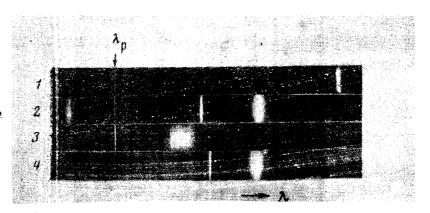
Stimulated-emission lasers with spectra of Nd $^{3+}$ -doped crystals: 1) KY(WO $_{4}$ )2, 2) Lu $_{3}$ Al $_{5}$ Ol $_{2}$ , 3) LiGe(MoO $_{4}$ )2, 4) Y $_{3}$ Al $_{5}$ Ol $_{2}$ . The arrow indicates a reference line with  $_{D}^{\lambda}$  = 10 561.5 A



progress attained in the development of effective parametric generators and harmonic generators with almost 100% conversion of IR into visible light, and their use in such basic physical research as laser photochemistry, molecular spectroscopy, or Raman spectroscopy. Of course, the high-power laser applications still remain a timely topic.

We report here three new cw lasers based on the crystals  $KY(WO_4)_2$ ,  $Lu_3Al_5O_{12}$ , and  $LiGd(MoO_4)_2$ , activated with  $Nd^{3+}$  ions. Their main spectroscopic and lasing characteristics are gathered in the table. For comparison, the table lists also the parameters of the widely used crystal  $Y_3Al_2O_{12}:Nd^{3+}$ , which is now widely used in quantum electronics. All the data are given for room temperature.

The crystals used in the lasing experiments were polished rods 25 mm long and of  $^{\circ}5$  mm diameter. The end faces were plane-parallel within about 10". The optical resonator was made up of confocally-mounted external multilayer dielectric mirrors ( $^{\tau}_{mir} > 1\%$  at 1.06  $^{\mu}$  wavelength). In the cw regime, the crystals and the lamp were cooled with running water. At an excitation power 2000 W and at  $^{\tau}_{mir} = 3\%$ , using the crystals KY(WO<sub>4</sub>)<sub>2</sub> and Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> with near-optimal activator contents and Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> with  $^{\circ}$ O<sub>16</sub> at  $^{\circ}$ NNd<sup>3+</sup>, we registered output powers 550, 600, and 350 mW, respectively. The figure shows the generation spectra of the investigated lasers, obtained at 300°K and with an exciting energy three times threshold.

Since the wavelengths of ordinary lasers with Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Nd<sup>3+</sup> and Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>: Nd<sup>3+</sup> are close to each other at 300°K, the spectra are shown for the case of lasers with a combined active medium. The spectrum of the Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Nd<sup>3+</sup> crystal contains in addition to the 10 642.5 Å line also two lines with wavelengths  $\sim 10$  535 and  $\sim 10$  610 Å.

In conclusion, we add that we have also discovered pulsed stimulated emission of a number of other  ${\rm TR}^{3+}$  ions in these crystals. Their properties will be reported in separate articles.

## TRANSFER OF VIBRATIONAL ENERGY FROM OD TO CO2

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We report here for the first time observation of effective energy transfer from the radical OD to  $CO_2$  molecules. This transfer leads to a laser effect in a mixture of  $O_3$ ,  $O_2$ , and  $CO_2$  at a wavelength  $O_3$ . Using a simple analytic model of the reaction and the measured temporal characteristics of the

laser pulse, we estimate the rate constant of the vibrational-vibrational energy exchange between OD and  ${\rm CO}_2$ . We used two measurement procedures, one based on the time delay of the lasing relative to the triggering of the pump, proposed in [1], and the other based on the attenuation of the generation signal of a chemical laser and heretofore apparently never employed for this purpose.

A laser tube 80 cm long and 1.5 cm in diameter was used in the experiment. The pumping was with two IFP-20 000 lamps. At a bank capacitance 1  $\mu$ F, the flash duration in the ultraviolet region of the spectrum was 7  $\mu$ sec at the half-intensity level. The laser radiation was registered with a Ge-Au receiver cooled with liquid nitrogen.

The predominant procesees that take place in the photolysis of a mixture of  $O_3$ ,  $D_2$ , and  $CO_2$  are represented by the scheme

0. 
$$O_3 + h\nu$$
 ( < 2500 Å)  $\frac{K_o(t)}{O}$  O('D) +  $O_2$ ,  
1.  $O(TD) + D_2$   $\frac{K_1}{O}$  OD\* +  $O_2$ ,  
2.  $D + O_3$   $\frac{K_2}{K_3}$  OD +  $O_2$ ,  
3.  $OD^* + CO_2$   $\frac{K_3}{K_3}$  OD +  $O_2^*$ ,  
4.  $O(TD) + M$   $\frac{K_4}{O}$  O +  $M$ ,  
5.  $OD^* + O_3$   $\frac{K_5}{O}$  products  
6.  $O(TD) + O_3$   $\frac{K_6}{O}$  O<sub>2</sub> + O<sub>2</sub>,

where an asterisk indicates vibrational excitation of the molecules, and CO $_2^*$  is the CO $_2$  molecule in the 00 $^0$ l state. The time dependence of K $_0$ (t) is given with sufficient accuracy by the relation K $_0$ (t) =  $\gamma_1$ t exp(- $\gamma_2$ t).

