# Gas lasers with solar excitation

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# 1. INTRODUCTION

In this review we present the results of a study of the possibility of direct conversion of solar radiation into laser radiation in molecular gases, i.e., the development of gas lasers with solar excitation, and we also consider natural laser effects in the atmospheres of planets illuminated by the sun.

The possibility of making lasers based on halogen-containing gases with solar excitation was first reported in 1976 at a seminar of the Department of Wave Processes of Moscow University, directed by Rem Viktorovich Khokhlov (see Ref. 1).

Among the wide range of scientific interests of R. V. Khokhlov, an important place was held by the investigation of nontraditional means of excitation of the active media of gas lasers, the development of high-power laser systems, and the search for new ways of controlling physico-chemical processes with the use of laser radiation.<sup>2-18</sup> R. V. Khokhlov and his coworkers carried out work on the analysis of new schemes for chemical lasers<sup>3,4</sup> and gasdynamic lasers based on the detonation products of solid materials,<sup>5</sup> and on the use of "hot" atoms to produce population inversion in gaseous media.<sup>6,7</sup> These workers also investigated the then-new "radiation-collisional" class of processes for producing molecular lasers.<sup>8</sup> R. V. Khokhlov was one of the founders of the new field-the initiation and control of chemical processes in molecular gases<sup>9</sup> and on surfaces<sup>10,11</sup> with the use of laser radiation. At the Moscow State University nonlinear optics laboratory, headed by R. V. Khokhlov, investigations were carried out on the interaction of powerful laser radiation with resonance absorbing gases and the propagation of the radiation in these media, on the cooling of molecular gases by laser radiation,<sup>12,13</sup> vibrational relaxation in strong laser fields,<sup>14</sup> the dissociation of polyatomic molecules, the shift of chemical equilibrium and the separation of isotopes in strong infrared fields,<sup>15-17</sup> and the propagation of ultraviolet light in the ozone sphere of the Earth.<sup>18</sup>

One of the nontraditional ways of exciting laser media, that was of interest to R. V. Khokhlov, is the pumping of gaseous media by solar radiation. This review is devoted primarily to an exposition of this topic.

Progress in the field of laser physics and technology, as well as the problems of the energy resources of the Earth and the development of ecologically clean energy sources, generate interest in the study of the possibility of direct conversion of solar radiation into the energy of laser radiation. The creation of powerful solar lasers would contribute greatly to such important areas of technology as energy-consuming laser chemistry, laser strengthening of materials, ultrapure high-temperature melting, and many others. High-power solar lasers would also be of great importance in power engineering. For instance, at the present time plans are being made to create powerful electric power stations in outer space, operating on solar batteries. The transmission to Earth for its subsequent utilization of energy, generated by photoelements, is intended to be accomplished with the use of microwave generators.<sup>19</sup> The use of a solar laser in these plans would confer a number of advantages associated with the high concentration of the energy in a laser beam and the small divergence of the beam. When such a beam is received on Earth, it can be converted into various forms of energy, including mechanical energy.20

# 2. GENERAL REQUIREMENTS FOR LASER MEDIA USING SOLAR EXCITATION

The first lasers with direct solar pumping, and the only ones achieved experimentally to the present time, have been solid state lasers<sup>21–26</sup> based on CaF<sub>2</sub>:Dy<sup>2+</sup> and YAG:Nd<sup>3+</sup> crystals. With these media generation was obtained at power up to 18 W.<sup>26</sup> However, further increase in the power of solid state solar lasers and the use of them in solar energy applications has met with a number of difficulties due to the following causes:

a) the necessity of using a high degree of solar light concentration ( $\xi > 10^3$ ) and consequently, the use of complicated solar light concentrators. This is because of the high threshold pumping power required to produce population inversion of the energy levels.

b) the necessity of operating at low media temperatures (in most cases, at cryogenic temperatures $^{21-24}$ ).

c) the difficulty of using large active medium volumes.d) the low optical and thermal strength of the crystals.

For the purpose of obtaining cw generation at substantial power levels ( $\sim 1 \text{ kW}$  and greater) under relatively simple conditions, the use of gaseous active media in solar lasers holds a great deal of promise. They have in principle a number of advantages over solid state media: They require lower pump power thresholds (because of the long energy level lifetimes and the small linewidths of the radiation), they permit the use of large volumes of the active media, which is important in order to achieve high generation power, and they allow operation at moderate temperatures (around room temperature and even higher). Liquid laser media also have some of these advantages. A comparative analysis of the potentialities of solid state and gas solar lasers has been carried out in Ref. 27.

All the above-mentioned considerations allow us to look forward to obtaining in the future cw generation at substantial power with the use of relatively simple solar energy systems. It is for these reasons that definite expectations for creating high-power energy and technological systems in space are coupled to plans for developing gas lasers with solar excitation, that is, the transformation of solar energy into laser radiation. For instance, in the USA investigations are being actively pursued under the aegis of NASA for the purpose of finding laser media that will be candidates for space gas lasers that utilize solar excitation.<sup>28,29</sup>

It should be noted here that there exist rather simple indirect means of transforming the energy of solar radiation into energy of laser radiation with the use of intermediate elements for transforming solar energy into heat or electrical energy. For example, it is possible to use gas-dynamical lasers in which the working medium is heated by solar radiation prior to the gas-dynamical expansion. Because of the high efficiency ( $\eta \simeq 100\%$ ) of converting solar energy into heat, this kind of solar laser can provide an efficiency of conversion of solar energy into laser energy equal to the efficiency of gas-dynamical lasers, i.e., around a few percent. At present this scheme has been realized experimentally<sup>30</sup> by heating nitrogen to 1473 K and then mixing it with CO<sub>2</sub>. Generated power of ~ 1.5 W was attained on the lasing transition of  $CO_2$  (00°1  $\rightarrow$  10°0) with an efficiency of ~0.7%. It is also possible to use solar batteries, which can convert solar energy to electrical energy with an efficiency of ~10%, and then convert this energy into laser radiation with the use of electric-discharge  $CO_2$  or CO lasers with an efficiency of 10% or 30%, respectively.

However, a more attractive option is to develop a type of solar laser in which it would not be necessary to convert the energy of the solar radiation into any form other than that of coherent laser radiation. An analysis of the possibilities of developing such schemes is the topic of the rest of this article.

We shall now formulate the general requirements for gases that would be candidates for the working media of lasers for the direct conversion of solar radiation. These requirements are the following:

a) a broad (and as continuous as possible) absorption band in the region of the spectrum where the main part of the pumping radiation is concentrated;

b) the existence of processes which, upon absorption of radiation, produce a level population inversion;

c) regeneration of the chemical composition of the laser mixture, in which various chemical reactions may occur under the action of the radiation.

If the pumping of the active medium is done directly with concentrated solar light, then these requirements must be satisfied, obviously, for systems that absorb radiation predominantly in the visible region of the spectrum. However, it is possible in principle to create a solar laser in which the pumping radiation that is used has been transformed from the visible to the infrared by means of a black body (see Section 4, below). In this case the requirements enumerated above apply to gases that absorb infrared radiation.

For optical pumping of gases by concentrated solar radiation, these requirements are satisfied to various degrees by halogen molecules  $(Br_2, I_2, Cl_2)$ , interhalogen and halogen-containing molecules (IBr, C<sub>3</sub>F<sub>7</sub>I, etc.) and molecules of alkali metals (Na<sub>2</sub>, Cs<sub>2</sub>, Rb<sub>2</sub>). We note here that these gases have previoulsy been suggested for use as heat conductors in solar energy systems.<sup>31</sup> For the problem we are considering, however, it is important that the absorption of solar light should lead both to the excitation of electronic states, with subsequent collisional and radiative relaxation and transition back to the initial ground electronic state of the molecule, and to the dissociation of the molecules with the formation of electronically excited atom products. Both these processes can, in principle, produce population inversion. Thus, in the first case, as a result of vibrational-rotational relaxation, the electronically excited molecules can be accumulated in the lower vibrational levels of the electronic term with an equilibrium internuclear spacing that is shifted relative to the internuclear spacing of the lowest term (Fig. 1a). According to the Franck-Condon principle, the most efficient transitions will be the radiative transitions from these lower vibrational levels of the excited term to the upper vibrational levels of the lower term. The latter states, because of rapid collisional relaxation at relatively low gas temperatures, can be only slightly populated, i.e., a population inversion can be attained in the system. To the present

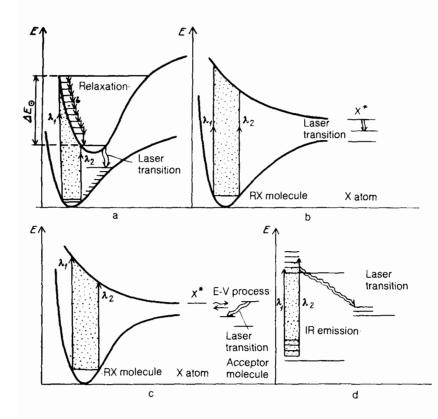


FIG. 1. Excitation diagrams of energy levels in gas lasers with solar pumping.

time, however, an analysis of the conditions for the formation of inversion and calculations of the characteristics of solar lasers of this type have not been carried out.

In the second case, a population inversion can be brought about by either the accumulation of electronically excited atoms formed during photodissociation (of course, without the accumulation of atoms in the lowest state) (Fig. 1b), or the transfer of energy from these electronically excited atoms to other acceptor molecules that have energy levels that are favorable for the formation of a population inversion among them (Fig. 1c).<sup>1)</sup>

In the scheme for a solar laser with pumping by infrared radiation from a black body heated by concentrated solar light, it is of course appropriate to use as the active medium molecules that are optically active in the infrared and that have vibrational-rotational infrared absorption bands. Population inversion in this scheme can be obtained in principle on the various vibrational-rotational levels of the absorbing molecule itself (Fig. 1d) or of an acceptor molecule.

