Gain Saturation Effects of a Flowing Chemical Oxygen–Iodine Laser *

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A new oxygen–iodine medium gain model is developed to include pumping and deactivation of the upper laser levels of the iodine atoms, hyperfine and translation relaxation, as well as the flowing effect. The rate equations for gain of a supersonic flowing cw oxygen–iodine laser (COIL) are described when the medium is stimulated by a single-mode field. The general solution of the self-consistency integral equation is obtained. The result shows that the saturation behaviour in low pressure of the COIL differs from both the inhomogeneous and homogeneous broadening, and exhibits an 'anomalous' saturation phenomenon.

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A flowing chemical oxygen–iodine laser (COIL) has attracted extensive interest in recent decades because of its near-infrared wavelength, high-power and potential applications. Zagidullin *et al.*[1] and Copeland *et al.*[2] examined the optical saturation properties of the iodine transition in oxygen–iodine medium. They have considered the pumping, quenching, and hyperfine and velocity cross-relaxation of the medium. They also have assumed that the total number density of excited iodine atoms is independent of the streamwise distance. Fan and Gao *et al.* have examined the saturation properties in low pressure supersonic cw HF chemical laser by considering flowing field effect.[3−7] In this Letter, we examine the saturation behaviour of the COIL in the pressure governed by Doppler broadening of the transition line.

The $^2P_{3/2}$ and $^2P_{1/2}$ electronic levels of atomic iodine in the oxygen–iodine medium are split into two and four hyperfine sublevels, respectively.[1] Since the hyperfine splitting energy is much less than the average thermal energy k_BT, the equilibrium population of the hyperfine sublevels is proportional to their statistical weights g_f^* $(f = 2, 3)$ and $g_{f'}$ $(f' = 1, 2, 3, 4)$. The operation of the COIL is in the low active-medium pressure that does not exceed a few torr.[8−10] Under this condition the spectral lines of the $^2P_{1/2} \rightarrow {}^2P_{3/2}$ transition in atomic iodine are broadened inhomogeneously, and do not overlap. Thus a monochromatic field interacts with just one of these transitions and only with those iodine atoms for which the Doppler frequency shifts (relative to the field frequency) are within the limits of homogeneous line width.

The kinetics model of the COIL gain generalizes the gain models of Zagidullin *et al.*[1] and Copeland *et al.*[2] It is described as follows: (1) The energy stored in the flowing excited metastable oxygen O_2 $(^1\Delta)$ is transferred to the atomic iodine $^2P_{3/2}$ states with the energy-transfer rate constant k_f. This results in the pumping of the upper laser level of the iodine atoms. (2) The deactivation of the I $(^2P_{1/2})$ states is due to the energy transfer from excited iodine atoms to oxygen molecules O_2 $(^1\Delta)$ with the transfer rate constant $k_r = k_f/K$ (K is the equilibrium constant) and the quenching of excited iodine atoms by water and O_2 $(^1\Delta)$ with the quenching rate constants k_{H_2O} and k_Δ, respectively. (3) The hyperfine relaxation of the upper and lower iodine hyperfine sublevels with the rate constants k_h^* and k_h, respectively. (4) The translational relaxation of the upper and lower iodine hyperfine sublevels with the rate constant k_t^* and k_t, respectively. (5) The magnetic dipole transition between the $f = 3$ and $f' = 4$ hyperfine levels of $^2P_{1/2}$ and $^2P_{3/2}$, respectively, the corresponding wavelength $1.315\,\mu$m has sufficient gain to laser. (6) The number density of excited oxygen is related to the streamwise distance.

We consider the single longitudinal-mode operation and assume the gas flow to be along the x-direction with a velocity u. Let $n_f(v')$ and $n_{f'}(v')$ denote the number densities of the distribution of the iodine atoms in terms of the Doppler frequency shift in states $^2P_{1/2}$ and $^2P_{3/2}$, respectively. The particles with Doppler frequency ν' emit an homogeneous broadening profile with the central frequency ν' and the homogeneous width $\Delta\nu_c$. The transition cross section reads as

$$\sigma(\nu') = \sigma_0 L(\nu'), \quad \sigma_0 = \frac{\lambda_0^2}{8\pi}A,$$
$$L(\nu') = \frac{(\Delta\nu_c/2)^2}{(\nu-\nu')^2 + (\Delta\nu_c/2)^2}, \qquad (1)$$

where λ_0 and A denote the wavelength and Einstein's A-coefficient for the $f = 3 \rightarrow f' = 4$ transition, and