Integrating the balance equation in the linear approximation, we obtain the time dependence of the population of the  $00^{\,0}\,\mathrm{l}$  level

$$[CO_2^*](t) = \sigma_3 \int_0^t (e^{-(\sigma_3 + \sigma_5)t})^{t'} \int_0^t e^{(\sigma_3 + \sigma_5)t'} w(t'') dt'' dt'', \qquad (1)$$

where w(t) =  $\sigma_1[O('D)] + \sigma_2[D]$ ,  $\sigma_1 = K_1[D_2]$ ,  $\sigma_2 = K_2[O_3]$ ,  $\sigma_5 = K_5[O_3]$ , and  $\sigma_3 = K_3[CO_2]$ . We choose the experimental conditions such that the process (3) limits the excitation of the upper laser level. This takes place if

$$\sigma_1 >> \sigma_3, \quad \sigma_4 >> \sigma_3$$
 (2a)

$$\sigma_2 \gg \sigma_3 + \sigma_5$$
, (2b)

$$\gamma_2 \gg \sigma_3$$
, (2c)

where  $\sigma_4 = K_4[M] + K_6[O_3]$ . If the pump pulse is chosen such that

$$\sigma_1 \gg \gamma_2, \quad \sigma_4 \gg \gamma_2, \tag{3}$$

then the kinetics of the reaction admits of a simple treatment. Using the method of stationary concentrations for [O('D)] on the basis of (3), and taking (2a - 2c) into account, we obtain from (1)

$$[CO_2^*](t) = 2 \frac{\gamma_1}{\gamma_2^2} \frac{\sigma_1}{\sigma_1 + \sigma_4} \frac{\sigma_3}{\sigma_3 + \sigma_5} [O_3] \left(1 - e^{-(\sigma_3 + \sigma_5)t}\right). \tag{4}$$

Equating the population of the  $00^{\circ}1$  level at the instant  $\tau$  of the start of generation to the threshold value, which is equal to the population of the  $10^{\circ}0$  level, and solving the equation with respect to  $\sigma_3 + \sigma_5$ , we obtain

$$\sigma_3 + \sigma_5 = r^{-1} \ln (1 - X^{-1})^{-1}, \tag{5}$$

where

$$X = 2 \frac{\gamma_1}{\gamma_2^2} \frac{\sigma_1}{\sigma_1 + \sigma_4} \frac{\sigma_3}{\sigma_3 + \sigma_5} \frac{[O_3]}{[CO_2]} \exp(h\nu_{100}/kT)$$
 (6)

is the multiple by which the pump initiation exceeds the threshold.

Formulas (4) - (6) were used to reduce the experimental data. Since the generation power is proportional to the rate of formation of the excited molecules CO½, it follows from (5) that P  $\sim \frac{d}{dt} [\text{CO}_2^*] \sim \exp[-(\sigma_3 + \sigma_5)t]$ . From this, after determining the argument of the exponential in accordance with which the generation power decreases, we can determine the value of  $\sigma_3 + \sigma_5$ . On the other hand, if we measure the generation delay time  $\tau$ , then  $\sigma_3 + \sigma_5$  can be determined also from (5). In the latter case it is necessary to know the value of X, which depends on the efficiency of the photodissociation of O3. We shall show that X can be determined from the generation delay time in an experiment in which the rate of excitation of the O0°l level is limited by the process (0) and not by the process (3), i.e., instead of (2c) we should have

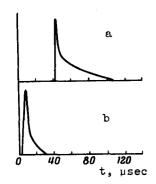
$$\sigma_3 + \sigma_5 >> \gamma_2. \tag{7}$$

Using the method of stationary concentrations for [O('D)], [D], and [OD\*], we get

$$[CO_2^*](r) = 2 \frac{\gamma_1}{\gamma_2^2} \frac{\sigma_1}{\sigma_1 + \sigma_4} \frac{\sigma_3}{\sigma_2 + \sigma_5} [O_3][1 - e^{-\gamma_2 r}(1 + \gamma_2 r)], \tag{8}$$

