Theoretical Study of a New Energy Extraction Scheme of a Chemically Pumped Pulsed Iodine Laser Amplifier

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Abstract. A new energy extraction scheme of a chemically pumped pulsed large-scale iodine laser based on a high-pressure pulsed singlet oxygen generator is proposed. In previous investigations only low-pressure oxygen generators have been considered. Since they require a high iodine density for an efficient amplifier operation, the lifetime of the stored energy is correspondingly small and thus only small-sized iodine amplifiers appear to be technically feasible. We found, however, that when the singlet oxygen is generated at high-pressure, the iodine density required can be considerably reduced so that the lifetime of the stored energy becomes sufficiently long to fill up large amplifier cells. A numerical model is developed and the extractable energy is theoretically estimated. It is shown that 0.2 J/l · pass can be extracted when an input pulse of 20 ns duration (FWHM) and 1 J/cm^2 fluence is fed into the amplifying medium.

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A theoretical study of a new energy extraction scheme suited for a chemically pumped oxygen–iodine laser amplifier operated in the pulsed mode is conducted. An essential prerequisite of the scheme proposed to become efficient is the disposition of singlet oxygen at high pressure. We recently succeeded in the development of a chemical singlet-oxygen generator meeting this requirement. It is capable of delivering singlet oxygen at a pressure of 46 hPa [1]. We think that its scalability potential will be sufficient to use it as a pump source for the final power amplifier of a large iodine-laser system whose smaller amplifiers are photolytically pumped and that is suited as a fusion driver. A few theoretical studies in this direction [2, 3] were already conducted in the early 80's. The main question concerned the suitability of an oxygen–iodine mixture as an amplifying medium for powerful short pulses meeting fusion requirements. It was generally believed that an oxygen–iodine medium is a good candidate for this purpose. However, a few shortcomings were pointed out later by Basov et al. [4]. The most difficult problem of an oxygen–iodine amplifier arises from the relation between the extractable energy and the lifetime of the energy stored in the oxygen molecules. When the oxygen–iodine medium contains no water vapor, this lifetime is controlled by the reaction (see Table 1).

$$O_2(a^1\Delta_g) + I(^2P_{1/2}) \xrightarrow{k_1} O_2(b^1\Sigma_g^+) + I(^2P_{3/2}), \qquad (1)$$
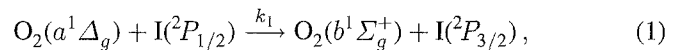
where $k_1 = 7.8 \times 10^{-14}$ cm^3/s. The stored energy lifetime τ_{rel} is then expressed as

$$\tau_{rel} = 1/\{k_1[I(^2P_{1/2})]\}. \qquad (2)$$

It becomes shorter than 1 ms for oxygen–iodine amplifier media which have been studied previously. Therefore, the characteristic length of the amplifier has to be shorter than 30 cm, even at an acoustic stream velocity. As the lifetime of the stored energy is inversely proportional to the iodine density, it will be lengthened by decreasing the iodine concentration. However, an efficient energy extraction is possible only when the iodine density is of the order of 10^{16} cm^{-3}.

On the other hand, we have found that the high iodine concentration is no longer indispensable when the density of the singlet oxyen is of the order of 10^{18} cm^{-3}. The use of an oxyge–iodine medium with thin iodine is then allowed and a large volume can thus be filled with the amplifying medium without major deactivation. In this paper, a new energy-extraction scheme based on a mixture containing oxygen at high pressure and iodine at low pressure is presented in detail as well as the results of model calculations. The input data necessary hereto were derived from our present apparatus.

1 Theory

1.1 Quasi-CW Energy Extraction

Generally, energy extraction from an oxygen–iodine medium comprises two steps. In the first step, the energy stored in the singlet oxygen molecules is transferred to excited state iodine atoms. In the second step, the input pulse extracts the energy corresponding to the population inversion existing

Table 1. Reactions and rate constants of the oxygen-iodine chemical system[a]

