RESONANCE SELF–FOCUSING OF RADIATION DUE TO LASER–INDUCED NONEQUILIBRIUM VELOCITY DISTRIBUTION OF GAS MOLECULES

A.E. Bazelyan, S.V. Ivanov, M.N. Kogan, and V.Ya. Panchenko

Scientific–Research Center for Technological Lasers, Russian Academy of Sciences, Troitsk
N.E. Zhukovskii Central Aerogidrodynamical Institute, Zhukovskii
Received November 11, 1991

The curves of anomalous dispersion of a minor impurity of a molecular gas in a buffer gas are calculated taking into account laser–induced distorsions of Maxwell's molecular velocity distribution at the levels resonant with laser radiation. It is shown that account of this nonequilibrium may lead to an increase in the absolute values of variations in the refractive index by several times. In this case the result depends strongly on the ratio of the velocities of elastic (T–T) and rotational (R–T) relaxations of molecules. In the limiting case of slow elastic relaxation a simple formula for the refractive index is obtained and its range of applicability is analyzed. A parameter of resonance self–focusing of the CO_2*–laser radiation in the atmosphere is estimated taking the laser–induced distortion of the Maxwellian distributions into account.*

1. It is well known that laser–induced variation in the refractive index of media in the region of anomalous dispersion leads to the resonance self–focusing (self–defocusing) of a laser beam.[1,2] This effect manifests itself within the small time of radiation exposure when the thermal effects are insignificant and the mechanism of thermal self–action has not yet worked.[3–5] The resonance self–focusing in a gas is usually analyzed either without consideration of the distortion of Maxwell's velocity distribution of molecules in the radiation field[1,2,6] or by means of qualitative description of such a nonequilibrium within the scope of the model of relaxation constants.[7] The quantitative description of the laser–induced velocity nonequilibrium in molecular gases requires the use of a more complicated kinetic model taking into account the processes of collisional elastic ($T–T$) and rotational ($R–T$) relaxations.[8] In this paper the curves of anomalous dispersion of a molecular gas are calculated for various ratios of the velocities of elastic and rotational relaxations. It is shown that an account of the laser–induced velocity nonequilibrium of molecules may increase the absolute value of variation in the refractive index of a gas by several times and, as a result, affect strongly the resonance self–focusing of the radiation.

2. Lef us consider a minor impurity of resonance absorbing molecules in a buffer gas. We assume that the monochromatic radiation interacts noncoherently with an isolated vibrational–rotational transition $|V, J> - |V', J'>$ distoring Maxwell's velocity distribution of molecules at the levels $|V, J>$ and $|V', J'>$. Here we restrict ourselves to an analysis of a situation in which the resultant velocity distribution function of molecules remains Maxwellian. Taking into consideration the processes of optical excitation and rotational and elastic relaxations of gas molecules the absorption coefficient has the form[8]:

$$\alpha_{VJ}^{V'J'}(\omega-\omega_{VJ}^{V'J'}) = \frac{\frac{\pi}{2}\alpha_0\Delta\omega_L g_V(x, a^*)}{\sqrt{1+k}+\frac{\pi\tau_{RT}}{2\tau_e}\kappa\Delta\omega_L g_V(x,a^*)}, \quad (1)$$

$$\alpha_0 = \frac{2}{\pi}\sigma_0(N_V q_J - N_{V'}q_{J'}); \quad \sigma_0 = \frac{4\pi^2}{3c\hbar^2}|d_{VJ}^{V'J'}|^2\frac{\hbar\omega_{VJ}^{V'J'}}{\Delta\omega_L};$$

$$\kappa = \frac{16\pi}{3c\hbar^2}|d_{VJ}^{V'J'}|^2 I\frac{\tau_p}{\Delta\omega_L}; \quad \tau_p = \frac{\tau_e\tau_{RT}}{\tau_e+\tau_{RT}};$$

$$g_V(x,a^*) = 2\sqrt{\frac{\ln 2}{\pi}}\frac{a^*}{\pi\Delta\omega_D}\int_{-\infty}^{\infty}\frac{\exp(-y^2)\,dy}{(x-y)^2+a^{*2}};$$

$$x = 2\sqrt{\ln 2}\frac{(\omega-\omega_{VJ}^{V'J'})}{\Delta\omega_D}; \quad a^* = \frac{\Delta\omega_L\sqrt{\ln 2}\sqrt{1+\kappa}}{\Delta\omega_D}.$$