## Experimental conditions and results

1	2	3	4	5	6
2:8:1	1:8:1	1:8:1	1:8:0,5	2:8:4	1:8:4
2.7	2	1,35	1,35	14	28
13	20	34	30	7	8
2,95	2,78	2,78	2.95	2,5	2,1
0.68 · 10 5	0,31 • 10 5	0,2 <b>5 • 10</b> <sup>5</sup>	0,21·10 <sup>5</sup>		_
0,24 · 10 <sup>-12</sup>	0.39 • 10-12	0.76 · 10 <sup>-12</sup>	0.36·10 <sup>-12</sup>	_	_
0,5 · 10 <sup>-12</sup>	0,53 • 10-12	0.64 • 10-12	0.54 · 10 <sup>-12</sup>	_	_
	2.7 13 2.95 0.68 • 10 <sup>5</sup> 0.24 • 10 <sup>-12</sup>	2:8:1     1:8:1       2,7     2       13     20       2,95     2,78       0,68:105     0,31:105       0,24:10-12     0,39:10-12	$2:8:1$ $1:8:1$ $1:8:1$ $2,7$ $2$ $1,35$ $13$ $20$ $34$ $2,95$ $2.78$ $2.78$ $0.68 \cdot 10^5$ $0.31 \cdot 10^5$ $0.25 \cdot 10^5$ $0.24 \cdot 10^{-12}$ $0.39 \cdot 10^{-12}$ $0.76 \cdot 10^{-12}$	$2:8:1$ $1:8:1$ $1:8:1$ $1:8:0.5$ $2.7$ $2$ $1,35$ $1,35$ $13$ $20$ $34$ $30$ $2.95$ $2.78$ $2.78$ $2.95$ $0.68 \cdot 10^5$ $0.31 \cdot 10^5$ $0.25 \cdot 10^5$ $0.21 \cdot 10^5$ $0.24 \cdot 10^{-12}$ $0.39 \cdot 10^{-12}$ $0.76 \cdot 10^{-12}$ $0.36 \cdot 10^{-12}$	$2:8:1$ $1:8:1$ $1:8:1$ $1:8:0,5$ $2:8:4$ $2,7$ $2$ $1,35$ $1,35$ $14$ $13$ $20$ $34$ $30$ $7$ $2,95$ $2,78$ $2.78$ $2.95$ $2.5$ $0,68 \cdot 10^5$ $0,31 \cdot 10^5$ $0,25 \cdot 10^5$ $0,21 \cdot 10^5$ $ 0,24 \cdot 10^{-12}$ $0,39 \cdot 10^{-12}$ $0,76 \cdot 10^{-12}$ $0.36 \cdot 10^{-12}$ $-$



- a) Generation pulse,
- b) pump pulse, sweep
- 20 usec/div.

From the threshold condition and (8) it follows that

$$X^{-1} = 1 - e^{-\gamma_2 r} (1 + \gamma_2 r)$$

where  $\gamma_2$  and  $\tau$  are measurable quantities.

To perform experiments whose results could be processed by the described procedure, we chose conditions under which the chemical laser pulse occurs 20 - 30 µsec after triggering the pump lamp. This situation is observed at D2 pressures close to 1 Torr and O3 and CO2 pressures near O.1 Torr. Since there is no chain reaction, the presence of a prolonged delay suggests that the excitation of the CO2 molecules is indeed limited under these conditions by the energy-transfer process. The validity of relations (2a - 2c) is proved a posteriori, on the

basis of the measured value of  $K_3$  and the known values  $K_1 = 2 \times 10^{-10}$  cm<sup>3</sup>/sec [2],  $K_2 = 2.6 \times 10^{-11}$  cm<sup>3</sup>/sec [2],  $K_4 = 10^{-10}$  cm<sup>3</sup>/sec [3], and  $K_6 = 3 \times 10^{-12}$  cm<sup>3</sup>/sec [2]. At a mixture pressure 10 - 20 Torr, the delay of the generation becomes much shorter and does not exceed the length of the pump pulse. Measurement of the delay in such experiments (5 and 6 in the table) was used to determine the values of X by means of formula (9). From the known two values of X corresponding to different partial compositions of the mixture, we determined the ratio  $K_5/K_3$  = 8 and the value of  $\gamma_1$ . This has made it possible to find the values of X for experiments at low pressure, using formula (6). The experimental conditions and the measurement results are given in the table. Both procedures of determining K<sub>3</sub> lead to results that agree within a factor 2. The mean value of the transfer constant is  $(0.5 \pm 0.25) \times 10^{-12}$  cm<sup>3</sup>/sec. The employed procedure made it also possible to determine the constant  $K_5$ . The obtained value,  $K_5 = 4 \times 10^{-12}$ cm3/sec, is approximately equal to the rate constant of the reaction OH\* + O3 [4].

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EFFECT OF RADIO-FREQUENCY FIELD ON THE DISCRETE SATURATION SPECTRUM IN EPR

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We have observed an influence of a radio-frequency (RF) field on the discrete-saturation (DS) spectrum [1, 2]. This influence is manifest in a resonant attenuation of the DS spectrum. The RF influences the DS spectrum in a frequency interval on the order of the NMR line width.

The effects of an RF field under conditions of pulsed saturation of an EPR line were discussed in [3]. In that reference, in contrast to our results, they predicted the appearance of a spectrum of induced holes and selective