Plasma lasers involving molecules with a dissociative ground state

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The theory of plasma (recombination) lasers is developed for the case of amplification based on the transition of dimer molecules to the ground dissociative electron state. Conditions of predominance of the photodissociative transitions over the inverse photoassociative transitions are considered, as well as the kinetics of population of the upper working state under recombination conditions and generation under conditions of afterglow and of quasistationary production of supercooled (recombining) plasma. The results of experiments with plasma lasers based on molecules with an uncoupled ground state are discussed.

1. INTRODUCTION

The purpose of this paper is the development of a theory of plasma lasers (i.e., lasers whose active medium is recombining plasma[1]), as applied to amplification based on the transition of a dimer molecule to the ground dissociative electron state. In the past the theory of plasma lasers was developed mainly for transitions in atoms and atomic ions. It was noted that, from the viewpoint of the high pumping rates and efficiencies required to achieve high power output and of the feasibility of laser action in vacuum-ultraviolet and X-ray wavelength ranges, the recombination-nonequilibrium dense plasma has a number of advantages over the ionization-nonequilibrium plasma that is the working medium of electronic transition gas lasers[2].

The advantages of the recombination principle turn out to be decisive for plasma lasers based on the photodissociative transitions of molecules into ground (dissociative) state. The possibility of generation in "dissociative" molecules was suggested in print by Houtermans[3] back in 1960, but all attempts to achieve such a laser have so far been unsuccessful (see[4] for example). The failures were primarily due to the fact that the experiments were based on the concept of an ionization pumping regime (see[5] for example). Papers discussing generation in the photodissociative transition from the converse "recombination" viewpoint appeared relatively recently[6-10]. Reports of the first successful experimental results confirmed the expectations placed in the recombination principle: amplification in the dissociative transition of the Xe_2 molecule[11-14] was obtained in a decaying plasma.

Plasma lasers based on dissociative molecules can now be considered as some of the most promising laser types for the following reasons:

1. The working transition lies in the vacuum ultraviolet range for many dissociative molecules.

2. The achievement of population inversion with respect to the ground state in a dense medium allows one to expect high power levels and high efficiency.

3. The unusually broad homogeneous line of the corresponding spontaneous transition promises both tunable-frequency lasers and ultrashort-pulse generators.

The present paper contains an analysis of the problems that are basic to amplification in a gas of dissociative molecules.

2. AMPLIFICATION CONDITIONS

A pair of states of a diatomic molecule, the upper of which is bound and the lower is dissociative, is usually considered to be "automatically inverted," i.e., it is assumed that the photodissociative transition (into the dissociative state) predominates over the converse photoassociative transition. The criterion of "population inversion" is actually fairly rigid and in fact reduces to the requirement that the gas temperature T be sufficiently low. We now consider briefly the problems associated with the amplification and absorption of light quanta by dimer molecules X_2, and for the time being refrain from discussing the kinetics of populating the upper working state.

Let light of frequency ω_0 be amplified (and absorbed) in the transitions (see Fig. 1):

$$\begin{aligned} &X_2(2, n, l) \rightleftarrows X_2(1, E_{\text{diss}}) \\ &+\hbar\omega_0 = X(1) + X(1) + \hbar\omega_0. \end{aligned} \tag{2.1}$$

Here n and l are the vibrational and rotational quantum numbers respectively, E_{diss} is the energy of dissociation, and 1 and 2 are electron excitation numbers. In addition we should in general take the absorption in the photoionization of excited states of atoms and molecules into account. Assuming that the state $X_2(2)$ has the largest population, we can assume that the working emission losses in photoionization are determined by the process

$$X_2(2) + \hbar\omega_0 \rightarrow X_2^+ + e. \tag{2.2}$$

The amplification of light in photodissociative transitions, taking photoassociation into account, was

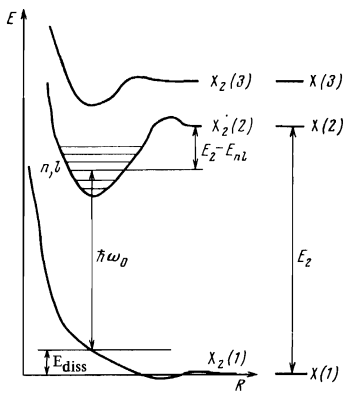

FIG. 1. Term diagram for atom X and molecule X_2.

