DISCUSSION OF THE EFFECT OF THE INTENSITY OF RADIATION AND THE PARAMETERS OF A MEDIUM ON THE CHANGE IN THE REFRACTIVE INDEX ACCOMPANYING THE ABSORPTION OF HF-LASER RADIATION BY WATER VAPOR

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The absorption of HF-laser radiation by water vapor leads to thermodynamic disequilibrium between the translational and vibrational degrees of freedom, which depending on the frequency of the radiation can be accompanied by both temporary cooling and heating of the gas [1]. In the process, owing to the excitation of different types of vibrations of the H_2O molecule, the polarizability of the medium changes. Both these effects bring about a change in the refractive index n in the beam channel. The character of the change in n depends on the radial distribution of the intensity in the beam, on the magnitude of the intensity, and also on the parameters of the medium. We shall analyze the dynamics of the change in the refractive index for square pulses ($I = I_0$ for $0 < t \leq \tau_u$, I is the intensity of the radiation), whose duration τ_u satisfies the condition

$$\max(\tau_{RT}, \tau_{VV}) < \tau_u \ll \min(\tau_T, \tau_C, \tau_D),$$

where τ_{RT} and τ_{VV} are the characteristic rotational–translational R–T relaxation time and the intramode vibrational–vibrational V–V exchange time in the H_2O molecule; and, τ_T, τ_C and τ_D are the heat-conduction, convection, and diffusion times.

Let the time of the induced processes τ_I be much greater than τ_{RT} and τ_{VV}. We shall study a mixture of the gases H_2O, N_2, and O_2. To determine the state of the medium in the beam channel, in this case, we shall employ the equations of hydrodynamics for a nonviscous, thermally nonconducting gas, and we shall employ the model of a harmonic oscillator and local vibrational temperatures to describe the kinetics of the vibrational energy exchange:

$$\partial\rho/\partial t + \operatorname{div}(\rho\mathbf{v}) = 0; \tag{1}$$

$$\partial\mathbf{v}/\partial t + (\mathbf{v}\nabla)\mathbf{v} + (1/\rho)\nabla p = 0; \tag{2}$$

$$\rho C_V \partial T/\partial t + \rho C_V \mathbf{v}\nabla T + p\nabla\mathbf{v} = Q_V; \tag{3}$$

$$\partial\varepsilon_j/\partial t + \mathbf{v}\nabla\varepsilon_j + \varepsilon_j\nabla\mathbf{v} = q_j,$$

$$Q_V = k_\nu I - \rho\partial l_V/\partial t - \rho\mathbf{v}\nabla l_V - \rho l_V\nabla\mathbf{v},$$

$$l_V = \gamma_1 \frac{R}{\mu}\sum_{j=1}^{3}\frac{h\nu_j}{K}\varepsilon_j + \sum_{j=3}^{4}\frac{R}{\mu}\frac{h\nu_j}{K}\varepsilon_j\gamma_j,\ p = \frac{\rho RT}{\mu}, \tag{4}$$

$$C_V = \frac{R}{\mu}\left(\frac{3}{2} + \sum_{i=1}^{3}\gamma_i + \frac{1}{2}\gamma_1\right),\ \varepsilon_j = r_j\left[\exp\left(\frac{h\nu_j}{KT_j}\right) - 1\right]^{-1}.$$

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Here ρ, T, p, and **v** are the density, temperature, pressure and velocity of the gas; k_ν is the absorption coefficient ($k_\nu = f(r, t)$); γ_i is the molar fraction of the i-th component in the mixture (i = 1, 2, and 3 for the gases H_2O, O_2, and N_2); ν_j is the normal frequency; T_j is the local vibrational temperature of the j-th mode; r_j is the degree of degeneracy of the j-th mode (j = 1, 2, and 3 correspond to symmetric, bending, and asymmetric vibrations of H_2O, while j = 4 and 5 correspond to vibrations of O_2 and N_2 molecules): K is Boltzmann's constant; h is Planck's constant; μ is the molecular mass of the mixture; R is the universal gas constant; and the term q_j characterizes the change in the stored vibrational quanta in the mode j owing to radiation and collisional energy-exchange processes (the specific form of q_j for j = 1, ..., 5 is presented in [1]).