No.	Reaction	Rate constant
1	$O_2(a^1\Delta_g) + I(^2P_{1/2}) \rightarrow O_2(b^1\Sigma_g^+) + I(^2P_{3/2})$	$k_1 = 7.8 \times 10^{-14}$
2	$O_2(a^1\Delta_g) + I(^2P_{3/2}) \rightarrow O_2(X^3\Sigma_g^-) + I(^2P_{1/2})$	$k_2 = 7.6 \times 10^{-11}$
3	$O_2(X^3\Sigma_g^-) + I(^2P_{1/2}) \rightarrow O_2(a^1\Delta_g) + I(^2P_{3/2})$	$k_3 = 2.621 \times 10^{-11}$
4	$O_2(a^1\Delta_g) + O_2(a^1\Delta_g) \rightarrow O_2(b^1\Sigma_g^+) + O_2(X^3\Sigma_g^-)$	$k_4 = 2.0 \times 10^{-17}$
5	$O_2(b^1\Sigma_g^+) + O_2(X^3\Sigma_g^-) \rightarrow O_2(a^1\Delta_g) + O_2(X^3\Sigma_g^-)$	$k_5 = 4.6 \times 10^{-17}$
6	$O_2(b^1\Sigma_g^+) + H_2O \rightarrow O_2(a^1\Delta_g) + H_2O$	$k_6 = 6.7 \times 10^{-12}$
7	$O_2(b^1\Sigma_g^+) + Cl_2 \rightarrow O_2(a^1\Delta_g) + Cl_2$	$k_7 = 4.5 \times 10^{-16}$
8	$O_2(b^1\Sigma_g^+) + Ar \rightarrow O_2(a^1\Delta_g) + Ar$	$k_8 = 5.8 \times 10^{-18}$
9	$O_2(a^1\Delta_g) + O_2(X^3\Sigma_g^-) \rightarrow O_2(X^3\Sigma_g^-) + O_2(X^3\Sigma_g^-)$	$k_9 = 1.7 \times 10^{-18}$
10	$O_2(a^1\Delta_g) + H_2O \rightarrow O_2(X^3\Sigma_g^-) + H_2O$	$k_{10} = 5.6 \times 10^{-18}$
11	$O_2(a^1\Delta_g) + Cl_2 \rightarrow O_2(X^3\Sigma_g^-) + Cl_2$	$k_{11} = 3.0 \times 10^{-18}$
12	$O_2(a^1\Delta_g) + Ar \rightarrow O_2(X^3\Sigma_g^-) + Ar$	$k_{12} = 2.1 \times 10^{-19}$
13	$O_2(a^1\Delta_g) + I(^2P_{3/2}) \rightarrow O_2(X^3\Sigma_g^-) + I(^2P_{3/2})$	$k_{13} = 3.0 \times 10^{-14}$
14	$I(^2P_{1/2}) + O_2(X^3\Sigma_g^-) \rightarrow I(^2P_{3/2}) + O_2(X^3\Sigma_g^-)$	$k_{14} = 4.6 \times 10^{-14}$
15	$I(^2P_{1/2}) + H_2O \rightarrow I(^2P_{3/2}) + H_2O$	$k_{15} = 1.9 \times 10^{-12}$
16	$I(^2P_{1/2}) + Cl_2 \rightarrow I(^2P_{3/2}) + Cl_2$	$k_{16} = 2.0 \times 10^{-14}$
17	$I(^2P_{1/2}) + Ar \rightarrow I(^2P_{3/2}) + Ar$	$k_{17} = 2.0 \times 10^{-18}$
18	$I(^2P_{1/2}) + I(^2P_{3/2}) \rightarrow I(^2P_{3/2}) + I(^2P_{3/2})$	$k_{18} = 1.7 \times 10^{-13}$

[a] See [10]

between the excited and ground state iodine atoms, and afterwards the gain decreases to zero. Because only a portion of the energy stored in the oxygen molecules is transferred to the iodine atoms, an oxygen–iodine amplifier requires a multiple-pass extraction scheme with sufficient time between the passes for reequilibration of the energy transfer process. A high iodine-atom density is thus desirable to reduce the pass number. However, the maximum aperture of the amplifier is restricted by the lifetime of the stored energy, which is inversely proportional to the iodine density [see (1,2)]. This is the reason why the conventional oxygen–iodine medium was said to be not suitable for an amplifier of large-scale pulsed iodine lasers. Nevertheless, it will be shown that an interesting phenomenon occurs when the singlet oxygen density is in the range of 10^{18} cm^{-3}.

The energy transfer from singlet oxygen to iodine and the backward reaction,

$$I(^2P_{3/2}) + O_2(a^1\Delta_g) \underset{k_3}{\overset{k_2}{\rightleftarrows}} I(^2P_{1/2}) + O_2(X^3\Sigma_g^-), \tag{3}$$
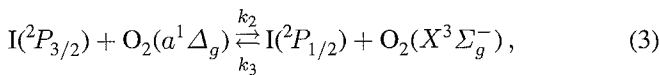
where $k_2 = 7.6 \times 10^{-11}$ cm^3/s and $k_3 = 2.6 \times 10^{-11}$ cm^3/s, are outstandingly higher than any other reaction in the oxygen–iodine chemical system. Therefore, the densities of $O_2(a^1\Delta_g)$, $O_2(X^3\Sigma_g^-)$, $I(^2P_{1/2})$, and $I(^2P_{3/2})$ reach the equilibrium state