studied by one of us[6], who obtained the following expression for the gain:

$$\varkappa=\sum_{n,l}\sigma_d^f(nl;E_{diss})\left[N_{2nl}^{X_2}-(2l+1)\left(\frac{2\pi\hbar^2}{\mu T}\right)^{3/2}\exp\left(-\frac{E_{nl}-\hbar\omega_0}{T}\right)(N_1^X)^2\right]. \quad (2.3)$$

Here $\sigma_d^f(nl;E_{diss})$ is the photodissociation cross section in the process (2.1), E_{nl} is the energy and $N_{2nl}X_2$ the population of state $x_2(2,n,l)$, μ is the relative mass of the molecule, and N_1^X is the population of the ground state X(1) of the atom. In (2.3) the first term in the square brackets takes the photodissociative transitions into account, while the second term pertains to photoassociative transitions. The photoionization absorption (2.2) could have been taken care of simply by introducing the factor $1-\sigma_i^f/\sigma_d^f$ in front of the first term, where σ_i^f is the cross section of the process (2.2). It is clear that amplification can take place only on condition that $\sigma_i^f<\sigma_d^f$, which is generally met with something to spare[1)]. From now on, assuming that $\sigma_i^f \ll \sigma_d^f$, the contribution from photoionization (2.2) will be neglected.

We consider the role of photoassociation. The main channel of populating the upper working state $X_2(2)$ is usually (see Sec. 3) association in the collision of a single excited X(2) and two unexcited X(1) atoms. The maximum possible population of the $X_2(2,n,l)$ state is given by the Saha-Boltzmann formula

$$N_{2nl}^{X_2}=(2l+1)\left(\frac{2\pi\hbar^2}{\mu T}\right)^{3/2}\exp\left(\frac{E_2-E_{nl}}{T}\right)N_2^X N_1^X, \quad (2.4)$$

where E_2 is the energy of excitation of an atom to the X(2) state and N_2^X is its population. Substituting (2.4) in (2.3) we have

$$\varkappa=(N_2^X-N_1^X e^{-\Delta E/T})N_1^X\left(\frac{2\pi\hbar^2}{\mu T}\right)^{3/2}\sum_{n,l}(2l+1)\sigma_d^f(nl;E_{diss}), \quad (2.5)$$

where $\Delta E \equiv E_2-\hbar\omega_0$. It follows from (2.5) that amplification in photodissociative transitions can be achieved only if the temperature is low enough:

$$T<T_{cr},\quad T_{cr}=\Delta E/\ln(N_1^X/N_2^X). \quad (2.6)$$

Since the population N_1^X of the ground state of the atom usually exceeds the population N_2^X of the excited state by several orders, the condition $T \ll \Delta E$ should hold. This leads to the requirement of sufficiently high heat-removal rate in continuous generation (Sec. 5) or to a limitation on the energy input in pulsed generation (Sec. 4). Condition (2.6) states that to reach population inversion in states participating in the (2.1) transition it is not enough to have a short decay time of the ground state $X_2(1, E_{diss})$ relative to the lifetime of the excited state $X_2(2)$, as suggested in a number of papers[3,5,15,16]. It is also erroneous to assume[16] that acts of emission and absorption of light by atoms moving past each other make a positive contribution to the amplification.

The photodissociation cross section can be represented in the form[6] $\sigma_d^f(nl;E_{diss})=(1/4)\lambda_0^2 A Q_l(n, E_{diss})$, where $\lambda_0=c/2\pi\omega_0$ is the wavelength of the amplified quantum, $A=4\omega_0^3 d_{12}/3\hbar c^3$ is the Einstein coefficient for spontaneous decay and is associated in the usual manner with the matrix element d_{12} of the transition expressed in terms of the electron wave functions, and $Q_l(n, E_{diss})$ is the squared overlap integral of the nuclear wave functions. The dependence of Q_l on E_{diss} determines the line shape. In the center of the line $Q_l(n)\approx 1/\Delta\omega_{nl}$, where $\Delta\omega_{nl}$ is the width of the wave function of the excited state projected onto the dissociative term and then onto the energy axis.