#### 3. LASERS WITH DIRECT EXCITATION BY SOLAR LIGHT

#### 3.1. Basic characteristics of laser media

The important parameters for the choice of the working medium of a gas solar laser are: the wavelength  $\lambda_m$  at the absorption maximum, the absorption band width  $\Delta\lambda$  that determines the efficiency  $\eta$   $\odot$  of the absorption of solar light, the absorption cross section  $\sigma$ , the quantum efficiency  $\eta_q$  that is, the ratio of the laser energy to the energy of the absorbed solar radiation, the possible operating temperature  $T_{\rm op}$  of the active medium, and the lasing wavelength  $\lambda_1$ .

The values of these parameters, taken from the litera-

ture,<sup>27,29-47</sup> for candidate media that have attracted the greatest interest among investigators are shown in Fig. 2 and Table I. From the data presented it can be seen that the halogen molecules I<sub>2</sub> and Br<sub>2</sub> have a definite advantage over molecules of type RI with respect to the efficiency of absorption of solar radiation, but they are inferior to the latter in their quantum efficiency. The reason is that in the use of the halogens, it is proposed that generation be obtained using the acceptor molecules in the middle infrared (according to the diagram of Fig. 1c). In the IBr and RI molecules, however, generation is also possible in principle according to the diagram of Fig. 1b on the electronic transitions of Br( $4^2P_{1/2} \rightarrow 4^2P_{3/2}$ ) and I( $5^2P_{1/2} \rightarrow 5^2P_{3/2}$ ) in the near infrared. By a similar scheme (but with a different mechanism of quenching the lower laser level) it has been suggested that generation can be obtained in a Cs<sub>2</sub>-Xe mixture.

We note that media in which the solar light is absorbed by homonuclear molecules have another important advantage—it is easy to regenerate the initial chemical composition.

Table I also includes estimated data for a  $Na_2$  laser. The development of lasers based on the vapors of the metals  $Na_2$ ,  $K_2$ , and  $Rb_2$  and operating according to the diagram of Fig. 1a are of undoubted interest for the conversion of solar light into laser radiation in the near infrared or the visible region with a high quantum efficiency. Moreover, dimers of the type  $Rb_2$  can also in principle be used in the scheme shown in Fig. 1c. However, the practicability of laser schemes based on vapors of the alkali metals has not yet been investigated. One of the impediments to theoretical modeling of schemes for solar lasers is the scarcity of reliable information on the rate constants of the various physicochemical processes that

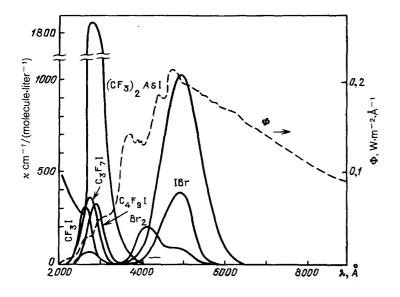


FIG. 2. Absorption coefficient x for various gaseous working media for solar lasers and spectral distribution of the solar radiation.

take place in a mixture of gases with the products of photochemical reactions. This is why only a few specific schemes for solar lasers have been investigated. We present below a short analysis of these schemes.

# 3.2. Photodissociation Br<sub>2</sub>-CO<sub>2</sub> lasers

In this scheme of a gas laser with solar pumping<sup>1</sup> the use of a Br<sub>2</sub>:CO<sub>2</sub>:He gas mixture was suggested. The Br<sub>2</sub> molecules absorb the solar light in the range 3800-5300 Å with a cross section (at the half-maximum of the absorption curve)  $\sigma \approx 3 \cdot 10^{-19}$  cm<sup>2</sup>. About 25% of the energy of the solar radiation lies in this spectral range. Accompanying the absorption is the dissociation of Br<sub>2</sub> molecules, with the formation of the excited atom Br\*(4<sup>2</sup>P<sub>1/2</sub>) with an energy 3685.2 cm<sup>-1</sup>. The collision

$$Br^* (4^2 P_{1/2}) + CO_2 \xrightarrow{K_1} CO^*_a (00v_3) + Br \qquad (3.1)$$

causes the excitation of the asymmetric type of vibrations of the  $CO_2$  molecules, and under certain conditions this can lead to the formation of a population inversion on the traditional laser transition  $CO_2 (00^01) \rightarrow CO_2 (10^00)$ . The chemical stability of the mixture is provided by the regeneration of the molecular bromine in the three-particle bulk recombination reaction  $Br + Br + M \rightarrow Br_2 + M$  and in the heterogeneous recombination reaction, which is controlled by the diffusion of atoms to the walls of the cell.

In addition to (3.1), important channels for the deactivation of Br\* atoms are the collisions

$$\operatorname{Br}^* + \operatorname{CO}_2 \xrightarrow{K_2} \operatorname{Br} + \operatorname{CO}'_2,$$
 (3.2)

$$Br^* + Br_2 \xrightarrow{K_0} Br + Br_2;$$
 (3.3)

where in (3.2) the events of (3.1) are not included, i.e.,  $CO'_2 \neq CO_2(00v_3)$ . Conditions that are adverse for the formation of population inversion are also heating of the gas and deactivation of the upper working level in collisions (here  $M = CO_2$ ,  $Br_2 Br$ , or He)

$$\operatorname{CO}_{2}(001) + \mathrm{M} \xrightarrow{K_{1}^{M}} \operatorname{CO}_{2}(\mathbf{v}_{1}, \mathbf{v}_{2}, 0) + \mathrm{M}.$$
(3.4)

In order to improve the thermal conditions of the mixture and to achieve substantial absorption of the solar light, helium is added to the gas and a rectangular cell is used, with three characteristic dimensions (the smallest dimension is the width l, which determines the outflow of heat to the walls; the depth H allows the absorption of a substantial fraction of the solar radiation in a single pass, and the length L allows for sufficient gain and generation on the working transition).

It has been shown<sup>32</sup> that with this choice of medium, and with such a cell, it is possible to obtain generation with solar pumping with realistic degrees of concentration of the solar radiation. If it is assumed, for instance, that the popula-

TABLE I. Principal parameters of gas lasers with solar pumping.

Working mixture	λ <sub>m</sub> , Å	<b>Δ</b> λ, Å	$\sigma$ , cm <sup>2</sup>	n⊙, %	$\eta_q, \%$	$\eta = \eta_{\odot} \eta_{q},$	T <sub>op</sub>	$\lambda_{gen}$
Br <sub>2</sub> -CO <sub>2</sub>	4889 4141	1500	3.10-19	25	4	1	<b>35</b> 0	9—12 µm
I2-HF	5000	1000	2.10-18	45	2	0.9	300	4.7 μm
IBr	5072 2682	1100	6·10 <sup>-19</sup> 1.3·10 <sup>-19</sup>	13	18.5	2,4	600	2.715 µm
С <sub>3</sub> F <sub>7</sub> I Сs <sub>2</sub> — Хө	2002 2745 5000	270 100	$6 \cdot 10^{-19}$ $10^{-18}$	1,2 0,1	20 0,4	0, <b>24</b> 0, <b>04</b>	650 650	1.315 μm 1.38 μm
Na <sub>2</sub>	4850	150	3.10-14	2	90	1,8	800	5250 A

tion of the lower CO<sub>2</sub> laser level (10<sup>0</sup>0) is close to equilibrium (which is attained by accelerating the relaxation of this level with helium), one can show that for the mixture  $N_{\rm Br_2}:N_{\rm CO_2}:N_{\rm He} = 1:a:b$  inversion occurs under the following conditions

$$\varphi \xi \Phi: N_{\text{Br}_{s}} > a \exp\left(-\frac{E_{100}}{T_{w}}\right): A,$$
  
$$\varphi \xi \Phi: N_{\text{Br}_{s}} \ll \mu \nu \left(h \overline{\nu} \sigma \Delta \lambda \cdot l^{2} \frac{E_{100}}{T_{w}}\right)^{-1}, \qquad (3.5)$$

where  $\Phi$  is the photon flux of solar radiation per angstrom wavelength,  $\xi$  is the degree of concentration of the radiation,  $\varphi$  is the number of passes of the radiation through the cell,  $E_{100}$  is the energy of the CO<sub>2</sub>(10°0) level,  $T_{\rm W}$  is the temperature of the cell walls,  $\mu$  is the thermal conductivity of the gas, and  $h\bar{\nu}$  is the average photon energy of the absorbed solar radiation, and

$$A = K_{4}\sigma\Delta\lambda \left[a \left(K_{4} + K_{2}\right) + K_{3}\right] \left(K_{4}^{CO} + \frac{b}{a} K_{4}^{He}\right)^{-1}.$$

The second inequality of (3.5) expresses the condition that the gas is not heated very much.