$$\frac{\left[O_2(X^3\Sigma_g^-)\right]\left[I(^2P_{1/2})\right]}{\left[O_2(a^1\Delta_g)\right]\left[I(^2P_{3/2})\right]} = \frac{k_2}{k_3} = K_{eq}, \tag{4}$$

where $K_{eq} = 2.9$ at room temperature. The equilibration time τ_{trans} is connected to the rate constant of the energy transfer reaction k_2 by,

$$\tau_{trans} = \left\{k_2\left[O_2(a^1\Delta_g)\right]\right\}^{-1}. \tag{5}$$

When the density of the excited oxygen is $\gtrsim 10^{18}$ cm^{-3}, τ_{trans} becomes of the order of nanoseconds, which is comparable to the width of the pulses considered in fusion laser systems. It is shown that if the pulse width is in the 10 ns range, the O_2^*–I energy transfer, where O_2^* refers to $O_2(a^1\Delta_g)$,

occurs several times during the pulse duration, and it looks like the pulse accepts energy from $O_2(a^1\Delta_g)$ directly. Thus the high iodine concentration is no longer indispensable. The reduced iodine pressure helps lengthening the stored energy lifetime so that a large aperture cell can be filled up without difficulties. As this energy-extraction scheme is similar to that of the cw chemical oxygen–iodine lasers, we call it "quasi-cw regime". In the next paragraph, the energy extractable from the oxygen–iodine medium in this regime is estimated.

1.2 Estimation of the Extractable Energy

For simplicity, we consider the iodine atom as a two level system. The upper and the lower state have then degeneracy factors of 12 and 24, respectively. This model holds if the sublevel relaxation is the fastest of all processes being considered. The extractable energy can be divided into two portions. The first is stored in the excited iodine atoms. When the medium is irradiated with a pulse whose energy density is close to the saturation energy density, the energy stored in the iodine atoms is completely extracted through stimulated emission, as in photolytic iodine lasers. In this phase, the extracted energy is described by

$$E_1 = 2/3\left\{\left[I(^2P_{1/2})\right] - (1/2)\left[I(^2P_{3/2})\right]\right\}h\nu, \tag{6}$$

where $h\nu = 1.51 \times 10^{-19}$ J is the energy of a photon corresponding to the iodine laser transition $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$. After the extraction of E_1, the medium gain falls to zero and, generally, extraction does not occur any more. However, this is not the case in the high-density oxygen–iodine system. Here, the O_2^*–I energy transfer and the backward reaction cannot be ignored any more. The forward reaction is more likely than the backward reaction while the medium is irradiated by a high intensity photon flux, because the ratio of $[I(^2P_{1/2})]/[I(^2P_{3/2})]$ is forced to be kept at a value of 1/2 by the photon–iodine interaction. The excess pumping is compensated by stimulated emission to keep the gain at

zero. In this phase, the iodine can be regarded as a kind of catalyst and the energy is, so to speak, directly extracted from $O_2(a^1\Delta_g)$. In this phase, the energy extractable from the $O_2(a^1\Delta_g)$ (second portion of the overall extractable energy) is then described by

$$E_2 = \left\{ k_2 \left[I(^2P_{3/2}) \right] \left[O_2(a^1\Delta_g) \right] \right.$$
$$\left. - k_3 \left[I(^2P_{1/2}) \right] \left[O_2(X^3\Sigma_g^-) \right] \right\} \Delta t h\nu , \qquad (7)$$

where Δt is the pulse duration. Note that Δt does not mean the full pulse width at half maximum but the width at which the intensity corresponds to the saturation intensity of the medium. Assuming a value of 1/2 for the ratio $[I(^2P_{1/2})]/[I(^2P_{3/2})]$, expressing the ratio of excited oxygen to total oxygen by α, and the ratio of k_2/k_3 by 2.9 as given in (4), (7) becomes, after some rearrangement,

$$E_2 = (6.8\alpha - 1)[I][O_2]k_3\Delta t h\nu/3 , \qquad (8)$$

where [I] and $[O_2]$ refer to the total number densities of iodine and oxygen, respectively. The benefit of this scheme is that the extractable energy is proportional not only to the iodine density but also to the oxygen density. Thus, the extractable energy can be increased by the use of a high-density singlet oxygen source.

