As discharge laser operating on the iodine 1315-nm transition

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Arguments are presented which indicate the feasibility of a gas-discharge iodine laser based on the 1315-nm transition, using a laser tube filled with a mixture of oxygen and iodine vapor. Underlying the operation of the laser is the experimental fact that the transfer of electron excitation energy between the iodine atoms and the oxygen molecules proceeds with a high efficiency. The construction of such a laser depends on how high the excitation temperature can be made by electron impact, while the translational temperature of the atoms and molecules is kept at a sufficiently low level. Experiments are briefly described in which an oxygen-induced increase of population of the metastable level $I(^2P_{1/2})$ is observed.

1. In 1971, Derwent and Thrush [1] established that quasi-resonant transfer of the energy of electron excitation between oxygen molecules and iodine atoms in the process

$$O_2(^1\Delta_g)+I(5^2P_{1/2})\rightleftarrows O_2(X,^3\Sigma_g^-)+I(5^2P_{3/2})+\Delta E(=279\,\text{cm}^{-1}) \qquad (1)$$

is characterized by an extremely high efficiency. This uncovers the possibility of producing a gas laser in which the overpopulation of the laser levels is maintained by transferring excitation energy from the molecules to the atoms. An interesting feature of this laser is that in principle one cannot exclude the possibility of its operating in the continuous regime, in spite of the fact that the lower laser level is the ground state of the iodine atom.

Several variants can be proposed for the use of the process (1) to pump a laser on the optical transition $5^2P_{1/2} - 5^2P_{3/2}$ in iodine atoms, with wavelength 1315 nm. We consider here the variant that is simplest and easiest to verify experimentally, in which it is proposed that the threshold overpopulation of the iodine laser levels be obtained in an electric-discharge laser tube containing oxygen with iodine vapor as an impurity. Owing to the process

$$I_2(X,\ ^1\Sigma_g^+)+O_2(^1\Sigma_g^+)\to 2I(5^2P_{1/2})+O_2(X,\ ^3\Sigma_g^-), \qquad (2)$$

which was investigated in [2,3], it would apparently be easy to reach almost complete dissociation of the iodine molecules in the positive column of the glow discharge, and also in an electrodeless high-frequency discharge. As we shall show, to obtain the population redistribution of the laser levels of iodine it is necessary to satisfy the following two conditions simultaneously:

1) The total population of the states $O_2(^1\Delta_g)$ and $I(^2P_{1/2})$ is determined by the collisions of the heavy particles (oxygen molecules and iodine atoms) with the electrons. If the electron velocity distribution is close to Maxwellian, the corresponding "excitation temperature" T_{ex} should be close to the electron temperature T_e. It is important here that the latter be high enough.

2) The relative populations of the states $O_2(^1\Delta_g)$ and $I(^2P_{1/2})$ are determined by the collisions of the heavy particles with one another and become redistributed in accordance with the translational gas temperature T_{tr}, which should be low enough.

The first condition determines the lower limit of the electron density (n_e^{min}), and the second the upper limit (n_e^{max}). The possibility of simultaneously satisfying both conditions reduces in fact to the requirement

$$n_e^{min} \leqslant n_e^{max}. \qquad (3)$$

The value n_e^{min} is obtained from the relation

$$\left(\langle\sigma_2^- v_e\rangle+\frac{n_{12}}{n_{22}}\langle\sigma_1^- v_e\rangle\right) n_e^{min} \gg \sum k_i n_i+\frac{n_{12}}{n_{22}}\sum k_i' n_i. \qquad (4)$$

Here and throughout $\langle\sigma_2^- v_e\rangle$ and $\langle\sigma_1^- v_e\rangle$ are the products of the electron velocities v_e by the effective cross sections for the quenching of the oxygen molecule by the electrons (σ_1^-) and of the iodine atom by the electrons (σ_2^-), averaged over the electron velocity; n_{11} and n_{12} are the populations of the ground and metastable ($^1\Delta_g$) states of the oxygen molecule; n_{21} and n_{22} are the populations of the ground and metastable states of the iodine atom; k_i and k_i' are the quenching constants of the iodine atoms and the $O_2(^1\Delta_g)$ molecules, respectively, in collisions with particles of sort i; n_i is the density of these particles.