A detailed numerical analysis of this scheme has been carried out in Refs. 32-34. In these investigations the output power P, the gain  $\varkappa_i$  and the efficiency  $\eta$  are given as functions of the degree of concentration of the solar radiation, the wall temperature, and the partial composition of the mixture, for rectangular and cylindrical cells. An example of these calculations is shown in Fig. 3. It can be seen that for a coefficient of solar energy concentration,  $\xi \approx 160$  and two passes of this flux ( $\varphi = 2$ ) through a cell with a gas mixture  $Br_2:CO_2:He = 1:2:6$  (the cross section of the rectangular cell is  $30 \times 3$  cm<sup>2</sup>, the total gas pressure is  $\sim 3$  torr, and the wall temperature is  $T_w = 230$  K) it is possible to obtain a

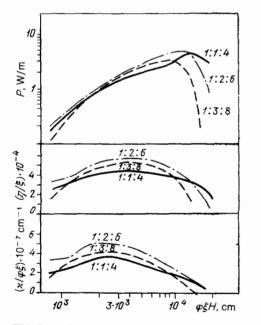


FIG. 3. Specific output power P, relative efficiency  $\eta/\xi$ , and relative gain  $\varkappa/\varphi\xi$  as functions of the parameter  $\varphi\xi H$  for various mixtures of Br<sub>2</sub>:CO<sub>2</sub>:He (total pressure 3 torr) in a rectangular cell of dimension H = 30 cm,  $\delta/H = 0.1$  for a wall temperature  $T_w = 320$  K.

gain of  $\kappa \approx 10^{-4}$  cm<sup>-1</sup>. For a laser length  $L \approx 30$  m, and 98% reflection at the mirrors, the generation power is  $P \approx 5$  W/m and the efficiency is  $\eta \approx 0.1\%$ . With a set of ten such cells 30 m long giving a total cross section  $30 \times 30$  cm<sup>2</sup>, the total output power will be about 1.5 kW, with a gain  $\kappa \approx 30\%$  in a single pass.

The practicability of a  $Br_2$ :CO<sub>2</sub> mixture in cw operation has been demonstrated experimentally<sup>35</sup> in the observation of gain in a probe beam from a CO<sub>2</sub> laser in a  $Br_2$ :CO<sub>2</sub> mixture with lengthwise excitation by the radiation from a cw  $Ar^+$  laser. The experimental conditions reported in Ref. 35 in fact simulated the excitation of the gas by solar radiation with a degree of concentration  $\xi \approx 10^2$ .

## 3.3. Interhalogen molecule lasers

Molecules of the interhalogens, IBr, ICl, etc., also efficiently absorb solar light, with the formation of electronically excited products in the photodissociation process. For this reason, molecules of this type are at present being studied as candidates for the working medium of gas solar lasers.

A laser based on IBr molecules operates according to the following scheme:

$$\odot \rightarrow \operatorname{IBr} \stackrel{\longrightarrow}{\underset{\longrightarrow}{\operatorname{Br}}^{*}} \stackrel{\operatorname{Iaser,}{\underset{\lambda=2,715}{\operatorname{\mum}}}}{\underset{\mu=3.6}{\operatorname{Iaser,}}}.$$
 (3.6)

In the photodissociation of IBr molecules by visible light in the range  $\Delta \lambda = 450-550$  nm, excited bromine atoms Br\* ( ${}^{2}P_{1/2}$ ) are formed with a quantum yield  $\gamma \approx 0.7$ . In this process a population inversion on the transition Br\*( ${}^{2}P_{1/2}$ )  $\rightarrow$  Br( ${}^{2}P_{3/2}$ ) is formed. The lower laser level, which is the Br electronic ground state, is quenched by means of the chemical reaction

$$Br + IBr \to I + Br_2, \tag{3.7}$$

for which the rate constant is  $K = 3.5 \cdot 10^{-11} \text{ cm}^3/\text{s}$ , thirty five times greater than the rate constant of the analogous reaction involving  $\text{Br}^*({}^2P_{1/2})$ .

A detailed analysis of the kinetics of an IBr solar laser has been carried out in Refs. 36–38. In Ref. 38, twenty six photochemical reactions that bear on laser operation were examined and the rate equations for the concentrations of the molecules and atoms IBr,  $I_2$ ,  $Br_2$ , I, Br,  $I^*$ , and  $Br^*$ , and for the lasing power were solved. The calculation showed, for instance, that at a gas pressure of 5 torr and a temperature of 300 K the gain  $\varkappa$  is related to the degree of concentration of the solar radiation by the relation

$$\kappa (\mathrm{cm}^{-1}) \approx 1.3 \cdot 10^{-7} \xi,$$
 (3.8)

Relation (3.8) is valid for  $\xi > \xi_{\text{th}}$ , where  $\xi_{\text{th}}$  is the threshold value of  $\xi$  at which population inversion occurs. According to the calculations,  $\xi_{\text{th}} \approx 800$  and the efficiency of conversion of solar into laser radiation energy is estimated<sup>38</sup> to be

$$\eta = 5 \cdot 10^{-4} \ pH, \tag{3.9}$$

where p is the IBr pressure in torr, and H is the depth of the cell for the absorption of the light. Expression (3.9) is valid for  $pH \leq 50$  torr cm, where the medium is still optically thin

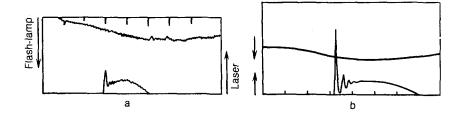


FIG. 4. Time dependence of output power of an IBr laser, obtained in Ref. 39. a) experimental; b) theoretical. Pressure  $p_{1Br} \approx 3 \text{ torr} (+4\% \text{ Br}_2, \text{ I}_2)$ , time scale, 20  $\mu$ s/div.

to the solar radiation, and it assumes a maximum value  $\sim 10^2$  for pH > 50 torr cm.

An IBr laser has been realized experimentally, as reported in Refs. 38 and 39. In these experiments the solar radiation was simulated by that of a xenon lamp at a power corresponding to a degree of concentration  $\xi = (0.5-5) \cdot 10^3$ .

In Ref. 38, for a laser 1 m long with a radius 2.22 cm, at a gas pressure of 4 torr and  $\xi = 5 \cdot 10^3$ , laser pulses were measured with a peak power of 2 kW. The calculation as a matter of fact gave a value of 1.7 kW. A comparison of the experimental and calculated laser pulses is given in Fig. 4. The experimentally determined power and efficiency of the laser, as well as the threshold value of the degree of concentration  $\xi$  for the onset of lasing agrees with the calculated values to within a factor of two.

One of the important factors that limit the operation of an IBr laser is heating of the active medium during its irradiation. This is why the laser action is pulsed, and for experimentally attainable parameters (gas pressure, tube radius, and the degree of concentration) the laser pulse length is  $\sim 40-80 \,\mu s$ . To be sure, as  $\xi$  decreases the heating rate also decreases and the laser pulse length increases. Nevertheless, to obtain cw generation it is necessary to use a gas flow to avoid overheating. A gas flow is also necessary to maintain the required composition of the gas, in which over a period of time I<sub>2</sub> and Br<sub>2</sub> molecules are produced and the concentration of IBr decreases.

### 3.4. lodine lasers

Photodissociation lasers based on the laser transition  $I(5^2 P_{1/2} \rightarrow 5^2 P_{3/2})$  operate according to the scheme shown in Fig. 1b, where the medium that absorbs the solar radiation can be polyatomic molecules RI, where  $R = C_3 F_7$ ,  $(CF_3)_2$ As,  $CF_3$ , or  $C_4F_9$ . The excited iodine atom  $(5^2P_{1/2})$  is formed by photodissociation, and its deactivation occurs mainly in collisions with RI and other reaction products (the radiative lifetime of  $I^*(5^2P_{1/2})$  is long: 0.15 s). The lower laser level is quenched by chemical reactions. The rate constants of these reactions are usually greater than the constants for the deactivation of the upper laser level and therefore in a flow system it is easy to obtain population inversion and the gain can be very large at comparatively small  $(\sim 10^2)$  degrees of concentration  $\xi$  of the solar light. A detailed analysis of the kinetics of the processes in solar lasers based on RI molecules has been carried out in Ref. 40.

A number of publications<sup>29,40–42</sup> have reported experiments which utilize solar simulators to pump a  $C_3F_7I$  laser. The simulator used was a xenon lamp, which has a radiation

spectrum close to that of the sun. With the use of these simulators C<sub>3</sub>F<sub>7</sub>I laser generation was observed for a duration of more than an hour with an output power  $P \approx 1$  W.<sup>42</sup> In an experiment reported in Ref. 40, a laser tube 10 cm long and  $0.7 \,\mathrm{cm}$  in diameter containing  $C_3 F_7 I$  gas was irradiated using a conical reflector which gave a degree of solar light concentration that varied  $(1-1.4) \cdot 10^3$  along the length of the tube. At the optimum gas pressures, 15-25 torr, a generation power ~4 W was obtained at a pulse length ~10  $\mu$ s. The coefficient of conversion of the energy of the simulator lamp radiation into laser energy was  $\eta \approx 0.2\%$ , i.e., close to the limiting value (see Table I). In Ref. 41 a laser cell of length L = 40 cm was used. In this case, with 99% reflectance of the laser mirrors and a C<sub>3</sub>F<sub>7</sub>I gas pressure of 10 torr, a threshold value of  $\xi$  of 150 for the onset of generation was obtained. This means that for this value of  $\xi$  the gain is  $\sim 2.5 \cdot 10^{-4} \text{ cm}^{-1}$ . In Ref. 41, for L = 40 cm and  $\xi = 200$ , a value  $\eta \approx 0.13\%$  was obtained. As we see, the degree of concentration  $\xi$  of the solar radiation is comparatively small, which gives grounds for anticipating that it will be possible in practice to use uncomplicated cylindrical solar concentrators for pumping these lasers. The operating degree of concentration  $\xi$  may be further reduced, evidently, by using, instead of  $C_3F_7I$ , molecules of  $(CF_3)_2AsI$ , which have an absorption cross section that is seven times greater in the same region of the solar spectrum (see Fig. 2).

Besides  $C_3F_7I$  and  $(CF_3)_2AsI$ , the gas  $CF_3I$  can be used as the active medium for a solar laser.<sup>38,45</sup> At a flow rate of this gas of 3 m/s and with excitation from two mercury lamps, laser action was sustained for 83 s at a power of 0.2 W.<sup>45</sup> The time of operation was limited only by the gas supply.