However, the saturation behaviour of this scheme is different from that of the conventional two level system. In the latter case the relevant parameter is the saturation energy density $E_{sat} = h\nu/(2\sigma)$, where σ is the stimulated emission cross section. To meet the quasi-cw condition the inequality $\tau_{trans} \gg \tau_{ex}$, where τ_{ex} is the characteristic energy extraction time from the iodine atoms through stimulated emission, must be satisfied because of the following reason. The relationship between photon density and population inversion is approximated by the rate equation

$$d\Delta n/dt \approx -\sigma\Delta n n_p c , \qquad (9)$$

where Δn is the inversion density, n_p is the photon number density, and c is the velocity of light. The right-hand side of this equation becomes zero after E_1 has been extracted from the medium, and the equilibrium is established. The O_2^*–I energy transfer reaction must not disturb this equilibrium to maintain the quasi-cw energy extraction. Therefore, the above mentioned inequality $\tau_{trans} \gg \tau_{ex}$ must be satisfied. From (9), τ_{ex} can be approximated by $(\sigma n_p c)^{-1}$. Since σ is of the order of 10^{-18} cm^2 under our conditions, τ_{ex} is then $\sim 3 \times 10^7 (n_p)^{-1}$ s, where n_p is measured in cm^{-3}. Noting that τ_{trans} is in the range of 10 ns, $n_p \sim 10^{16}$ cm^{-3} is required. This corresponds to a pulse of 50 MW/cm^2 peak intensity and of 1 J/cm^2 fluence when the pulse duration (FWHM) is assumed to be 20 ns. These values exceed the corresponding values of the saturation intensity and saturation energy density of the above mixture by nearly an order of magnitude. Hence, the quasi-cw energy-extraction scheme requires quite high-power input pulses.

2 Numerical Calculation Model

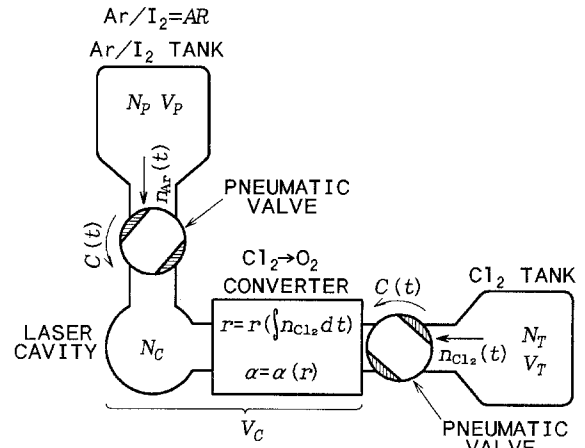We combined a suitable set of rate equations simulating the chemical kinetics of the laser medium and the Maxwell-

Fig. 1. Schematic drawing of the model used for the numerical calculations

Bloch equations for the description of the light–matter interaction to find out how the quasi-cw energy extraction occurs in a real singlet-oxygen generator. The calculation model is divided into two parts. In the first part, the time history of the amplifier medium is calculated until the pulse input time. The set of rate equation used for this is adapted to the characteristics of our apparatus. Figure 1 shows schematically how this calculation is carried out. The apparatus to be simulated comprises a laser cavity, a Cl_2 tank, a $Cl_2 \rightarrow O_2$ converter connecting the cavity and the Cl_2 tank, and an Ar/I_2 tank which contains a mixture of argon and gaseous iodine molecules. Two pneumatic valves are simultaneously opened at $t = 0$, thus allowing that the laser cavity is filled up with the oxygen–iodine mixture. The detailed description of the model can be found in [5]. The time dependence of the species in the laser cavity are described by the system of rate equations:

$$\frac{d\left[O_2(b^1\Sigma_g^+)\right]}{dt} = \chi^{pro}\{O_2(b^1\Sigma_g^+)\} - \chi^{dis}\{O_2(b^1\Sigma_g^+)\} , \quad (10)$$

$$\frac{d\left[O_2(a^1\Delta_g)\right]}{dt} = \chi^{pro}\{O_2(a^1\Delta_g)\} - \chi^{dis}\{O_2(a^1\Delta_g)\}$$
$$+ \alpha(r)r(t)\frac{n_{Cl_2}}{V_C} - 3\frac{n_{Ar}}{V_C AR} , \qquad (11)$$

$$\frac{d\left[O_2(X^3\Sigma_g^-)\right]}{dt} = \chi^{pro}\{O_2(X^3\Sigma_g^-)\} - \chi^{dis}\{O_2(X^3\Sigma_g^-)\}$$
$$+ \left[1 - \alpha(r)\right]r(t)\frac{n_{Cl_2}}{V_C} + 3\frac{n_{Ar}}{V_C AR} , \qquad (12)$$

$$\frac{d\left[I(^2P_{1/2})\right]}{dt} = \chi^{pro}\{I(^2P_{1/2})\} - \chi^{dis}\{I(^2P_{1/2})\} , \qquad (13)$$