We shall consider the case when

$$E_n \ll H,\ E_n = \int_0^{\tau_u} \frac{k_\nu I}{\rho} I dt,\ H = C_V T + l_V + \frac{p}{\rho}. \tag{5}$$

The system (1)-(4) can be simplified. Since the perturbations of the parameters of the gas owing to the interaction with laser radiation are small when (5) holds, the values of T, p, and ρ can be represented in the form [2].

$$\xi = \xi_0[1 + \xi' \exp(ikr - i\omega t)],\ \xi = p, \rho, T, \tag{6}$$

while the velocity of the gas is determined by the perturbation relative to the velocity of sound c_0:

$$v = c_0 v' \exp(ikr - i\omega t),\ c_0 = \sqrt{\varkappa \frac{R}{\mu} T_0},\ \varkappa = 1 + \frac{R}{\mu C_V}. \tag{7}$$

Here v' and ξ' are dimensionless amplitudes of the perturbations v and ξ; k is the complex wave vector; and, $\omega = c_0 \operatorname{Re} k$. Substituting (6) and (7) into (1)-(4) and neglecting terms which are first-order infinitesimals we obtain the equations for the dimensionless perturbations p', ρ', and v':

$$\partial\rho'/\partial t + c_0 \operatorname{div} v' - i\omega\rho' + 2ikc_0 v' = 0; \tag{8}$$

$$c_0 \frac{\partial v'}{\partial t} + \frac{p_0}{\rho_0} \nabla p' - i\omega c_0 v' + 2ik \frac{p_0}{\rho_0} p' = 0; \tag{9}$$

$$C_V \frac{\partial T'}{\partial t} + \frac{p_0}{\rho_0} c_0 \nabla v' - iC_V \omega T' + 2ik \frac{p_0}{\rho_0} c_0 v' = Q_V. \tag{10}$$

Setting, as usual, $k = k_0 - i\delta$, where δ is the coefficient of absorption (amplification) of sound, and separating the real parts in (8)-(10), we find

$$\frac{\partial\rho'}{\partial t} + c_0 \operatorname{div} v' + 2\delta c_0 v' = 0; \tag{11}$$

$$c_0 \frac{\partial v'}{\partial t} + \frac{p_0}{\rho_0} \nabla p' + 2v \frac{p_0}{\rho_0} p' = 0; \tag{12}$$

$$C_V \frac{\partial T'}{\partial t} + \frac{p_0}{\rho_0} c_0 \nabla v' + 2\delta \frac{p_0}{\rho_0} c_0 v' = Q_V. \tag{13}$$

Analysis of (11)-(13) shows that the acoustic approximation with

$$2|\delta|a \ll 1 \tag{14}$$

(a is the characteristic radius of the beam) can be used to describe the change in the gas-dynamic parameters in the beam channel. In this work we study beams with $a \lesssim 10$ cm. For

them (14) holds with $\delta \ll 5\cdot10^{-2}$ cm^{-1}. In [2] it was found that for a vibrationally nonequilibrium gas in the presence of an external source of excitation $\delta \leq 10^{-4}$ cm^{-1} for $T_j \leq k\nu_j/K$. When the HF-laser radiation is absorbed by water vapor $T_j \leq 0.1h\nu_j/K$ [1] and therefore the acoustic approximation holds, the equations (11)-(13) assume the form

$$\left(\frac{\partial^2}{\partial t^2} - c_0^2\Delta_\perp\right)\frac{\partial}{\partial t}\rho' = (\varkappa - 1)\Delta_\perp\left[k_\nu I - \rho_0\frac{\partial l_V}{\partial t}\right]; \tag{15}$$

$$\rho_0 C_V\frac{\partial T'}{\partial t} - \frac{R}{\mu}T_0\frac{\partial\rho'}{\partial t} = \left[k_\nu I - \rho_0\frac{\partial l_V}{\partial t}\right],\ \Delta_\perp = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial}{\partial r}\right). \tag{16}$$

