

# Investigation of Vibrational Relaxation of Excimers

## Quantum Yield of UV Emission and Amplification of Far Infrared Radiation

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**Abstract.** The excimer vibrational relaxation have been studied in the diffusional approximation, and stationary substantially non-equilibrium populations of high vibrational levels have been found. New formulae have been derived for quantum yield of UV lasing. Taking into account the vibrational relaxation the new effect of far-IR radiation amplification in the active media of excimer lasers is predicted.

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The present paper aims at investigating the electron-vibrational relaxation in the active media of excimer lasers (EL). In ELs the formation of electron-excited molecules is due to recombination and/or exchange reactions on upper vibrational levels. Thus, the vibrational relaxation is the essential intermediate stage of laser levels pumping. In the course of vibrational relaxation, a portion of the excited molecules is quenched and this affects the laser energy-extraction efficiency.

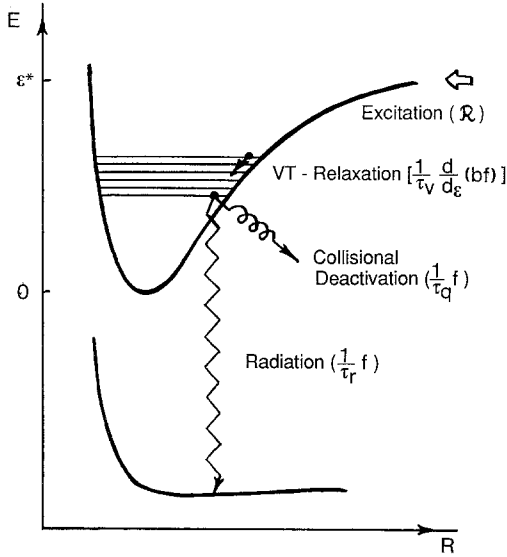
The influence of electron kinetics of ELs parameters has been studied by many researchers. However, there are only simplified theoretical models that simultaneously account for both the electron deactivation and the vibrational relaxation. In the models [1–3] the populations of those levels which do not contribute to the lasing were assumed in thermal equilibrium. There are also several experimental papers dealing with relaxation processes in excimers. In [4–7] the propagation of saturating picosecond or subpicosecond light pulses through active media of excimer lasers and gain dynamics have been studied. The data on the gain recovery made it possible to determine the time scales of the rotational and vibrational relaxation processes. The fraction of the excimers lying on the low vibrational levels and contributing to picosecond lasing have been determined [5]. In our paper the stationary substantially non-equilibrium populations of high vibrational levels have been found theoretically. New formulae have been derived for the quantum yield of lasing with allowance for the excitation of higher vibrational levels, the VT-relaxation, and the electron deactivation. The formulae allow one to calculate the quantum yield of laser emission and the EL internal efficiency depend-

ing on the vibrational energy of excimers excitation, the intracavity laser flux, excimers kinetic parameters, and the EL active media composition. The excimer distributions over vibrational levels have been studied using concept of the electron-vibrational relaxation developed in this work. The existence of distributions with higher population of upper vibrational levels is demonstrated.

As it is known, heteronucleus excimers (for example, the inert-gas halides) are characterised by ionic binding. The excimer's dipole moment may be found using the known analogy of ionic states of inert-gas halides and the ground state of alkali-halides. The dipole moment depends near-linearly on internuclear distance, so the optical transitions between neighbouring vibrational levels (that is far-IR emission) should exist. The excimers' IR activity (emission, absorption) has not been examined in the literature before. The occurrence of the inverse population of the vibrational-rotational levels and the existence of IR phototransitions may possibly lead to new phenomena, such as light amplification and generation in the far-IR spectral region. In this work it is shown for the first time that the IR-radiation gain can be obtained in the active medium of rare-gas-halide ELs.