We assume that a discharge regime can be realized wherein the decisive term in the right-hand side of the inequality (4) is $k_q n$, corresponding to the quenching of the iodine atoms by the iodine molecules, and that 99% of the iodine molecules have become dissociated in this case. Then, neglecting the second term of the left-hand side of the inequality (4) (and by the same token only strengthening this inequality), we obtain

$$n_e^{min} \gg k_q n/\langle\sigma_2^- v_e\rangle.$$

We present a numerical estimate for the case when the initial pressure of the iodine molecules is 1 Torr and the average electron velocity is 10^8 cm/sec ($T_e \sim 2$ eV). According to Tolmachev's definition [4], the effective cross section σ_2^- at $T_e \sim 6$—8 eV is equal to $(1.0 \pm 0.5) \times 10^{-16}$ cm^2. According to the data of Donovan and Husain [5], $k_q = 5 \times 10^{-12}$ cm^{3}sec. Assuming that in the range $T_e = 2$—8 eV the cross section σ_2^- is approximately proportional to $1/T_e$, we obtain for the indicated data the condition

$$n_e \geqslant n_e^{min} \gg \frac{5\cdot10^{-12}\cdot3.3\cdot10^{16}\cdot10^{-2}}{3.5\cdot10^{-16}\cdot10^{8}} = 4.7\cdot10^{10}\ \text{cm}^{-3}.$$

When the discharge current is increased, the degree of dissociation of the iodine molecules increases, but this may not lead to a decrease of n_e^{min}, inasmuch as when the degree of dissociation of the oxygen molecules is noticeably increased, the value of n_e^{min} will, apparently, be determined by the quenching of the iodine atoms by the oxygen atoms.

The quantity n_e^{max} is determined by the condition

$$k_{-1}n_{11} \gg n_e^{max}\langle\sigma_2^- v_e\rangle. \qquad (5)$$

Using the value of the rate constant of the reverse of process (1), $k_{-1} = 2.7 \times 10^{-11}$ cm^{3}sec^{-1} [6], and assuming that the concentration of the unexcited oxygen molecules is 3×10^{17} cm^{-3}, we obtain

$$n_e \ll n_e^{max} \ll \frac{2.7 \cdot 10^{-11} \cdot 3 \cdot 10^{17}}{3.5 \cdot 10^{-16} \cdot 10^{8}} = 2.3 \cdot 10^{14} \text{ cm}^{-3}.$$

Thus, in this case the two indicated conditions (4) and (5) are sufficiently well satisfied in the range of concentrations n_e from $n_e^{min} \sim 5 \times 10^{11}$ cm^{-3} to $n_e^{max} \sim 2 \times 10^{13}$ cm^{-3}.

At the accuracy defined by condition (4), the following balance equations are valid for a stationary discharge:

$$\dot{n}_{12} = -\dot{n}_{11} = k_{-1} n_{11} n_{22} - k_1 n_{12} n_{21} + k_1^{+} n_e n_{11} - k_1^{-} n_e n_{12} = 0, \quad (6)$$

$$\dot{n}_{22} = -\dot{n}_{21} = -k_1 n_{11} n_{22} + k_1 n_{12} n_{21} + k_2^{+} n_e n_{21} - k_2^{-} n_e n_{22} = 0. \quad (7)$$

Here $\dot{n}_{12}$, $\dot{n}_{11}$, $\dot{n}_{22}$ and $\dot{n}_{21}$ are the derivatives of the corresponding variables with respect to time; k_1 is the rate constant of process (1); $k_j^{\pm} = \langle \sigma_j^{\pm} v_e \rangle$, where j = 1 or 2; σ_1^{+} and σ_2^{+} are the effective cross sections of electron-impact excitation of the metastable states of $O_2(^1\Delta_g)$ and $I(^2P_{1/2})$, respectively. Adding (6) and (7), we obtain

$$n_{12} + \gamma n_{22} = b_1 n_{11} + \gamma b_2 n_{21};$$
$$b_1 = \frac{g_{12}}{g_{11}} \exp\left(-\frac{E_1}{kT_{ex}}\right), \quad b_2 = \frac{g_{22}}{g_{21}} \exp\left(-\frac{E_2}{kT_{ex}}\right), \quad \gamma = \frac{\langle \sigma_2^{-} v_e \rangle}{\langle \sigma_1^{-} v_e \rangle}; \quad (8)$$

