\cdot10^{-27}\,T_e$ cm^6/s, $2\cdot10^{-7}\,T_e^{-0.5}$ cm^3/s, $2\cdot10^{-7}\,T_e^{-0.5}$ cm^3/s, and $2\cdot10^{-6}$ cm^3/s at 2 atm total pressure of the buffer gas. The electron and gas temperatures entering the constants are in units of electron-volts. Then we have assumed in our model that 50% of the total outcome of reactions (5) to (7), 60% of that for the reaction (8), and 10% of the reaction (9) contribute to the population of the upper level of the transition lasing at 1.59 μm. For the level $3d^4D_{7/2}$ those values are 10, 20, and 0%, respectively.

The molecular ions CCl_2^{+*} and CCl_2^+ are the products of Penning reactions of the CCl_4 and CCl_2 molecules with the excited atoms and molecules of a buffer gas, as well as of recharging reactions between the atomic and molecular ions of the inert gases and CCl_4 and CCl_2. Strictly speaking the effective reaction (9) is the process in which first unstable excited complexes are formed whose fragmentation yields the formation of chlorine atoms in various excited states.

The fractional contributions coming from the processes (5) to (9) to pumping the upper lasing levels depend on the CCl_4 concentration in the mixture. Figure 1 presents the pump fluxes as functions of the CCl_4 partial pressure. It is seen from this figure that at pressures below 10 mTorr the processes (5) and (6) dominate. In the pressure interval from 10 to 100 mTorr the process (7) takes most active part in pumping the upper levels, with the pump flux from it being maximum at CCl_4 of 30 mTorr. At the pressure of 60 mTorr the process (8) starts to contribute into the pumping and it becomes dominating at partial pressures above 100 mTorr.

The unloading of the lower lasing level occurs due to the radiation decay and inelastic collisions with helium and neon. The constant of quenching by the buffer gas atoms was taken to be 10^{-12} cm^3/s.

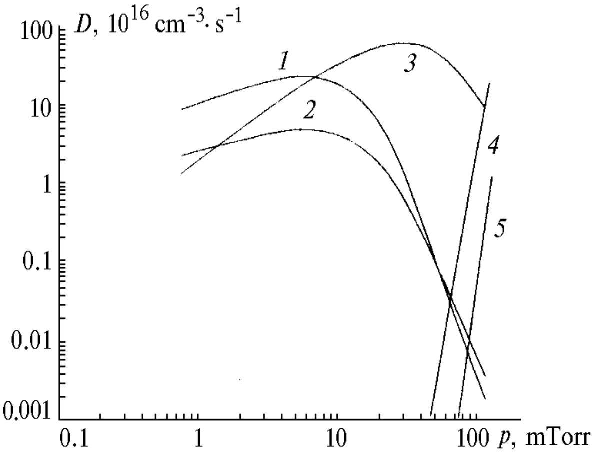

FIG. 1. Total fluxes D from the reactions (1) to (5), that contribute to the upper lasing levels pumping as functions of the CCl_4 *partial pressure. Total pressure of the mixture is* 2 atm; He:Ne = 1:1; λ = 1.59 μm, $r_1 = r_2$ = 0.998. *1* – (5)*; 2* – (6)*; 3* – (7)*; 4* – (8)*; 5* – (9).

The constants of the de-excitation rates by electrons have been estimated using the van-Redgemorter approximation. In so doing we related the rates of the direct and inverse processes based on the principle of detailed balancing. It should be noted, however, that under experimental conditions described in Ref. 1 the electron density is not high (10^{12} to 10^{13} cm^{-3}). As a result, the mixing of working levels by the electrons does contribute into the population distribution only weakly as compared to other relaxation processes.

