

$$I(c) \geq \sum_{n=-1}^{-\infty} \kappa_n c_n^2 - \lambda \sum_{n=-1}^{-\infty} c_n^2, \quad (\text{A. 3})$$

a sufficient condition for this is the compactness of the set of sequences that are such that $\sum \kappa_n c_n^2 < A$. The latter assertion can easily be verified with the aid of the criterion for compactness^[8] if we take into account the monotonic increase of the coefficients κ_n .

Similar arguments enable us to prove the existence of bound states in the case of two delta-wells.

Notice that the analog of the considered situation in the case of differential equations is the assertion that the spectrum of systems with a potential that increases without restriction is discrete.^[9]

¹⁾It is convenient to employ the following procedure in practical computations. At sufficiently large $|n|$, we can use the asymptotic form (A.1) for the recurrence relations. Then, with the aid of the recurrence relations, we successively find the coefficients c_n , moving from the two sides (positive and negative n) right up to $n = n_0$, where we carry out a "matching" with allowance for the incident wave. After this a re-

verse run over the recurrence relations gives all the amplitudes F_{n_0-n} .

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Translated by A. K. Agyei.

Investigation of the possibility of designing a recombination gasdynamic O₂ laser

A. Yu. Volkov, A. I. Demin, E. M. Kudryavtsev, and N. N. Sobolev

P. N. Lebedev Physics Institute, USSR Academy of Sciences, Moscow
(Submitted August 15, 1975)
Zh. Eksp. Teor. Fiz. **70**, 503-510 (February 1976)

An experimental and theoretical study was made of the possibility of designing a recombination gasdynamic oxygen laser operating in the visible range, as suggested by A. S. Biryukov et al. [*Zh. Eksp. Teor. Fiz.* **67**, 2064 (1974), [*Sov. Phys.-JETP* **40**, 1025 (1975)]]. The negative results obtained in the search for laser action with the aid of a shock tube indicated that the gain of transitions in the first positive system of the Herzberg bands was less than 10^{-3} cm^{-1} . The reported calculations indicated that although the population inversion in these transitions could reach values of 10^{18} cm^{-3} , the gain should not exceed $2 \times 10^{-6} \text{ cm}^{-1}$, which was in agreement with the results of an experimental check.

PACS numbers: 42.60.Cz

1. A recently published paper^[1] is concerned with one of the possibilities of solving the pressing problem of development of chemical gas lasers emitting in the visible range. The main idea behind these lasers is the utilization of various chemiluminescence reactions as a result of which molecules are formed in excited electronic states. However, in spite of the fact that there is a large number of such reactions with a considerable quantum efficiency (see, for example^[2,3]), nobody has yet been able to build a chemical laser emitting in the visible range.

Fairly attractive chemical reactions which can be used in such a laser are those involving recombination reactions producing molecules with a small number of atoms. The simplest reactions of this type are those

involving three-particle recombination of atoms producing diatomic molecules. A specific example is



where $\text{O}_2(i)$ is an oxygen molecule in an i -th bound electronic state correlated with the ground state of 3P of the oxygen atom. Six such states are known: $X^3\Sigma_g^-$, $a^1\Delta_g$, $b^1\Sigma_g^+$, $C^3\Delta_u$, $A^3\Sigma_u^+$, $c^1\Sigma_u^-$ (see Fig. 1 in Ref. 1); we shall denote these by $i=1, \dots, 6$, respectively. The states can be divided arbitrarily into two groups. The minima of the potential curves of the states $i=4, 5, 6$, located higher than those in the other group ($i=1, 2, 3$), are shifted toward greater internuclear distances.