Here g_{ik} are the statistical weights of the corresponding states; k is Boltzmann's constant; E_1 and E_2 are the energies of the levels $O_2(^1\Delta_g)$ and $I(^2P_{1/2})$, respectively; T_{ex} is the "excitation temperature" determined by the electrons, and is close to T_e in value.

It follows from (7) that

$$\frac{n_{12}}{n_{11}} = \frac{n_{22}}{n_{21}} a(1+\delta) = \frac{n_{22}}{n_{21}} a', \quad (9)$$

where

$$a = \frac{g_{12} g_{21}}{g_{11} g_{22}} \exp\left(-\frac{\Delta E}{kT_{tr}}\right), \quad \Delta E = E_1 - E_2,$$

T_{tr} is the "translational" temperature of the gas, and

$$\delta = \frac{k_2^{-} n_e}{k_{-1} n_{11}} \left(1 - b_2 \frac{n_{21}}{n_{22}}\right). \quad (10)$$

Taking into account the relation

$$\frac{1 - b_2 n_{21}/n_{22}}{1 - b_1 n_{11}/n_{12}} = -\frac{n_{12}}{\gamma n_{22}} < 0,$$

which follows from (8), it can be shown that the inequalities $b_2 n_{21}/n_{22} > 1$, $0 < b_1 n_{11}/n_{12} < 1$ and

$$\beta = \frac{b_2 n_{21} n_{12}}{b_1 n_{11} n_{22}} > 1 \quad (11)$$

hold true at $\delta < 0$, and that the inverse inequalities hold at $\delta > 0$. It follows from (9), however, that in general

$$\beta = (1+\delta) \exp\left\{\frac{\Delta E}{k}\left(\frac{1}{T_{ex}} - \frac{1}{T_{tr}}\right)\right\}. \quad (12)$$

We are interested only in the case when $T_{tr} < T_{ex}$. For this case we obtain from (11)

$$0 < \beta < 1 + \delta. \quad (13)$$

Inasmuch as the conditions (11) and (13) cannot be satisfied simultaneously at $\delta < 0$, the case $T_{tr} < T_{ex}$ corresponds to the condition $\delta > 0$ and accordingly $\beta < 1$ (it is obvious that $\delta = 0$ at $T_{tr} = T_{ex}$).

By virtue of the condition (5) we obtain at $T_{tr} < T_{ex}$, in accordance with (10),

$$|\delta| < \frac{k_2^{-} n_e}{k_{-1} n_{11}} \leqslant \frac{k_2^{-} n_e^{max}}{k_{-1} n_{11}} \ll 1.$$

Defining the effective value of T_r' by means of the relation $a(T_{tr})(1 + \delta) = a' = a(T_{tr}')$, and recognizing that $\ln(1 + \delta) \approx \delta$, we get

$$T_{tr}' \simeq T_{tr} \frac{1}{1 - \delta k T_{tr}/\Delta E}. \quad (14)$$

It is seen from (14) that under the condition (5) and $kT_{tr} \lesssim \Delta E$, the interaction of the heavy particles with the electrons increases the effective translational temperature of the gas only insignificantly.