The change in n in the beam channel, according to [3], can be found from the equation

$$\frac{\partial n}{\partial t} = \frac{n_0 - 1}{\rho_0}\frac{\partial\rho}{\partial t} + \frac{2\pi\rho_0 R}{\mu K}\sum_{j=1}^{M}\frac{g_j}{h\nu_j a_j^2}(K_j + 3L_j + 2\varepsilon_j L_j)\frac{\partial\varepsilon_j}{\partial t}\gamma_j, \tag{17}$$

where $K_j = \left(\frac{\partial\mu}{\partial Q_j}\right)_0^2 + \frac{h\nu_j}{2}\left(\frac{\partial^2\alpha}{\partial Q_j^2}\right)_0$; $L_j = \frac{1}{8a_j^2}\left(\frac{\partial^2\mu}{\partial Q_j^2}\right)_0^2$; $a_j = 2\pi\sqrt{\frac{m_j\nu_j}{h}}$; g_j is the degree of degeneracy; and m_j is the reduced mass of the j-th oscillator. The solution of the system (15)-(17) together with the equations of the vibrational kinetics (4), in which under the assumptions made the terms $(\mathbf{v}\cdot\mathrm{grad}\,\varepsilon_j)$ can be neglected, and the equation of state for an ideal gas permits determining, for a fixed value of I, the dynamics of the change in n taking into account the excitation of vibrations of molecules of the mixture as well as the change in the density and temperature in the beam channel.

We shall first study some limiting cases. Combining (15) and (16) and introducing the new variables $t' = t/\tau_u$ and $r' = r/a$ (the prime is dropped in what follows) it is not difficult to derive an equation that relates the change in density in the beam channel and the change in the temperature:

$$\frac{\partial}{\partial t}\left(\varkappa\frac{\partial^2\rho}{\partial t^2} - \frac{\tau_u^2}{\tau_s^2}\Delta_\perp\rho\right) = \frac{\tau_u^2}{\tau_s^2}\Delta_\perp\frac{\partial T}{\partial t}. \tag{18}$$

Here τ_s is the propagation time of sound vibrations along the radius of the beam ($\tau_s = a/c_0$) Let $\tau_u \ll \tau_s$. Then after taking the limit with $\rho(t=0) = \partial\rho(t=0)/\partial t = \partial^2\rho(t=0)/\partial t^2 = 0$ we obtain from (17) $\partial\rho/\partial t = 0$, $\rho = \rho_0$. The change in n depends, in this case, only on the change in the polarizability of the medium (the second term on the right side of Eq. (17)) owing to excitation of molecular vibrations $(\varepsilon_j > \varepsilon_j^0,\ \varepsilon_j^0 = \varepsilon_j(t=0))$.

When the intensity distribution is symmetric relative to the center of the beam, for example, for a Gaussian distribution $I = I_0\exp(-r^2/a^2)$ (I_0 is the intensity on the beam axis), a converging lens forms in the beam channel. In the other limiting case, for $\tau_u \gg \tau_s$ we obtain from (16) and (18)

$$\frac{d\rho}{\rho_0} = -\frac{dT}{T_0},\quad \rho C_V^{RT}\varkappa\frac{dT}{dt} = k_\nu I - \frac{\rho R}{\mu K}\sum_i\sum_j h\nu_j\frac{\partial\varepsilon_j}{\partial t}\gamma_i.$$

As follows from (17) the change $\Delta n = n - n_0$ depends here on the temperature and polarizability of the medium in the beam channel.

It is shown in [1] that when spectral lines with close frequencies do not overlap (radiation with frequency ν_I is absorbed in an isolated vibrational–rotational transition line or in the band $0 \to \nu_1$, or $0 \to 2\nu_2$, or $0 \to \nu_3$ of the H_2O molecule) the gas in the beam channel can be cooled only if

$$E_{j''} - E_{j'} + h\Delta\nu < 0, \tag{19}$$

where E_j'' and E_j' are the rotational energies of the upper and lower states of the absorbing transition, and $\Delta\nu$ is the difference of the frequency of the acting radiation and the frequency at the center of the line of this transition. The effect of cooling of the medium

persists up to the time $t \approx \tau_r$ (τ_r is the relaxation time of the energy absorbed by H_2O molecules). At times $t \geq \tau_r$ the gas starts to heat up. The medium is also heated owing to the intramode V–V exchange. As calculations showed, however, the contribution of this process to ΔT for the conditions studied is not significant. If $\tau_I < \tau_r$ (τ_I is the time of the induced transitions), then for $t < \tau_r$ a focusing lens forms in the beam channel owing to the change in the polarizability of the medium and the cooling of the medium. For $t < \tau_r$ the increase in T and the corresponding decrease in ρ can lead to a change in the sign of Δn and the formation of a diverging lens.

For radiation that is absorbed in the transitions with $E_j'' - E_j' + h\Delta\nu > 0$, even for $t < \tau_r$, the gas in the beam channel will be heated up; this decreases Δn owing to the change in the polarizability of the medium accompanying excitation of the molecular vibrations of H_2O.