$$\frac{d\left[I(^2P_{3/2})\right]}{dt} = \chi^{pro}I(^2P_{3/2})\} - \chi^{dis}\{I(^2P_{3/2})\}$$
$$+ 2\frac{n_{Ar}}{V_C AR} , \qquad (14)$$

$$\frac{d[Cl_2]}{dt} = \left[1 - r(t)\right]\frac{n_{Cl_2}}{V_C} , \qquad (15)$$

$$\frac{d[\text{Ar}]}{dt} = \left[1 - \frac{1}{AR}\right]\frac{n_{\text{Ar}}}{V_{\text{C}}}, \qquad (16)$$

$$\frac{d[\text{H}_2\text{O}]}{dt} = 0, \qquad (17)$$

$$N_{\text{C}} = \left[O_2(b^1\Sigma_g^+)\right] + \left[O_2(a^1\Delta_g)\right] + \left[O_2(X^3\Sigma_g^-)\right]$$
$$+ \left[I(^2P_{1/2})\right] + \left[I(^2P_{3/2})\right]$$
$$+ [\text{Ar}] + [\text{Cl}_2] + [\text{H}_2\text{O}], \qquad (18)$$

$$\frac{dN_{\text{P}}}{dt} = -\frac{n_{\text{Ar}}}{V_{\text{P}}}, \qquad (19)$$

$$\frac{dN_{\text{T}}}{dt} = -\frac{n_{\text{Cl}_2}}{V_{\text{T}}}, \qquad (20)$$

$$n_{\text{Cl}_2} = C(t)(N_{\text{T}} - N_{\text{C}}), \qquad (21)$$

$$n_{\text{Ar}} = C(t)(N_{\text{P}} - N_{\text{C}}) \qquad (22)$$

with

AR [Ar]/[I$_2$] ratio,
n_{Cl_2} Cl$_2$ molecules fed to SOG in unit time [s^{-1}],
n_{Ar} Ar/I$_2$ mixture fed to laser cavity in unit time [s^{-1}],
V_{C} Volume of the laser cavity [cm^3],
V_{P} Volume of the Ar/I$_2$ tank [cm^3],
V_{T} Volume of the chlorine tank [cm^3],
N_{C} Number density in the laser cavity [cm^{-3}],
N_{P} Number density in the Ar/I$_2$ tank [cm^{-3}],
N_{T} Number density in the chlorine tank [cm^{-3}],
$C(t)$ Conductance [cm^3/s],
$r(t)$ Cl$_2 \rightarrow$ O$_2$ conversion efficiency,
$\alpha(r)$ Transient yield of O$_2(a^1\Delta_g)$.

χ^{pro} and χ^{dis} represent the production or dissipation reactions listed in Table 1. For example,

$$\chi^{\text{pro}}\{O_2(a^1\Delta_g)\} = \left[O_2(b^1\Sigma_g^+)\right]\left\{k_5\left[O_2(X^3\Sigma_g^-)\right]\right.$$
$$+ k_6[\text{H}_2\text{O}] + k_7[\text{Cl}_2] + k_8[\text{Ar}]\}$$
$$+ k_3\left[O_2(X^3\Sigma_g^-)\right]\left[I(^2P_{1/2})\right], \qquad (23)$$

$$\chi^{\text{dis}}\{O_2(a^1\Delta_g)\} = \left[O_2(a^1\Delta_g)\right]\left\{k_9\left[O_2(X^3\Sigma_g^-)\right]\right.$$
$$+ k_{10}[\text{H}_2\text{O}] + k_{11}[\text{Cl}_2] + k_{12}[\text{Ar}]$$
$$+ (k_2 + k_{13})\left[I(^2P_{3/2})\right] + k_1\left[I(^2P_{1/2})\right]\}$$
$$+ 2k_4\left[O_2(a^1\Delta_g)\right]^2. \qquad (24)$$

The expressions of $C(t)$, $r(t)$, $\alpha(r)$, and related constants were derived from the experimental results of our prototype apparatus [1]. The iodine molecule is assumed to be dissociated instantaneously and completely. The water vapor pressure is assumed to be constant at the saturation pressure in accordance with the observed results. The calculation starts at $t = 0$ with the initial conditions [O$_2$] = [Cl$_2$] = [Ar] = [I] = 0, and [H$_2$O] = 200 Pa. AR and the intial pressure of both the Cl$_2$ and Ar/I$_2$ tanks are determined prior to the beginning of the calculation. The calculation of the medium properties is stopped at the time when the pulse to be amplified enters the laser cavity. Then the second part of the model, namely, the pulse amplification is started. Now we can choose between two approaches for the pulse propagation. One is the rate-equation approximation and the other is the semi-classical approach in form of the Maxwell-Bloch

equations. We employed the latter because it can describe the effects of finite bandwidth of the medium.