Adding to (8) and (9) the equations that follow from the conservation law for the particles (the oxygen molecules and iodine atoms):

$$n_{11} + n_{12} = n_1, \quad (15)$$

$$n_{21} + n_{22} = n_2, \quad (16)$$

where n_1 and n_2 are respectively the concentrations of the oxygen molecules and iodine atoms, we obtain a closed system of equations (8), (9), (15), and (16). Substituting the solutions of these equations into the expression for the overpopulation of the laser levels $N = n_{22} - (g_{22}/g_{21}) n_{21}$, we obtain the following expression:

$$N = [2(1-a')(1+b_2)]^{-1} \{n_1'(1+g_2)(a'+b_1)(1 \pm D) + n_2[1 - g_2 + 2g_2 a' + 2b_2 - a' b_2 (1-g_2) \mp (1+g_2)(1+a'b_2) D]\}, \quad (17)$$

where

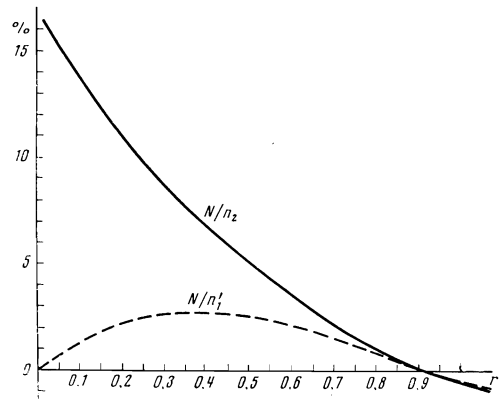
$$D = \{1 + 4a'(1+b_1)(1+b_2) r [r(1+a'b_2) - a' - b_1]^{-2}\}^{1/2},$$
$$g_2 = g_{22}/g_{21}, \quad r = n_2/n_1', \quad n_1' = n_1/\gamma.$$

The upper sign of D is taken at $r > (a' + b_1)/(1 + a'b_2)$ and the lower sign at $r < (a' + b_1)/(1 + a'b_2)$.

Expression (17) is suitable for any plasma that can be simulated with sufficient accuracy by a system consisting of two-level particles of two types and free electrons and distinguished by the fact that the exchange of excitation energy in collisions between particles of different types is much more rapid in the exchange of that energy with the free electrons that determine the excitation temperature of the metastable states. It should be noted that a similar model was in fact proposed by G. Gould for the description of collision lasers operating with the vapor of manganese and several other metals [7]. Insofar as we know, however, none of the subsequently developed metal-vapor lasers agreed with this model. Thus, for example, the manganese-vapor laser generates at self-limited transitions, not the ones predicted, and only in the pulsed regime [8]. Thus, inelastic collisions between the atoms did not play the required role in the maintenance of the overpopulation of the laser levels.

The laser-system model proposed in this paper differs essentially from Gould's model only in that we use not one but two different types of particles with relatively close ($\Delta E \sim kT_{tr}$) metastable levels. This yields an additional "controllable" parameter (r), the correct choice of which can facilitate the overpopulation of the laser levels. In particular, it is of interest to apply this conclusion to mixtures of metal vapors. It is not excluded that if the pair of metals is properly chosen, new forbidden (but not excessively so) optical quantum transitions will become accessible to laser radiation.

Let us consider the O_2 + I system in greater detail. In this case $g_{11} = 3$, $g_{12} = 2$, $g_{21} = 4$, $g_{22} = 2$, $E_2/k = 10\,900$°K and $\Delta E/k = 401$°K. If an electron temperature $T_e \approx T_{ex} = 2(E_2/k) = 1.9$ eV/k is reached in the discharge and the translational gas temperature is $\Delta E/k = 401$°K, then a positive excess population is reached in accordance with (17) in a rather wide range of values $r < 0.9$.



Plots of N/n_1' and N/n_2 against $r = n_2/n_1'$, calculated from formula (17) ($T_{ex} = 2(E_2/k)$, $T_{tr} = \Delta E/k$).

The plots of $N(r)/n_1'$ and $N(r)/n_2$ calculated for this case from formula (17) are given in the figure. They show that the optimal laser discharge regime should be sought at initial oxygen-molecule pressures from 1 to 20 Torr (if $\gamma \sim 1$), and at iodine pressures from 0.3 to 3 Torr (if the iodine is completely dissociated, the corresponding pressures of the iodine atoms range from 0.6 to 6 Torr). At higher initial gas pressures, the discharge may turn out to be inhomogeneous and unstable, and the energy consumed in maintaining the necessary degree of iodine dissociation may be too large.