Finally, we should point out another possible medium for a solar laser. In Ref. 30 the use of the  $C_4F_9I$  molecule was suggested; these molecules have an important advantage in that after photodissociation they are regenerated with an efficiency of almost 100%.

#### 3.5. Aikali metal vapor lasers

One of the possible schemes for lasers of this sort, using a  $Cs_2$ -Xe mixture, was suggested in Ref. 46. As in the case of lasers based on RI molecules, pumping of the upper operating level is produced according to the diagram of Fig. 1b via the photodissociation of the  $Cs_2$  dimers, while the principal difference is that the lower operating level is an excited state and is quenched not by a chemical reaction, but rather as a result of radiative or collisional transitions into the ground state.

The  $Cs_2$  molecules dissociate upon the absorption of

solar radiation in the region of 4500 Å. The usable fraction of the solar spectrum is equal to  $\eta \odot \approx 0.009 P$ , where P is the total gas pressure in atmospheres. One of the photodissociation channels can be the formation of excited Cs atoms  $(7^2S_{1/2})$  so that it is possible, in principle, to obtain a population inversion of the  $7^2 S_{1/2}$  and  $6^2 P_{3/2}$  levels if, of course, the  $6^2 P_{3/2}$  level can be quenched. The usual spontaneous radiative transition from this state to the ground state is, however, not effective in quenching this state because of reabsorption of the radiation. Therefore the authors of Ref. 47 suggested the tactic of adding a buffer gas of xenon to "drain off" the  $Cs(6^2P_{3/2})$  in a reaction involving the formation of the exiplex molecule  $Cs(6^2P_{3/2})$ Xe and the subsequent radiative decay of this molecule to the ground states of Cs and Xe. Reabsorption following this radiative transition into the Cs-Xe repulsive term is insignificant because of the broad bandwidth of the radiation. Another mechanism for quenching the  $6^2 P_{3/2}$  level—its deactivation via collisions with Cs atoms in the ground state—has also been analyzed.<sup>47</sup> This deactivation can be efficient because of the nonadiabatic nature of the interaction.

A detailed analysis of the operating conditions of dissociation solar lasers based on mixtures of the type Cs<sub>2</sub>-Xe has been carried out in Refs. 46 and 47 (see also Ref. 27). This analysis showed that there are a number of advantages associated with this type of laser: the possibility in principle of operating at rather high gas temperatures (~650 K), a comparatively high efficiency ( $\sim 3\%$ ), and minimal operating requirements as regards the degree of concentration of the solar radiation ( $\sim$  30). However, the practical implementation of this scheme for the specific mixture  $Cs_2$ -Xe is problematical, since experiments have shown<sup>48</sup> that there is a high degree of correlation of the excited state Cs<sub>2</sub>\* with the lower Cs levels. This means that in the photodissociation of the Cs<sub>2</sub> these levels will be preferentially populated instead of the  $7^2 S_{1/2}$  level, and the formation of a population inversion will be inhibited. Nevertheless, the above-noted advantages of this laser scheme remain an attractive concept, and to realize it a further search for specific new gaseous media is necessary.

Investigations into the possibility of using alkali metal vapors in other schemes for producing population inversion (for example that shown in Fig. 1a and b) are also of great interest. Such investigations have not yet been carried out.

# 4. LASERS WITH THERMAL CONVERSION OF SOLAR PUMPING

#### 4.1. General considerations

The search for new active media for gas solar lasers has led to the idea of using infrared-active molecules excited by radiation from a black body heated to 1000–3000 K by concentrated solar light.<sup>49-55</sup> This heating effect can convert the spectrum of the solar radiation from the visible to the infrared with a high (close to 100%) efficiency. The peak in the spectral density of this radiation is at a wavelength  $\lambda(\mu m) = 5.08 \cdot 10^4/T$ , i.e., for T = 1000-3000 K the peak is in the absorption region of the combination-rotational bands of many molecules that are active in the infrared, including CO and CO<sub>2</sub> molecules, which form the basis of operation of other successfully operating high-power and highly efficient lasers that employ other means of excitation (electrical discharge, for example). In this case the limiting quantum efficiency of such laser systems (that is the ratio of the quantum energy of the laser radiation to that of the absorbed radiation) is very high (~43% for the 10  $\mu$ m CO<sub>2</sub> laser and ~80% for the CO laser, which absorbs energy on the 0 $\rightarrow$ 1 vibrational transition and generates on the 10 $\rightarrow$ 9 transition). This, of course, is a very important advantage over the photodissociation solar lasers considered above that use transfer of excitation (the Br<sub>2</sub>-CO<sub>2</sub> type) (see Table I).

There is, however, one important drawback to the method of optical pumping by infrared radiation. This drawback involves the fact that at moderate pressures the vibrational-rotational absorption bands of many molecules consist of a set of separate, very narrow vibrational-rotational lines. Therefore, the fraction of the energy of the infrared radiation from the black body that is absorbed will, in the general case, be small, and this, naturally, will decrease the efficiency of this type of laser. For example, for the 4.3  $\mu$ m band of CO<sub>2</sub>, which has  $\sim 30$  absorption lines, taking into account their Doppler broadening the fraction of infrared radiation absorbed from a black body at 2000 K is only  $\sim 2.5 \cdot 10^{-5}$ . However, it is possible to cope with this drawback in principle by choosing molecules with a small rotational constant, by using a mixture of isotopes, and by increasing the pressure of the gas (thereby increasing the total number of absorption lines and their widths as a result of pressure broadening). All these considerations suggest that the development of a solar laser with pumping by the radiation from a black body is highly attractive.<sup>2)</sup>

# 4.2. $\text{CO}_2$ laser with excitation in a black body cavity and with gas flow

The solar laser proposed in Ref. 45 is shown schematically in Fig. 5. An extended cavity 1 is coated on the inside with a light-absorbing material (for instance, graphite) and a flux of solar radiation, concentrated by a factor  $10^3-10^4$ enters it through a small hole 2. Because of the use of thermally insulating walls for the cavity, the heat loss through the walls is kept to a minimum, so that it is easy to heat the interior to a temperature  $T_b \approx 1200-3000$  K, depending on

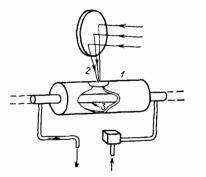


FIG. 5. Conceptual diagram of a solar laser with excitation by infrared radiation in a black body cavity and with gas flow.

TABLE II. Characteristics of active media of solar lasers with pumping by black body radiation.

Ire No.				Gas layer thick-	Black body radiation	Number of iso-	Power ab- sorbed in the	
Mixture	со	CO <sub>2</sub>	Не	ness, cm	tempera- ture, K	topes	center of the layer, W/cm <sup>3</sup>	
1 2 3 4 5 6 7	72 72 10 0 0 760	4 0 10 12 12 0	0 0 0 12 12 12 0	3 10 1 1 1 1 3	2000 2000 2000 2000 2000 3000 2000	6-1 6-1 1 12-1 12-1 12-1 6	$\begin{array}{c} 0,172\\ 0.081\\ 0.0044\\ 0,03\\ 0,268\\ 0.569\\ 1,36\end{array}$	

the degree of concentration  $\xi$  of the solar radiation:

$$T_{\rm b} \approx \left(\frac{P_{\odot}}{\sigma}\xi\right)^{1/4},\tag{4.1}$$

where  $P_{\odot}$  is the solar constant and  $\sigma$  is the Stefan-Boltzmann constant. The equilibrium radiation from the heated walls will pump the laser levels of the molecules of the gas, which, to prevent its heating, is flowed continuously through a tube 3 placed within the cavity. In order to reduce energy loss, the tube is made out of a material with a high transmission in the infrared, such as sapphire, MgO, CsI, or KCl. What should we expect from a solar laser of this construction? Table II, taken from Ref. 51, shows, for various gas mixtures and for different isotopic content, one of the important characteristics of a laser medium—the infrared power absorbed in the center of the tube.

According to the estimates,<sup>51</sup> for example, for case 5 of Table II, 35% of this power can be converted into laser radiation. Simple estimates show that the efficiency of conversion of all the black body radiation energy into laser energy will in this case be  $\sim 0.1\%$ . As we see, the efficiency of this solar laser is very low.

In order to determine more fully the potential of these systems, we have carried out a numerical modeling of the kinetic processes in the active medium of a laser based on a  $CO_2$ -He-Ar mixture with optical pumping of energy into the asymmetric types of  $CO_2$  vibrations by the infrared radiation from a black body. In the modeling we used the vibrational kinetic equation for the  $CO_2$  asymmetric mode and the rate constants of the processes as given in Ref. 58. The effect of diffusion on the vibrational kinetics was taken into account, and the optical pumping was calculated allowing for the finite optical density of the medium in the "radiative removal" approximation.<sup>59,60</sup> In this approximation the probabilities of optical excitation,  $W_3$ , and spontaneous decay,  $A_3$ , of the  $00^01$  level of the  $CO_2$  molecule can be written in the form

$$W_{3} = \beta M(\tau) A_{3}^{\bullet} \frac{\exp(-h\nu_{3}/kT_{b})}{1 - \exp(-h\nu_{3}/kT_{b})},$$

$$A_{3} = M(\tau) A_{3}^{\bullet},$$
(4.2)