### 1. Equation of Electron-Vibrational Kinetics

The electron-vibrational kinetics of excimers covers the population processes of high-vibrational levels, VT relaxation, quenching, state conversion, spontaneous and stimulated phototransitions (Fig. 1). At pressures of  $P \gtrsim 0.05$  atm, the collisional mixing of electron states proceeds



**Fig. 1.** Schematic presentation of electron terms of the excimer and the main kinetic processes. The values in parentheses are the process rates per unit excimer energy interval, i.e. the numbers of excimer transitions from unit vibrational energy interval per 1 s

at a much higher rate in comparison with other processes (see, for example, [8]). One can, therefore, derive a distribution function  $f(\varepsilon)$ , such that  $f(\varepsilon) d\varepsilon$  is the number of molecules in the electron-excited states with the vibrational energy  $\varepsilon$  in the range of  $\varepsilon, \varepsilon + d\varepsilon$ . The vibrational distributions in specific states are characterized by the functions of the type  $f_i(\varepsilon) = g_i f(\varepsilon) / \sum_i g_i$ , where  $g_i$  is the electron degeneracy of state  $i$ .

In excimers, the VT transitions are the multiquantum ones [9], and the distribution function  $f(\varepsilon)$  varies smoothly. In this case, the diffusional approximation shall be valid [10], and the relaxation shall be described by the Fokker-Planck equation which has the following form for the condition of spontaneous luminescence in ELs.

$$\frac{dJ(\varepsilon)}{d\varepsilon} + \frac{1}{\tau_u} f(\varepsilon) = \sum_l R_l(\varepsilon), \quad (1)$$

where  $J(\varepsilon) = -B(\varepsilon) \varrho(\varepsilon) \left[ \frac{d}{d\varepsilon} \left( \frac{f}{\varrho} \right) + \frac{f}{T\varrho} \right]$ ,  $B(\varepsilon) = \frac{Tb(\varepsilon)}{\tau_v}$  is the coefficient of energy diffusion,  $0 < \varepsilon < D$ ,  $D$  is the excited state dissociation energy,  $\tau_v$  is the VT-relaxation time, the multiplier  $b(\varepsilon)$  allows for the dependence of  $B$  on  $\varepsilon$ . For instance, for the harmonic oscillator  $b(\varepsilon) = \varepsilon$ .  $\varrho(\varepsilon)$  is the density of vibrational states,  $T$  is the temperature,  $R_l(\varepsilon)$  is the function characterising the formation of species during the reaction 1,  $\tau_u^{-1} = \tau_r^{-1} + \tau_q^{-1}$ ,  $\tau_r, \tau_q$  are the radiative and collisional lifetimes, respectively. For collisionally-mixed states  $\tau_r = \sum_i g_i / \sum_i g_i \tau_{ri}^{-1}$ ,  $\tau_{ri}$  is the radiative life time of state  $i$ . For ELs  $\tau_v / \tau_u \equiv \kappa \ll 1$  normally holds.

## 2. Vibrational States Distribution

Let the function  $R_l(\varepsilon) d\varepsilon$  equal to the number of nascences of excimers with the energy in the range of  $\varepsilon, \varepsilon + d\varepsilon$  per 1 s,

be different from zero over the relatively narrow region from  $\varepsilon_l^*$  to  $\varepsilon_l^* + \Delta\varepsilon_l$  ( $\Delta\varepsilon_l \ll \varepsilon_l^*$ ). Integration of (1) yields

$$J(\varepsilon_l^*) = - \int_{\varepsilon_l^*}^{\varepsilon_l^* + \Delta\varepsilon_l} R(\varepsilon) d\varepsilon \equiv -J_l^*, \quad (2)$$

where  $J_l^*$  denotes the pumping rate in the reaction 1. The second boundary condition follows from the law of conservation of species number.

