OPERATION OF A FOUR-LEVEL OPTICAL QUANTUM GENERATOR

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The properties of a four-level optical generator are studied theoretically and experimentally. The conditions for transition from four-level to three-level generation are found for the case of $CaF_2:U^{3+}$.

THIS paper presents an analysis of the effect of the parameters of the resonator, the working substance, and the pump intensity on the properties of an optical quantum generator (laser), on the basis of the steady-state theory of a four-level laser.[1,2] Experiments on $CaF_2:U^{3+}$ and $CaF_2:Sm^{2+}$ laser systems are reported.

In order to determine the density of generated radiation u_{32} (transition between levels 3 and 2) and the threshold density of the pump radiation $(u_{14})_n = u_n$, corresponding to the initiation of generation ($u_{32} = 0$), the following system of equations can be established for the steady-state population of the levels:

$$N_1 + N_2 + N_3 + N_4 = N_0,$$
$$u_{14}B_{14}(N_1 - N_4) - N_4(d_{43} + A_{41} + A_{42}) = 0,$$
$$u_{32}B_{23}(N_2 - N_3) + d_{43}N_4 - N_3(A_{31} + A_{32}) = 0,$$
$$d_{12}N_1 - d_{21}N_2 + u_{32}B_{23}(N_3 - N_2) + A_{32}N_3 + A_{42}N_4 = 0,$$
$$k = N_3 - N_2 = c\Delta\nu_{23}[\ln(1/R) + \sigma]/2hn\nu_{23}B_{23}l, \tag{1}$$

where N_1, N_2, N_3, and N_4 are the populations of the levels, N_0 is the total concentration of particles, A_{41}, A_{42}, A_{32}, A_{31}, B_{23}, and B_{14} are the Einstein coefficients for the corresponding transitions, d_{43}, d_{21}, and d_{12} are probabilities for radiation-less transitions, $\Delta\nu_{32}$ is the luminescence line width, R is the reflection coefficient of one of the resonator mirrors (the reflection coefficient of the second mirror is taken equal to 1), $\sigma = 2\sigma_0 l$ is the loss in the crystal (losses in the dielectric mirrors can be neglected), l is the length of the crystal.

Upon satisfying the inequalities

$$d_{21} \gg A_{31} + A_{32} + u_{14}B_{14} + A_{42},$$
$$d_{43} + A_{42} + A_{41} \gg u_{14}B_{14}, \qquad d_{43} \gg A_{31} + A_{32},$$
$$N_2/N_1 = d_{12}/d_{21} = \exp(-h\nu_{12}/kT) \ll 1,$$

we obtain the following solution to the system (1):

$$u_{32} = \frac{\eta_1 B_{14}}{B_{23}}\frac{N_0 - k}{k}(u_{14} - u_n), \tag{2}$$
$$u_n = \frac{A_{32}}{\eta B_{14}}\frac{N_0\exp(-h\nu_{12}/kT) + k}{N_0 - k}, \tag{3}$$

where

$$\eta_1 = d_{43}/(d_{43} + A_{42} + A_{41}), \qquad \eta = \eta_1 A_{32}/(A_{31} + A_{32})$$

is the quantum yield of the luminescence.

From the expressions for the radiation densities we can transform to expressions for laser power P_{32} and pump power P_{14}:

$$P_{32} = \frac{c}{2n}\beta\Delta\nu_{32}\int_s u_{32}\,ds$$
$$= \frac{c}{2n}\beta\Delta\nu_{32}\frac{\eta_1 B_{14}}{B_{23}}\frac{N_0 - k}{k}\left(\int_s u_{14}\,ds - \int_s u_n\,ds\right), \tag{4}$$
$$P_{14} = B_{14}N_1 h\nu_{14} l\int_s u_{14}\,ds = B_{14}h\nu_{14}(N_0 - k)\,l\int_s u_{14}\,ds, \tag{5}$$

where s is the cross-section area of the crystal and $\beta = 1 - R$ is the transparency of the mirror.

The expression (5) is obtained under the assumption that the crystal is optically dense.