It is suggested in^[1] that the reaction (1) be used in

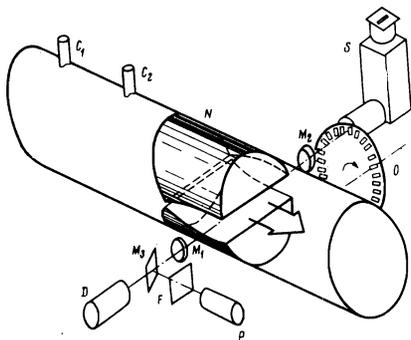


FIG. 1. Block diagram of the apparatus. Here, C_1 and C_2 are ionization probes for measuring the shock-wave velocity; N is the nozzle; the rest of the apparatus is explained in text.

the process of flow of dissociated oxygen from a nozzle in a laser based on vibronic transitions between the upper and lower groups of states. The above-mentioned shift of the potential curves makes it more likely that vibronic transitions occur from the lower most densely populated vibrational levels of the group of states $i=4, 5, 6$ to higher states and, therefore, there should be a strong deactivation of the vibrational levels of the group $i=1, 2, 3$.

An estimate of the gain α was obtained in^[1] for one of these vibronic transitions, which gives rise to the first system of the Herzberg bands $A^3\Sigma_u^+(v'=0) \rightarrow X^3\Sigma_g^-(v''=10)$ when the initial gas temperature is 4000°K and the initial pressure is 10 atm. This estimate gave $\alpha = 10^{-4} \text{ cm}^{-1}$ and it was pointed out that α could rise by an order of magnitude by increasing the initial pressure to 30–50 atm.

The value $\alpha = 10^{-3} \text{ cm}^{-1}$ obtained from these estimates would seem to be sufficient to achieve stimulated emission in an optical path of $\sim 10 \text{ cm}$. This path is compatible with the above initial parameters of the gas (4000°K and 30–50 atm) and it should be possible to establish this path in shock-tube experiments.

The present study^[4] was undertaken to check experimentally the possibility of attaining laser action predicted in^[1] and also to obtain more accurate estimates of the gain and to find the optimal experimental conditions.

2. The experiments were carried out using a shock tube with an internal diameter of 90 mm; the apparatus used is shown schematically in Fig. 1. The use of two- and three-diaphragm shock tubes, of hydrogen as the driver gas, and (in some cases) of reflection from a corner, which acted as a converging guide for the nozzle, made it possible to establish the required initial parameters of the gas ($3800^\circ\text{K} < T_5 \lesssim 4500^\circ\text{K}$, 10 atm $< p_5 \lesssim 40 \text{ atm}$).

A photomultiplier P with a set of filters F recorded the radiation in the range of wavelengths of interest to us ($5000 \pm 200 \text{ \AA}$). This photomultiplier was used to determine the $x-t$ diagram of the reflection of a shock wave: the time before the arrival of perturbations at the end, approximately equal to the steady-flow time, was about $\lesssim 100\text{--}200 \text{ \mu sec}$.

A spectrograph S (with time scanning provided by a disk O) was used in recording the emission spectra of the gas in the working part of the nozzle over the wavelength range $4000\text{--}6500 \text{ \AA}$. No molecular oxygen bands were observed in the emission; the main impurity spectra were the D sodium line and the AlO bands. The presence of these impurities complicates realization of laser systems of this kind when the temperature of the working mixture is high and the gain is small.

The spontaneous radiation power was measured with a detector D and this provided an additional control.

Mirrors M_1 and M_2 had multilayer dielectric coatings with a transmission of $\sim 0.2\%$ in the region of 4800 \AA and they acted as filters in recording of the spontaneous radiation. They replaced the tube windows in experiments in which attempts were made to detect laser action; therefore, these unique mirrors had to be replaced after every such experiment. The mirrors were aligned with an auxiliary He–Ne laser (they were transparent to the radiation of this laser).

We carried out five experiments attempting to detect laser action. In these experiments all three recording methods were used simultaneously and they all gave a negative result.

In view of the fact that the losses in each mirror did not exceed $\sim 0.5\%$, we concluded that the gain was $\alpha \lesssim 10^{-3} \text{ cm}^{-1}$.

3. We carried out a detailed calculation dealing with the processes occurring in partially dissociated oxygen in the course of its passage through a nozzle ahead and past the nozzle throat. This was done in order to find the reasons for the failure to achieve laser action when it was expected on the basis of the estimates given in^[1] and to establish optimal conditions for subsequent experiments. We considered chemical dissociation reactions and three-particle recombination making allowance for the heat evolved in these reactions.