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ν is the frequency of the stimulating field. After the iodine atoms come into an energy exchange reaction with $O_2(^1\Delta)$, the velocity distribution of the iodine atoms becomes Maxwellian. The Maxwellian distribution with frequency ($\nu' = \nu_0(1+v/c)$ (v is the thermal velocity) is expressed by

$$g(\nu', \nu_0) = \frac{1}{\Delta\nu_D}\sqrt{\frac{4\ln 2}{\pi}}\exp\Big[4\ln 2\Big(\frac{\nu'-\nu_0}{\Delta\nu_D}\Big)^2\Big], \quad (2)$$

Let $F(\nu)$ denotes the intensity of the stimulating field travelling in the positive or negative z directions. Then the master equations describing the steady-state change rate of the atomic iodine are expressed by

$$u\frac{\partial n_3^*(\nu')}{\partial x} = \alpha\tau_f^{-1}ng(\nu',\nu_0) - \tau_d^{-1}n_3^*(\nu') - \tau_h^{*-1}[n_3^*(\nu') - \alpha n^*(\nu')] - \tau_t^{*-1}[n_3^*(\nu') - n_3^* g(\nu',\nu_0)] - \sigma_0\frac{F}{hv}L(\nu')\Delta n(\nu'), \quad (3)$$

$$u\frac{\partial n_2^*(\nu')}{\partial x} = \alpha\tau_f^{-1}ng(\nu',\nu_0) - \tau_d^{-1}n_2^*(\nu') - \tau_h^{*-1}[n_2^*(\nu') - \alpha' n^*(\nu')] - \tau_t^{*-1}[n_2^*(\nu') - n_2^* g(\nu',\nu_0)], \quad (4)$$

$$u\frac{\partial n_4(\nu')}{\partial x} = \beta\tau_d^{-1}n^* g(\nu',\nu_0) - \tau_f^{-1}n_4(\nu') - \tau_h^{*-1}[n_4(\nu') - \beta n(\nu')] - \tau_t^{*-1}[n_4(\nu') - n_4 g(\nu',\nu_0)] + \sigma_0\frac{F}{hv}L(\nu')\Delta n(\nu'), \quad (5)$$

$$u\frac{\partial n_{f'}(\nu')}{\partial x} = \beta_f\tau_d^{-1}n^* g(\nu',\nu_0) - \tau_{f'}^{-1}n_{f'}(\nu') - \tau_{f'}^{*-1}n_{f'}(\nu') - \tau_h^{-1}[n_{f'}(\nu') - \beta_{f'}n(\nu')] - \tau_t^{-1}[n_{f'}(\nu') - n_{f'}g(\nu',\nu_0)], \quad f' = 1,2,3, \quad (6)$$

$$\alpha = g_3^*/g^* = 7/12, \quad \alpha' = g_2^*/g^* = 5/12,$$
$$\beta = g_4/g = 9/24, \quad \beta_{f'} = g_{f'}/g \quad (f' = 1,2,3),$$
$$\gamma = g_3^*/g_4 = 7/9, \quad \tau_f^{-1} = k_f n_\Delta,$$
$$\tau_d^{-1} = k_r n_\Sigma + k_\Delta n_\Delta + k_{H_2O}n_{H_2O},$$
$$\tau_h^{*-1} = k_h^* n_{O_2}, \quad \tau_h^{-1} = k_h n_M,$$
$$\tau_t^{*-1} = k_{t\,He}n_{He} + k_{t\,O_2}n_{t\,O_2},$$
$$\tau_t^{-1} = k_{t\,He}n_{He} + k_{t\,O_2}n_{t\,O_2},$$
$$n(\nu') = n_3^*(\nu') - \gamma n_4(\nu'), \quad (7)$$
$$n_f^* = \int n_f^*(\nu')d\nu', \quad n^*(\nu') = \sum n_f^*(\nu'),$$
$$n^* = \sum n_f^* = \int n^*(\nu')d\nu' = \sum\int n_f^*(\nu')d\nu', \quad (8)$$