It appears unlikely that continuous lasing could be counted on, owing to the danger of overheating and dissociation of the oxygen molecules. It would probably be much easier to realize a laser with periodically repeating pulses. Inasmuch as T_{ex} may remain high for a long time after the discharge current is turned off, in spite of the rapid decrease of T_e and n_e, lasing may continue (or begin) even during the afterglow stage.

The gain α can be determined from the formula $\alpha = \lambda^2 AN/8\pi\Delta\nu$, where λ is the wavelength (1315 nm) and A is the Einstein coefficient (7.7 sec^{-1}). From the width of the laser-line contour $\Delta\nu = 3 \times 10^9$ sec^{-1} and $n_2 = 6 \times 10^{16}$ cm^{-3} (corresponding to an initial iodine-vapor pressure ~1 Torr) we obtain in the case r = 0.2 (in which case $N/n_2 \sim 0.1$ according to the figure) a gain $\alpha = 0.01$ cm^{-1}, which is perfectly adequate to produce lasing even in tubes longer than 10—20 cm.

The per-unit power of the laser radiation w can easily be estimated if the unsaturated gain greatly exceeds the threshold. A sufficiently accurate formula for this case is

$$w \approx E_2 \langle \sigma_1^+ v_e \rangle \frac{n_e n_1}{b_1} \left[\frac{b_1 - g_2 a'}{1 + g_2 a'} - r \frac{g_2 - b_2}{1 + g_2} \right]. \qquad (18)$$

According to Trajmer, Williams, and Kupperman [9], at an electron energy 20 eV, the cross section σ_1^+ amounts to 4.2×10^{-18} cm^2 and is approximately equal to the effective cross section σ_V of vibration excitation in the O_2 molecule ($\sigma_V = 4.0 \times 10^{-18}$ cm^2). The fact that σ_1^+ is quite large, even through the corresponding optical transition is triply forbidden (in spin, parity, and angular momentum), can be attributed to the exchange mechanism [9]. The effective value of σ_1^+ at $kT_e \sim 2$ eV may therefore turn out to be much higher. Assuming it to be 10^{-17} cm^2, we obtain with the aid of formula (18) for the conditions corresponding to the figure, at $n_1 = 3 \times 10^{17}$ cm^{-3}, r = 0.2, and $n_e = 10^{12}$, the value $w \approx 9.6$ W/cm^3.

To estimate the efficiency η, we can use the formula $\eta = f_p h\nu/E_2$ [8]. Inasmuch as in this case the ratio of the quantum energy at the working transition to the energy of the upper laser level is $h\nu/E_2 = 1$, the value of η is determined entirely by the energy fraction f_p used to excite the upper working level. Numerical estimates seem to show that the only process that can complete to any degree and lead to dissipation of the energy and to heating of the gas is the excitation of nuclear vibrations in the O_2 molecule by electron impact. If it is assumed that the relation $\sigma_V \sim \sigma_1^+$ remains in force also at an electron energy ~2 eV, then the per-unit power dissipated in this channel is approximately equal to the power consumed in excitation of the upper laser level. The per-unit power w_r dissipated when the iodine atoms recombine under the conditions of the given laser is relatively small. Defining it by the formula $w_r \approx E_{diss} k_r n_{21}^2 n_1$, where E_{diss} is the binding energy of the iodine atoms in the I_2 molecule (1.56 eV), and recognizing that at $T_{tr} = 400°K$ the recombination constant is $k_r = 9 \times 10^{-33}$ $cm^6 sec^{-1}$ [10], we obtain a value $w_r = 0.87$ W/cm^3 in the case when $n_1 = 3 \times 10^{17}$ cm^{-2}, r = 0.2, $n_2 = 6 \times 10^{16}$ cm^{-3}, and $T_{tr} = 400°K$.

According to our estimates, other possible channels that lead to quasistationary dissipation of the power input to the discharge (the excitation of other electronic states, electron-ion and ion-ion recombination, and other processes) are also relatively inessential. It is difficult as yet to identify the reasons why the efficiency of the given laser should drop to values lower than 30—50%, if no account is taken of the previously considered general conditions that determine its realizability. It must be emphasized, in particular, that the realizability of the proposed laser depends to a great degree on the possibility of obtaining a discharge with a plasma in which the iodine molecules and oxygen atoms have sufficiently low concentrations. Nor is the danger of quenching by the O_3 and IO molecules, which are quite possibly produced in the discharge, quite clear.