The hyperfine structure must be considered when short pulses are amplified. Due to the specific hyperfine splitting of the excited $^2P_{1/2}$ state, the relaxation time T_{U}, between the upper-state sublevels is four to five orders of magnitude longer than that of the lower-state sublevels [6]. The dominant mixing process of the upper-state sublevels is collisional energy transfer between two iodine atoms, one of them being in the ground state and the other being in the excited state. Consequently, the relaxation time T_{U} can be shown to be only dependent on the iodine density [7] according to

$$T_{\text{U}} = (1.0 \times 10^{-9}\,[\text{I}])^{-1}, \qquad (25)$$

where [I] is in units of [cm^{-3}]. Under our conditions, T_{U} is of the order of hundreds of nanoseconds. In contrast, the mixing time of the lower-state sublevels is of the order of the inverse of the medium linewidth because Van der Waals interaction occurs in almost every collision involving the lower-state sublevels.

The derivation of the one-dimensional, plane-wave Maxwell-Bloch equations in the semi-classical approach starts from the definition of the electromagnetic field of the incident beam in the form

$$E(z,t) = 1/2\left\{e(z,t)\exp\left[i(\omega t - kz)\right] + c.c.\right\}, \qquad (26)$$

where $e(z,t)$ is the slowly varying electric field envelope, ω the circular carrier frequency, k the wave number, and z the propagation direction. The induced medium polarization has to account for the six individual $(l \rightarrow j)$ transitions and is thus given by

$$P(z,t) = -1/2\sum_{l=2}^{3}\sum_{j=l-1}^{l+1}\left\{ip_{lj}^*(z,t)\right.$$
$$\times \exp\left[-i(\omega t - kz)\right] + c.c.\}, \qquad (27)$$

where $*$ denotes the complex conjugate ($c.c.$). The subscript l refers to the upper-state sublevels and j to the lower-state sublevels. Further derivations for the degeneracy-summed treatment, applicable to the iodine laser, were already given by Uchiyama et al. [8]. We simplified their results according to our conditions. Firstly, the transition terms except that for the $(3 \rightarrow 4)$ transition are neglected because the pressure is too low to overlap the six transition lines and the carrier frequency is set at the center of the $(3 \rightarrow 4)$ transition. Secondly, the O$_2^*$–I energy transfer and the backward reaction must be considered since this process participates in the population of the upper- and lower-state sublevels. The Maxwell-Bloch equations can thus be written in the form

$$\frac{\partial e}{\partial z} + \frac{1}{c}\frac{\partial e}{\partial t} = \frac{k}{2\epsilon_0 c}p_{34}, \qquad (28)$$

$$\frac{\partial p_{34}}{\partial t} = -\frac{p_{34}}{T_2} + \frac{\epsilon_0 c}{kT_2}\sigma_{34}\left\{N_{l=3} - \frac{7}{9}N_{j=4}\right\}e, \qquad (29)$$

$$\frac{\partial N_l}{\partial t} = -\frac{1}{T_{\text{U}}}\left\{N_l - \frac{g_l}{12}\sum_{l=2}^{3}N_l\right\} + \frac{g_l}{12}\sum_{j=1}^{4}\chi_{\{N_j\}}^{\text{dis}}$$
$$- \chi_{\{N_l\}}^{\text{dis}} - \chi_{lj}, \qquad (30)$$

$$\frac{\partial N_j}{\partial t} = -\frac{1}{T_L}\left\{ N_j - \frac{g_j}{24}\sum_{j=1}^{4} N_j \right\} + \frac{g_j}{24}\sum_{l=2}^{3} \chi_{\{N_l\}}^{dis}$$
$$- \chi_{\{N_j\}}^{dis} - \chi_{lj}\,, \tag{31}$$

$$\chi_{lj} = \begin{cases} \dfrac{1}{4\hbar c}(ep_{34}^* + e^*p_{34}) & (l = 3 \text{ or } j = 4) \\ 0 & (\text{else})\,, \end{cases}$$

where ϵ_0 is the dielectric permittivity of vacuum, h Planck's constant, and σ_{34} the stimulated emission cross section of the $(3 \rightarrow 4)$ transition. The χ^{dis} represent the dissipation of the population due to the reactions listed in Table 1 and are described by