$$J(0) = 0 \quad (3)$$

Solution of (1) with the boundary conditions (2) and (3) yields

$$f(\varepsilon) = \sum_l J_l^* \tau_u f_l(\varepsilon),$$

$$f_l(\varepsilon) = \begin{cases} \frac{\kappa}{b(\varepsilon)} \exp \left( -\tau_v \int_{\varepsilon}^{\varepsilon_l^*} \frac{d\varepsilon}{\tau_u b(\varepsilon)} \right), & T \ll \varepsilon < \varepsilon_l^* \\ \frac{\hbar\omega}{T} e^{-\frac{\varepsilon}{T}} \theta_l \Gamma(1 + \kappa) F \left( \kappa, 1; \frac{\varepsilon}{T} \right) \varrho(\varepsilon), & \varepsilon \ll D \end{cases} \quad (4)$$

where  $\theta_l \equiv \exp \left\{ -\tau_v \int_T^{\varepsilon_l^*} d\varepsilon [\tau_u b(\varepsilon)]^{-1} \right\}$ ,  $\hbar\omega$  is the energy of the excimer's vibrational quantum,  $F$  denotes the degenerate hypergeometric function of the first order. When deriving (4) it was assumed that with  $\varepsilon \ll D$  the form of  $b(\varepsilon)$  coincided with that of the harmonic oscillator,  $b = \varepsilon$  and  $\tau_u = \text{const}$ . Using the approximation obtained in [10] for the Morse oscillator,  $b(\varepsilon) = 2D(1 - \sqrt{1 - \varepsilon/D}) \sqrt{1 - \varepsilon/D}$ , for  $\varepsilon \gg T$  one can obtain

$$f_l(\varepsilon) = \kappa \frac{[1 - (1 - \varepsilon/D)^{1/2}]^{\kappa-1}}{[1 - (1 - \varepsilon_l^*/D)^{1/2}]^{\kappa}} \frac{\hbar\omega}{2D} \varrho(\varepsilon) \quad (5)$$

and  $\theta_l \equiv [T / (2D(1 - \sqrt{1 - \varepsilon_l^*/D}))]^{\kappa}$  will be included in (4).

Let us determine the quantum yield of the excimer's spontaneous luminescence from the vibrational level with the energy of  $\varepsilon_0 - \eta_{sp}$  as the ratio between the rate of radiative decay of this level,  $\tau_r^{-1} f(\varepsilon_0) \varrho^{-1}(\varepsilon_0)$ , and the pumping rate  $J^* = \sum_l J_l^*$ . Making use of (4) with  $\varepsilon_0 \lesssim T$  yields

$$\eta_{sp} = \frac{\tau_u}{\tau_r} \frac{\hbar\omega}{T} e^{-\frac{\varepsilon_0}{T}} F \left( \kappa, 1; \frac{\varepsilon_0}{T} \right) \sum_l \beta_l \theta_l, \quad (6)$$

where  $\beta_l \equiv J_l^* / J^*$ . Equation (6) indicates the importance of mixture composition. Specifically, the nonequilibrium nature of vibrational population causes the nonmonotonic (with the maximum) dependence of  $\eta_{sp}$  on buffer gas pressure.

## 3. Quantum Yield of Laser Emission

Let us consider the condition of stimulated radiation of ELs. Let stimulated phototransitions occur in the energy range of  $0 < \varepsilon < \varepsilon'$  ( $\varepsilon' \ll \varepsilon_l^*$ ) where the reverse lifetime

equal  $\tilde{\tau}_u^{-1} = \tau_u^{-1} + \sigma\varphi/h\nu$  with  $\sigma$  being the stimulated transition cross-section,  $\varphi$  is the intracavity laser flux,  $h\nu$  is the photon energy [11]. Solution of (1) with  $\tilde{\tau}_u$  in the interval  $0 < \varepsilon < \varepsilon'$  yields

$$f(\varepsilon) = \frac{\sum_l J_l^* \tau_u \eta_l^v}{1 + \varphi/\varphi_s} \frac{\hbar\omega}{T} \left(\frac{T}{\varepsilon'}\right)^{\tilde{\kappa}} \Gamma(1 + \tilde{\kappa}) e^{-\frac{\varepsilon}{T}} \times F\left(\tilde{\kappa}, 1; \frac{\varepsilon}{T}\right) \varrho(\varepsilon), \quad (7)$$

where

$$\tilde{\kappa} \equiv \kappa(1 + \varphi/\varphi_s), \quad \eta_l^v \equiv \exp\left(-\tau_v \int_{\varepsilon'}^{\varepsilon_l^*} \frac{d\varepsilon}{\tau_u b(\varepsilon)}\right), \quad \varphi_s \equiv \frac{h\nu}{\sigma\tau_u}$$

is the saturation laser flux.