Considering the above, we shall use the following expression in the computation of gain

$$\varkappa=1/4\lambda_0^2 A N_2/\Delta\omega, \quad (2.7)$$

where $N_2 \equiv \Sigma N_{2n}^{X_2}$ is the population of the $X_2(2)$ state and $\Delta\omega$ is the effective line width. It is understood here that the inversion condition (2.6) has been met. The line width of the photodissociative transition, $\Delta\omega \sim 1$ eV $\approx 10^{15}$ sec^{-1}, is large in comparison to the line widths of electron transitions in atoms (it is three or four orders larger than the characteristic Doppler line width). This calls for an intensive pumping of the $X_2(2)$ state, which is feasible if the recombination is fast enough.

3. RELAXATION CHANNELS

We first inquire which processes fill and deplete the upper working state $X_2(2)$ in a recombining dense plasma. The problems associated with the preparation of strongly supercooled (intensely recombining) plasma are considered in Secs. 4 and 5.

In a highly dense gas (> 1 atm) under conditions of intense recombination, there are two channels of electron outflow: (a) via excited states of atoms, and (b) via excited states of molecules. It is important that both channels lead to the population of the upper working state $X_2(2)$.

The first relaxation channel can be described as follows: Triple recombination of atomic ions

$$X^+ + e + e \to X(m) + e \quad (3.1)$$

produces atoms X(m) in highly excited states. Flow over the energy levels is ensured by electron collisions of the second kind

$$X(m) + e \to X(n) + e, \quad E_m > E_n. \quad (3.2)$$

Relaxation from the excited levels continues down to the first excited state X(2). We assume here that the energy of this state is high enough; in that case the deexcitation acts by electrons

$$X(2) + e \to X(1) + e \quad (3.2a)$$

have a low probability. Since emission via $X(2) \to X(1)$ transitions is forbidden in dense gas, the relaxation from excited atomic states "turns" toward the formation of excited molecules $X_2(2)$, i.e., a collision association takes place

$$X(2) + X(1) + X(1) \to X_2(2) + X(1). \quad (3.3)$$

The other channel is the relaxation from the excited $X_2(m)$ states of molecules, beginning with the formation of a molecular ion X_2^+ as a result of conversion of an atomic ion

$$X^+ + X(1) + X(1) \to X_2^+ + X(1). \quad (3.4)$$

The atomic and molecular relaxation channels become intermixed because of the collision association and dissociation, for example:

$$X(m) + X(1) + X(1) \rightleftarrows X_2(m) + X(1) \quad (3.5)$$

or because of the dissociation of a molecule by electron impact, in particular by transition to one of the dissociative terms:

$$X_2(m)+e\to X(n)+X(1)+e. \quad (3.6)$$

In a number of cases the relation among the relaxation flows is determined by the dissociative recombination process

$$X_2^++e\to X(m)+X(1), \quad (3.7)$$

so that the main flow of electrons switches over from the molecular channel to the atomic channel. The remainder of the flow over the excited states of the molecules also reaches the $X_2(2)$ state with no perceptible additional losses[2)].

We now consider those processes that disrupt the upper working state. One of these is spontaneous photodissociative transition (2.1) to the ground dissociative state. The situation in which every act of ionization an atom X is accompanied by the formation of a molecule $X_2(2)$ and its subsequent radiative dissociation is from now on called the ideal situation. The real situation differs usually from the ideal one in that the decay of the upper working state is not always radiative. Among other ("parasitic") reactions that disrupt the upper state, the most energetic appears to be a variant of the Penning effect. This is ionization via collision of excited molecules:

$$X_2(2)+X_2(2)\to\begin{cases}X_2^++2X(1)+e\\ X^++3X(1)+e\end{cases}. \quad (3.8)$$

The process of de-excitation by an electron

$$X_2(2)+e\to X(1)+X(1)+e \quad (3.9)$$

just as process (3.2a), has a low probability if the first excited state $X_2(2)$ is far enough from the ground state. We also neglect the process of aggregation of excited associates

$$X_2(2)+X(1)+X(1)\to X_3(2)+X(1). \quad (3.10)$$

Summing up the above, we can accept the following scheme: The recombination flow passes almost entirely through the upper working state $X_2(2)$ of the molecule that decays via radiative dissociation (2.1) and reaction (3.8) occurring in the collision of two excited molecules.