where n_Δ, n_Σ, n_{O_2}, n_{H_2O}, n_{He} and n_M denote the number densities of O_2 ($^1\Delta$) and $O_2(^3\Sigma)$, total molecular oxygen, water, helium and total gas, respectively; $\Delta n(\nu')$ is the inversion of the oxygen iodine medium; $n_f(\nu')$, $n(\nu')$, n_f, and n have the similar relationships. The following nondimensional quantities are introduced:

$$R_f = \frac{\tau_d}{\tau_f}, \ R_h^* = \frac{\tau_d}{\tau_h^*}, \ R_h = \frac{\tau_d}{\tau_h}, \ R_t^* = \frac{\tau_d}{\tau_t^*},$$
$$R_t = \frac{\tau_d}{\tau_t}, \ \zeta = \frac{x}{u\tau_d}, \ I = \frac{F}{(h\nu/\sigma_0\tau_t)}, \quad (9)$$

where R_f is the energy-transfer intensity, R_h^* and R_h are the hyperfine relaxation intensities of the upper and lower iodine hyperfine manifolds, and R_t^* and R_t are the translational cross-relaxation intensities of the iodine-atom velocity manifolds; ζ and I are the dimensionless streamwise distance and intensity of the stimulating field, respectively.

Since the total number density of iodine atoms is conserved, i.e. $n^* + n = n_I$ =const, we have

$$\frac{\partial n^*}{\partial\zeta} = -\frac{\partial n}{\partial\zeta} = R_f n - n^* - R_t I\int_0^\infty L(\nu')\Delta n(\nu')d\nu'. \quad (10)$$

Considering that R_h and R_t are larger than R_f, R_h^* (their values are calculated in the following), we can find the self-consistency integral equation for $\Delta n(\nu')$,

$$\frac{\Delta n(\nu')}{\Delta n} = \frac{g(\nu',\nu_0)}{1 + A_0 + (1+\gamma)IL(\nu')} \cdot\Big[1 + A_0 + (1+\gamma)I\Big]\int_0^\infty L(\nu')\frac{\Delta n(\nu')}{\Delta n}d\nu', \quad (11)$$

where $A_0 = (R_h^* + R_h + R_t^*)/R_t$, $\Delta n = \int\Delta n(\nu')d\nu'$. In order to obtain the analytic solution of Eq. (11), we make use of the iteration method and approximately obtain

$$\Delta n(\nu') = \Delta n\frac{g(\nu',\nu_0)}{1 + A_0 + (1+\gamma)IL(\nu')} \cdot(1 + A_0)[1 + (1+\gamma)IP(\nu)], \quad (12)$$

$$P(\nu) = \int_0^\infty\frac{L(\nu')g(\nu',\nu)}{1 + A_0 + (1+\gamma)IL(\nu')}d\nu' = \frac{1}{1+A_0}\frac{1}{\sqrt{\pi}}\int_{-\infty}^\infty\frac{\eta^2 e^{-t^2}}{(\xi - t)^2 + \eta_I^2}dt, \quad (13)$$

where ξ, t, η, and η_i read as

$$\xi = \sqrt{4\ln 2}\frac{\nu-\nu_0}{\Delta\nu_D} \quad t = \sqrt{4\ln 2}\frac{\nu'-\nu_0}{\Delta\nu_D},$$
$$\eta = \sqrt{\ln 2}\frac{\Delta\nu_c}{\Delta\nu_D} = \frac{r}{\sqrt{\pi}}, \quad \eta_I = \eta\sqrt{1 + \frac{1+\gamma}{1+A_0}I}. \quad (14)$$

Multiplying Eq. (11) by R_f^{-1} and neglecting the term with R_f^{-1}, we have

$$n^* = n_0^* e^{-\zeta}, \quad n = -n_0^* e^{-\zeta}. \quad (15)$$

Then n_3^*, n_4 and Δn are obtained. With these results and considering $\eta \ll 1$, we obtain the gain of the medium at ν, i.e.