The quantum yield of EL generation  $\eta_{st}$  shall be defined as the ratio between the induced radiation rate and the pumping rate. It can be found from (7) with allowance for the nonsaturated absorption that

$$\eta_{st} = \left(\sum_l \beta_l \eta_l^v\right) \eta_s, \quad \eta_s = \frac{\varphi}{\varphi + \varphi_s} - \frac{\varphi}{\gamma\varphi_s}, \quad (8)$$

where  $\gamma \equiv g_0/\alpha_0$ ,  $g_0 \equiv \sum_l J_l^* \eta_l^v h\nu/\varphi_s$  is the small-signal gain,  $\alpha_0$  is the nonsaturated absorption coefficient. Therefore, the quantum yield of lasing can be represented as a product of two multipliers, the first allowing for effects of excitation, VT relaxation and quenching, and the second having the standard form, such as in [3], and related to photoabsorption. Assuming that  $b(\varepsilon) = 2D(1 - \sqrt{1 - \varepsilon/D}) \sqrt{1 - \varepsilon/D}$ ,  $\tau_u = \text{const}$  one obtains

$$\eta_l^v = \left(\frac{\varepsilon'}{2D(1 - \sqrt{1 - \varepsilon_l^*/D})}\right)^{\kappa}. \quad (9)$$

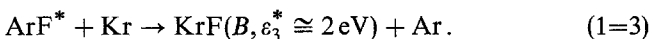
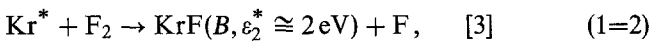
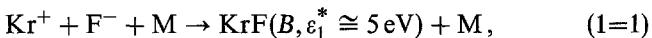
The internal laser efficiency  $\eta_{int}$  can be found from

$$\eta_{int} = \sum_l \beta_l \frac{h\nu}{\xi_l} \eta_l^v \eta_s, \quad (10)$$

where  $\xi_l$  is the energy expended for the formation of a excimer during the reaction 1. It is to be noted that (10) with (9) is radically different from the simplified formulae in [3].

#### 4. KrF-Laser Study

We apply the results obtained to the study of the excimer KrF laser. The upper laser state of  $\text{KrF}^*(B)$  is formed according to the below processes:



5 lower vibrational levels participate in the generation with  $\lambda = 248 \text{ nm}$ , hence  $\varepsilon' = 5\hbar\omega$  ( $\hbar\omega = 310 \text{ cm}^{-1}$ ) [9]. Figure 2 shows the values of internal efficiency of sponta-

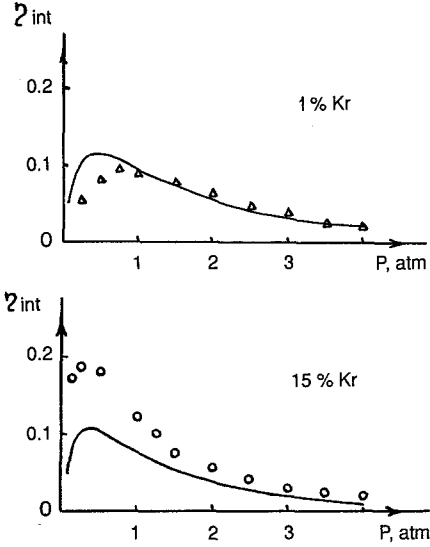


Fig. 2. Internal efficiency of the spontaneous luminescence of KrF in the mixture of 0,3% F, Kr, Ar;  $\Delta$ ,  $\circ$  denote experimental values [13]