2. In the first attempt to realize the discussed laser (in the form of a quantum amplifier), which was undertaken by the present author together with S. S. Polikarpov, it turned out that it is difficult to ensure simultaneous maintenance of the optimal values of n_e and T_e under the conditions of an autonomous discharge through an ordinary pulsed gas-discharge tube for the time necessary to accumulate sufficiently high concentrations of the metastable molecules $O_2(^1\Delta_g)$ and atoms $I(^2P_{1/2})$. (Just as for n_e, it is possible to determine for T_e an optimal value, apparently lying in the region 1.5—2.5 eV, above which the danger of oxygen-molecule dissociation increases.) The experiments have shown that in a mixture of oxygen and iodine vapor (0.2—1.0 Torr), at initial $O_2 : I_2$ partial-pressure ratios from 6 : 1 to 45 : 1 and with different amounts of argon added (from 0 to 50 Torr), the breakdown values of the parameter $\mathscr{E}/p$ ($\mathscr{E}$ is the electric field intensity and p is the gas pressure) under the conditions of an ordinary pulsed discharge tube are too large (~36 $V\text{-}cm^{-1}Torr^{-1}$ if no argon is added) and correspond to initial values of T_e exceeding the optimal value. Depending on the capacitance discharged through the tube with the gas, T_e passes more or less rapidly through the optimal value and then drops to values on the order of T_{tr}.

A signal from a sounding beam (λ = 1315 nm) passing through a cylindrical discharge region 16 mm in diameter and 50 cm long, in the case, say, of the discharge

of a 1-μF capacitor charged to 30 kV through a mixture of oxygen (10 Torr) and iodine (0.3 Torr) (maximum current 250 A, current-pulse duration ~150 μsec), was used to observe the following phenomena:

1) During the first 100 μsec—a sharply pronounced attenuation of the beam, dependent on the receiver aperture and independent of the type of gas, having a relatively small value (20—30%) at the maximum aperture;

2) During the next 100—150 μsec—almost complete (with accuracy ±5—10%) induced transparency of the medium;

3) During the following 600—800 μsec (the subsequent behavior of the signal from the sounding beam was not investigated)—a gradual (over ~200 μsec) attenuation of the signal to a practically stationary level that depended on the initial iodine concentration in the mixture and corresponded to the calculated absorption of the beam by the iodine atoms in the case of complete dissociation of the iodine molecules into atoms.

The sounding-beam source was a CF_3I photodissociation laser with a semiconcentric resonator and a square-wave pump-radiation pulse, ensuring a spikeless lasing regime and an approximately constant intensity of the sounding beam for 1.2 μsec. In control experiments performed on a mixture of iodine vapor with argon (without oxygen), the observed picture differed from the preceding one only in that the second phenomenon of the described sequence was missing. These results seem to offer unequivocal evidence that: a) a radial inhomogeneity sets in during the initial stage of the discharge and leads to scattering of the sounding-beam photons; b) complete or almost complete dissociation of the iodine molecules is attained in the discharge; c) additional population of the metastable level $^2P_{1/2}$ of the iodine atom is produced by the presence of oxygen in the discharge.

An attempt to obtain lasing under the described conditions, using a high-Q resonator, was unsuccessful.

The negative results obtained in these experiments with autonomous discharge do not appear to mean that the gas-discharge plasma parameters required to trigger this type of laser are not attainable. Much more independent are the parameters n_e and T_e under the conditions of non-autonomous discharge, which has recently come into extensive use for pumping high-pressure gas lasers with transverse arrangement of the electrodes. This type of discharge, using, for example, spark (photoionization) preionization, seems to us to be the most suitable for obtaining a sufficiently high electron density (10^{12}—10^{13} cm^{-3}) at a relatively moderate electron temperature (1.5—2.5 eV) in gases where the breakdown is determined by the trapping of the electrons. This is precisely what is required in our case.

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