AMPLIFICATION OF IR RADIATION IN ACTIVE MEDIA OF EXCIMER LASERS

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Abstract—There are predicted 1) the existence of the far IR optical activity of the heteronuclear excimer molecules (EM's); 2) the formation of an inverse population of certain vibrational-rotational levels of EM's; and 3) the amplification of the IR radiation corresponding to transitions between those levels.

1. -Introduction.

Excimer UV lasers are among the most studied and promising ones. One of their peculiarities is that the upper vibrational levels of EM's are pumped in physico-chemical processes, while only the ground level or the few low vibrational levels contribute to the generation. A strongly non-equilibrium distribution of EM's over vibrational levels may be formed as a result of vibrational relaxation which is accompanied by radiative decay, collisional deactivation, and coupling between excited states. As examples of such non-equilibrium distributions of EM's we can mention the experimental papers [1,2]. In Ref. 2 the inverted populations of vibrational levels with numbers $v = 0 + 5$ have been observed in mercury halide discharge lasers.

2. -Inverted population of vibrational-rotational levels.

We have investigated the kinetics of electronic-vibrational relaxation of molecules in [3]. The distribution of EM's over vibrational levels is governed by the processes of population, deactivation and VT-relaxation. We have found the distribution function f over vibrational energy ε when the energy of EM formation ε^* are large,

$$f(\varepsilon) = \begin{cases} N \frac{[1 - (1 - \varepsilon/D)^{1/2}]^{\alpha-1}}{[1 - (1 - \varepsilon^*/D)^{1/2}]^{\alpha}} \frac{\hbar\omega}{2D} \rho(\varepsilon), & \varepsilon \gg T, \\ N \frac{\hbar\omega}{T} e^{-\frac{\varepsilon}{T}} g \Gamma(1-\alpha) F(\alpha, 1; \frac{\varepsilon}{T}) \rho(\varepsilon), & \varepsilon \ll D, \end{cases} \quad (1)$$

Here N is the EM volume density, $\alpha \equiv \tau_v/\tau_q$, τ_v - is the VT-relaxation time, τ_q is the lifetime, $\hbar\omega$ is the vibrational quantum, D is the dissociation energy, ρ is the density of vibrational states of the EM, T is the temperature in energy units, F denotes the degenerate

hypergeometric function of the first order, $g = \{T/[2D(1 - \sqrt{1 - \varepsilon^*/D})]\}^{\alpha}$.

In the region of upper vibrational levels even in the case of fast vibrational relaxation the distribution function becomes strongly non-equilibrium. For the value of τ_v we have estimated $\tau_v \geq 10^2 \tau$, where τ is the collision time. Meanwhile, rotational relaxation time τ_r is equal approximately $\tau/3$. That is why the population inversion should exist between certain vibrational-rotational levels.

With distribution (1) the inversion density on the vibrational-rotational transition $v, j \rightarrow v-1, j \pm 1$ equals $/3/$

$$\Delta_{v-1, j \pm 1}^{v, j} = \left[\pm \frac{j}{j_m^2} - \frac{1-\alpha}{v} \right] \frac{(2j+1)B}{T} \frac{\hbar\omega}{2D} \frac{f(\varepsilon_v)}{\rho(\varepsilon_v)} e^{-\frac{Bj(j+1)}{T}},$$

where B is the EM rotational constant, ε_v is the energy of the v -th vibrational level, $j_m = \sqrt{T/2B}$ is the number of the most populated rotational level. With $\alpha > 1$, complete inversion is implemented, and the gain can be attained for all transitions in the P-branches of vibrational-rotational spectrum as well as those transitions of R-branches, for which $j < j_m^2 \frac{1-\alpha}{v}$. If $\alpha < 1$, only the partial inversion exists and the gain is possible for transitions of P-branches with $j_m^2 \frac{1-\alpha}{v} < j < j_m^2 \frac{\hbar\omega}{T}$.

3.-Far IR-radiation gain.