neous luminescence given in [13],  $\eta_{int}$ . In addition, Fig. 2 presents the nonmonotonic dependence of  $(h\nu/\xi)\eta^v$  on the pressure of Ar which was calculated by means of (9). During the calculations we assumed that  $h\nu/\xi = 0.25$ ,  $\varepsilon^* = 1.7 \text{ eV}$  ( $\varepsilon^* \cong \varepsilon_2^* \cong \varepsilon_3^*$ ) and made use of formulae for  $\tau_u$  and  $\tau_v$  from [3]. Obviously, the relationship  $\eta_{int} \leq (h\nu/\xi)\eta^v$  is to be satisfied. But, as it is seen from Fig. 2, the theoretical and experimental values of  $\eta_{int}$  do not always agree. There should be nothing surprising about it, because the rate constants of the processes were obtained in [13, 3] as a result of experimental data based on simplified models. Thus, in [13] it was assumed that the excimer's vibrational relaxation occurred at an infinite rate. The model in [3, 8] suggests an infinitely fast VT relaxation of excimer levels, which do not take part in laser transitions. Consequently, the generally accepted values of KrF relaxation rate constants invite re-examination. However, they can be used for rough estimates. This can be illustrated by the example of KrF-EL internal efficiency calculations.

Table 1 lists the parameters of KrF lasers pumped by an electron beam. In compliance with the measurements [14, 3], lasing with  $\eta_{int} = 5.7\%$  and  $\eta_{int} = 11.0\%$  was obtained for similar conditions. Let us assess  $\eta_{int}$  according to (10). The quantum yield of lasing output was found by the assumption that the function  $\eta_s(\varphi)$  attains its maximum value of  $(\eta_s)_m = (1 - \gamma^{-1/2})^2$ . Calculating the values  $(\eta_s)_m$  in Table 1 we used the values of  $\gamma$  given in [14, 3],  $\tau_u$  and  $\tau_v$  were found from formulae of [3], and thus we have derived  $\kappa \cong 0.2$ . Then, using (9) we have found  $\eta_1^v(\varepsilon_1^* \cong D = 5.3 \text{ eV})$  and  $\eta_2^v(\varepsilon_2^* \cong 1.8 \text{ eV})$  [12]. The values of  $\eta_{int}$  were calculated according to (10) with  $(\eta_s)_m$  for  $\beta_1 = 1(\xi_1 = 24.1 \text{ eV})$  and  $\beta_2 = 1(\xi_2 = 20 \text{ eV})$ . Since, actually,  $\beta_1 + \beta_2 < 1$ , it is obvious that for EL [14]  $\eta_{int}(\%) < 5.9\beta_1 + 10.0\beta_2$  while for EL [3]  $\eta_{int}(\%) < 6.6\beta_1 + 10.5\beta_2$ .

Thus, the internal EL efficiency is strongly dependent on the excitation processes and the VT relaxation. Formulae (8) to (10) allow to calculate the quantum yield

**Table 1.** Parameters of KrF-ELs

Reference	Mixture [Torr]	$W^a$ [MW/cm <sup>3</sup> ]	$\eta_{\text{int}}^a$ [%]	$\gamma$	$(\eta_s)_m$	$\tau_v$ [ns]/ $\tau_u$ [ns] $\kappa$	$\eta^{v,b}$		$\eta_{\text{int}}^c$ [%]	
							$l=1$	$l=2$	$\beta_1=1$	$\beta_2=1$
[14]	F <sub>2</sub> (3) Kr (76) Ar (681)	1.0	5.7	25	0.64	1.0/4.9 0.20	0.45	0.63	5.9	10.0
[3]	F <sub>2</sub> (3) Kr (76) Ar (654)	0.77	11.0	23	0.63	1.0/5.8 0.17	0.51	0.67	6.6	10.5

<sup>a</sup> Experimental values,  $W$  is the pump power

<sup>b</sup> Calculation according to (9)

<sup>c</sup> Calculation using (10) with  $(\eta_s)_m$

and the internal lasing efficiency as functions of vibrational energy of excimer excitation,  $\varepsilon^*$ , the cavity flux  $\varphi$ , the kinetic parameters and the composition of an active mixture, which determine the values of  $\kappa$  and  $\gamma$ .

