SOME SELF-FOCUSING MECHANISMS FOR ABSORPTION ON ROTATIONAL TRANSITIONS

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There is considerable interest in nonlinear effects in gases on radiation absorption. The refractive index is affected and this governs laser beam propagation [1]. The radiation frequency governs the type of internal motion excited. For example, IR radiation is absorbed by vibrational—rotational or purely rotational transitions. The main mechanisms whereby the refractive index is altered on vibrational excitation have been considered in [2-4]. No such analysis has been performed for absorption in rotational bands. Many of the frequencies radiated by a CO_2 laser are absorbed by purely rotational transitions in H_2O [5, 6], and it is assumed that the absorption at 9.4-10.6 μm by water vapor causes heating and thus reduces the refractive index and results in a divergent thermal lens in the beam channel [7, 8]. It has recently been shown [9] that a temporary temperature reduction can occur when there is absorption on a rotational transition. Here we consider the changes in refractive index for molecular gases showing absorption on transitions between rotational levels of the ground vibrational state, from which we deduce the conditions for the formation of a focusing lens in the beam channel.

We consider molecules in the ground electronic and vibrational states, while the frequency of the input radiation ν_I is in resonance with the center of the absorption line for the m → n rotational transition, where the state energies are E_m and E_n ($E_n > E_m$) correspondingly: $\nu_I = (E_n - E_m)/h + \Delta\nu$. Here $\Delta\nu = \nu_I - \nu_{nm}$, while ν_{nm} is the frequency corresponding to the line center for the absorbing transition and h is Planck's constant. The refractive index n is governed by the resonant part α_{nm} of the molecular polarizability and the nonresonant (static) part α_0 [10]:

$$n - 1 = 2\pi N(\alpha_0 + \mathrm{Re}\,\alpha_{nm}). \tag{1}$$

The imaginary part of α_{nm} is related to the absorption coefficient k_ν [11]:

$$\mathrm{Im}\,\alpha_{nm} = ck_\nu/(\pi^2\nu_I N), \tag{2}$$

in which c is the velocity of light in vacuum, $N = \rho N_A/\mu$, ρ is gas density, μ molecular weight, and N_A Avogadro's number; k_ν is [12] defined by

$$\begin{gathered} k_\nu = S_{nm}H(x,\ a),\ S_{nm} = \sigma_{nm}(g_n N_m/g_m - N_n), \\ H(x,\ a) = \frac{a}{\pi}\int\limits_{-\infty}^{\infty}\frac{\exp(-y^2)}{a^2 + (x-y)^2}\,dy,\quad \sigma_{nm} = \frac{\lambda_{nm}^2 A_{nm}}{4\pi b_D}\sqrt{\frac{\ln 2}{\pi}}. \end{gathered} \tag{3}$$

Here $a = b_C\sqrt{\ln 2}/b_D$; $x = 2\Delta\nu\sqrt{\ln 2}/b_D$; $\lambda_{nm} = c/\nu_{nm}$, while b_C and b_D are the collisional and Doppler line widths at half height, A_{nm} is the Einstein coefficient, g_q (a = n, m) is the degeneracy of state q, and N_q the molecular concentration in that state. We use (2) with (3) and the Kramers—Kronig formulas to get [13]

$$\mathrm{Re}\,\alpha_{nm} = \frac{n_0 c\sqrt{\ln 2}\mu}{4\pi^3\rho N_A b_D}S_{nm}H_{nm}(x,\ a,\ \xi_0), \tag{4}$$

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$$H_{nm}(x, a, \xi_0) = \int\limits_{-\infty}^{\infty} \frac{\left[(\xi_0 - y)(x - y) - a^2\right] \exp(-y^2)}{\left[(\xi_0 - y)^2 + a^2\right]\left[(x - y)^2 + a^2\right]}\, dy,$$

in which $\xi_0 = 2\sqrt{\ln 2}\,\nu_{nm}/b_D$, while n_0 is the refractive index for the unperturbed medium.