where  $\nu_3$  is the frequency of the transition in the middle of the 4.3  $\mu$ m absorption band,  $T_b$  is the radiation temperature of the black body,  $\beta$  is the factor that takes into account the "attenuation" of the radiation at the boundary (for example as a result of the absorption of radiation on passing through the wall of the tube; we have taken  $\beta = 0.85$ ),  $M_3^0 = 2.5 \cdot 10 \text{ s}^{-1}$  is the probability of spontaneous decay of the 00°1 level, and  $M(\tau)$  is a function that, within the framework of the radiative removal approximation that we are using, takes into account the decrease in the probability of optical excitation and of radiative decay of the level on account of the imprisonment of the radiation in a vibrationalrotational band with optical thickness  $\tau$ . The form of the function  $M(\tau)$  depends on the shape of the spectral vibrational-rotational lines. In the case of a Voigt lineshape we may, with an accuracy sufficient for our purposes, assume that

$$M(\tau) = \begin{cases} M_{D}(\tau) + M_{L} \text{ for } M_{D}(\tau) + M_{L}(\tau) < 1, \\ 1 & \text{ for } M_{D}(\tau) + M_{L}(\tau) \ge 1; \end{cases}$$
(4.3)

where  $M_{\rm D}(\tau)$  and  $M_{\rm L}(\tau)$  are the values of the function  $M(\tau)$  for pure Doppler and pure Lorentzian lineshapes, respectively. The quantities  $M_{\rm D}(\tau)$  and  $M_{\rm L}(\tau)$  have been published<sup>59</sup> and their asymptotic expressions for  $\tau \ge 1$  are given in Refs. 61 and 62. To an accuracy no worse than  $\approx 15\%$  the data of Refs. 59 and 61 can be approximated by the following expressions

$$M_{D}(\tau) = \begin{cases} 1 - 0.336\tau & \text{for } \tau \leq 2.5, \\ \frac{0.4}{\tau} & \text{for } \tau > 2.5, \end{cases}$$
(4.4)

$$M_{L}(\tau) = \begin{cases} 1 & \text{for } \tau \leq 0.202a, \\ \left(\frac{0.202a}{\tau}\right)^{1/2} & \text{for } \tau > 0.202a; \end{cases}$$
(4.5)

where *a* is a parameter that is defined by the ratio of the Lorentzian linewidth  $\Delta v_L$  to the Dopper linewidth  $\Delta v_D$  at the middle of the band:  $a = (\Delta v_L / \Delta v_D) (\ln 2)^{1/2}$ . The optical thickness  $\tau$  in (4.4) and (4.5) is understood to mean the optical thickness for absorption at the middle of the line with pure Doppler lineshape at the middle of the P and R branches of the vibrational-rotational band. For example, at the center of a tube of radius  $R, \tau = [CO_2]\sigma_D R$ , where  $\sigma_D$  is the absorption cross section:

$$\sigma_{\rm D}[\rm cm^2] = 9 \cdot 10^{-7} \frac{MB_0}{Tv^3} \frac{g_u}{g_1} A_{\rm g}^0; \qquad (4.6)$$

where  $g_{u}$  and  $g_{l}$  are the statistical weights of the upper and

lower vibrational levels,  $B_e$  is the rotational constant in  $cm^{-1}$ ,  $\nu$  is the frequency of the transition in  $cm^{-1}$ , M is the mass of the molecule in atomic units, T is the gas temperature in K, and  $A_3^0$ , the probability of a radiative transition is expressed in  $s^{-1}$ .

In the calculation of the laser characteristics it was assumed that the vibrational temperature of the deformation and symmetrical modes of CO<sub>2</sub> are equal to the gas temperature. This condition is easy to attain by an admixture of helium, which substantially increases the relaxation rate of the vibrational energy of these modes. The gas temperature is assumed to be constant and independent of any other parameters, that is, in the calculations we are in fact ignoring the possibility of heating of the gas. It is clear that in this approximation the calculation gives only the maximum possible values of the laser parameters (since heating of the gas always degrades them). The results of the calculation for the gain of a weak signal, the generation power, and the efficiency of conversion of the energy of black body radiation into laser energy are given in Fig. 6. In the calculation of the gain it was assumed that the threshold gain is equal to  $\kappa_{\rm th} = 10^{-4} \, {\rm cm}^{-1}$ .

From Fig. 6 it can be seen that even under optimum operating conditions of the laser, the values of its most important characteristics, such as the weak signal gain and the efficiency, remain small. The reason is that here we are in the position of having to satisfy conflicting requirements. Thus,

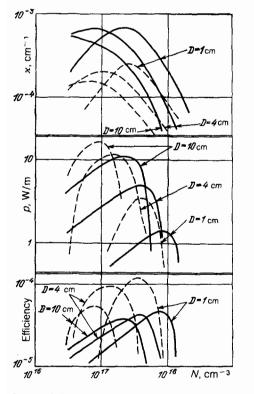


FIG. 6. Gain x, generation power P, and efficiency of conversion of radiation of a black body at T = 2000 K into energy of laser radiation, all as a function of concentration of the mixture 16% CO<sub>2</sub> + 4% He + 80% Ar, for various tube diameters, for the scheme shown in Fig. 4, with gas flow. Solid curves: mixture with single-isotope CO<sub>2</sub>; dashed curves: with 12 isotopes. Gas temperature T = 300 K.

the necessity of increasing the efficiency of conversion of solar into laser radiation requires, as noted above, the use of isotopes and an increased gas pressure (in order to absorb efficiently the black body infrared radiation). However, other conditions being equal, an increase in the number of isotopes leads to a corresponding decrease in the gain, while an increase in the gas density brings about an increase in the collisional relaxation rate of the  $(00^{\circ}1)$  upper laser level and a decrease in the probability of optical excitation of this level as a result of the increased optical density of the gas. The latter circumstance is an important obstacle to the development of an efficient CO gas laser based on the scheme discussed here or gas lasers proposed in Ref. 51.

We note that the necessity of maintaining a gas flow is the main feature of the laser suggested in Refs. 49–51 and 53–54. It follows from the last section that to obtain cw generation it is also necessary to maintain a gas flow in the majority of RI molecule lasers. This, of course, complicates the construction, and a closed cycle scheme requires the use of compressors and pumps. Laser operation thus requires either a supplemental source of energy or conversion of solar energy not only into laser radiation but also into another form of energy. Of the schemes discussed above, only lasers based on a  $Br_2$ -CO<sub>2</sub>-He mixture or on alkali metal vapors (and possibly C<sub>4</sub>F<sub>9</sub>I molecules) do not require a gas flow. Continuous generation without a gas flow can also be obtained in principle with pumping by the radiation of a black body.

### 4.3. cw CO<sub>2</sub> laser without gas flow

In using the idea, which we have considered above, of pumping a laser with the radiation from a black body heated by the sun, the scheme that is employed may in principle, be that illustrated in Fig. 7. Here the solar radiation is concentrated by a long cylindrical mirror 1 onto a long cylindrical black body 2, heating the latter to  $T_b \approx 800-1000$  K, where this temperature is determined principally by radiation losses and is given by the relation [compare (4.1)]

$$T_{\rm b} = \left(\frac{P_{\odot}}{\pi\sigma}\xi\right)^{1/4}.\tag{4.7}$$

The infrared radiation from this black body is focused by means of an elongated but smaller mirror 3 onto a tube 4 which is structurally connected to the main solar concentrator 1. The laser medium is inside the tube, and the laser mirrors as usual are placed on the ends of the tube. In order

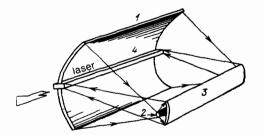


FIG. 7. Conceptual diagram of a solar laser with pumping by black body radiation, a cold laser tube, and no gas flow.

to facilitate the removal of the heat from the gas to the walls of the tube, the latter can be divided into sections. In this system the radiation loss of energy from the black body is of course greater than in the scheme with the enclosed cavity discussed above (see Fig. 5), but it is simpler by not requiring a gas flow. Here the gas is cooled by the outflow of heat to the walls and to the inner partitions of the tube. By virtue of its construction this kind of design is suitable for placing a laser in space, since in this case absorption of the infrared radiation on its way from the second mirror to the tube, which would be deleterious to the operation of the laser, does not occur. An important merit of this construction when located in space is the possibility of holding the walls of the tube at a very low temperature ( $\sim 200-300$  K). This is attained by the structural connection (and hence the strong thermal connection) with the solar concentrator, whose temperature can be low in outer space as a result of radiative cooling. If the reflectivity of the mirror is  $\alpha$  while its rear side radiates and absorbs as a black body, then this temperature Tfor a cylindrical solar concentrator with a transverse dimension l, a total length L, and a radius of curvature R, can be estimated from the radiation energy balance equation:

$$(2-\alpha) 2RL\sigma T^{4} \arcsin \frac{l}{2R} = (1-\alpha+\delta) P_{\odot}Ll + S_{\text{eff}} F_{3};$$

$$(4.8)$$

where  $\sigma$  is the Stefan-Boltzmann constant  $P_{\odot}$  is the solar constant,  $\delta$  is the fraction of the energy absorbed in the laser tube, and  $S_{\rm eff}$  is the effective area of this design for irradiation by thermal radiation of energy flux  $F_3$  from the Earth. For the appropriate orientation of the laser in space relative to the sun and the Earth, the second term on the right hand side of (4.8), which describes the heating by the thermal radiation from the Earth, can obviously be made smaller than the first term, which describes the heating by the solar radiation. In this case for l/2R = 0.867,  $\alpha = 0.95$ , and  $\delta = 0.1$ , for instance, we have T = 234 K.