### 5. Vibrational-Rotational Levels Inversion Population

As was demonstrated above, the distribution of excimers over the vibrational levels is determined by population processes, deactivation and the VT relaxation. Even despite the fast vibrational relaxation in the region of upper vibrational levels, the function of distribution in the vibrational energy,  $f(\varepsilon)$  is highly non-equilibrium, see (7) and Fig. 3.

Let us determine the nature of rotational-level population of excimers. Noteworthy is that the VT-relaxation time in the light rare-gas atmosphere is  $\tau_v > 10^2 \tau$ , where  $\tau$  is the time of gas-kinetic collisions. At the same time

the rotational relaxation time is  $\tau_{RT} \cong \tau$  [4–7, 15]. If the following conditions are satisfied

$$\tau_{RT} \ll \tau_u, \quad \tau_{RT} \ll v^{-1}\tau_v \quad (11)$$

the distribution of excimers over rotational levels,  $j$ , should be equilibrium.

If the inequalities (11) imposing constrains on the inert-gas pressure and the number of the rotational level,  $v$ , are fulfilled, then the population inversion should exist between certain vibrational-rotational levels. With the distribution (7) the inversion density on the vibrational-rotational transition  $v, j \rightarrow v-1, j \pm 1$  equal [16]

$$A_{v-1,j \pm 1}^{v,j} = \left( \pm \frac{j}{j_m^2} - \frac{1-\kappa}{v} \right) \frac{(2j+1)B}{T} e^{-\frac{Bj(j+1)}{T}} \frac{f(\varepsilon_v)}{\varrho(\varepsilon_v)},$$

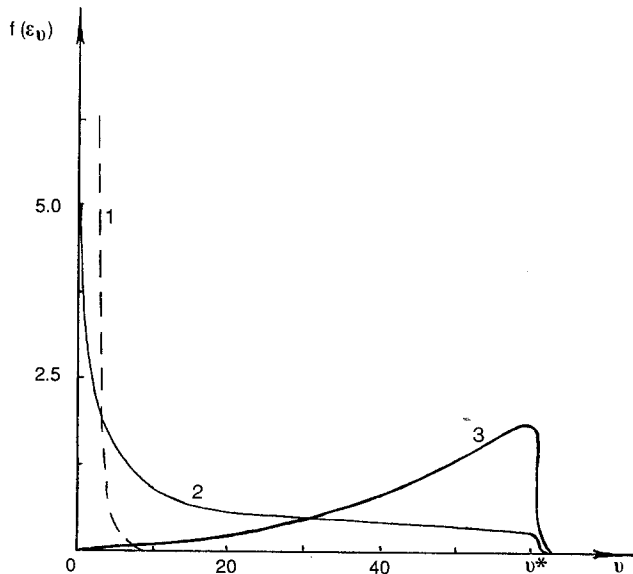
where  $B$  is the excimer's rotational constant,  $\varepsilon_v$  is the energy of the  $v$ -th vibrational level,  $j_m = (T/2B)^{1/2}$  is the number of the most populated rotational level. With  $\kappa > 1$ , complete inversion is implemented, and the gain can be attained for all transitions in the  $P$ -branches of vibrational-rotational spectrum as well as those transitions of  $R$ -branches, for which  $j < j_m^2(\kappa - v/1)$ . If  $\kappa < 1$ , there exists the partial inversion and the gain is possible for transitions of  $P$ -branches with  $j_m^2(1 - v/\kappa) < j < j_m^2(\hbar\omega/T)$ .

### 6. Far IR-Radiation Gain

Heteronuclear excimers are characterized by the ion coupling, and the radiative IR transitions shall take place between their vibrational levels. The Table 2 lists the optical parameters of some excimers that we have calculated. In the course of calculations we have employed the known similarity between the excited ion states of excimers and the ground states of alkali-halide molecules. The collisional width of an IR-radiation lines was assumed equal to 4 GHz/atm [15].