If, however, $\tau_u \leqslant \tau_s$, then the change Δn is determined by more complicated processes. Here Δn is strongly affected by, aside from the change in the polarizability of the medium, the propagation of acoustic disturbances across the beam, owing to the nonuniform temperature profile. Let the frequency of the radiation be such that (19) holds. For $t < \tau_r$ and for a Gaussian intensity distribution along the radius, stronger cooling of the medium in the region near the axis (it is assumed that the absorbing transition is not saturated) will lead to lower pressure than at the periphery of the beam and to propagation of a density from the periphery to the center. If, however, the frequency of the radiation is such that $E_{j''} - E_{j'} + h\Delta\nu > 0$, then even for $t > \tau_r$ the heating of the medium increases the pressure in the region near the axis. The density wave propagates, in this case, from the axis to the periphery of the beam. The value of n for $\tau_u \leq \tau_s$ will change depending on the ratio of τ_r and τ_s.

In the general case a quantitative analysis of the effect of the radiation intensity and the parameters of the medium on the change in n in the beam channel must be conducted based on the solution of the complete system of equations (14), (15), and (16). For a Gaussian distribution of intensity over the radius of the beam the following boundary and initial conditions must hold:

$$T(r = R,\ t) = T_0,\ \varepsilon_j(r = R,\ t) = \varepsilon_{j0},\ \chi(r = R,\ t) = 0,$$
$$\frac{\partial\chi}{\partial r}(r = 0,\ t) = 0,\quad T(r,\ t = 0) = T_0,\quad \chi(r,\ t = 0) = 0,$$
$$\frac{\partial\chi}{\partial t}(r,\ t = 0) = 0,\quad \varepsilon_j(t = 0) = \varepsilon_{j0},\quad \rho(r,\ t = 0) = \rho_0.$$

Here $\chi = \partial\rho/\partial t$; $R = \infty$; and $\varepsilon_{j_0} = \varepsilon_j(T)$. In this work the equations (4), (15), and (16) were integrated numerically using the absolutely stable, symmetric, implicit, second-order difference schemes [4]. The rate constants of the V–V' and V–T exchange processes and the molecular constants, required for the calculations, were taken from [1], and the coefficients

$$(\partial\mu/\partial Q_j)_0,\quad (\partial^2\alpha/\partial Q_j^2)_0,\quad (\partial^2\mu/\partial Q_j^2)_0,$$

determining the polarizability and hyperpolarizability of the molecules, were taken from [5, 6]. The reduced masses of the oscillators are: $m_1 = 2.1$, $m_2 = 7.0$, $m_3 = 2.3$, $m_4 = 8.0$, $m_5 = 7.0$.

We shall make a concrete analysis for radiation with frequency $\nu_I = 3427.98$ cm^{-1}, absorbed in the transition $000(6_{34}) \rightarrow 001(5_{15})$ of the H_2O molecule (for $E_j'' - E_j' + h\Delta\nu < 0$), and for radiation with $\nu_I = 3759.1$ cm^{-1}, absorbed in the transition $000(6_{34}) \rightarrow 001(6_{33})$ (for $E_j'' - E_j' + h\Delta\nu > 0$) and with a Gaussian radial distribution of the intensity. Figure 1 shows the results of the calculation of the change Δn over the radius of the beam with $a = 0.2$ cm for $\tau_u < \tau_s$ at $t = 0.2\tau_r$ (a) and $t = 1.5\ \tau_r$ with radiation with frequency $\nu_I = 3427.98$ cm^{-1} acting on the medium H_2O–N_2–O_2 ($\gamma_{H_2O} = 0.01$, $\gamma_{O_2} = 0.2$) with $T_0 = 300$ K and $p_0 = 0.01$ MPa for $I_0 = 20$, 100, and 500 MW/cm^2 (the lines 1-3). The broken line shows the change Δn owing solely to the change in the density of the medium Δn_ρ. Under the indicated conditions $\tau_r = 1$ μsec and $\tau_s = 5.7$ μsec. One can see that increasing the intensity of the radiation, first of all, increases Δn and, second, leads to a significant change $\Delta n = f(r)$.