From (1) and (4) we represent the refractive index change $\delta n = n - n_0$ as

$$\delta n = \delta n_\rho + \delta n_R,$$
$$\delta n_\rho = \frac{\delta\rho}{\rho_0}(n_0 - 1), \quad \delta n_R = \frac{n_0 c H_{nm}}{2\pi^2 b_D}\,\delta S_{nm}. \tag{5}$$

This shows that δn may be calculated if one can determine not only the change in density but also the changes in the populations in the absorbing-transition levels. We perform the analysis for pulses whose length τ_p satisfies $\max(\tau, \tau_t) \ll \tau_p \ll \min(\tau_T, \tau_F, \tau_c)$ in which τ, τ_t, τ_T, τ_F, and τ_c are the characteristic times for polarization relaxation, translational relaxation, thermal conduction, diffusion, and convection. To determine the state of the medium under irradiation, we use the hydrodynamic equations for a nonviscous gas with zero thermal conductivity. We also assume that

$$\int\limits_0^{\tau_p} k_\nu I\, dt \ll \rho H_0\ ,$$

in which I is the radiation intensity and H_0 is the specific enthalpy. The hydrodynamic equations can then be linearized. We neglect the density change due to striction and the temperature change arising from the electrocaloric effect (with the parameters used here, those effects are small and do not influence the essence of the treatment), when the equations for $\delta\rho$ and δT become

$$\left(\frac{\partial^2}{\partial t^2} - c_0^2 \Delta_\perp\right) \frac{\partial}{\partial t}\,\delta\rho = (\alpha - 1)\,\Delta_\perp Q; \tag{6}$$

$$\rho_0 C_V \frac{\partial \delta T}{\partial t} - \frac{R}{\mu} T_0 \frac{\partial \delta\rho}{\partial t} = Q, \tag{7}$$

$$Q = k_\nu I - \frac{de_r}{dt}, \quad C_V = \left(\frac{3}{2} + \bar{C}_r\right)\frac{R}{\mu},$$

$$\alpha = 1 + \frac{R}{\mu C_V}, \quad \Delta_\perp = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial}{\partial r}\right).$$

Here c_0 is the speed of sound in the medium, R the universal gas constant, e_r the rotational energy of the molecules in unit volume, and RC_r/μ the rotational specific heat.

We determine $\delta\rho$ for two limiting cases. We introduce the variables $t' = t/\tau_p$ and $r' = r/a$ (a is the characteristic beam radius) and combine (6) and (7) to get

$$\frac{\partial}{\partial t'}\left(\alpha \frac{\partial^2 \rho'}{\partial t'^2} - \frac{\tau_p^2}{\tau_s^2}\Delta_\perp \rho'\right) = \frac{\tau_p^2}{\tau_s^2}\Delta_\perp \frac{\partial T'}{\partial t'}, \tag{8}$$

in which τ_s is the propagation time for sound waves along the beam radius ($\tau_s = a/c_0$); $\rho' = \delta\rho/\rho_0$; $T' = \delta T/T_0$.

Let $\tau_p \ll \tau_s$, and then with $\rho'(t = 0) = \partial\rho'/\partial t\ (t = 0) = 0$ we get from (8) that $\delta\rho \sim (\tau_p/\tau_s)^2$ and $\delta\rho \to 0$ for $\tau_p/\tau_s \to 0$, so $\delta n = \delta n_R$. Now let $\tau_p \gg \tau_s$, when (7) and (8) imply $\delta\rho/\rho_0 = -\delta T/T_0$, $\rho_0 \alpha C_V \partial\delta T/\partial t = Q$. Then one determines δn along with δn_R and the temperature change:

$$\delta n = -(\delta T/T_0)(n_0 - 1) + \delta n_R. \tag{9}$$

Now let the rotational relaxation time τ_R be greater than τ_t, which occurs for a gas in which the molecules have large rotational quanta (H_2O, HF, HCl, H_2S, etc.). Then one can consider the rotational kinetics on the basis of equilibrium for the translational degrees of freedom, while the equations describing the numbers of particles in the various rotational states m, n, k ($k \neq m, n$) can be put [12] as

$$\frac{dN_m}{dt} = -\tau_I^{-1}\left(\frac{g_n}{g_m}N_m - N_n\right) + \sum_j F_j^m + \sum_k F_{\text{sp}}^{mk}; \tag{10}$$

$$\frac{dN_n}{dt} = \tau_I^{-1}\left(\frac{g_n}{g_m}N_m - N_n\right) + \sum_j F_j^n + \sum_k F_{\text{sp}}^{nk}; \tag{11}$$