4. PULSED GENERATION

We consider the conditions of generation in the photodissociative transition (2.1) and in the afterglow of an ionization pulse with a short trailing edge. We note that the initial method of ionization, i.e., a fast beam of charged particles, a transverse (relative to the amplification direction) electric field pulse, or laser radiation, is not particularly important to the amplification in the afterglow regime. Each of these methods has its own advantages. For example, gas breakdown by a laser pulse focused with cylindrical optics (see[17]) is convenient in testing the composition of the active medium, while the transverse electric field can be used in high-power high-energy laser oscillators.

In accordance with the recombination channels discussed above, we write the balance equations for the densities N_+ of the atomic ions X^+ and N_2 of the excited molecules in the state $X_2(2)$:

$$dN_+/dt=-k_rN_+^3+qN_2^2, \quad (4.1a)$$

$$dN_2/dt=k_rN_+^3-2qN_2^2-AN_2. \quad (4.1b)$$

Here $N_+\approx N_e$; $q=\langle\sigma v\rangle$ is the rate constant for reaction (3.8); and k_r is a coefficient characterizing the rate of recombination of ions X^+ into atomic and molecular states (see Sec. 3) and representing a complicated function of the concentration N_e and temperature T_e of electrons as well as the concentration $N\equiv N_1^X$ and temperature T of the heavy particles.

For the case in which dissociative recombination (3.7) leads to the population of atomic levels X(m) which lie substantially below the "flow throat" of the atomic relaxation channel, we can write

$$k_r=k_eN_e^2+k_cN^2, \quad (4.2)$$

where $k_e[cm^6\cdot sec^{-1}]\approx 8\times10^{-27}(T_e[eV])^{-9/2}$ is the coefficient of triple recombination (for details, see[18] for example), $k_c[cm^6\cdot sec^{-1}]\approx 10^{-31}\beta^{5/4}M^{-1/2}(T[eV])^{-3/4}$ is the coefficient of the conversion (3.4), β is the polarizability of atom X in atomic units, and M is its atomic weight (see[19], p. 332, for example).

In general, Eqs. (4.1) should be supplemented with equations for T_e and T, whose form depends significantly on the method of energy loading. If the energy is introduced by a pulse with a sufficiently short trailing edge, the degree of ionization of the medium is low ($\alpha\lesssim10^{-3}$), and the characteristic cooling time τ_{T_e} of the free electrons is short enough (see below for specific details), then T_e and T can be considered constant for the duration of the light-amplification pulse in the aferglow state. It follows from the plasma relaxation scheme discussed in Sec. 3 that the state $X_2(2)$ usually represents the bottleneck for the recombination flow. Therefore at sufficiently high initial concentrations $N_2(0)+N_e(0)\equiv N_{20}+N_{e0}$, $N_e(t)$ follows $N_2(t)$ in a quasi-stationary manner after a short interval following the pulse, i.e., we can assume that $dN_+/dt=0$ and consider the equations

$$dN_2/dt=-qN_2^2-AN_2, \quad k_rN_e=qN_2^2. \quad (4.3a)$$

The solution of the nonstationary Eq. (4.3a) is of the form

$$N_2(t)=N_{20}e^{-At}\left[1+\frac{q}{A}N_{20}(1-e^{-At})\right]^{-1}. \quad (4.3b)$$

The characteristic time $\tau_2(t)$ during which N_2 varies increases monotonically with time:

$$\tau_2(t)\equiv\left|\frac{1}{N_2}\frac{dN_2}{dt}\right|^{-1}=(qN_2+A)^{-1}=\begin{cases}(qN_{20}+A)^{-1} & \text{as } t\to0\\ A^{-1} & \text{as } t\to\infty\end{cases}.$$

The applicability of the quasi-stationary approximation (4.3) requires that

$$\tau_{Ne}=\left|\frac{1}{N_e}\frac{dN_e}{dt}\right|^{-1}\ll\tau_2$$

and consequently that $N_{e0}\ll N_{20}$.