We note that a design with a "cold" laser tube, similar to the one discussed above, can also be used for lasers in space that are pumped by directly concentrated solar radiation (a laser of the Br<sub>2</sub>-CO<sub>2</sub>-He type), for which it is usually desirable to have a low temperature of the working medium. In this case it is simply necessary to design good thermal contact between the laser tube and the solar concentrator. Unfortunately, the main difficulty at the present time in developing solar lasers is a different one, namely, the difficulty in finding a suitable active medium. For instance, in the use of  $CO_2$  gas with pumping by the radiation from a black body according to the scheme of Fig. 7, the power and efficiency of a cw solar laser without gas flow are as small as in the scheme of Christiansen with a black body cavity and gas flow (see Fig. 5). This fact is illustrated in Fig. 8, which gives some results of our calculations for a 50%  $CO_2 + 50\%$  He mixture placed in a tube of rectangular cross section lh with a wall temperature  $T_{\rm w} = 200$  K and irradiated with the radiation from a black body at a temperature of 1000 K (which corresponds to a degree of concentration  $\sim$  130 of solar radiation on a long cylindrical black body). As in the ordinary electrical discharge laser, the helium serves to increase the

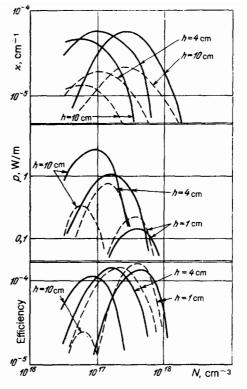


FIG. 8. Gain x, generation power P, and efficiency of a solar laser as functions of the concentration of the mixture 50%  $CO_2 + 50\%$  He for a solar laser (the scheme of Fig. 7) with pumping by the radiation from a black body, for various transverse tube dimensions. Solid curves: mixture with single-isotope  $CO_2$ ; dashed lines: with 12 isotopes. Wall temperature  $T_w = 200$  K.

relaxation rate of the lower laser level and increase the thermal conductivity of the gas mixture. The radiation from the black body is incident on one side of a tube of dimension h, while the other dimension l is chosen so that in the entire volume of the tube the weak signal gain on the  $CO_2(00^{\circ}1) \rightarrow CO_2(10^{\circ}0)$  laser transition is greater than the threshold value (because of the increase in the optical thickness  $\tau_{\rm gas}$  with respect to absorption of the black body radiation, the excitation probability W [see (4.2)] of the  $00^{\circ}$  l upper laser level decreases with distance from the side of the tube on which the radiation is incident, and consequently the gain decreases). A calculation was carried out for a black body radiation attenuation factor  $\beta$  equal to 0.5 [see (4.2)] and the power and efficiency were obtained for a threshold gain  $x \approx 10^{-5}$  cm<sup>-1</sup>. In the calculations, the gas temperature was also obtained in a "self consistent" manner.

From a comparison of Figs. 6 and 8 it can be seen that the scheme with a black body cavity with gas flow and that with a "cold" tube without gas flow give about the same efficiency, but in the latter case the gain and the generation power are approximately an order of magnitude lower. The reason for this is that in the cold tube scheme the attenuation factor  $\beta$  is smaller and the temperature of the black body is lower.

We should note that our conclusion that the efficiency is low for solar lasers with pumping of  $CO_2$  molecules by black body radiation has in fact been confirmed by experiments<sup>53,54,57</sup> in which a single-isotope mixture of CO<sub>2</sub> at a pressure 5–50 torr was irradiated with black body radiation and laser generation was obtained with an efficiency  $\sim 10^{-6}$ – $10^{-4}$ . Nevertheless, it would appear to be premature to give up the idea of using this radiation in solar lasers. Experiments<sup>53,54,55</sup> which have in fact simulated such a solar laser have demonstrated in principle the practicability of such a scheme in spite of the low laser energy characteristics obtained. It is necessary to seek gas mixtures that absorb thermal radiation efficiently at moderate pressures.

## 5. SPACE LASER MEDIA WITH SOLAR EXCITATION

In addition to the study of the development of highpower gas lasers with solar excitation, that can be used in solar power applications, there is considerable scientific interest in the experimental search for, and the theoretical investigation of, natural media with level population inversion and natural lasers excited by natural radiation, and in particular by the radiation of the stars and the sun. Already in the 1960s natural cosmic objects which amplified electromagnetic radiation in the centimeter wavelength range were well known to astrophysics. These are OH and H<sub>2</sub>O molecule masers existing in the extremely extended media of deep space objects (see, e.g., Refs. 63 and 64). It has recently been hypothesized that a maser effect is possible on the lines of H<sub>2</sub>O inside the coma of some comets.<sup>65</sup> At the beginning of the 1980s natural lasers were observed in the infrared region.<sup>66-68</sup> In the search for lasers of this type, close attention was paid to the upper atmospheres of planets. These atmospheres, which are rarified gases that are subject to the action of corpuscular and electromagnetic radiation from the sun, are highly nonequilibrium molecular media (nonequilibrium in terms of chemical composition, temperatures of the components, and populations of the excited states).

# 5.1. Population inversion of molecular levels in the outer atmosphere of the Earth

The possibility of this effect in the Earth's atmosphere was considered for the first time<sup>69</sup> in the context of infrared emission of the vibrational-rotational bands of hydroxyl OH in the mesosphere at altitudes of 85–90 km. It was shown that there is a substantial population inversion of the vibrational-rotational levels of OH excited in the process of the exothermic reaction

$$H + O_3 \rightarrow OH \ (v \leqslant 9) + O_2. \tag{5.1}$$

This reaction is responsible for the vibrational-rotational emission bands, well known in atmospheric optics, which were discovered and identified for the overtone frequencies already in 1950.<sup>70–72</sup> As a result of the chemical reaction and spontaneous radiative transitions (at these altitudes collisional relaxation is insignificant), the vibrational levels of the hydroxyl with v = 1-9 have populations that vary slowly with v, and to this situation corresponds a high effective vibrational temperature  $T_v \gtrsim 7000$  K. However, the temperature of the rotational degrees of freedom of the molecule, because of the rapid rotational-translational relaxation, is close to the gas temperature, and therefore low (~180–250)

K). This departure of the vibrational temperature from the rotational temperature leads to a population inversion of the vibrational-rotational levels on the P-branch of the transition  $(v, i \rightarrow v - 1, i + 1)$  (this case, which is well known in laser physics as the so called "partial" inversion, can be obtained, for instance, in laboratory CO lasers). We note that although the excitation of the vibrational levels of the OH occurs as a direct result of a chemical reaction (5.1), the original cause of this pumping is, of course, the solar radiation. The H atoms are formed by photodissociation of H<sub>2</sub>O vapor, the O<sub>3</sub> molecules are formed in a triple recombination  $O + O_2 + M \rightarrow O_3 + M$  while the atomic oxygen is supplied to the 85-90 km level from higher altitudes, where it is formed by photodissociation of O2 by the ultraviolet radiation from the sun. Estimates<sup>69</sup> have shown, however, that although amplification of the radiation occurs, it is extremely weak. For instance, at the maximum possible length of the active medium (horizontal line of sight to the limb of the planet) the gain in a single pass is  $(2-5) \cdot 10^{-3} \%$ .

A similar mechanism for the formation of population inversion also occurs on the vibrational-rotational levels formed as a result of the chemical reaction

$$N (^{2}D) + O_{2} \rightarrow NO + O.$$
(5.2)

This reaction proceeds most efficiently in the polar aurora at altitudes of 105-120 km, with 1-3% of the total energy of the fast electrons formed in the polar aurora and poured out of the magnetosphere being transformed into nonequilibrium infrared emission of NO on the  $v \rightarrow v - 1$  vibrational transitions (the 5.3  $\mu$ m band).<sup>73,74</sup> However, in this case the gain is very low. Estimates of the amplification of the infrared emission, given in Ref. 75 have also given negative results.

## 5.2. Laser effect in the atmospheres of Venus and Mars

Very attractive objects for the search for natural laser media are the atmospheres of Venus and Mars. This is because these atmospheres consist mainly of carbon dioxide gas, the molecules of which, as is well known, are extremely "convenient" for obtaining population inversion of the vibrational levels. Moreover, in an atmosphere of such a gas, which is optically active in the infrared, even at high altitudes, where the medium is in a nonequilibrium condition and it is consequently reasonable to look for laser effects, there is, besides the ultraviolet and corpuscular radiation from the sun, another important source of optical pumping-the solar infrared radiation. References 67 and 68 contained the first detailed theoretical investigation of the nonequilibrium infrared radiation in the 10.6 and 9.4  $\mu$ m bands of CO<sub>2</sub> in the outer atmospheres of Venus and Mars. The properties of these atmospheres were studied as active laser media and it was shown that there is a substantial amplification of infrared radiation on the  $00^{\circ}l \rightarrow 10^{\circ}0$  transition of the CO2 molecule. Subsequently, similar theoretical investigations were carried out<sup>76-79</sup> and they confirmed the conclusions reported in Refs. 67 and 68.

Investigations have been carried  $out^{67,68}$  for the regions illuminated by the sun, at an altitude of 80–130 km, for Venus and at 35–120 km for Mars. The lower boundaries of the

regions correspond approximately to the level at which the condition of local thermodynamic equilibrium no longer holds for the vibrational state of the asymmetric mode of the  $CO_2$  molecule. At these altitudes the main mechanisms for excitation and deactivation of the  $CO_2$  vibrational levels are absorption of infrared radiation from the sun, spontaneous radiative vibrational-rotational transitions (with allowance for radiation imprisonment) and vibrational transitions as a result of collisions. The fastest of these processes is the vibrational-vibrational exchange of energy during collisions (the VV processes). This circumstance allows us to simplify the problem of finding the populations of the various vibrational states by reducing it to a determination of the energy (or the average amount of vibrational quanta) of the various vibrational modes of the  $CO_2$ .