The population and gain were found for the above-mentioned transitions on the following four assumptions.

1) The probability of formation of an oxygen molecule as a result of the three-particle recombination was assumed to be the same for any one of the six coupled electronic states, correlated with the ground state of the oxygen atom.

2) The rotational and vibrational degrees of freedom of all the electronic states of the oxygen molecules were taken to be in equilibrium with the translational degrees of freedom.

3) The three higher electronic states $C^3\Delta_u$, $A^3\Sigma_u^+$, $c^1\Sigma_u^-$ were assumed to be in equilibrium with each other because of the proximity and intersection of the electron terms of these states.

4) The rate of deactivation of the upper electronic states was ignored in the kinetics of the population of the upper active levels.

Clearly, the assumptions made above should give us the upper limits of the values of the population inver-

sion and gain.

We used the following system of equations:

$$\rho v f = \rho^* v^*, \quad \rho v \frac{dv}{dx} = -\frac{dp}{dx}, \quad \mathcal{H} + \frac{1}{2} v^2 = \text{const}, \quad (2)$$

which describes a quasiuniform flow of a gas through a nozzle. Here, ρ is the gas density; v is the velocity of the gas stream (ρ^* and v^* are the corresponding values in the throat part of the nozzle); p is the pressure in the gas mixture; $f = S(x)/S^*$, where $S(x)$ and S^* are the cross-sectional areas at a point x on the axis of the nozzle and in its throat part; \mathcal{H} is the enthalpy of a unit mass in the gas mixture. The system of gasdynamic equations (2) was solved together with the chemical kinetics equation describing the chemical composition of the gas:

$$\frac{d\kappa_o}{dt} = (2 - \kappa_o) N [\kappa_o (K_{d,1}\kappa_o + K_{d,2}\kappa_o) - N\kappa_o^2 (K_{r,1}\kappa_o + K_{r,2}\kappa_o)], \quad (3)$$

where κ_o is the degree of dissociation of oxygen, equal to the ratio of the partial pressure of the atomic oxygen to the total pressure in the mixture; $\kappa_o + \kappa_{O_2} = 1$; N is the total number of particles per unit volume [$N = \rho N_0 / 16(2 - \kappa_o)$, where N_0 is the Avogadro number]; $K_{d,1}$, $K_{d,2}$, $K_{r,1}$, and $K_{r,2}$ are, respectively, the rate constants of the dissociation and recombination reactions. The indices 1 and 2 mean that the third particle M [see Eq. (1)] is either an atom or a molecule of oxygen. We assumed the following rate constants in our calculations:^[5]

$$K_{d,1} = 1.5 \cdot 10^{-4} \cdot T^{-1} \exp(-59400/T) \text{ [cm}^3/\text{sec]},$$

$$K_{d,2} = 18 \cdot T^{-2.5} \exp(-59400/T) \text{ [cm}^3/\text{sec]},$$

$$K_{r,1} = 1.3 \cdot 10^{-29}/T \text{ [cm}^6/\text{sec]},$$

$$K_{r,2} = 2.7 \cdot 10^{-32} \cdot T^{-0.44} \text{ [cm}^6/\text{sec]},$$

where T is the temperature of the gas mixture.

The gain of a rovibronic transition was calculated from the formula

$$\alpha(\nu) = \frac{\lambda^2}{8\pi} A_{ik} g_k \left(\frac{N_k}{g_k} - \frac{N_i}{g_i} \right) F(\nu - \nu_0), \quad (4)$$

where λ is the wavelength of the transition; A_{ik} is the Einstein coefficient of the probability of a spontaneous transition; N_i , g_i and N_k , g_k are—respectively—the populations and degrees of degeneracy of the upper and lower active levels; $F(\nu)$ is the line profile factor. The Einstein coefficient (see, for example,^[6]) is

$$A_{j',j''} = \frac{64\pi^4}{9h\lambda^3} S_{j',j''}^{nm} q_{v',v''} S_{j',j''} / (2J'+1),$$

where $S_{j',j''}^{nm}$ is the strength of an electronic transition; $q_{v',v''}$ is the Franck-Condon factor; $S_{j',j''}$ is the Hönl-London factor, which satisfies the sum rule

$$\sum_{j''} S_{j',j''} = 3(2J'+1),$$

which is due to the fact that in our case the levels are

degenerate only in respect of the magnetic quantum number.