We present some estimates. Let δW be the fraction of total energy W introduced into the gas and spent on ionization and excitation of particles of the medium. Then $N_{20}\approx\delta W/JSl$, where J is the energy of ionization of the particles (of atom X), S is the transverse cross section, and l is the length of the active region. For the amplification of light in a single pass through the resonator, at $t\approx0$, we have

$$\frac{I}{I_0}=e^{\varkappa l}(1-\gamma)=(1-\gamma)\exp\left\{\frac{\lambda_0^4}{4}\frac{A}{\Delta\omega}\frac{\delta W}{JS}\right\},$$

where γ is the energy loss coefficient in the resonator. The generation condition $I/I_0>1$ therefore becomes a lower bound of the pumping energy referred to the transverse cross section:

$$\delta W/S>U_t, \quad U_t=4|\ln(1-\gamma)|\Delta\omega J/\lambda_0^2A. \quad (4.4a)$$

We note that our approximations omitted the length l from (4.4a); in the general case the product κl depends weakly on l. Substituting typical parameters for the xenon Xe_2 molecule into (4.4a) ($\lambda_0 = 1730$ Å, $A = 5 \times 10^7$ sec^{-1}[11,12], $\Delta\omega \approx 10^{15}$ sec^{-1}, $J = 12$ eV), we obtain $U_t = 0.4$ J/cm^2 for $\gamma \sim 0.5$.

There is an upper bound of the introduced energy. According to Sec. 2 the gas should not be overheated, i.e., its temperature should not exceed a certain critical value T_{cr}. Consequently, the condition

$$(1-\delta)W/Sl < {}^3/_2 NT_{cr}. \tag{4.5}$$

should hold. In conjunction with (4.4a) we have

$$\frac{U_t}{\delta} < \frac{W}{S} < \frac{l}{1-\delta} NT_{cr}, \quad l > \frac{1-\delta}{\delta}\frac{U_t}{NT_{cr}}. \tag{4.4b}$$

The quantity T_{cr} can be evaluated from (2.6) with the aid of (4.3b). For the initial stage of the pulse (at $N_2(t) = N_{20}$) we use the estimate

$$T_{cr} = \Delta E / \ln\left(\frac{JSNl}{\delta W}\right). \tag{4.6}$$

Substituting in (4.6) $\delta W/S = U_t$ and $\Delta E \sim 1$ eV, we obtain from (4.4b) for xenon ($\delta = 0.5$):

$$l[\text{cm}]N[3\cdot 10^{19}\ \text{cm}^{-3}] > 0.5. \tag{4.5a}$$

Here $T_{cr} \approx 0.1$ eV within a broad range of values of the product $Nl \sim 1 - 10^3$.

Let us consider the limits of the requirement for a rapid cooling of free electrons. Assuming that electrons are cooled via elastic collisions, the inequality $\tau_{Te} < \tau_2$ yields the following limitation on the gas concentration:

$$N > \left(q\frac{U_t}{Jl} + A\right)\left(\frac{2m}{\mu}\langle\sigma_{elst} v\rangle\right)^{-1}, \tag{4.7}$$

where $\langle \sigma_{elst} v \rangle$ is the product of cross section and velocity averaged over the Maxwellian distribution, and m/μ is the ratio of electron mass to atomic mass. Substituting xenon parameters into (4.7) we obtain a requirement that is more stringent than (4.5a):

$$N[3\cdot 10^{19}\ \text{cm}^{-3}] > 2(1 + 2/l[\text{cm}]). \tag{4.7a}$$

According to the above assumptions, the characteristic length of the trailing edge of the pulse should satisfy the condition

$$\tau_{fr} < \tau_2 \approx (qU_t/Jl + A)^{-1}, \tag{4.8}$$

i.e., the time τ_{fr} should remain shorter than A^{-1} for any length of the active medium.

Consequently, in order to obtain generation in the pulsed regime, one should satisfy the following conditions, which are fairly liberal although unusual from the viewpoint of gas-laser practice[2]:

a. The trailing edge of the heating pulse should be sufficiently short (4.8);

b. The electrons should be cooled sufficiently rapidly (4.7).

In addition, (4.4)—(4.5) indicate the desirability of changing over to larger resonator lengths l while ensuring a minimal density U_t of the energy input. Helium is suitable as a buffer gas for fast cooling of electrons (and consequently for the intensification of generation). HeXe* helium compounds may prove to be of interest for the purpose of lasing.

The above conditions are most readily realized in laser breakdown. Therefore we once more stress the possibilities of this method, both in the search for optimal amplifying media and from the viewpoint of the pulsed relaxation process whose study is important to quantum electronics.