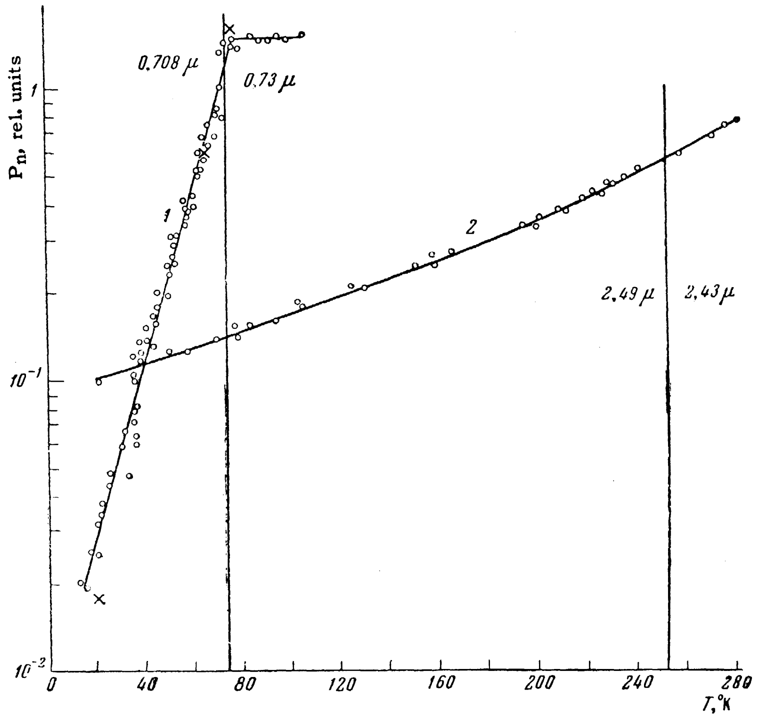
From Eqs. (2) and (5):

$$P_{32} = \eta_1\frac{\nu_{32}}{\nu_{14}}\beta(P_{14} - P_n)/(\ln\frac{1}{R} + \sigma), \tag{6}$$
$$P_n \sim \frac{A_{32}}{\eta N_0}\left[N_0\exp\left(-\frac{h\nu_{12}}{kT}\right) + \frac{c\Delta\nu_{32}(\ln R^{-1} + \sigma)}{2hn\nu_{32}B_{23}l}\right]. \tag{7}$$

It is not difficult to see that P_{32} attains its maximum value for some value of the transmission coefficient of the mirror β_0. Expressions for β_0 and the value $P_{32} = P_0$ corresponding to it are particularly lucid in the case when we can take $\ln(1/R) \approx 1 - R = \beta$ ($R \gtrsim 0.7$) and can neglect the first term in brackets in Eq. (7) (the case of sufficiently low temperature). As will be seen below, the experimental conditions correspond well to these assumptions. Then we obtain:

$$\beta_0 = \sigma(\sqrt{n_0} - 1), \tag{8}$$

FIG. 1. Dependence of threshold intensity on crystal temperature: 1 – $CaF_2:Sm^{2+}$, 2 – $CaF_2:U^{3+}$; points – experimental data, crosses – calculated data.



$$P_0 = \eta_1 \frac{\nu_{32}}{\nu_{14}} P_{14} \left(1 - \frac{1}{\sqrt{n_0}}\right)^2, \quad (9)$$

where $n_0 = P_{14}/(P_n)_{\beta=0}$ is the excess over threshold for $\beta = 0$; $(P_n)_{\beta=0}$ is the threshold pump power for $\beta = 0$.

It follows from (8) that the optimum value for transmission depends on the loss in the crystal and on the pump power.[1]

In order to check these relations, we investigated the dependence of threshold power and generated power in $CaF_2:U^{3+}$ and $CaF_2:Sm^{2+}$ lasers on the crystal temperature and the reflection coefficient of the resonator mirror, as well as the dependence of the generated power on the pumping power.

The experiments were carried out on crystals in the shape of cylinders whose end surfaces were covered with a dielectric reflecting layer. Excitation was effected by tubular flash lamps. The temperature of the crystals was varied between the limits 8—300°K. Temperatures below 80°K were obtained by cooling the crystals in a stream of gaseous helium obtained by vaporization of the liquid phase. The pass band of the measuring system permitted the recording of the instantaneous intensity of generation averaged over the individual flashes.

The threshold pump intensity was determined by measuring the pump intensity at the moment the generation disappeared.