Here $|d_{VJ}^{V'J'}|$ and $\omega_{VJ}^{V'J'}$ are the matrix element of the dipole moment and the frequency of transition $|V, J> - |V', J'>$, ω and I are the frequency and the intensity of laser radiation, $\Delta\omega_L$ and $\Delta\omega_D$ are the Lorentz and Doppler widths of the absorption line, τ_{RT} and τ_e are the characteristic times of rotational and elastic relaxations between the molecular levels being considered (τ_{RT}^{-1} is the total velocity of withdrawal of the particles onto the other rotational sublevels and τ_e^{-1} is the frequency of thermalized collisions with unchanged rotational molecular state[8]), N_V and $N_{V'}$ and q_J and $q_{J'}$ are the vibrational populations and equilibrium fractions of molecules at rotational sublevels, c is the light velocity, and $\hbar$ is Planck's constant. The function $g_V(x, a^*)$ describes the Voigt line profile with the radiation intensity–dependent parameter a^*.

In accordance with the Kramers–Kronig relations[9] the refractive index of a gas is

$$n = 1 + \frac{c}{2\pi}\int_{-\infty}^{\infty}\frac{\alpha_{VJ}^{V'J'}(\tilde\omega-\omega_{VJ}^{V'J'})}{\tilde\omega(\tilde\omega-\omega)}d\tilde\omega. \quad (2)$$

0235-6880/92/04 262-03 $02.00

In writing down Eq. (2) we neglect the contribution of far–line wings. Hereafter it is also assumed that the pulse duration of laser radiation is much less than the characteristic time of change in the population of the lower vibrational level. In this case we assume with a good accuracy the value of N_V be equilibrium and $N_V q_J \gg N_{V'} q_{J'}$. This assumption allows us to consider the quantity $(N_V q_J - N_{V'} q_{J'})$ entering into Eq. (1) to be independent of the radiation frequency which considerably simplifies the integration of Eq. (2).

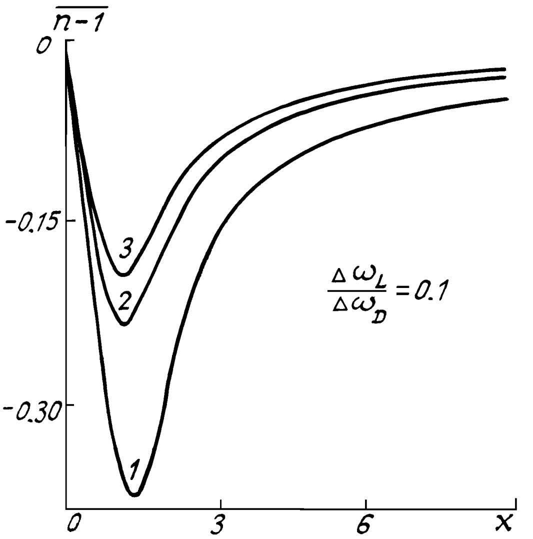

FIG. 1. Lower halves the curves of anomalous dispersion of a molecular gas calculated according to Eqs. (1) and (2) for a fixed value of the radiation intensity ($\beta^* = 5$) and for different values of τ_e/τ_{RT}: 1) 0, 2) 1, and 3) ∞. The quantity $\overline{n-1} = \dfrac{2(n-1)\,\omega_{VJ}^{V'J'}\,\Delta\omega_D}{c\alpha_0\Delta\omega_L}$ *is laid off as ordinate.*

Shown in Fig. 1 are the curves of anomalous dispersion calculated from Eqs. (1) and (2) for different values of the parameter τ_e/τ_{RT}. The dependences of the normalized variations in the refractive index $\delta\overline{n} = \dfrac{\delta n}{1 - n(I = 0)}$ on the radiation intensity (on the parameter $\beta^* = \dfrac{\kappa\tau_{RT}}{2\tau_p} \sim I$) are shown in Fig. 2, where $\delta n = n(I) - n(I = 0)$. The curves with $\tau_e/\tau_{RT} = \infty$ correspond to the maximum velocity nonequilibrium, the curves with $\tau_e/\tau_{RT} = 0$ correspond to the absence of distortions of Maxwellian molecular velocity distribution. It can be easily seen that the account of laser–induced velocity nonequilibrium does not alter the qualitative behavior of anomalous dispersion and of $\delta\overline{n}(I)$. However, an account of this nonequilibrium increases, as it can be seen from Fig. 2, the magnitude of variation in the refractive index $\delta\overline{n}$ almost by a factor of three in some cases.