5. STATIONARY AMPLIFICATION

Stationary generation requires, in the first place, the maintenance of intensive recombination in plasma. For example, a high-current electron beam can be used for this purpose[1,9,20-22]. The method of feeding the energy to a dense gas should ensure an anomalously large (for a Maxwellian distribution with a temperature T_e) quantity of fast penetrating electrons (current electrons), which ionize and excite the atoms. As a result, the active medium acquires a high concentration of secondary electrons (plasma electrons) cooled-down in collisions with the atoms (first inelastic, then elastic). For a moderate degree of ionization α the temperature of heavy particles rises comparatively slowly. With a sufficiently fast heat removal, stationary amplification can be achieved in principle[3)].

We consider briefly how the parameters of electron-beam generated plasma are connected with the beam characteristics. If the beam introduced into a dense medium fails to excite plasma instabilities, which is apparently the case in a sufficiently dense medium[20,22], stationary operation is possible only when the recombination flow of plasma electrons neutralizes the ionization flow of current electrons.

If the degree of ionization is not too high ($\alpha \lesssim 10^{-3}$), the density of the excited molecules and the temperature and concentration of plasma electrons take on quasistationary (dependent on T and not explicitly dependent on the time t) values in times (τ_2, τ_{Te}, and τ_{Ne}, respectively) that are short in comparison to the characteristic time τ_T of gas temperature variation. The quasistationary equations for N_e, T_e, and T then take the form

$$dN_e/dt = -k_r N_e^2 + qN_2^2 + \nu_i N = 0, \tag{5.1a}$$

$$dN_2/dt = k_r N_e^2 + \nu_2 N - 2qN_2^2 - AN_2 = 0, \tag{5.1b}$$

$$\frac{3}{2}N_e\frac{dT_e}{dt} = \frac{3}{2}E^*\nu_{inl}N - Q_{\Delta T} = 0, \quad \frac{3}{2}N\frac{dT}{dt} = Q_{\Delta T} - Q_{cool}. \tag{5.1c}$$

Here $\nu_{inl} \approx \nu_2 + \nu_i$, ν_i, and ν_2 characterize the corresponding velocities of inelastic interaction, ionization, and excitation of atoms to the X(2) state by beam (current) electrons and fast secondary electrons before they are cooled by inelastic collisions. In the computation, ν_{inl}, ν_2, and ν_i can be related to the corresponding cross sections σ_{inl}, σ_2, and σ_i and to the current density j by $\nu \approx (1-2)\sigma j/e$, where e is the electron charge. The quantity $E^* + \hbar\omega_0$ yields the average energy expended on the formation of a bound state of a molecule. The cooling of plasma electrons due to elastic collisions with atoms is characterized by the quantity

$$Q_{\Delta T} = \frac{3m}{\mu}\langle\sigma_{elst} v\rangle N_e N(T_e - T); \tag{5.2}$$

Q_{cool} effectively takes gas cooling into account.

The following relations between characteristic times follow from (5.1)

$$\alpha\nu_{inl}^{-1} \sim \tau_{Te}\frac{E^*}{T_e} \approx (\tau_{Ne} + \tau_2), \quad \frac{TN}{T_e N_e}(\tau_{Te}) = \tau_T \sim \frac{T}{E^*}\nu_{inl}^{-1}. \tag{5.3}$$

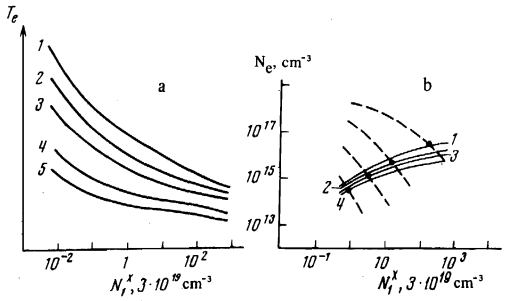

FIG. 2. Dependence of temperature (a) and concentration (b) of electrons on the density of xenon-helium mixtures for $\nu_i = 10^3$ sec^{-1}. Dashed lines represent equilibrium values of N_e^0 for the corresponding T_e; 1–0% He; 2–7.4% He; 3–20% He; 4–50% He; 5–100% He.