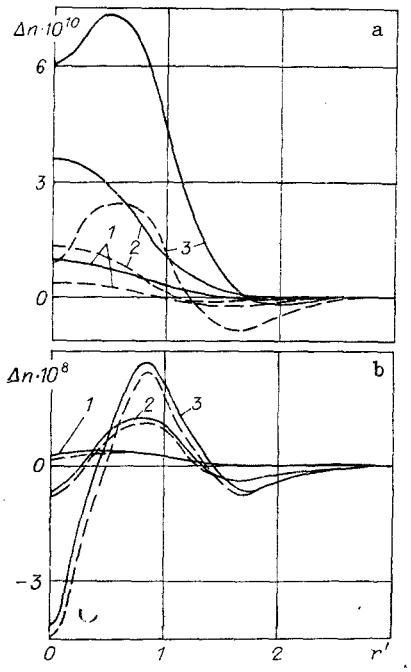

Fig. 1

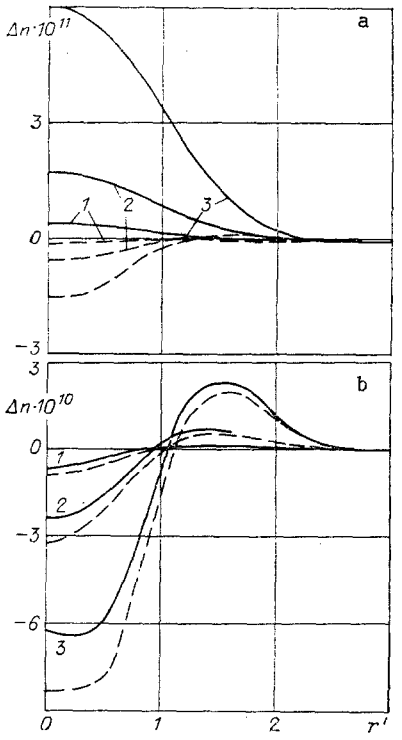

Fig. 2

For $t < \tau_r$ (Fig. 1a) as I is increased Δn increases primarily owing to the change in the polarizability of the medium accompanying excitation of molecular vibrations of H_2O, while for $t > \tau_r$ (Fig. 1b) Δn increases owing to the increase in the relative fraction of the energy released in the translational degrees of freedom, and the amplification of the density wave propagating along the axis to the periphery of the beam. For $I = 500$ MW/cm^2 and $t < \tau_r$ the absorbing transition saturates on the axis of the beam. This accelerates thermalization of the energy absorbed by the H_2O molecules and, as a consequence, leads to a decrease in Δn_ρ on the beam axis. In the process the maximum of the dependence $\Delta n(r)$ shifts from the axis to the periphery of the beam. The change Δn along the beam radius ($a = 1$ cm) for radiation with $\nu_I = 3759.1$ cm^{-1}, absorbed on the transition with $Ej'' - Ej' + h\Delta\nu > 0$ by atmospheric water vapor ($\gamma_{H_2O} = 0.01$), with $p_0 = 1$ kPa and $T_0 = 300$ K for $\tau_u < \tau_s$ at $t = 0.1\tau_r$ and $t = 0.3\tau_r$ ($\tau_r = 10$ μsec and $\tau_s = 28.5$ μsec) is illustrated in Fig. 2a and b, where the broken lines correspond to Δn_ρ while the lines 1-3 correspond to $I = 4$, 20, and 100 MW/cm^2 (the values of I were chosen so that τ_I would be the same as for radiation with $\nu_I = 3427.98$ cm^{-1} in the preceding case). Increasing I also increases Δn. However by virtue of the fact that $\Delta n_\rho < 0$, Δn for times $t < \tau_r$ is significantly smaller. At $t = 0.1\tau_r$ Δn is determined primarily by the change in the polarizability of the medium owing to the excitation of vibrations of H_2O molecules, while at $t = 0.3\tau_r$ the change in the density Δn_ρ makes the main contribution to Δn. It should be noted that the lifetime of the focusing lens ($\Delta n > 0$) is much shorter than for radiation absorbed on the transitions with $Ej'' - Ej' + h\Delta\nu < 0$. For $\tau_s > \tau_r$ the change in Δn_ρ owing to the change in the density of the medium makes a determining contribution to Δn for transitions with $Ej'' - Ej' + h\Delta\nu < 0$ only for $t \geq \tau_r$, when the gas starts to heat up, while for $\tau_s < \tau_r$ this also happens in the regime of kinetic cooling, when $t \leq \tau_r$. The dependence of Δn for this case with different values of the intensity of the acting radiation with $\nu_I = 3427.98$ cm^{-1} ($a = 0.2$ cm) for an atmosphere with $\gamma_{H_2O} = 0.01$, $T_0 = 300$ K, $p_0 = 1$ kPa is shown in Fig. 3, where the lines 1-3 correspond to $I = 50$, 100, and 200 MW/cm^2, while the broken and solid lines correspond to the times $t = 0.2\ \tau_r$ and τ_r ($\tau_r = 10.1$ μsec and $\tau_s = 5.7$ μsec). One can see that owing to the propagation of the density wave from the periphery to the center of the beam (kinetic cooling is observed at times $t < \tau_r$) in the region near the axis a converging lens, whose optical power increases as I is increased, forms near the axis even for $t = \tau_r$. Saturation of the absorbing transition with $I = 200$ MW/cm^2 and $t = \tau_r$ in the region of the axis, as in the case when $\tau_s > \tau_r$, leads to a displacement of the maximum of the dependence $\Delta n(r)$ from the axis to the periphery of the beam. We note that Δn and hence the optical power of the converging lens formed in the case under study ($\tau_s < \tau_r$) is much greater than for $\tau_s > \tau_r$, where Δn for $t < \tau_r$ is determined