The effect of elastic relaxation becomes pronounced only for $\tau_e/\tau_{RT} < 1$. If $\tau_e/\tau_{RT} > 1$, the approximation $\tau_e/\tau_{RT} \gg 1$ can be used with satisfactory accuracy. In this case the simple formula for the refractive index can be derived from Eqs. (1) and (2)

$$n = 1 + \frac{1}{2\sqrt{1+\kappa}}\frac{c\alpha_0}{\omega_{VJ}^{V'J'}}\frac{\Delta\omega_L}{\Delta\omega_D}\sqrt{\frac{\ln 2}{\pi}}\int_{-\infty}^{\infty}\frac{(y-x)\exp(-y^2)\,dy}{(y-x)^2 + a^{*2}}, \quad (3)$$

$$\kappa = \frac{16\pi}{3c\hbar^2}\,|d_{VJ}^{V'J'}|^2\,I\tau_{RT}/\Delta\omega_L .$$

This formula becomes simpler for $a^* \gg 1$:

$$n = 1 - \frac{c\alpha_0}{\omega_{VJ}^{V'J'}\sqrt{1+\kappa}}\,\frac{(\omega - \omega_{VJ}^{V'J'})\,\Delta\omega_L}{4\,(\omega - \omega_{VJ}^{V'J'})^2 + \Delta\omega_L^2\,(1+\kappa)} . \quad (4)$$

Note that Eq. (4) for $a^* \gg 1$ also follows immediately from Eqs. (1) and (2) regardless of the ratio of τ_e and τ_{RT}.

The saturation parameter κ in Eqs. (3) and (4) is independent of the time τ_e and is determined by the characteristic time τ_{RT}. It is physically obvious since in the former case the elastic relaxation can be neglected and in the latter case all molecules interact with radiation regardless of their velocities. It should be noted that in the particular case of $\kappa = 0$ Eq. (3) transforms into the relation which was derived in Ref. 6 and used to estimate the variation in the refractive index of water vapor in the field of the CO_2–laser radiation.

3. Let us estimate the parameter of self–action of the beam $Z_f = \dfrac{d}{4}\left(\dfrac{1}{\delta n}\right)^{1/2}$ (d is the initial beam diameter) in the case of resonance self–focusing of the CO_2–laser radiation in the atmosphere. Recall that the effect of resonance self–focusing is observed when $\omega > \omega_{VJ}^{V'J'}$. The detuning of the cyclic frequency of the CO_2–laser radiation from the center of the corresponding absorption line of the atmospheric CO_2 can be produced not only by the intraresonator methods[10] but also as a result of motion of the radiation source.[11] The calculated parameters of the curves $\delta\overline{n}(I)$ in Fig. 2 correspond to the absorption of the radiation at the 10 P (20) line of the CO_2 laser for $x = 1$ (the cyclic radiation frequency $\omega = \omega_{VJ}^{V'J'} + \dfrac{\Delta\omega_D}{2\sqrt{\ln 2}}$) for the mid–latitude atmosphere at an altitude of about 45 km (see Ref. 12): $p \approx 1.76\cdot10^{-3}$ atm, $T \approx 270$ K, and $\Delta\omega_L/\Delta\omega_D \approx 0.158$; in addition, $1 - n(I = 0) \approx 1.85\cdot10^{-11}$ and $\beta^* \approx 2\cdot10^{-3}I$ (W/cm^2). The exact value of τ_e/τ_{RT} for the levels of the transition 10 P (20) of CO_2 in air (N_2) is unknown. However, simple estimates on the basis of the experimental data of Ref. 13 yield $\tau_e/\tau_{RT} > 1$. As has already been noted above, in this case Eq. (3) can be used with satisfactory accuracy. For the radiation intensity $I = 500$ W/cm^2 ($\beta^* \approx 1$) $Z_f \approx 8.7\cdot10^4 d$. Disregarding the velocity nonequilibrium this value would be greater by a factor of 1.5. The beam is focused when $Z_f < \dfrac{\pi d^2}{2\lambda}$ ($\lambda \approx 10.6$ μm is the radiation wavelength). Under these conditions with an account of the velocity nonequilibrium this inequality is fulfilled for the beams with diameter $d > 0.6$ m. It must be stressed that the inequality $(\alpha_{VJ}^{V'J'})^{-1} > Z_f$ is valid for $d < 1$ m, i.e., the beam's energy dissipation can be ignored when estimating the self–action parameter.