The high degree of imprisonment of the infrared radiation in the principal 4.3  $\mu$ m CO<sub>2</sub> band (the 00<sup>0</sup>1  $\rightarrow$  00<sup>0</sup>1 transition in the main isotopic component  $C^{12}O_2^{16}$ ) makes it necessary to take into account optical excitation and radiative decay of the asymmetric mode vibrations of the  $CO_2$ , and because of the weaker bands at 2.7 and 2  $\mu$ m it is also necessary to take into account a series of hot bands (i.e., transitions between excited vibrational states) and transitions in the less abundant isotopic mixtures.<sup>3)</sup> In Refs. 68 and 69, to find the amount of energy in the CO<sub>2</sub> asymmetric mode, five isotopically distinct CO2 molecules were studied  $(C^{12}O_2^{16}, C^{13}O_2^{16}, C_{12}O^{16}O^{18}, C^{12}O^{16}O^{17}, and C^{13}O^{16}O^{18})$ with the natural abundances, and 17 infrared bands in each of the isotopic components (five bands in the 4.3  $\mu$ m region, four in the 2.7  $\mu$ m region, six in the 2  $\mu$ m region and two in the 10  $\mu$ m region for the transitions 10°0–00°1 and 02°0–  $00^{01}$ ). In the calculations it was assumed that the separate vibrational-rotational lines in all the bands do not overlap. This assumption is valid for the range of altitudes investigated.

Another considerable simplification was achieved<sup>67,68</sup> by using in the description of the transport of the infrared radiation the "radiative removal" approximation<sup>59,60</sup> mentioned in a previous section. A more rigorous solution of the transport equation carried out subsequently<sup>78,79</sup> has given similar results. In the radiative removal approximation, however, the average store of vibrational quanta in the asymmetric CO<sub>2</sub> mode can be described approximately by the equation

$$\frac{d\alpha}{dt} = 0 = P_{10} (\alpha^0 - \alpha) + \sum_{\substack{j=1\\j=1}}^{5} \gamma_j \sum_{\substack{i=1\\i=1\\i=1}}^{17} W_i M(\tau_{ij}^{\odot}) \exp\left(-\frac{n_i E_{010}}{kT_{010}} - \frac{\Delta E_i'}{kT}\right) - \sum_{\substack{j=1\\j=1}}^{5} \gamma_j \sum_{\substack{i=1\\i=1}}^{17} A_i L(\tau_{ij}^{\odot}) \exp\left(-\frac{m_i E_{010}}{kT_{010}} - \frac{\Delta E_i'}{kT}\right) \frac{\alpha}{1 + \alpha}$$
(5.3)

Here the first term on the right hand side describes collisional relaxation with probability  $P_{10}$ , the second term describes the excitation by the absorption of solar infrared radiation in the various bands and the various isotopic components; the third term describes spontaneous radiative

transitions (taking into account imprisonment of the radiation), the quantity  $\alpha^0$  is the equilibrium value of  $\alpha$  corresponding to the gas temperature T,  $E_{010}$  and  $E_{001}$  are the energies of the 01°0 and 00°1 states,  $\gamma_i$  are the relative isotopic concentrations of the  $CO_2$  molecules,  $A_i$  is the probability of spontaneous radiative transitions for the band i, and  $W_i$  is the probability of excitation of the levels as a result of absorption of solar radiation in these bands (for an optically thin medium). The exponential factors of  $W_i$  and  $A_i$  in (5.3) determine the relative populations of, respectively, the lower and upper vibrational levels for the infrared bands. These combination levels include various states of the symmetric and deformation CO<sub>2</sub> modes having the temperature  $T_{010}$ . The energies of these modes are represented in the form  $n_i E_{010} + \Delta E'_i$  (for the lower levels of the bands), or  $m_i E_{010} + \Delta E_i''$  (for the upper levels), where  $n_i$  and  $m_i$  are integers.

The functions  $M(\tau_{ij}^{\odot})$  and  $L(\tau_{ij}^{\odot})$  in Eq. (5.3) within the framework of the radiative removal approximation being used take into account the decrease of the probabilities of excitation of the levels by solar infrared radiation and of the probabilities of their radiative decay as a result of radiation imprisonment in the vibrational-rotational bands. In these expressions  $\tau_{ij}$  is the optical thickness for absorption, by the *j*th isotopic component, of infrared radiation in the *i*th band, and  $\tau_{ij}^{\odot}$  is the analogous optical thickness for absorption of solar radiation. The function  $M(\tau_{ij}^{\odot})$  is given by expressions (4.3)-(4.5), and  $L(\tau_{ij}^{\odot})$  has a similar form (the small difference is due to averaging over angles).

In the general case, Eq. (5.3) has to be solved simultaneously with the equations for the vibrational temperature  $T_{010}$ , the gas temperature T, and the gas density  $\rho$  at a given altitude. In Refs. 68 and 69, however, T and  $\rho$  were taken as known, and to find their altitude profiles in the atmosphere of Venus the daily-average model of Dickenson<sup>60</sup> was used, while for the atmosphere of Mars, the model with a moderate temperature, recommended by COSPAR was used.<sup>80</sup> In addition, models were analyzed in which the temperature at

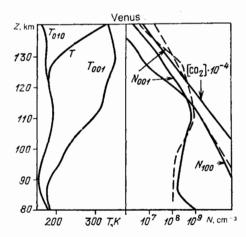


FIG. 9. Altitude dependence of the gas temperature T, the vibrational temperatures  $T_{010}$  and  $T_{001}$  of the CO<sub>2</sub> molecule, the populations of the 10<sup>0</sup>0 and 00<sup>0</sup>1 levels, and the total concentration of CO<sub>2</sub> molecules in the atmosphere of Venus. Solid lines: data of Refs. 67 and 68; dashed lines: data of Ref. 76.

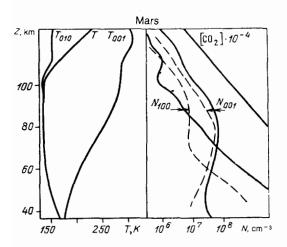


FIG. 10. Same data as Fig. 9, but for Mars.

all altitudes studied differed from the temperature of the main profile by  $\pm 20^{\circ}$  for Venus and  $\pm 10^{\circ}$  for Mars.

The equation for the vibrational temperature  $T_{010}$  was also not specially analyzed, and to find  $T_{010}$  the results of a calculation<sup>58</sup> of the source function for the fundamental 15  $\mu$ m band (the 00°0  $\rightarrow$  01°0 transition) in the atmosphere of Venus were used. To find the altitude profile of  $T_{010}$  in the atmosphere of Mars this source function was rescaled to the altitude scale of Mars.

On the basis of this model a calculation was carried out<sup>67,68</sup> for the average supply of vibrational quanta in the asymmetric  $CO_2$  mode and the vibrational temperature of this mode. Some of the results of the calculation are shown in Figs. 9 and 10. It can be seen that at the lower bound of the altitude region studied breakdown of local thermodynamic

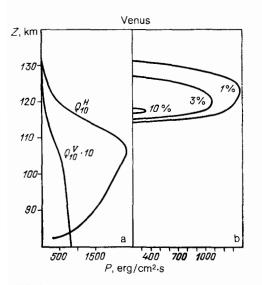


FIG. 11. a) Infrared radiation flux in the 10.6 and 9.4  $\mu$ m CO<sub>2</sub> bands in the atmosphere of Venus.  $Q_{10}^{H}$  is the flux at the limb of the planet, i.e., for a horizontal line of sight from space;  $Q_{10}^{V}$  is the vertical flux into space from a column of atmosphere with its base at the altitude Z. b) Laser generation power in the 10.6  $\mu$ m line in the atmosphere of Venus as a function of the altitude of the mirror axis above the surface and for three values of the loss coefficient at the mirrors.

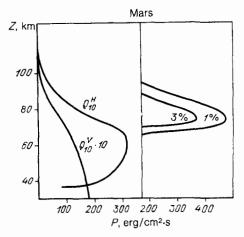


FIG. 12. The same as Fig. 11, but for Mars.

equilibrium for the asymmetric mode sets in and  $T_{001}$  becomes greater than T or  $T_{100}$ . One of the reasons for the increase of  $T_{001}$  is a decrease with altitude of the optical thicknesses  $\tau_{ij}^{\odot}$ , which leads to an increase in the effective probabilities of excitation of the mode by solar infrared radiation. Here an extremely important role is played by the absorption of solar radiation in the 2.7 and  $2\mu$ m bands of the main component  $C^{12}O_2^{16}$  (including some hot bands) as well as in the 4.3 and 2.7  $\mu$ m of the low-abundance isotopic molecules.

One of the interesting results of the analysis was the observation of direct (without the involvement of the translational degrees of freedom) conversion of absorbed solar radiation in the near infrared ( $\lambda < 4.3 \,\mu$ m) to intrinsic emission by the atmospheres in the 10.6 and 9.4  $\mu$ m bands. It was found that in the atmospheres irradiated by the sun there is a layer of nonequilibrium infrared emission in these bands with intensities that substantially exceed (by 10<sup>3</sup> to 10<sup>4</sup>) the equilibrium values and have maxima at altitudes ~ 108 km for Venus and ~ 60 km for Mars, for line of sight to the limb

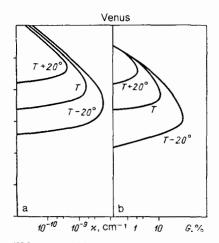


FIG. 13. a) Altitude dependence of the infrared emission gain per unit length and b) the gain in a single pass along a horizontal line of sight, for the  $00^{0}1 \rightarrow 10^{0}0$  transition, in the atmosphere of Venus. The calculation was carried out for a model atmosphere with a "moderate" temperature, as well as for a "cold" and a "hot" atmosphere.

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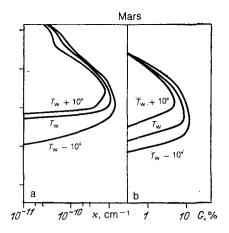


FIG. 14. The same as Fig. 13, but for Mars.

of the planet. This fact is illustrated in Figs. 11 and 12.