$$\frac{dN_k}{dt} = \sum_j F_j^k + \sum_q F_{\text{sp}}^{kq}. \tag{12}$$

Here $\tau_I^{-1} = W_{nm}I/h\nu_I$; $W_{nm} = \sigma_{nm}H(x, a)$, while $F_j{}^q$ is the term responsible for the collisional relaxation of molecules in state q during rotational–rotational R–R and rotational–translational R–T exchange, while $F_{sp}{}^{kq}$ is related to the spontaneous decay of state k. As $e_r = \sum\limits_j E_j N_j$ (E_j is the rotational energy of a molecule in state j), we use (10)-(12) to get the equation describing the temperature change in the beam channel as

$$\rho_0 C_T \frac{dT}{dt} = \frac{\Delta\nu}{\nu_I}k_\nu I - \Phi_R - \Phi_{\text{sp}},$$
$$\Phi_R = \sum_{i=n,m,k} E_i \sum_j F_j^i, \quad \Phi_{\text{sp}} = \sum_{i=n,m,k} E_i \sum_q F_{\text{sp}}^{iq}. \tag{13}$$

For $\tau_p \ll \tau_s$ we have $C_T = C_V$ and for $\tau_p \gg \tau_s$ we have $C_T = \alpha C_V$. Let $|k_\nu \Delta\nu| I \gg \nu_I |\Phi_R + \Phi_{sp}|$, which is so for $\tau_I \ll h\Delta\nu \min(\tau_R{}^j/E_j)$, in which $\tau_R{}^j = \{\tau_{RR}{}^j, \tau_{RT}{}^j, \tau_{sp}{}^j\}$, and τ_{RR}, τ_{RT}, and τ_{sp} are the characteristic times for R–R and R–T exchange and for spontaneous decay. Here the gas is cooled when the radiation is absorbed ($k_\nu > 0$) if $\Delta\nu < 0$. As the intensity increases in an inverted medium ($k_\nu < 0$), cooling will occur for $\Delta\nu > 0$. That mechanism for cooling on absorption has been analyzed [9] for vibrational–rotational and purely rotational transitions in molecular gases. One can solve (13) with (10)-(12) for each particular gas mixture only by numerical methods, but if the numbers of particles in states m and n are conserved, (10)-(12) and consequently (13) will allow an analytic solution in fairly general cases. That condition is met by a two-level model for rotational relaxation, for which the equations follow from (10) and (11) for $\tau_R{}^{nm} \ll \tau_R{}^{mk}$, $\tau_R{}^{nk}$. In that case, we have the interval (τ_t, $\tau_R{}^{nm}$) that

$$\frac{dN_m}{dt} = -\tau_I^{-1}\left(\frac{g_n}{g_m}N_m - N_n\right) + \tau_R^{-1}\left[N_n - \frac{g_n}{g_m}N_m \exp\left(-\frac{\Delta E_{nm}}{KT}\right)\right] + A_{nm}N_n; \tag{14}$$

$$\frac{dN_n}{dt} = \tau_I^{-1}\left(\frac{g_n}{g_m}N_m - N_n\right) - \tau_R^{-1}\left[N_n - \frac{g_n}{g_m}N_m \exp\left(-\frac{\Delta E_{nm}}{KT}\right)\right] - A_{nm}N_n. \tag{15}$$

Here τ_R is the collisional R–T relaxation time, K is Boltzmann's constant, and $\Delta E_{nm} = E_n - E_m$. The solutions to (14) and (15) are

$$N_m = \frac{B}{A} + \left(N_m^0 - \frac{B}{A}\right)\exp(-A\bar{t}); \tag{16}$$

$$N_n = N_n^0 + \left(N_m^0 - \frac{B}{A}\right)[1 - \exp(-A\bar{t})], \tag{17}$$

$$A = \left(1 + \frac{g_n}{g_m}\right)\frac{\tau_R}{\tau_I} + A_{nm}\tau_R + 1 + \frac{g_n}{g_m}\exp\left(-\frac{\Delta E_{nm}}{KT}\right),$$

$$B = (N_m^0 + N_n^0)(1 + A_{nm}\tau_R + \tau_R/\tau_I) \quad (N_q^0 = N_q(t = 0),\ \bar{t} = t/\tau_R).$$

We use (16) and (17) to get

$$\delta S_{nm} = \sigma_{nm}\left(1 + \frac{g_n}{g_m}\right)\left(\frac{B}{A} - N_m^0\right)[1 - \exp(-A\bar{t})].$$