Heteronuclear EM's are characterized by the ion coupling, and the radiative IR-transitions should take place between their vibrational levels. To estimate the optical activity of the EM's in far IR spectral region one can use the well known analogy between the ionic states of the EM's and the ground states of the alkali halide molecules (see also /4/). Optical parameters of certain EM's that we have calculated are given in the table,

Table. Optical parameters of excimers .

Excimer	KrF	XeF	XeCl
$\lambda, \mu\text{m}$	35 - 50	35 - 60	55 - 70
A, s^{-1}	0.8	0.9	0.2
$p\delta_v/\nu$	0.5	0.6	0.3

Here λ is the wavelength of radiation corresponding to transitions from some vibrational level ($v = 10 + 50$), A is the Einstein coefficient for the transition $v = 1 \rightarrow v = 0$, $\delta_v (10^{-16} \text{ cm}^2)$ is the induced phototransition cross-section for $v \rightarrow v-1$ transition, $p(\text{atm})$ is the gas pressure. In the course of calculations the collisional width of an IR-radiation line was assumed equal to 4 GHz/atm $^{1/3}$.

For the IR-radiation gain at the transition $v, j_m \rightarrow v-1, j_m \pm 1$ ($\epsilon_v \gg T$) the following expression was obtained:

$$g_v = \delta_v \left[\pm \frac{1}{j_m} - \frac{1-x}{v} \right] \frac{x N}{\sqrt{e} j_m v} \left[\frac{\epsilon_v}{\epsilon^*} \right]^x \quad (2)$$

Let us consider the mixture of 1.5 % F_2 , 10 % Kr, 89.5 % He, at $p = 0.05 \text{ atm}$, $T = 300 \text{ K}$, as an example. The medium with these parameters was excited by an electric discharge in the volume 2.8 cm^3 /5/. Ref. 5 gives the measurement of spontaneous radiation energy of KrF which equals 32.5 mJ at the luminescence pulse width 100 ns. For this case we have estimated the volume density of the EM's $N = 3 \cdot 10^{15} \text{ cm}^{-3}$. In the active medium under study $x \approx 1$, $\epsilon^* \approx 1.8 \text{ eV}$. For EM at the transition $v = 25$, $j_m = 22 \rightarrow v = 24$, $j = 23$, we obtain $\delta_v = 2.5 \cdot 10^{-14} \text{ cm}^2$, $\lambda = 40 \mu\text{m}$; and $\delta_v = 5 \cdot 10^{-14} \text{ cm}^2$, $\lambda = 50 \mu\text{m}$ for the transition $v = 50 \rightarrow v = 49$. Substituting the values of parameters into (2), we obtain $g_{25} = 2 \cdot 10^{-3} \text{ cm}^{-1}$, $g_{50} = 4 \cdot 10^{-3} \text{ cm}^{-1}$.

4. -Absorption of IR-radiation by plasma.

It is well known that a plasma absorbs an IR radiation. Thus, for the active medium excited by an electron beam or electric discharge it is necessary to estimate this absorption. Let us consider the dissipation of IR-radiation in the plasma. We will calculate the absorption coefficient γ for the radiation with angular frequency Ω : $\gamma = \frac{\Omega}{c} \epsilon' / \sqrt{\epsilon''}$, where ϵ' and ϵ'' are the real and imaginary part of the complex electric permittivity, c is the velocity of light. If Ω is considerably higher than the plasma frequency Ω_0 , we have

$$g' = 1 - \frac{\Omega_0^2}{\Omega^2 + \nu^2}, \quad g'' = -\frac{\nu}{\Omega} (1 - g'), \quad \Omega_0^2 = 4\pi e^2 \frac{n_e}{m_e},$$