The increase of the vibrational temperature above Tand  $T_{010}$  not only causes the nonequilibrium nature of the infrared emission but also brings about a population inversion of the 00°1 and the 10°0 and 02°0 levels (see Figs. 9 and 10) and an amplification of the emission in the 10.6 and 9.4  $\mu$ m bands. The calculated altitude profiles of the gains and of the total gain in one pass are shown in Figs. 13 and 14. It can be seen that the gain has a maximum at 110–125 km for Venus and 70–80 km for Mars, and at these maxima the gain reaches values of 3–50% for Venus and 5–15% for Mars. These gains are quite sufficient for experimental observation.

From the data of Refs. 59 and 81, where the vibrational temperature  $T_{001}$  of the asymmetrical mode in the meso-sphere and the lower thermosphere (60–140 km) of the Earth were calculated, it can also be concluded that there is a population inversion of the 00<sup>0</sup>1 and 10<sup>0</sup>0 levels in the region of 85–95 km. However, in this case, because of the small concentration of CO<sub>2</sub> the gain is negligibly small.

# 5.3. Terrestrial experimental technique for observing infrared emission in the atmospheres of planets

The first experimental observation of population inversion of the vibrational-rotational levels of CO<sub>2</sub> in the atmosphere of Mars was reported in Ref. 66. The authors used an Earth-based infrared telescope-spectrometer to observe the solar-illuminated disk of the planet and measure the intensity and shape of the individual vibrational-rotational lines of the 10.6  $\mu$ m band incident on the window of transparency of the Earth's atmosphere. From the measured line widths the temperature of the gas in an emitting column of the atmosphere was determined and from the intensity of a single line the authors of Ref. 66, by summing over the rotational sublevels determined the total energy flux in all the lines in the 10.6 and 9.4  $\mu$ m CO<sub>2</sub> bands and found the total population of the 00°1 level, which turned out to be greater than that of the lower 10°0 level at equilibrium corresponding to the measured gas temperature. Similar measurements were also made for Venus.<sup>82</sup> For the subsolar point the measurements gave a vertical flux in the 10.6 and 9.4  $\mu$ m bands of ~ 80 erg/

cm<sup>2</sup> s for Venus and  $\sim 18 \text{ erg/cm}^2$  s for Mars, values which agree with the calculated vertical fluxes from a column with the lower base at an altitude Z below  $\sim 40 \text{ km}$  for Mars and  $\sim 85 \text{ km}$  for Venus (see Figs. 11 and 12).

Measurement of the profiles of the individual vibrational-rotational lines in the atmospheric infrared emission requires development of spectrometers with a resolving power  $\lambda / \Delta \lambda \approx 10^7$  (which is two orders of magnitude greater than that obtained in modern high-resolution infrared spectrometers of the Fourier spectrometer or Fabry-Perot etalon type). Such high values of  $\lambda / \Delta \lambda$  can be attained with the use of laser infrared heterodyne spectrometers (LIRHS). Detailed information on the technique of LIRHS and a review of the literature can be found in Ref. 83.

Let us briefly describe the LIRHS at the Goddard Space Flight Center of NASA, which has been used now for a number of years in investigations of various spectral lines in the atmospheres of the Earth, Venus, Mars, Jupiter<sup>66,83,84</sup> as well as in the tails of comets.<sup>85</sup> This device was also used for the first observations of natural laser media in the outer atmosphere of Mars.<sup>66</sup> A schematic diagram of the LIRHS is shown in Fig. 15.

A signal from a source of infrared radiation,  $E_s \exp(i\omega_s t)$ , admitted through a telescope, is mixed with a reference signal  $E_L \exp(i\omega_L t)$  from a high-stability CO<sub>2</sub> laser. The photomixer (a crystal of HgCdTe) produces a difference signal  $2E_L E_s \cos[(\omega_L - \omega_s)t]$ , which can be amplified and processed by means of rf electronics. In the diagram of Fig. 15 the difference signal is fed into the input of a 128 channel rf spectral line receiver, where it is integrated, synchronously detected, and processed by computer. With the laser infrared heterodyne spectrometer described above a resolution of 0.00017 cm<sup>-1</sup> for the CO<sub>2</sub>R8 line at

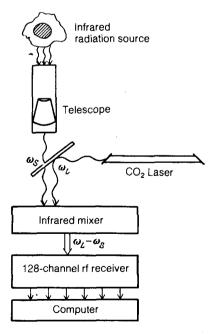


FIG. 15. Block diagram of a laser infrared heterodyne spectrometer (from Ref. 83).

967 707 cm<sup>-1</sup> ( $\lambda / \Delta \lambda \approx 5.7 \cdot 10^6$ ) was attained. The sensitivity of an infrared spectrometer can be characterized by the minimum temperature of a black body whose radiation is detectable for a given signal/noise ratio. In the case described above, for  $\lambda_L = 10.6 \ \mu\text{m}$ , the sensitivity of the LIRHS permitted detection of the radiation from a black body with a temperature of 130 K for a signal/noise ratio  $\approx 1$ . A similar system with  $\lambda_L \approx 30 \ \mu\text{m}$  could be used to detect radiation of a black body with  $T = 50 \ \text{K}$  (for a signal/noise ratio  $\approx 10^3 \ (\text{Ref. 83})$ .

From the data on the shape of the line intensities, obtained with the LIRHS described above, in addition to the measurements of the translational and vibrational temperatures of  $CO_2$  in the atmospheres of Venus and Mars and the observation of population inversion of the molecular levels, the wind speeds in the atmospheres of these planets have been determined (with an accuracy to a few meters per second), the concentration of  $C_2H_6$  molecules in the atmosphere of Jupiter has been measured,<sup>85</sup> an upper limit has been placed on the concentration of NH<sub>3</sub> in the atmosphere of a comet, and the molecules C10, COS, O<sub>3</sub>, NH<sub>3</sub>, HNO<sub>3</sub>, and  $C_2H_4$  have been detected in the atmosphere of the Earth.<sup>83-85</sup>

### 5.4. Designs for laser systems in the atmospheres of Venus and Mars

The observation of population inversion and very pronounced amplification of 10.6  $\mu$ m radiation in tangential directions in the atmospheres of Venus and Mars prompt us to propose a design for a space laser generator in these atmospheres.<sup>67,68,86</sup> By placing two artificial satellites above these planets and setting up the necessary mirror orientation, it is possible to obtain a laser resonator system. The height of the orbits of these satellites and the distance between them should be chosen so that the line joining the axis of the mirrors runs along the altitude at which the gain is maximum. The generation power of such lasers has been calculated<sup>67,68</sup> under the assumption that one of the mirrors is perfectly reflecting and the loss at the other mirror is due to transmission necessary to extract the radiation from the resonator. The results of the calculation are shown in Figs. 11 and 12. It can be seen that the generation power of the Venus laser can amount to  $10^{-4}$  W/cm<sup>2</sup> and that of the Mars laser to  $4 \cdot 10^{-5}$  $W/cm^2$ . The mirror area, which determines the total generation power, can be more than 100 km<sup>2</sup>. These lasers could, in principle, be used as energy stations or serve as unique cosmic civilization beacons.

# 6. CONCLUSIONS

The investigations that have been carried out over the past nearly ten years have led to definite progress and have enabled investigators to discover gaseous media which can provide direct conversion of solar radiation into laser radiation. In experiments using solar simulation a conversion efficiency  $\sim 0.2\%$  has been obtained. However, for the practical use of lasers in power systems this is not sufficient. Therefore the search for new media continues. Here there

are very good prospects in the search for molecules of the RI type, with a broad absorption spectrum in the visible (and not in the near ultraviolet, as for  $C_3F_7I$ ) and with complete regeneration after photodissociation (similar to  $C_4F_9I$ ). Also holding out promise is the study of the potentialities of media containing alkali metal vapor, and the search for molecules that absorb infrared radiation efficiently at moderate pressures (for schemes of pumping with black body radiation). If such media are found, then there is no doubt that high-power gas lasers with solar excitation will take a respectable place in solar power and laser technology.

Earth-based measurements of nonequilibrium infrared emission from the atmospheres of Mars and Venus, carried out with the use of laser infrared heterodyne spectrometers, in combination with theoretical investigations allow us to relate the outer atmospheres of these planets to the first cosmic laser objects in the infrared region whose existence has become known to us. Subsequently, in this connection it appears to be of interest to carry out a space experiment (the technology presently exists) in laser probing and direct measurement of the emission gain in these media, and, in the more distant future, in the development of space lasers.

In conclusion the authors consider it their pleasant duty to express appreciation to S. A. Akhmanov for support and constant attention to this work, and many fruitful discussions. The authors are grateful to V. S. Zuev and M. J. Mumma (USA) for helpful discussions.

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<sup>&</sup>lt;sup>1</sup>The transfer of the excitation to the acceptor molecule can, in principle, occur not only from the excited atom products of the photodissociation, but also from molecules excited by the sun.

<sup>&</sup>lt;sup>2)</sup>Here it is appropriate to note that the idea of using infrared radiation from a black body for pumping a CO<sub>2</sub> laser was first suggested<sup>56</sup> in the USSR in 1969 and was subsequently implemented experimentally.<sup>57</sup>

<sup>&</sup>lt;sup>3)</sup>Under laboratory conditions at CO<sub>2</sub> concentrations greater than  $10^{15}$  cm<sup>-3</sup> the absorption in all the bands except the  $00^{\circ}0 \rightarrow 00^{\circ}1$  cannot produce any substantial deviation of the vibrational temperature of the asymmetric mode from that of the translational modes, and therefore in the balance of energy this mode may be ignored (see the previous section)

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