We now determine the temperature change on the basis that the states m and n have nonequilibrium populations in the range (τ_t, τ_R^{nm}) in this model. The specific heat is

$$\begin{gathered} C_V = C_V^e - \frac{1}{\rho T^2}\sum_{j=m,n} E_j N_j^e\left[\frac{E_j}{K} - \bar{C}_r^e T\right], \\ C_V^e = \left(\frac{3}{2} + \bar{C}_r^e\right)\frac{R}{\mu}, \quad N_j^e = \frac{\rho N_A}{\mu}\gamma_k \frac{g_j}{Z_r}\exp\left(-\frac{E_j}{KT}\right), \end{gathered} \tag{18}$$

in which $(R/\mu)\bar{C}_r^e$ is the specific heat due to the rotational degrees of freedom in equilibrium with the translational ones ($\bar{C}_r^e = 1$ for linear molecules and $\bar{C}_r^e = 1.5$ for nonlinear ones), N_A Avogadro's number, γ_k the molar fraction of the absorbing component in the mixture, and Z_r the rotational sum.

We integrate (13) with (16)-(18) to get for $\delta T = T - T_0$ that

$$\begin{gathered} \rho_0 C_T \delta T = h\nu_I \frac{\tau_R}{\tau_I}\left[\left(1 + \frac{g_n}{g_m}\right)\frac{B}{A} - (N_m^0 + N_n^0)\right]\bar{t} - \\ - \left[\frac{h\nu_I}{A}\frac{\tau_R}{\tau_I}\left(1 + \frac{g_n}{g_m}\right) - \Delta E_{nm}\right]\left(\frac{B}{A} - N_m^0\right)[1 - \exp(-A\bar{t})]. \end{gathered} \tag{19}$$

For $A_{nm}\tau_R \ll 1$, which applies for not very low densities, (19) implies that $\delta T < 0$ only if

$$\begin{gathered} \frac{\tau_R}{\tau_I} h\Delta\nu\left(\frac{g_n}{g_m}N_m^0 - N_n^0\right) + h\nu_{nm}(N_n^0 - N_n^e) < 0, \\ N_n^e = \frac{g_n}{g_m}N_m^0 \exp(-\Delta E_{nm}/KT). \end{gathered} \tag{20}$$

If $N_n^0 = N_n^e$ (the gas is in equilibrium at $t = 0$) or $\tau_R/\tau_I \to \infty$, (20) becomes

$$\left(\frac{g_n}{g_m}N_m^0 - N_n^0\right)\Delta\nu < 0. \tag{21}$$

This coincides with the (13) cooling condition for $I \to \infty$. If on the other hand $N_n^0 > N_n^e$, (21) is accompanied by the additional condition

$$I > \frac{h\nu_0\nu_I}{|\Delta\nu k_\nu|\tau_R}(N_n^0 - N_n^e). \tag{22}$$

The maximum cooling ΔT_S subject to (21) and (22) occurs for $\bar{t} = \bar{t}_S$:

$$\bar{t}_S = \frac{1}{A}\ln\left\{\frac{\left[\left(1 + \frac{g_n}{g_m}\right)\nu_I - \frac{\tau_I}{\tau_R}A\nu_0\right]\left[N_m^0 - \frac{B}{A}\right]}{\nu_I\left[(N_m^0 + N_n^0) - \left(1 + \frac{g_n}{g_m}\right)\frac{B}{A}\right]}\right\}$$

and is defined by

$$\begin{gathered} \rho_0 C_T \Delta T_S = \frac{h\nu_I}{A}\frac{\tau_R}{\tau_I}\left\{\left[\left(1 + \frac{g_n}{g_m}\right)B - (N_m^0 + N_n^0)A\right]\bar{t}_S + \right. \\ \left. + \left(\frac{g_n}{g_m}N_m^0 - N_n^0\right)\right\} - h\nu_{nm}\left(N_m^0 - \frac{B}{A}\right). \end{gathered}$$