In the case of transitions satisfying the selection rules $\Delta J = 0$, $\Delta K = 0$, and $J = K \mp 1$ (here, K is the rotational quantum number and J is the quantum number of the total angular momentum), the expression for the maximum value of $S_{j',j''}$ is^[7]

$$S_{j',j''} = 3J'(J'+1)/(2J'+1).$$

Using the results in^[8,9], we find that in the case of the $v' = 0$, $v'' = 10$ transition,

$$A_{j',j''} = 0.505 \frac{3(J'+1)J'}{(2J'+1)^2} \text{ [sec}^{-1}\text{]}.$$

The profile factor for the central part of the line is

$$F(\nu_0) = \frac{2}{\Delta\nu_D} \sqrt{\frac{\ln 2}{\pi}} \frac{a}{\pi} \int_{-\infty}^{\infty} \frac{\exp(-z^2) dz}{a^2 + z^2}, \quad (5)$$

where $a = \Delta\nu_L \sqrt{\ln 2} / \Delta\nu_D$. Here, $\Delta\nu_L = \lambda c p (300/T)^{1/2}$ and $\Delta\nu_D = (RT \ln 2)^{1/2} / 2\lambda$ (sec⁻¹) are, respectively, the Doppler and Lorentz widths at line midamplitude; c is the velocity of light. Using an estimate obtained in^[9], we find that $\delta = 0.25$ cm⁻¹ · atm⁻¹. Possible overlap of neighboring lines is ignored in Eq. (5).

We shall assume a vibrational-rotational equilibrium within electronic states so that the population of vibrational-rotational levels can be found from the expression

$$N_j(\nu, J) = N_{j, v, J} \exp(-E_j(\nu, J)/kT) / Z_j.$$

Here, N_j is the total population of a j -th electronic state; Z_j is the partition function of the rotational and vibrational states; $E_j(\nu, J)$ is the energy of a j -th rovibronic level. The values of $E_j(\nu, J)$ were found by applying the Schlapp scheme^[10] to the case intermediate between Hund cases (a) and (b). The constants used in the calculation of $E_j(\nu, J)$ were taken from^[10].

The differential equation system (2) was solved in conjunction with (3) by the Runge-Kutta method on a M-220 computer.

Figure 2 gives the results of calculations for a wedge-shaped nozzle with an expansion angle $\theta = 22^\circ$, slit with $h_0 = 2$ mm, and initial parameters $T = 4000^\circ\text{K}$ and $p = 30$ atm. The abscissa gives the distance x along the nozzle axis. We can see that ahead of the nozzle the degree of dissociation is $\kappa_o = 0.27$, whereas 2.5 cm past the throat section the chemical composition is almost completely "frozen" with $\kappa_o = 0.12$. The maximum population of the upper electronic states ΔN is reached in the nozzle throat and is of the order of 10^{18} cm⁻³. This population of the upper electronic states corresponds to a population of 10^{13} cm⁻³ of the rotational levels involved. The initial rise of α is due to the continuing, after passing through the throat section, population of the upper levels because of recombination, reduction in the vibrational and rotational parti-

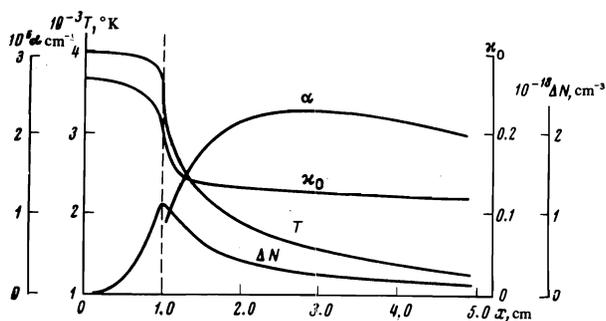


FIG. 2. Results of one of the variants of the calculation of the optimal (in respect of J) gain α , population inversion ΔN , and gas parameters (temperature T and degree of dissociation of oxygen x_0) in the case of flow of dissociated oxygen through a nozzle.

tion functions due to the fall of the translational temperature, and independence of the gain of the pressure in this region. The gain maximum corresponds to the condition when the Doppler line width becomes comparable with the Lorentz width.