$$\chi_{\{N_l\}}^{dis} = k_3 N_l \left[O_2(X^3\Sigma_g^-)\right] + N_l \left\{ k_1 \left[O_2(a^1\Delta_g)\right] \right.$$
$$+ k_{14}\left[O_2(X^3\Sigma_g^-)\right] + k_{15}[H_2O] + k_{16}[Cl_2]$$
$$\left. + k_{17}[Ar] + k_{18}\sum_{j=1}^{4} N_j \right\}\,, \tag{32}$$

$$\chi_{\{N_j\}}^{dis} = k_2 N_j \left[O_2(a^1\Delta_g)\right]\,. \tag{33}$$

T_2 denotes the phase coherence time related to the line width $\Delta\nu$ by

$$T_2 = 1/(\pi\Delta\nu)\,. \tag{34}$$

The relaxation time T_L of the lower-state sublevels is related to T_2 by

$$T_L = T_2/2\,, \tag{35}$$

in accordance with [8].

In the following calculations the intial pressure of both the Cl_2 tank and the Ar/I_2 tank were kept at 1 MPa, and the ratio $[Ar]/[I_2] = AR$ was selected such that the overall extractable energy $(E_1 + E_2)$ should reach 90% of its peak value during a 1.3 ms period. The time limit of 1.3 ms was chosen because it takes the medium 1.3 ms to travel radially with subsonic velocity through a cell of 5000 cm² aperture area.

3 Results and Discussion

3.1 Time History of the Species

Figure 2 shows the time history of various species prior to the arrival of the input pulse. The Cl_2 density begins to increase at $t \sim 1$ ms because the reagent upon the porous-pipe surface is almost run out within the first 3 ms. It can be seen that the population inversion is maintained during the first 4 ms. This is due to the relatively low iodine density compared to previous works [2, 3].
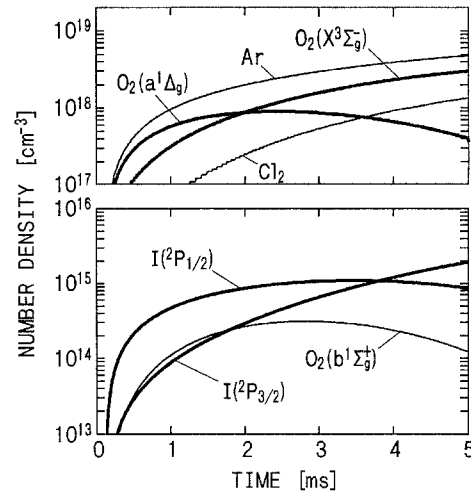

Fig. 2. Time history of the various species, calculated by the integration of the rate equations (10–22)

3.2 Saturation Intensity of the Medium

As stated above, the quasi-cw extraction scheme requires a high intensity input pulse compared to conventional iodine lasers. Figure 3 shows the energy extractable from the amplifier cell in one-pass operation as a function of the input pulse fluence for a triangular pulse of 20 ns duration (FWHM). The abscissa is scaled in units of peak intensity (W/cm²) and fluence (J/cm²). The vertical dashed line gives the saturation energy density of the iodine medium, given by $E_{sat} = h\nu/(2\sigma)$, and the horizontal line indicates the theoretical limit of the extractable energy expressed as $(E_1 + E_2)$. It can be seen that an efficient energy extraction requires a fluence 10 times higher than the saturation energy density, and 0.2 J/l is expected when the input fluence is higher than 1 J/cm².

Even in the highly saturated region, the extractable energy is only one half of the theoretical limit. This is due to the slow relaxation of the upper-state sublevels. The relaxation of these sublevels during amplification is therefore discussed in the next paragraph.
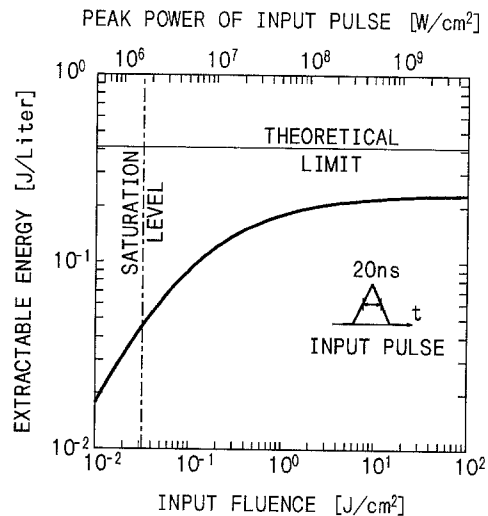

Fig. 3. Extractable energy per unit volume per pass as a function of input fluence, based on a numerical solution of the Maxwell-Bloch equations