For the IR-radiation gain at the transitions  $v, j_m \rightarrow v-1, j_m \pm 1$  the following expression was obtained:

$$\alpha_v = \sigma_v \left( \pm \frac{1}{j_m} - \frac{1-\kappa}{v} \right) \frac{\kappa N}{\sqrt{e} j_m v} \left( \frac{\varepsilon_v}{\varepsilon^*} \right)^\kappa, \quad (12)$$



**Fig. 3.** Dependence of the function of distribution in vibrational energy,  $f$ , on the number of the vibrational level,  $v$ . Curve 1 is the equilibrium distribution function,  $f(v=0) = 100$ ; curve 2 corresponds to  $\kappa = 0.5$ ,  $3 - \kappa = 3(\hbar\omega = T)$

**Table 2.** Optical parameters of excimers<sup>a</sup>

Excimer	KrF	XeF	XeCl
$\lambda$ [ $\mu\text{m}$ ]	35–50	35–60	55–70
$A$ [ $\text{s}^{-1}$ ]	0.8	0.9	0.2
$\sigma_v p/v$	0.5	0.6	0.3

<sup>a</sup>  $\lambda$  [ $\mu\text{m}$ ] is the wavelength of radiation corresponding to transition from  $v$ -th vibrational level ( $v \cong 10$ –50),  $A$  [ $\text{s}^{-1}$ ] is the Einstein coefficient for the transition  $v = 1 \rightarrow v = 0$ ,  $\sigma_v$ , ( $10^{-16} \text{ cm}^2$ ) is the induced phototransition cross-section for  $v$ -th vibrational level,  $p$  [atm] is the gas pressure

where  $N$  is the excimers concentration in the EL active medium,  $e = 2.72$ .

Let us consider the mixture of 1.5%  $\text{F}_2$ , 10%  $\text{Kr}$ , 89.5%  $\text{He}$ , 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 of  $V = 2.8 \text{ cm}^3$  [17]. Reference 17 gives the measurement of spontaneous radiation energy of  $\text{KrF}$  which equals  $E = 32.5 \text{ mJ}$ . The luminescence pulse width was  $\tau_p = 100 \text{ ns}$ . We have estimated the excimers concentration,  $N = 3 \times 10^{15} \text{ cm}^{-3}$ , from the formula  $N = E\tau_r/(\tau_p h\nu V)$ . We employed in our calculations the value of  $\tau_r = 20 \text{ ns}$  found with allowance for mixing of B- and C-states of  $\text{KrF}$  [8] and the dependence of  $\tau_r$  on  $\varepsilon$  [9]. In the active medium under study  $\kappa \cong 1$ ,  $\varepsilon^* = \varepsilon_2^* \cong 1.8 \text{ eV}$ , and the criteria (11) are fulfilled. For excimers at the transition  $v = 25$ ,  $j_m = 22 \rightarrow v = 24$ ,  $j = 23$ , one obtains  $\sigma_v = 2.5 \times 10^{-14} \text{ cm}^2$ ,  $\lambda = 40 \mu\text{m}$ ; for the transition  $v = 10 \rightarrow v = 49$   $\sigma_v = 5 \times 10^{-14} \text{ cm}^2$ ,  $\lambda = 50 \mu\text{m}$ . Substituting the values of parameters into (12), one obtains  $\alpha_{25} = 2 \times 10^{-3} \text{ cm}^{-1}$ ,  $\alpha_{50} = 4 \times 10^{-3} \text{ cm}^{-1}$ .

## 7. Conclusions

In this work the excimers distribution over vibrational levels have been investigated covering processes of excimers formation, deactivation, and vibrational relax-

ation. The vibrational energy distribution function was shown to be a nonequilibrium one. As a result, the allowance for processes of population and VT relaxation is important for determination of quantum yield of both the spontaneous and laser emission. The theory constructed have been illustrated for the example of the  $\text{KrF}$ -laser study. Taking into account the vibrational relaxation we predict the new phenomena for far-IR radiation amplification in the excimer lasers active media. 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 (Table 2).

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