Table 1 gives the maximum, along the x axis, values of the gain calculated for the same nozzle in a wide range of the initial parameters of the gas. We can see that when the initial pressure is increased from 10 to 50 atm, the gain increases approximately by a factor of 3 (at $T=4000^\circ\text{K}$, the value of α rises from 0.95×10^{-6} to $2.9 \times 10^{-6} \text{ cm}^{-1}$). These values of the gain are clearly insufficient for laser action and are much smaller than the estimate obtained in^[1].

Our calculations indicate that the inversion and gain depend weakly on the shape of the nozzle ahead of the throat section and are governed mainly by the shape of the diverging part of the nozzle. This is due to the fact that it is this part that governs the degree of dissociation of oxygen at which recombination reaction "freezes" and this governs the population of the upper active levels.

The results of our calculation clearly depend not only on the shape of the nozzle but also on the constants of the elementary processes (the probability of formation of O_2 in an excited electronic state by three-particle recombination, and rates of recombination, dissociation, and quenching of excited states), which are either unknown completely in the case of oxygen or are known

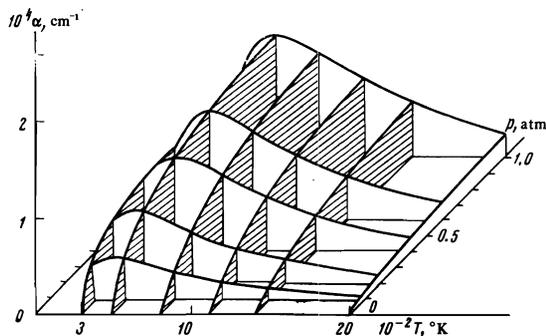


FIG. 3. Dependences of the maximum attainable gain on pressure and temperature.

TABLE 1. Calculated maximum (along x axis) values of gain $\alpha \times 10^6$ (cm^{-1}).

p , atm	T , $^\circ\text{K}$					
	3600	3700	3800	3900	4000	4200
10	0.66	0.71	0.80	0.88	0.95	1.2
30	1.3	1.6	1.8	2.0	2.3	2.7
50	1.8	1.9	2.3	2.6	2.9	3.4

very inaccurately.

We decided to obtain results independent of these constants by estimating the maximum gain for the $A^3\Sigma - X^3\Sigma$ transition in the O_2 molecule. The calculations were made on the assumption that, for given values of the pressure and temperature, all the oxygen molecules were at upper electronic states $i=4, 5, 6$, which were in equilibrium with one another. The results of these estimates are plotted in Fig. 3. We can see that the maximum attainable gain is $\sim 2 \times 10^{-4} \text{ cm}^{-1}$ and it corresponds to 500°K and $p > 0.5 \text{ atm}$. Even this value of the gain α is too low that it would be very difficult to achieve laser action. The value of α which follows from the above detailed calculation is, even under the optimal conditions, a hundred times smaller. This means that the proposed oxygen recombination laser is far from realistic.

4. We shall conclude by making the following point. A highly attractive feature of the proposed visible laser is that very high populations of the upper electronic active levels, $\sim 10^{18} \text{ cm}^{-3}$, are achieved in the working part of the nozzle (this is true to the extent that our assumptions are justified). On the other hand, our analysis shows that the gain is very low ($\sim 10^{-6} \text{ cm}^{-1}$), which is a direct consequence of the low probability of radiative transitions of the vibronic oxygen bands under consideration. Hence, it follows that in considering other possible lasers of this kind it is necessary to seek, on the one hand, ways of releasing the energy stored in metastable levels (for example, with the aid of stimulated emission from an impurity gas^[11]) and, on the other, to find suitable active molecules with laser transitions which are more strongly allowed.