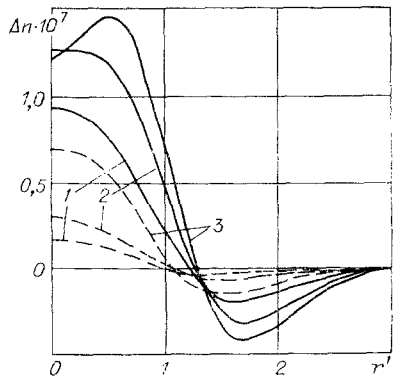

Fig. 3

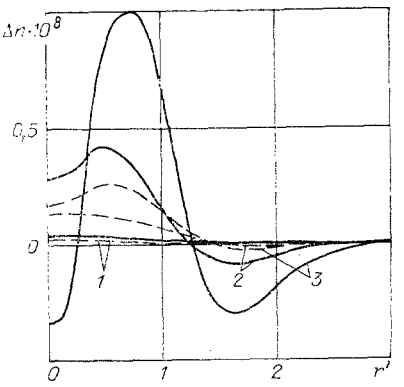

Fig. 4

primarily by the change in the polarizability of the medium owing to the excitation of the molecular vibrations of H_2O. However the region with $\Delta n > 0$ is significantly smaller, and for $r \geq 1.25$ even the case $\Delta n < 0$ is realized. For long times heating of the medium starts, and the region with $\Delta n < 0$ also occupies the zone near the axis. For radiation absorbed in transitions with $E_j'' - E_j' + h\Delta\nu > 0$ the character of the dependence $\Delta n(r)$ for $\tau_s < \tau_r$ remains the same as for $\tau_s > \tau_r$ (see Fig. 2b), though the absolute values of Δn are much larger. Since the ratio of the times τ_s and τ_r is determined both by the parameters of the medium and the characteristic size of the beam (the radius a) it is obvious that even for the same values of T_0, p_0, γ_{H_2O}, and I the character of the change in the refractive index along r for beams with different radii a can be substantially different. This is illustrated in Fig. 4, which shows the curves $\Delta n(r)$ for beams with $a = 0.2$ and 0.5 cm (broken and solid lines) at $t = 0.2 \cdot \tau_r$, $0.8\,\tau_r$, and $1.4\tau_r$ (curves 1-3) for radiation with $\nu_I = 3427.98\ cm^{-1}$, $I = 100\ MW/cm^2$, acting on a medium with $p_0 = 10^{-2}$ MPa, $T_0 = 300$ K, and $\gamma_{H_2O} = 0.01$. Here $\tau_r = 1$ μsec, $\tau_s = 5.7$ ($a = 0.2$ cm) and 14.3 μsec ($a = 0.5$ cm). For a beam with $a = 0.5$ cm the state of the medium with $\Delta n > 0$ is preseved in the region near the axis right up to $t = 1.4\tau_r$, while for $a = 0.2$ cm by this time near the axis $\Delta n < 0$.

Thus the calculations showed that when HF-laser radiation is absorbed by atmospheric water vapor a focusing, nonstationary lens can form in the beam channel. The lifetime of the lens depends on the ratio of the thermalization time of the radiation energy absorbed by the H_2O molecules and the propagation time of acoustic waves across the beam.

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