As noted above, all calculations have been done assuming the action of hard ionization source. In these calculations we did not take into account the effects of plasma track structure, formed by the decay fragments, on the lasing parameters. The matter is that the characteristic times of the relaxation processes in an active medium of a nuclear-pumped laser in the mixture of He–Ne–CCl_4 that result in populating of the upper lasing level (under conditions of experiments in Ref. 1) are much longer than the tracks' lifetimes. The latter, according to Ref. 9, are from 1 to 100 ns. Moreover, under optimal conditions for lasing in the mixture considered here, we may always neglect the track structure. The matter is that, in principle, its influence on the lasing parameters is only possible if the tracks do not overlap (i.e., at low pump energy) and the characteristic times of the upper level populating and lasing formation do not exceed the tracks' lifetimes, at the rate of the upper level populating being proportional to the pump power. This conclusion is evidently true regarding any laser with the nuclear pump in which the upper lasing level is populated due to recombination processes that involve electrons. As to other lasers where the active medium is excited by the fission fragments a more detailed analysis is needed based on the general statement formulated above. The above considerations have an indirect confirmation in the results from Ref. 9 where the account for diffusion of the He–Cd plasma components from the initial region of a track resulted in an essential decrease of the fluctuation amplitude, while the enhancement of the pump power weakened the effect of the track structure on the dynamic processes in plasma.

3. CALCULATED RESULTS AND DISCUSSION

The results of a comparison we have made between the calculated, using the above proposed model, and experimentally measured parameters of the output laser emission (the output power and threshold density of neutrons) are shown in Figs. 2 to 4.

As seen from these figures the output emission power, as a function of CCl_4 partial pressure in the He:Ne = 1:1 buffer gas mixture at a constant pressure of 2 atm, reaches its maximum at p_{CCl_4} = 30 mTopp (Fig. 2). The maximum occurs here due to a competition between the processes of the [Cl] and [Cl_2] concentration growth giving rise to an increase in the [Cl_2^+] concentration (reactions (3) and (4)), on the one hand, and the processes (1) and (2) causing a decrease in the electron density while increasing the concentration of [Cl^-], on the other hand. At CCl_4 partial pressures above 30 mTorr the lasing becomes poorer because of the charge exchange between Cl_2^+ and atomic carbon that is produced as an outcome of plasmochemical reactions. In addition, the quenching of the upper lasing level by the atomic and molecular chlorine also deteriorates lasing.

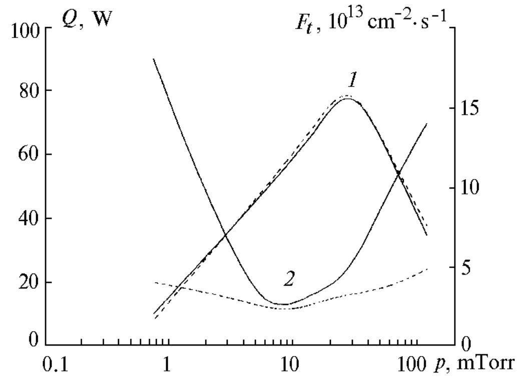

FIG. 2. The laser emission power, Q (curve 1), and threshold density of the neutron flux, F_t *(curve 2) as functions of the* CCl_4 *partial pressure. Total pressure of the mixture is* 2 atm; He:Ne = 1:1*;* λ = 1.59 μm, $r_1 = r_2$ = 0.998. *Solid lines present experimental data; dashed curves are the results of calculations.*

The minimum in the threshold density of the neutron flux occurs to be for the emission at 1.59 μm wavelength, at 7 to 8 mTorr partial pressure of CCl_4 (see Fig. 2). At a low, near threshold, pump power the upper lasing level $3d^4F_{9/2}$ is primarily populated due to the recombination of the excited qCl_2^{+*} molecular ions with the negative Cl^- ions (reaction (9)). The optimum in the dependence of the threshold neutron density of the CCl_4 partial pressure is a result of competition between the creation of q Cl_2^{+*} ions, their annihilation in the charge exchange reactions with CCl_4 molecules, and quenching of the upper lasing level by CCl_4 molecules in the reaction

$$Cl^* + CCl_4 = CCl_3 + Cl_2. \qquad (10)$$

It should be noted that the assumption on ion–ion recombination of the excited CCl_2^{+*} ions has been required just to describe the threshold lasing properties, though we have no any reliable information about them. However, one may suppose that a significant portion of CCl_2^{+*} molecular ions produced in the reactions of charge exchange between the atomic and molecular ions of the inert gases with CCl_4 molecules are being in different electron-excited states.