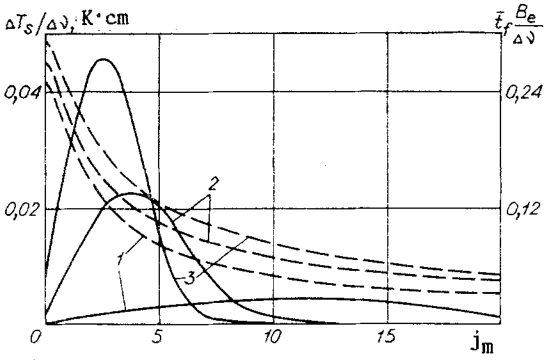

Fig. 1

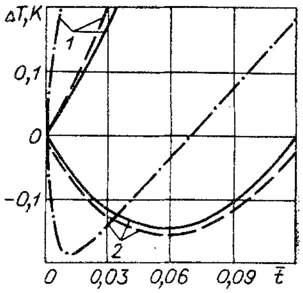

Fig. 2

The time for which the kinetic cooling exists $\bar t_f$ is found by solving the transcendental equation $\delta T = 0$.

In the strong-field approximation ($\tau_R/\tau_I \to \infty$), $\bar t_S = 0$,

$$\Delta T_S = \frac{h\Delta\nu\left(\frac{g_n}{g_m}X_m^0 - X_n^0\right)}{C_R K\left(1+\frac{g_n}{g_m}\right)}; \tag{23}$$

$$\bar t_f = -\frac{\Delta\nu}{\nu_I}F_{nm}^{-1}, \quad F_{nm} = \left[1+\frac{g_n}{g_m}\exp(-\Delta E_{nm}/KT)\right]. \tag{24}$$

Here $C_R = C_T\mu/R$; $X_q{}^0 = N_q{}^0/N_0$, and N_0 is the total number of molecules in unit volume. For $\tau_R \ll \tau_I$ (the weak-field approximation) and $N_n{}^0 = N_n{}^e$

$$\Delta T_S = \frac{\left(1+\frac{g_n}{g_m}\right)h\Delta\nu}{C_R K F_{nm}^2\sigma_{nm}}\left(\frac{\tau_R}{\tau_I}\right)^2; \tag{25}$$

$$\bar t_S = \frac{1}{A}\ln\left(1-\frac{\Delta\nu}{\nu_I}\right). \tag{26}$$

For $\Delta\nu \ll \nu_I$ and $A_{nm}\tau_R \ll 1$, (26) gives

$$\bar t_S = -\frac{\Delta\nu}{\nu_I}F_{nm}^{-1}, \quad \bar t_f = -2\frac{\Delta\nu}{\nu_{nm}}F_{nm}^{-1}.$$

These results may be illustrated on detailed examples. We first consider absorption on rotational transitions in diatomic molecules, for which $g_q = 2j_q + 1$, $Z_r = B_e/KT$, $E_q = B_e j_q \cdot (j_q + 1)$, in which j_q is the rotational quantum number, which defines the total angular momentum, and B_e is the rotational constant.

Figure 1 shows results from (23) and (24) for $\Delta T_S/\Delta\nu$ ($\Delta\nu < 0$) and the kinetic cooling existence time $\bar t_f B_e/|\Delta\nu|$ (solid and dashed lines) for CO, HCl, HF (lines 1-3) (B_e = 1.93, 10.59, 20.95 cm^{-1}) for T_0 = 300 K and various j_m ($j_m \to j_m + 1$ transition). The largest ΔT_S and $\bar t_f$ occur for molecules having large B_e (HF and HCl). For $\Delta\nu = B_e$, the largest ΔT_S for HF, HCl, and CO are 0.926, 0.239, 0.078 K, with the corresponding $\bar t_f$ = 0.27, 0.13, 0.025.

Figure 2 shows the temperature as a function of time on absorption on the 4 → 5 transition in HCl for $N_n{}^0 = N_n{}^e$, T_0 = 300 K and $\Delta\nu = +B_e, -B_e$ (curves 1 and 2), where the independent variable is τ_R/τ_I. Here the dot-dash, solid, and dashed lines correspond to τ_R/τ_I = 100, 1, 0.01 (for τ_R/τ_I = 1 and 0.01, the scale for ΔT has been reduced by factors of 10 and 100). Any reduction in τ_I, e.g., from intensity increase, for $\Delta\nu < 0$ will increase ΔT_S and reduce the existence time, while for $\Delta\nu > 0$, there will be more extensive heating.