For $\alpha \ll T/E^*$ their hierarchy is as follows:

$$\tau_{Te} \ll (\tau_{Ne}+\tau_2) \ll \tau_T \ll \nu_{inl}^{-1}. \qquad (5.4)$$

Amplification requires that $T \lesssim 0.1$ eV and E^* usually amounts to several eV; hence it follows that $\alpha \lesssim 10^{-3}$.

Equations (5.1) hold in the case of an intense recombination when it is possible to neglect the ionization of atoms by plasma electrons corresponding to the hot tail of the energy distribution. This may not hold in the case of a heavy gas such as xenon. In fact, the deviation of the electron temperature from the gas temperature, determined by the relation

$$T_e - T = E^* \nu_{inl} \Big/ \frac{2m}{\mu} \langle \sigma_{elst} v \rangle N_e,$$

increases with μ. The above can be illustrated by the results of simulation computation of N_e and T_e for xenon-helium mixtures shown in Fig. 2. The electron concentration was found from the relation $N_e = (\nu_i N/k_e)^{1/3}$, i.e., the triple recombination (see (4.2)) was assumed to be dominant.

As we know, dissociative recombination (see[23] for example) predominates in dense xenon at low temperatures ($T_e \sim 0.05$ eV). However, for the high temperatures obtained in the computation, the "flow throat" of the recombination lies practically between the states X(2) and X(1), and therefore triple recombination seems more justified. Nevertheless the computation, as mentioned above, is a simulation process and illustrates the analytic method and the qualitative aspect of the relationships rather than providing numerical results.

For the population N_2 we obtain from (5.1a) and (5.1b)

$$N_2 = \frac{A}{q}\left[\left(1+4q\nu_{inl}\frac{N}{A^2}\right)^{1/2}-1\right]. \qquad (5.5)$$

Thus an ideal situation takes place for $4qN\nu_{inl} \ll A^2$, and $\nu_{inl}N = AN_2$. With increasing pumping intensity $\nu_{inl}N$, the value of N_2 varies practically linearly up to the critical value $N_{2cr} = A/2q$ following which, at $4q\nu_{inl}N \gg A^2$, the function $N_2(\nu_{inl}N)$ changes over to a square-root relation $N_2 = (\nu_{inl}N/q)^{1/2}$. Consequently, molecules with a large value of A are of the greatest interest. Thus, for a xenon molecule ($A \approx 5 \times 10^7$ sec) at $\nu_{inl} \sim 10^3$ sec^{-1} the gain coefficient increases linearly $\kappa \propto \nu_{inl}N$ up to pressures of the order of 30 atm, reaching the value ~ 0.01 cm^{-1}. At the same time, for molecules with low values of the Einstein coefficient at the working transition ($A < 10^4$ sec^{-1}) effective amplification in stationary operation is fairly problematic.

The gas cooling problem is of basic importance to generation in stationary operation. According to (5.3), at $\nu_{inl} \sim 10^3$ sec, the cooling time τ_{cool} should be shorter than $\tau_T \sim 10^{-5}$ sec. Such time intervals are not readily achieved by means of thermal conductivity that requires unrealistic limitation of the transverse dimension of the active medium. Therefore it makes sense to consider a quasi-stationary generation of pulses with a length of $\tau_{Ne} + \tau_2 < \tau < \tau_T \sim 10^{-5}$ sec, or to search for more effective gas cooling methods such as the nozzle expansion method.

6. CONCLUSION

We now briefly compare the several published methods of dissociative molecular generation. We first consider the optical pumping methods proposed in the literature.

Borovich and Zuev[24] proposed to use optical pumping of the $X_2(2)$ state via the photo-associative transition (2.1) that is the reverse of the working transition. This idea is difficult to realize because of the low probability of such a process. Thus according to[24] a gain of $\kappa \sim 10^{-3}$ cm^{-1} would require a photo-associative transition absorption coefficient $\kappa_{abs} \sim 1-10$ cm^{-1}. In such a case, according to (2.3), the gas concentration should exceed 10^{23} cm^{-3}.

Photon absorption by an atom (including absorption at a frequency shifted by another passing atom) is more probable by many orders of magnitude. Therefore another method is more feasible: optical pumping of the excited state of the X(2) atom with a subsequent formation of molecules as a result of collision association (3.3). Gas heating is relatively moderate in schemes of this type. One-photon pumping appears less promising here, but even this is of interest, at least from the viewpoint of the opportunity to study the relaxation of vibrational-rotational degrees of freedom (see[25] for example). An attempt was made[26] to achieve generation by two-photon pumping, but so far it was not possible to reach the self-excitation threshold.