In Figure 3 one may see the dependence of the output laser emission power on the fraction content of neon in the mixture at its constant total pressure of 2 atm. The experiment has revealed an optimum regarding the neon partial pressure that is being achieved in a mixture of He:Ne = 1:1. At the further increase of neon pressure there occurs a fall off of the output laser power. Most likely this decrease of power observed in the experiment could be due to the nonuniform energy deposition across the lasing channel that increases with increasing pressure of neon. As it follows from Ref. 10 the efficiency of the energy deposition on the channel axis may decrease twice, in a pure neon, because of this factor.

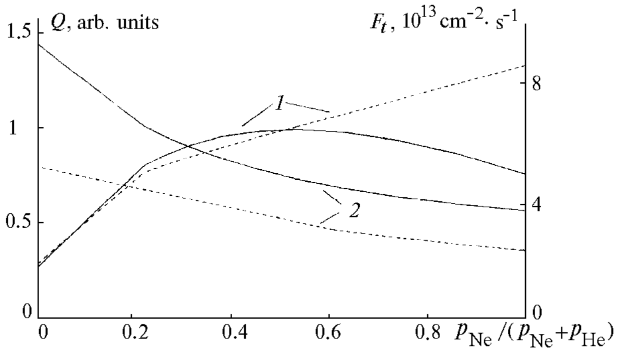

FIG. 3. The laser emission power, Q (curves 1), and threshold density of the neutron flux, F_t (curves 2) as functions of the fractional content of neon in the mixture of He–Ne–CCl_4. *Total pressure of the mixture is* 2 atm; CCl_4 *partial pressure is* 0.03 Torr; λ = 1.59 μm; $r_1 = r_2 = 0.998$.

In the course of this study we have also investigated numerically the dependence of the laser output power at λ = 1.59 μm on the coefficient of useful losses, $\varkappa = 1/2L\cdot\ln(1/(r_1\cdot r_2))$, where L is the resonator active length, r_1 and r_2 are the transmission coefficients of mirrors (see Fig. 4, curve *1*). The maximum in calculated power occurs at $\varkappa \approx 0.5\cdot10^{-3}$ cm^{-1} (the transmission of mirrors being about 12 to 14%) what is much lower than the experimental value so having higher amplitude. The fact that the concentration of impurities in the mixture is of the same order as that of the lasing component made us to assume that the differences in the purity of the laser chamber provided in different experiments could affect the shapes of the experimental curves along with certain inaccuracy in estimating the initial CCl_4 concentration. To check up this assumption we have calculated the output power, as a function of $\varkappa$, at different values of the CCl_4 initial concentration. Data of these calculations are shown in Fig. 4, curves *2* and *3*. From the calculated curves one can conclude that the experimental curve presented in this figure is constructed for the initial pressures of CCl_4 estimated with a certain error (up to a factor of two). Besides, one may calculate a curve that would be close to the experimental one by smoothly increasing the coefficient of parasitic absorption of laser radiation by the active medium and resonator mirrors, from $\varkappa^- = 1\cdot10^{-5}$ cm^{-1} to $\varkappa^- \approx 5\cdot10^{-4}$ cm^{-1}, while increasing the transmission of the resonator mirrors. It is quite probable that in our experiments both factors contributed.