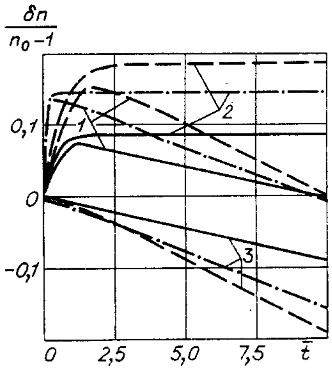

Fig. 3

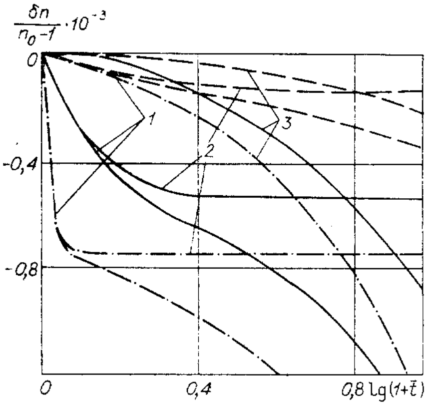

Fig. 4

The cooling in the beam increases n, as (9) shows, and heating reduces it. Cooling on absroption occurs for $\Delta\nu < 0$ and heating for $\Delta\nu \geq 0$. It is readily shown that $H_{nm} > 0$ for $\Delta\nu < 0$ and $H_{nm} < 0$ for $\Delta\nu > 0$, and as $\delta S_{nm} < 0$ in that case, saturation on the absorbing transition produced by radiation having $\nu_I < \nu_{nm}$ reduces n, while for $\nu_I > \nu_{nm}$, n is increased. $H_{nm} = 0$ for $\Delta\nu = 0$.

Then for $\tau_p \ll \tau_s$, the focusing lens is formed in the channel ($\delta n > 0$) only for radiation having $\nu_I > \nu_{nm}$. For $\tau_p \gg \tau_s$, there are two competing mechanisms that affect n. A region having $\delta n > 0$ can occur for $\Delta\nu \neq 0$ only if

$$\frac{\delta T}{T_0}(n_0 - 1) < \frac{n_0 c H_{nm}}{\pi^2 b_D}\,\delta S_{nm}.$$

Figure 3 shows how the refractive index of hydrogen chloride varies over time on absorption for $\nu_I = \nu_{nm} + B_e$ on the 4 → 5 transition in HCl at $T_0 = 300$ K for various τ_R/τ_I. Curves 1-3 correspond to δn, δn_R, and δn_ρ (for $\tau_p \gg \tau_s$), while the solid, dot-dash, and dashed lines are for $\tau_R/\tau_I = 1$, 100, 0.01 (for $\tau_R/\tau_I = 0.01$, the scale has been reduced by a factor of 100). The Einstein coefficient for the $j \to j + 1$ transition was calculated in accordance with [14] from $A_{j,j+1} = (E_{j+1} - E_j)^3\bar{\mu}^2/3\hbar^4c^3$, $E_j = B_e j(j + 1)$, $\bar{\mu} = 1.07$D, while we took $n_0 = 1 + 2\pi N\alpha_0$, in which $\alpha_0 = 2.637 \cdot 10^{-24}$ cm^3. The distributions show that the region with $\delta n > 0$ is formed in spite of the heating, whose existence time t_0 under these conditions is ~$9.5\tau_R$ for all τ_R/τ_I. For $p_0 = 1$ kPa, $\tau_R = 10^{-7}$ sec and $t_0 = 1$ µsec.

If $\Delta\nu$ is reduced to $0.5b_\ell$, in which b_ℓ is the width of the absorption line at half height, δn and t_0 are increased; $\delta n_R = 0$ for $\Delta\nu = 0$, and $\delta n < 0$ always. For $\Delta\nu = -B_e$, $\delta n_R \simeq 10^2\delta n_\rho$ for hydrogen chloride, and $\delta n < 0$ for virtually any $-B_e < \Delta\nu < 0$.