We now consider the attempts to use the traditional gas discharge methods. The ionization principle of a gas laser is analogous to the recombination principle of a plasma laser. Based on the ionization principle, a number of authors proposed the use of plasma with superheated electrons formed in the ordinary gas discharge[4,5,15], such as an arc[5], or in the leading front of a heating-field pulse[16]. Calculations based on idealized theory (according to which a significant fraction of the atomic excitation events is accompanied by the emission of a photo-dissociation quantum) give an encouraging result in this case. Actually, however, the idealized theory is much farther from reality in the ionization regime than in the recombination regime. This is basically due to the following two reasons. First, the ionization flow "running upwards" through the excited states (in contrast to the recombination flow "running downwards") fails to "stick" properly in the X(2) and $X_2(2)$ states, i.e., there is an additional parasitic channel due to further excitation and ionization. Second, it is difficult to ensure a significant difference between the electron and gas temperatures at a high concentration of electrons in a dense medium. At the same time, excitation and ionization involving a small number of fast current electrons lead to the recombination regime (see Sec. 5).

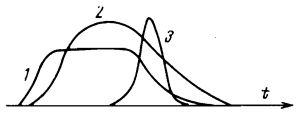

FIG. 3. Typical dynamics of the pulse development [12]; 1—pumping; 2—spontaneous luminescence; 3—laser emission.

A number of reports has recently appeared[11-14] (the number continues to increase) of successful realization of plasma lasers based on dissociative molecules and a relativistic electron beam. Thus generation at the molecular transition ($\lambda_0 \approx 1730$ Å) in dense (~10 atm) xenon ionized by an electron beam characterized by electron energy V = 1.5 MeV, current density $j \approx 200$ A/cm^2 (total current $J \approx 350$ A), and pulse length of ≈50 nsec, is reported in[11,12]. The oscillograms (see Fig. 3) published by Wayne Johnson and Gerardo[12] show that the medium begins to amplify weakly the radiation shortly before the end of the electron-beam pulse. The maximum gain occurs in the afterglow stage and appears after the maximum of spontaneous emission. It is thus clear that the generation proceeds in the recombination regime. After the beam is switched off (the trailing pulse edge is $\tau_{fr} \approx 2 \times 10^{-8}$ sec) the electrons cool down in a time $\tau_{Te} \sim 10^{-8}$ sec, the recombination intensifies, and the gain reaches the value required for generation. Consequently, the results of the experiments reported in[12] can be explained in terms of the recombination scheme[4)].

Generation by dissociative molecules of xenon and krypton in the recombination regime was reported by Hoff, Swingle, and Rhodes[13], while the preliminary theoretical investigation of feasibility of such a generation was performed by George and Rhodes[8].

The experiments cited above undoubtedly represent only the first step in the development of plasma lasers based on dissociative molecules. Further steps should be taken in the search for optimal media and new sources that would be more convenient and have a higher energy than the electron beams. The transverse pulsed electric discharge is of interest from the viewpoint of the possible high efficiency and the resulting high-power and high-energy lasers.

The above discussion shows that pure xenon is not the optimal gas. Apparently it makes sense to consider various helium-containing compositions and the possibility of generation based on asymmetric diatomic molecules such as HeXe* and XeH*.

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[1)]This is due to the fact that the wave function of the emitted electron is smeared out over a larger energy interval than the wave function of the nuclei.

[2)]Such losses are basically due to the radiative decay of highly excited molecules to the ground state. For the atomic channel, losses of this kind are even less significant because of the practically total reabsorption of the resonance emission of atoms in a dense medium.

[3)]The high heat capacity requirement of the pulsed regime is replaced by the requirement of a high heat transfer rate in the stationary regime.

[4)]We have recently learned of results by the same authors [27], who used much more intense pumping (V = 1.5 MeV, J = 3 × 10^4 A) of xenon by an electron beam in order to achieve the maximum generation energy yield. The time behavior of the beam current and of the spontaneous and stimulated emission intensity has a significantly different nature; the generation falls off as early as in the beam-current rise stage. According to (4.5) this is due to the rapid superheating of the gas (during a time interval that is one-third as short as the ionizing pulse length).

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