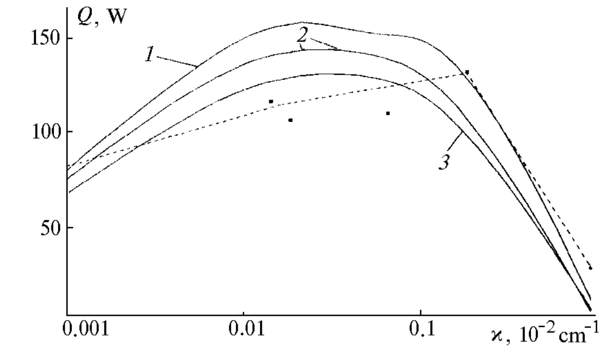

FIG. 4. The laser emission power, Q, as a function of useful losses at the CCl_4 *pressures of* 30 mTorr *(curve 1),* 15 mTorr *(curve 2), and* 60 mTorr *(curve 3). Solid lines present calculated data; dashed curves are the experimental results;* λ = 1.59 μm, *total pressure of the mixture is* 2 atm.

For the conditions of experiments described in Ref. 1 we have calculated the output power and efficiency of the lasing medium as functions of the neutron flux density F_t (pump power). In calculations made for the emission line at 1.59 micron wavelength we obtained that there should occur shortening of the laser pulse duration along with an increase of its peak power at increasing pump power. This circumstance explains the existence of an optimal efficiency η = 0.06% at $F_t \approx 5\cdot10^{15}$ neutrons/(cm^2·s).

Thus, we may state that we have constructed a detailed nonstationary kinetic model of a nuclear-pumped laser operating on the optical transitions in atomic chlorine and emitting at the wavelengths of 1.59 and 2.45 microns in the mixture of He–Ne–CCl_4.

From the results of numerical modeling we have established that the basic mechanisms of populating the upper lasing levels are dependent on the initial concentration of the lasing component (CCl_4) and on the pump power. Under optimal conditions for lasing ($p_{CCl_4} \approx 30$ mTorr) the dissociative recombination of the molecular chlorine ions is the main source of pump energy. The molecular ions of chlorine are an outcome of a complicated chain of plasmochemical reactions. At low concentrations of CCl_4, below 7 mTorr, the upper level pumping takes place due to triple recombination of the atomic chlorine ions, while at high concentrations, above 100 mTorr, the pumping mostly occurs due to the dissociative recombination of the molecular q Cl_2^+ ions. At low pump powers, near the threshold level, the ion–ion recombination of CCl_2^+ ions with the negative ions of chlorine plays the primary role in pumping. It is important that in all cases the low lasing levels are unloaded due to radiative processes and collisions with the buffer gas atoms.

The calculated optimal characteristics of lasing at 1.59 wavelength, i.e., the power $Q = 160$ W and the efficiency $\eta = 0.12\%$ may be achieved in a mixture of He–Ne–CCl_4 at the pressure of 2 atm and the composition He:Ne:CCl_4 = 25000:25000:1, the transmission of the resonator mirrors being $T = 12\%$. The minimum threshold density of the neutron flux required equals to $F_t \approx 2{\cdot}10^{13}$ neutrons per $cm^2{\cdot}s$.

REFERENCES

1. S.P. Mel'nikov and V.V. Porkhaev, Kvant. Elektron. **22**, No. 9, 891–894 (1995).
2. A.G. Zhidkov, S.V. Protopopov, O.V. Sereda, et al., Trudy IOFAN **21**, 116–138 (1989).
3. A.A. Vainshtein, I.I. Sobel'man, and E.A. Yukov *Cross-Sections of Exciting Atoms and Ions by Electrons* (Nauka, Moscow, 1973), 144 pp.
4. A.V. Karelin and S.I. Yakovlenko, Kvant. Elektron. **22**, No. 8, 769–775 (1995).
5. P. Spanel, et al. J. Phys. B. **28**, 2941–2957, (1995).
6. L.G. Rogoff, J.M. Kramer, and R.B. Piejak, IEEE Trans. on Plasma Science. **14**, 103–111 (1986).
7. A.P. Budnik and I.V. Dobrovol'skaya, Kvant. Elektron. **24**, No. 6 (1997).
8. A.V. Karelin, O.V. Sereda, V.V. Kharitonov, et al., Atomnaya Energiya **61**, No. 1, 44–46 (1986).