3.3 Energy Extraction and Sublevel Relaxation of the Medium

Figure 4 shows the population density of each sublevel of the iodine atoms as a function of time, when a triangular pulse whose carrier frequency coincides with the center frequency of the $(3 \rightarrow 4)$ transition is fed in. The gain of the $(3 \rightarrow 4)$ transition and the intensity of the pulse are also shown using the same time scale. The width and fluence of the pulse is 20 ns (FWHM) and $10 \, \mathrm{J/cm^2}$, respectively. During about 3 ns after the pulse was fed in, the gain falls to zero, indicated as "phase A" in this figure. It means that the energy stored in the iodine atoms is completely exhausted in this phase. The amount of extracted energy is described by E_1 in (6), and the period of this phase is characterized by τ_{ex}. It is seen that in phase A the sublevel relaxation of the ground state is so fast that the corresponding population distributions are in equilibrium at any time. In contrast to this, the relaxation of the upper-state sublevels is much slower than the time scale now being considered. Thus, the density of the $(F = 2)$ sublevel of the supper-state hardly changes in this phase. In the next 40 ns, indicated by "Phase B", although the gain is kept at zero, the energy stored in the oxygen is extracted in the quasi-cw manner. The amount of energy extracted in this phase is described by E_2 in (8). It should be noted that only the density of the $(F = 2)$ upper-state sublevel is gradually increasing while the density of the other sublevel $(F = 3)$ is decreasing. This is because there are no energy extraction processes for the $F = 2$ sublevel while the energy-transfer reaction pumps both the $F = 2$ and $F = 3$ sublevels corresponding to their statistical weights. Therefore, only 7/12 of the O_2^*-I energy transfer is utilized for energy extraction and useless iodine molecules are stacked in the $(F = 2)$ sublevel. This is the reason why the calculated extractable energy is roughly one

half of the theoretical limit in Fig. 3. After the pulse has passed by, as indicated by "Phase C" in Fig. 4, the density of the iodine atoms shifts to the O_2^*-I equilibrium point described by (4). The time constant of the reequilibration is shown to be 30 ns in this figure. This corresponds to the theoretical value indicated by (5). The iodine atoms are charged with energy in this phase and extracted in phase A of the next pulse. A multi-pass amplification repeats these processes.

4 Conclusion

A theoretical study of a new energy extraction scheme of a pulsed chemical oxygen–iodine laser amplifier was presented. A theoretical model was developed by combining a set of rate equations describing the high-pressure chemical singlet oxygen generator and the laser medium evolution with the one-dimesional Maxwell-Bloch equations. It was shown that an efficient energy extraction can be expected from an oxygen–iodine medium having a lifetime of several milliseconds. The energy extractable by single-pass amplification was estimated to be $0.2 \, \mathrm{J/l \cdot pass}$, when the input pulse had a full width of 20 ns at half maximum and a fluence of $1 \, \mathrm{J/cm^2}$. A 100 kJ-class amplifier appears to be possible when the medium is prepared for a cell of $5000 \, \mathrm{cm^2}$ cross section and 10 m length, and when hundred passes can be achieved. This relatively high number can be greatly reduced by using an $O_2(a^1 \Delta_g)$ source operated at even higher pressures than considered here. This will not cause any shortening of the stored energy lifetime. The experimental verification of the new scheme proposed is still to be expected. Presently, we have combined our high-pressure singlet oxygen generator with an iodine injector and have successfully demonstrated pulsed operation of an iodine laser oscillator [9]. In the next step, the quasi-cw energy-extraction scheme will be investigated.

Fig. 4. Population distribution of the iodine sublevels as a function of time. The input pulse intensity and the gain of $(3 \rightarrow 4)$ line is also shown using the same time scale

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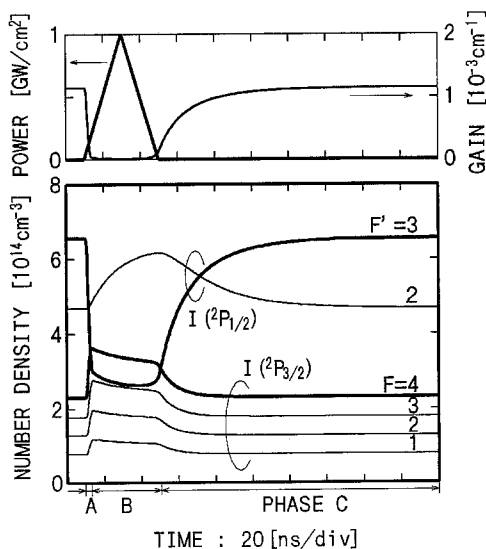10. A.I. Ddyukov, Yu.A. Kulagin, L.A. Shelepin, V.N. Yarygina: Sov. J. Quant. Electron. **19**, 578 (1989)