The progress in the development of chemical lasers emitting in the visible range can be accelerated by determination of the constants of the elementary processes governing the populations of excited electronic states of the products of reactions accompanied by chemiluminescence.

The authors are grateful to A. M. Prokhorov for his encouragement, to A. S. Biryukov and L. A. Shelepin for valuable discussions, to V. L. Bukhtiyarov, Yu. V. Chicheryukin, and N. A. Shubina for their help in the experiments, and to V. F. Kitaeva and I. L. Chisty for supplying high-quality mirrors.

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Translated by A. Tybulewicz

Photon statistics for the synchrotron radiation from electrons in a storage ring

I. S. Guk, N. N. Naugol'nyi, and A. S. Tarasenko

Physiotechnical Institute, Ukrainian Academy of Sciences

(Submitted August 22, 1975)

Zh. Eksp. Teor. Fiz. 70, 511-520 (February 1976)

Photon statistics have been investigated for the synchrotron radiation emitted by a bunch of relativistic electrons. Expressions are obtained for the variance of the photon number and the field density operator. The experimental method is described and experimental data confirming the theoretical analysis are reproduced.

PACS numbers: 29.20.Dh

1. INTRODUCTION

The solution of a series of new applied problems has recently led to a much greater interest in the statistical properties of electromagnetic fields of different origin. The theory of coherence, developed by many authors, has led not only to a successful description, but also to the prediction, of a number of new properties for the two main classical sources, namely, thermal and coherent sources.^[1] There is considerable interest in the statistics of synchrotron-radiation photons emitted by a bunch of relativistic charged particles because a source of this kind cannot be assigned to either of the above two classes. Moreover, detailed studies of the properties of synchrotron radiation are desirable in view of the increasingly extensive application of this radiation as a practical tool in physics generally.^[2]

2. PHOTON STATISTICS AND THE FIELD-DENSITY OPERATOR FOR SYNCHROTRON RADIATION

Since synchrotron radiation losses per orbit in the storage ring of the Physico-technical Institute of the Ukrainian Academy of Sciences^[3] are negligible in comparison with the energy of the radiating electron ($E = 100$ MeV), the recoil experienced by the electron can be neglected in the calculation of the synchrotron-radiation parameters. In this approximation, a relativistic

electron can be regarded as a classical current, and Glauber has shown^[4] that its radiation field is in a coherent state and is described by a set of complex amplitudes $\{\alpha_{1l}\}$.

The emission probability amplitude $\alpha_{1l}(t)$ for the l -th harmonic of orbital frequency ω and a charge e moving on a circular orbit of radius R is given by^[5]

$$\alpha_{1l}(t) = \frac{\pi^{1/2} e v \epsilon_{\perp}}{(2\omega l \hbar)^{1/2}} \sum_{p=-\infty}^{\infty} \frac{1}{p\omega + \omega_l} \exp \left[i \left(\frac{3}{2} \pi (p-1) - p\varphi_l + p\psi \right) \right] \times J(p) \exp[i(p\omega + \omega_l)t], \quad (1)$$

where ϵ_{\perp} is the projection of the polarization vector onto the plane of the orbit, $\varphi_l = \arctan(k_y/k_x)$, ψ is the initial phase of the particle, $\omega_l \approx \omega l$, $v = \omega R$,

$$J(p) = J_{p-1}(Rk_{\perp}) \exp[i(\varphi_l + \Delta_l)] + J_{p+1}(Rk_{\perp}) \exp[-i(\varphi_l + \Delta_l)];$$

J_p is the Bessel function, $\Delta l = \arctan(\epsilon_x^*/\epsilon_y^*)$, and k_{\perp} is the projection of the wave vector onto the plane of the orbit.

Consider the radiation emitted by a set of N noninteracting charges. For this ensemble,

$$\alpha_{1N}(t) = \sum_{v=1}^N \alpha_{1v}(t). \quad (2)$$