where n_e is the density of electrons, e , m_e are the charge and mass of electron in absolute units, ν is the effective frequency of electron collisions with ions and neutral molecules, $\nu = N \delta_{\text{eff}} v_e$, N is the number of particles in unit volume, δ_{eff} is the effective collision cross section, v_e is the velocity of electrons, $v_e^2 = 2k_B T_e / m_e$, T_e is the temperature of electrons, k_B is the Boltzmann constant. At electron collisions with ions the effective collision cross section equals $\delta^+ = 4\pi e^4 / (m_e^2 v_e^4)$. At collisions with neutral particles we have assume $\delta_{\text{eff}} = 0.1 \cdot \delta^+$. Therefore, at $p = 0.05$ atm, $n_e = 10^{14} + 10^{15} \text{ cm}^{-3}$, $T_e = 10$ eV we have $\Omega_0 = 6 \cdot 10^{11} + 2 \cdot 10^{12} \text{ Hz}$, and $\nu = 2 \cdot 10^{10} \text{ Hz}$. For the emission with the wavelength $\lambda = 35 \mu\text{m}$ ($\Omega = 5 \cdot 10^{13} \text{ Hz}$) the absorption coefficient equals $\gamma = 6 \cdot 10^{-5} + 6 \cdot 10^{-4} \text{ cm}^{-1}$. That means that under the conditions mentioned above the absorption coefficient γ is smaller than the gain g (see Sec.3).

5. -IR absorption spectroscopy of EM's.

In accordance with Eq. (1), there is no population inversion in the region of low vibrational levels. Therefore, only the absorption of IR-radiation can exist in this region. In this case for the phototransitions $v-1, j_m \rightarrow v, j_m \pm 1$ the absorption coefficient equals

$$\alpha_{v-1} = \delta_v \frac{\Delta N}{\sqrt{e} j_m} \quad (3)$$

where $\Delta N = N_{v-1} - N_v$ is the difference of populations of $(v-1)$ -th and v -th vibrational levels.

For the $B \rightarrow X$ transitions of KrF and XeF molecules various electronic-vibrational-rotational bands overlap with each other. The UV-spectroscopy of EM's is therefore complicated. In this case an IR-spectroscopy may serve as an additional tool for a study of EM's.

As an example, let us consider the absorption in the XeF-laser with photolytical pumping of XeF_2 [6]. In Ref. 6 the mixture of XeF_2 (at the volume density $7 \cdot 10^{16} \text{ cm}^{-3}$) and Ar (at a pressure p from 0 to 150 torr) was excited by the optical pumping with the wavelength $\lambda_0 = 172 \text{ nm}$. The length of active medium was equal $L = 30 \text{ cm}$. It is known that the first 6 vibrational levels of XeF(B) were primarily populated. Lasing has been observed for UV-transitions from the vibrational levels with $v = 1, 2$, and 3. The intensity of spontaneous luminescence was highest at $p = 15$ torr. At this pressure we have $\delta_4 = 1.2 \cdot 10^{-14} \text{ cm}^2$ for the transition from $v = 3$ to $v = 4$ (see the Table). For the XeF(B) molecule we have $B = 0.15 \text{ cm}^{-1}$, and $j_m = 27$. According

to Eq. (3) we shall have $\alpha L = 10\%$ at $\Delta N = 10^{13} \text{ cm}^{-3}$. Thus the coefficient of the IR absorption by the low vibrational levels of EM's is high enough at typical concentrations of EM's ($N = 10^{14} + 10^{15} \text{ cm}^{-3}$).

One can use the far IR spectroscopy as a new approach to an excimer's study. It permits to analyze the population of laser levels, to study details of the relaxation processes, and so on. On the other hand, the existence of the tunable laser sources for the spectral region $25 + 50 \text{ } \mu\text{m}$ (the semiconductor lasers) opens up new possibilities to influence on the vibrational distribution of the EM's and to control some features of UV lasers.

6.-Conclusions.

Taking into account the ionic character of the excited states of the heteronuclear excimers in this paper it is predicted the far IR optical activity for these molecules. It is shown that the new effect of the far IR-radiation amplification can exist in the active media of excimer lasers. According with our estimates, inversion and gain takes place for transitions from a great number of vibrational levels and, due to anharmonicity, one excimer radiates in a wide spectral range ($\lambda = 30 + 70 \text{ } \mu\text{m}$ for KrF, XeF, and XeCl). The absorption of IR-radiation by plasma should be taken into account. However, it can be small enough and the far IR radiation of EM's can be amplified in the media excited by an electric discharge.

The spectroscopy of IR-absorption can be applied for a study of low vibrational levels of EM's.

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