We now consider the refractive-index changes on absorption of a CO_2 laser beam by water vapor. For the H_2O molecule, $g_q = g_j^q g_a^q$, $g_j^q = 2j_q + 1$ and $g_a^q = 3$ for antisymmetric states and $g_a^q = 1$ for symmetric ones, with $Z_r = 2\sqrt{\frac{\pi T^3}{A_0 B_0 C_0}}$ (A_0, B_0, C_0 are the rotational constants). Table 1 gives ν_{nm}, E_j, A_{nm}, and g_a for several rotational transitions whose line flanks absorb CO_2 laser radiation (the A_{nm} have been derived from the [15] data on the line intensities). We also show the transitions in CO_2 that produce radiation with frequency $\nu_I = \nu_{nm} - \Delta\nu$. We calculated the CO_2 laser absorption for frequency ν_I from the tabulated data for various τ_R/τ_I and b_c/b_D (conditions close to atmospheric) and found that for all $\nu_I < \nu_{nm}$ one has $|\delta n_\rho| \ll |\delta n_R|$ even for $\bar{t} = \bar{t}_S$. For example, with $\nu_I = 947.742$ cm^{-1}, which is generated on the P16(00^{0}1 → 10^{0}0) transition in CO_2, with $\bar{t} = \bar{t}_S$ one has $\delta n_R/\delta n_\rho = 30$. That ratio is even larger for the other lines. Then a CO_2 laser beam giving $\nu_I < \nu_{nm}$ will always result in a region having $\delta n < 0$ in a medium containing H_2O molecules in spite of the kinetic cooling. After $\bar{t} > \bar{t}_f$, $|\delta n|$ increases even with the absorbing transition saturated on account of the heating ($\delta n_\rho < 0$), as Fig. 4 shows from the $\delta n(\bar{t})$, $\delta n_R(\bar{t})$, $\delta n_\rho(\bar{t})$ curves

TABLE 1

CO_2 laser line in 10.4 µm band		H_2O absorption lines						
branch of $00^01 \to 10^00$ transition	ν_l, cm^{-1}	Transition $j'k_a', k_c' \to j''k_a'', k_c''$	ν_0, cm^{-1}	$E_{j'}$, cm^{-1}	$E_{j''}$, cm^{-1}	A_{nm}, cm^{-1}	g_a	$\Delta\nu$, cm^{-1}
$P(40)$	924,974	$8_{2,7} \to 9_{7,2}$	924,988	885,60	1810,588	$1{,}47 \cdot 10^{-3}$	3	—0,014
$P(36)$	929,017	$13_{6,7} \to 14_{9,6}$	928,986	2756,418	3685,404	1,055	3	0,0314
$P(34)$	931,001	$9_{1,8} \to 9_{8,1}$	930,725	1079,080	2009,805	$5{,}96 \cdot 10^{-6}$	3	0,276
$P(28)$	936,804	$13_{6,8} \to 14_{9,5}$	937,308	2748,106	3685,414	0,35	3	—0,504
$P(16)$	947,742	$11_{0,11} \to 12_{5,8}$	948,263	1327,110	2275,373	0,0226	3	—0,521
$P(8)$	954,545	$15_{6,9} \to 16_{9,8}$	954,239	3472,880	4427,119	2,24	3	0,306
$R(12)$	970,547	$12_{7,5} \to 13_{10,4}$	970,268	2613,104	3583,372	0,445	1	0,279
		$12_{7,6} \to 13_{10,3}$	970,570	2612,801	3583,371	0,445	3	—0,0228
		$16_{5,12} \to 17_{8,9}$	973,253	3639,537	4612,790	3,22	3	0,036
$R(16)$	973,2885	$12_{1,11} \to 13_{6,8}$	973,487	1774,619	2748,106	0,0966	1	—0,198
$R(34)$	984,383	$10_{3,7} \to 11_{8,4}$	984,113	1538,15	2522,263	1,076	1	0,27

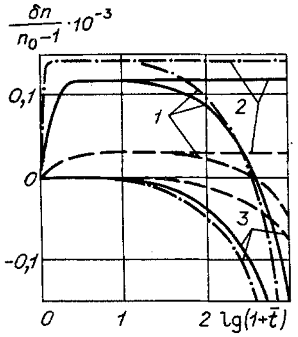

Fig. 5

(curves 1-3) for absorption of radiation having $\nu_I = 973.2885\ \text{cm}^{-1}$ (the P16($00^01 \rightarrow 10^00$) line from a CO_2 laser), which is absorbed by the $12_{1.11} \rightarrow 13_{6.8}$ ($\Delta\nu = -0.198\ \text{cm}^{-1}$) transition in H_2O for $b_c/b_D = 0.1, 1, 10$ (dashed, solid, and dot-dash lines). Those b_c/b_D correspond to $\tau_R/\tau_I = 0.1, 1, 10$. The change in the resonant part of the molecular polarizability on absorption has a marked effect on δn, which predominates over δn_ρ at the start.