$$G(v) = \sigma_0 \int L(\nu')\Delta n(\nu')d\nu'$$
$$= n_0^*\sigma_0\eta\sqrt{\pi}\frac{1}{\sqrt{1+\dfrac{1+\gamma}{1+A_0}I}}e^{-\xi^2}$$
$$\cdot\Big\{\frac{A_1 - A_1'}{R_h^* + R_h + (1+\gamma)(1+A_0)S_v}e^{-\zeta}$$
$$-\frac{A_2 - A_2^* - A_4\zeta}{R_h + (1+\gamma)(1+A_0)S_v}e^{-(1+R_h^*)\zeta}$$
$$+ O[e^{-(R_f+R_h)\zeta}, e^{-(1+R_h^*+R_h)\zeta}]\Big\}, \tag{16}$$

where

$$S_\nu = R_t IP(\nu),$$
$$A_1 = \alpha(R_h^* - R_f)\Big(1 + \frac{R_h}{R_h^*}\Big) + \beta\gamma(R_h - 1)\frac{R_h^* + R_h - 2}{R_f + R_h - 1},$$
$$A_2 = \frac{R_h}{R_h^*}\alpha(R_h^* - R_f),$$
$$A_3 = \beta\gamma(R_h^* - R_f + 1)(R_h - 1)\frac{1}{R_f + R_h - 1},$$
$$A_4 = (1 + A_0)S_\nu A_2,$$
$$A_1' = (1 + A_0)S_\nu\Big[(R_h^* - R_f + 1) \cdot\gamma\frac{A_1}{R_h^* + R_h}\cdot\frac{1}{R_f + R_h - 1} + \frac{R_h}{R_h^*}\cdot\frac{A_1}{R_h^* + R_h}\Big],$$
$$A_2' = (1 + A_0)S_v\Big[(R_h^* - R_f + 1)\cdot\gamma\frac{A_2}{R_h}\cdot\frac{1}{R_f + R_h - 1} + \frac{A_1}{R_h^* + R_h} - \frac{A_2}{R_h} + \frac{A_3}{R_f + R_h - R_h^* - 1}\Big]. \tag{17}$$

From $dG_0(v)/d\zeta = 0$, the characteristic streamwise distance ζ_D is obtained. The small-signal gain $G_0(\nu)$ has the maximum value at ζ_D,

$$\zeta_D = \frac{1}{R_h^*}\ln\Big[\frac{A_2}{A_1}(1 + R_h^*)\Big(1 + \frac{R_h^*}{R_h}\Big)\Big]. \tag{18}$$

Following Ref. [2], $k_f = 2.33 \times 10^{-8/T}\,\mathrm{cm^3/s}$, $k_r = k_f/K$, $K = 0.75e^{(401.42/T)}$, $k_{\mathrm{H_2O}} = 2.0 \times 10^{-12}\,\mathrm{cm^3/s}$, $k_\Delta = 1.1 \times 10^{-13}\mathrm{cm^3/s}$, $k_h^* = 1.0 \times 10^{-10}\mathrm{cm^3/s}$, $k_h = 4.0 \times 10^{-10}\mathrm{cm^3/s}$; $k_{t\,\mathrm{He}}^* = 1.8 \times 10^{-11}T^{1/2}\mathrm{cm^3/s}$, $k_{t\,\mathrm{O_2}} = 8.7 \times 10^{-11}\mathrm{cm^3/s}$, $k_{t\,\mathrm{He}} = 1.3\times10^{-10}T^{0.262}\mathrm{cm^3/s}$, $k_{t\,\mathrm{O_2}} = 2.7\times10^{-10}\mathrm{cm^3/s}$. The Mach number is 2. The cavity temperature is 175 K. The medium conditions at the cavity inlet are found to be [2] $\Delta\nu_D = 14.5\sqrt{T}$ MHz, $\eta = 0.04$ ($r = 0.0172$) at $P = 1.22$ Torr, $\eta = 0.06$ at $P = 2.2$ Torr. According to Ref. [1] we obtain $A = 5\mathrm{s}^{-1}$. Zagidullin *et al.* assumed[1] that the diffusion coefficient of iodine atoms in oxygen does not differ too greatly from the diffusion coefficient in argon and the diffusion coefficient for I ($^2P_{1/2}$) differs slightly from the value for I ($^2P_{3/2}$). Based on these the relaxation times calculated as a function of pressure are summarized Table 1.