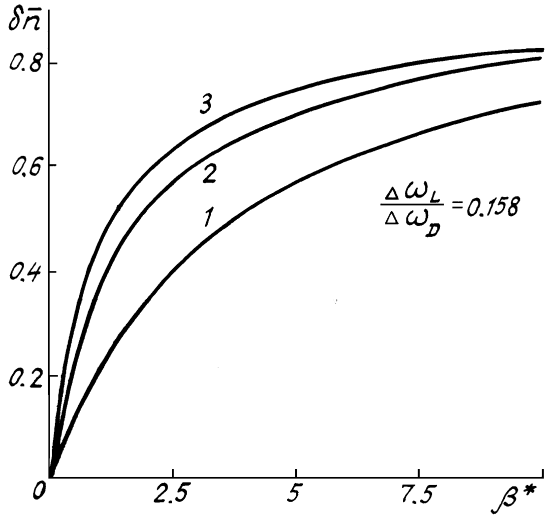

FIG. 2. Variation in the refractive index $\delta n = \frac{[n(I) - n\,(I = 0)]}{[1 - n\,(I = 0)]}$ vs *the radiation intensity (the parameter* β*) *for a fixed value of detuning* $x = 1$. *For notation see Fig. 1.*

The analysis of the curves of anomalous dispersion of CO_2 in the atmosphere calculated at different altitudes H shows that the effect of laser–induced velocity nonequilibrium of molecules on δn decreases rapidly as the altitude decreases (i.e. as $\Delta\omega_L/\Delta\omega_D \sim p \sim \exp(-H)$ grows).

The altitude $H = 45$ km was chosen for the estimates starting from the following compromise considerations:

1) altitude must be high enough in order to the Benneth dip and peak be sharply pronounced ($H > 35$ km);

2) altitude must not be very high to observe the radiation self–focusing along the path lengths of interest in practice.

Note in conclusion that the above–discussed consideration is applicable also for resonance self–defocusing of radiation when $\omega < \omega_{VJ}^{V'J'}$.

REFERENCES

1. A. Javan and P.L. Kelley, IEEE J. Quant. Electr. **QE–2**, No. 9, 470–473 (1966).
2. V.S. Butylkin, A.E. Kaplan, Yu.G. Khronopulo, and E.I. Yakubovich, *Resonance Interaction of Light with Matter* (Nauka, Moscow, 1977).
3. S.A. Akhmanov, R.V. Khokhlov, and A.P. Sukhorukov, Usp. Fiz. Nauk **93**, No. 1, 19–70 (1967).
4. A.I. Osipov and V.Ya. Panchenko, *Thermal Effects Attendant the Interaction of Laser Radiation with Molecular Gases* (State University Publishing House, Moscow, 1981).
5. V.E. Zuev, *Propagation of Laser Radiation in the Atmosphere* (Radio i Svyaz', Moscow, 1981).
6. V.V. Zhuravlev, A.A. Sorokin, and A.M. Starik, Khimich. Fizika, **9**, No. 2, 201–211 (1990).
7. S. Stenkhol'm, *Foundations of Laser Spectroscopy* [Russian translation] (Mir, Moscow, 1987).
8. A.E. Bazelyan, S.V. Ivanov, and M.N. Kogan, Zh. Eksp. Teor. Fiz. **99**, No. 4, 1088–1102 (1991).
9. V.M. Fain and Ya.I. Khanin, *Quantum Radiophysics* (Sov. Radio, Moscow, 1965).
10. G.W. Sutton and D.H. Douglas–Hamilton, Appl. Opt. **18**, No. 13, 2323–2328 (1979).
11. S.V. Ivanov, in: Trudy of the Central Aero–Hydrodynamics Institute, No. 2461, 15–27 (1990).
12. A.A. Mitsel and Yu.N. Ponomarev, *Optical Models of the Molecular Atmosphere* (Nauka, Novosibirsk, 1988).
13. R.R. Jacobs, K.J. Pettipiece, and S.J. Thomas, Appl. Phys. Lett. **24**, No. 8, 375–377 (1974).