A region having $\delta n > 0$ is formed only on absorption for $\nu_I > \nu_{nm}$. Figure 5 shows δn together with δn_R and δn_ρ (curves 1-3) as time functions on absorption of $\nu_I = 929.017\ \text{cm}^{-1}$ radiation generated by the P36($00^01 \rightarrow 10^00$) transition in CO_2 and absorbed by water vapor for $b_c/b_D = 0.1, 1, 10$ (dashed, solid, and dot-dash lines). Those b_c/b_D correspond to $\tau_R/\tau_I = 0.1, 1, 8.4$. There is a fairly long time interval ($t_0 \approx 500\tau_R$) during which a state with $\delta n > 0$ is realized. The following figures characterize the radiation intensities and t_0 corresponding to those parameters. For p = 1 kPa and T = 300 K, we have $\tau_R \approx 1.5\cdot 10^{-7}$ sec and $\tau_R/\tau_I = 1$ will correspond to $I \approx 10^5\ \text{W/cm}^2$, with $t_0 \approx 7.5\cdot 10^{-5}$ sec. Under those conditions, the absorption length is $L_\nu = 10^5$ m and the focal length of the positive lens formed in the channel with an inhomogeneous radial intensity distribution (e.g., gaussian) is $3\cdot 10^2$ m with the maximal δn.

Some general comments are that IR absorption by rotational transitions can cause δn as a function of time to be determined by the density change associated with heating or temporary cooling and with the change in the resonant part of the polarizability due to the excitation by the resonant radiation. δn is substantially dependent on the magnitude and sign of the frequency difference, the line strength, the exciting intensity, and the gas parameters. If τ_p is substantially less than the time for sound propagation across the beam, δn is governed by the change in the resonant part of the polarizability if $\left(\frac{\tau_p}{\tau_s}\right)^2 \frac{\delta T}{T_0}(n_0 - 1) \ll \frac{n_0 c H_{nm}}{\pi^2 b_D}\, \delta S_{nm}$, while if $\tau_p \gg \tau_s$, but is naturally less than the characteristic times for thermal conduction and diffusion, δn will be dependent on the change in density (temperature) and the change in the resonant part of the polarizability. The latter can be neglected in deriving δn only if $\pi^2 b_D \delta T(n_0 - 1) \gg n_0 c T_0 H_{nm} \delta S_{nm}$. A region having $\delta n > 0$ is produced for $\tau_p \ll \tau_s$ only if there is positive frequency deviation, $\nu_I > \nu_{nm}$, and it occurs for $\tau_p \gg \tau_s$ and $\nu_I \neq \nu_{nm}$ only in a time interval such that $\pi^2 b_D \delta T(n_0 - 1) < n_0 c T_0 H_{nm} \delta S_{nm}$.

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RADIATION CONTAMINATION OF DIELECTRICS IRRADIATED WITH A FAST-ELECTRON FLUX

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It is known that, in irradiating dielectrics and semiconductors with a fast-electron flux, they accumulate a space charge (SC), the electric field of which affects considerably the transport of primary particles, while it can also cause electric breakdown and disintegration of the dielectric [1-3]. Although this phenomenon has been investigated intensively during the past 20 years, a satisfactory mathematical description of it is still lacking, which sometimes hampers the interpretation of experimental data. The principal approximation inherent in the known solutions of the problem of SC dynamics [2, 4] consists in neglecting the distortion of the absorbed energy and thermalized electrons caused by the effect of the SC field on transport.

We have devised and realized a nonstationary mathematical model which describes in a self-consistent manner the SC build-up and the transport of fast electrons for different boundary conditions.

System of Equations. The mathematical model describing the space charge build-up in the dielectric is based on the phenomenological model [4]. In order to take into account the distortion in the distribution of absorbed energy and thermalized electrons due to the effect of the electric field on the past particle transport, the equations of space charge kinetics were supplemented with the kinetic equation accounting for the electric field for fast electrons. In the one-dimensional case, the system of equations has the following form:

$$\frac{\partial N}{\partial t} = Q_e + S - \alpha P N - \frac{\partial J_N(N, E)}{\partial z}; \qquad (1)$$

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