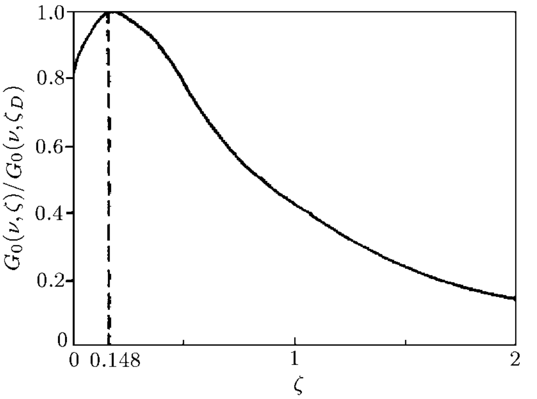

Fig. 1. The small-signal gain versus the streamwise distance.

Table 1. Parameters as a function of pressure P.

Parameter	Parameter values (for $P = 1.22$ Torr)	(for $P = 2.2$ Torr)	Lifetime
$q_f = 1.369 \times 10^6\,P\mathrm{s}^{-1}$	0.599	0.332	τ_f
$q_d = 1.861 \times 10^5\,P\mathrm{s}^{-1}$	4.404	2.442	τ_D
$q_h^* = 3.787 \times 10^6\,P_{\mathrm{O_2}}\mathrm{s}^{-1}$	0.398	0.221	τ_h^*
$q_h = 1.516 \times 10^7\,P\mathrm{s}^{-1}$	0.054	0.030	τ_h
$q_t^* = 5.705 \times 10^6\,P\mathrm{s}^{-1}$	0.144	0.080	τ_t^*
$q_t = 1.383 \times 10^7\,P\mathrm{s}^{-1}$	0.059	0.033	τ_t

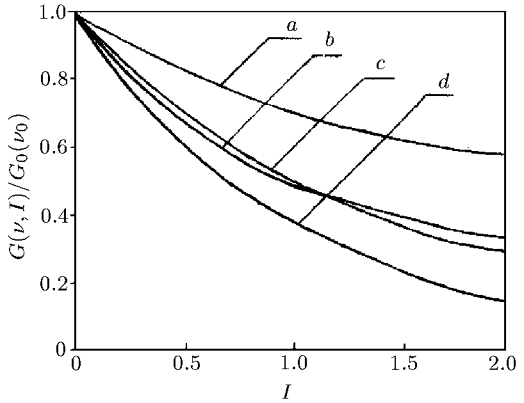

Fig. 2. The gain saturation behaviour: (*a*) $u = 0$, $\eta \ll 1$, $G(\nu,I)/G_0(\nu_0) = (1+I)^{-1/2}$; (*b*) $u = 0$, $\eta \gg 1$, $G(\nu,I)/G_0(\nu_0) = (1+I)^{-1}$; (*c*) $G(\nu,I,\zeta_D)/G_0(\nu_0,\zeta_D)$ with the Mach number 2, $P = 1.22$ Torr, $\eta = 0.04$, $\zeta_D = 0.148$, (*d*) $G(\nu,I,\zeta_D)/G_0(\nu_0,\zeta_D)$ with the Mach number 2, $P = 2.2$ Torr, $\eta = 0.06$, $\zeta_D = 0.148$.

The numerical calculations are carried out for ζ_D, $G_0(v)$, and $G(v)$ in terms of Eqs. (16) and (18) at the central frequency ($\xi = 0$). $\zeta_D = 0.148$, at $P = 1.22$ Torr and 2.2 Torr, corresponding the downstream distance $x = 0.044$ cm from the nozzle exit. The results for $G_0(\nu)$ and $G(\nu)$ are illustrated in Figs. 1 and 2.

It has been shown that the gain saturation behaviour in low pressure of the COIL differs from both the inhomogeneous and homogeneous broadenings, and exhibits the anomalous phenomenon with the mixing inhomogeneous–homogeneous broadening behaviour. This is due to the fact that in the low pressure level, the transition line is governed by the Doppler broadening and an inhomogeneous behaviour would be expected. However, the translational cross relaxation of the hyperfine sublevels of iodine atoms and the convection effect both can enhance the homogeneity of the transition line, which leads to the anomalous saturation phenomenon.

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