Figure 1 shows the dependence of the threshold intensity on crystal temperature. In the case of $CaF_2:Sm^{2+}$ there is an extremely strong dependence of the threshold power P_n on temperature. It was found that in some crystals an intermittent change of generated wavelength from 0.708 to 0.73μ occurs at about 75°K. For $CaF_2:U^{3+}$ the temperature dependence of P_n is decidedly weaker. At about 253°K there also occurs an intermittent change in wavelength from 2.49 to 2.43μ.[2]

It is possible to compute the temperature dependence of P_n for $CaF_2:Sm^{2+}$ from Eq. (7). The data used in this calculation are: $l = 3.3$ cm, $n = 1.42$, $\beta + \sigma = 0.3$, $A_{32} = 0.5 \times 10^5\ \text{sec}^{-1}$, $h\nu_{12} = 265\ \text{cm}^{-1}$, and $N_0 \approx 1.6 \times 10^{18}\ \text{cm}^{-3}$ (measured from the 6900 Å absorption line in the sample). The temperature dependence of the quantum yield η and the halfwidth of the line $\Delta\nu_{32}$ were taken from Kaiser et al.[3] As can be seen from Fig. 1, the correspondence between experimental and theoretical results is completely satisfactory.

The change in generated wavelength in $CaF_2:U^{3+}$ is not difficult to explain if one considers that the probability of a transition at wavelength 2.43μ, as is established from the luminescence data, is greater than the probability of a transition at wave-

1) Expressions analogous to (8) and (9) can be obtained for the three-level scheme, but in this case n_0 depends in a more complicated way on the excess over threshold.

2) In some samples simultaneous generation at two wavelengths – 2.49 and 2.61 μ – is observed at 77° K.

length 2.49μ, and the populations of the corresponding levels equalize at high temperature (Fig. 2).

Similar considerations can be used to explain the change in wavelength in $CaF_2:Sm^{2+}$ (see Fig. 2).

Figure 3 shows the dependence of generated intensity on pump intensity. This can be seen to be a linear dependence, in accordance with Eq. (6).

The dependence of the generated intensity P_{32} and the threshold pump intensity P_n on the transmission coefficient β of the mirror is given in Fig. 4. It is seen that P_{32} attains a maximum value at $\beta \approx 0.17-0.25$. Considering that the excess over threshold in the experiment was about 6.7 in regime 2 and about 10 in regime 1, we find from Eq. (8) σ to be 0.09 and 0.11, respectively. For $CaF_2:Sm^{2+}$, $\sigma \approx 0.15-0.2$ ($l = 33$ mm). It was established that at high transmission ($\beta \geq 0.4$) $CaF_2:U^{3+}$ emits at 2.22μ, which corresponds to the three-level scheme (Fig. 2). To get emission at λ_{31} it is necessary to satisfy the condition

$$N_3 - N_1 = c\Delta\nu_{13}\left(\ln\frac{1}{R} + \sigma\right)/2hn\nu_{13}B_{13}l.$$

Using the solution to the system of equations (1) for the populations of the levels $N_3 - N_1 = 2k - N_0$, the following relation can be obtained determining the value of the reflection coefficient for which three-level generation can be expected:

$$R \leqslant \exp\left\{\sigma\left[1 - \frac{N_0}{k_0}\left(2 - \frac{\nu_{31}^2\Delta\nu_{31}A_{32}}{\nu_{32}^2\Delta\nu_{32}A_{31}}\right)^{-1}\right]\right\}, \quad (10)$$

where k_0 is the value of k for $\beta = 0$.

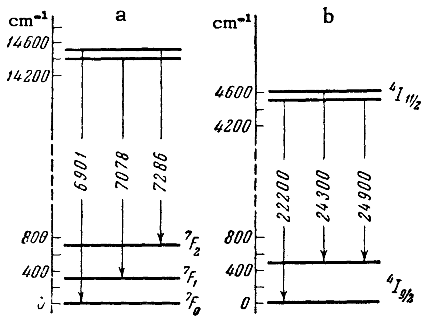


FIG. 2. Energy level schemes for $CaF_2:Sm^{2+}$ (a) and $CaF_2:U^{3+}$ (b); the numbers on the arrows refer to wavelengths in angstroms.

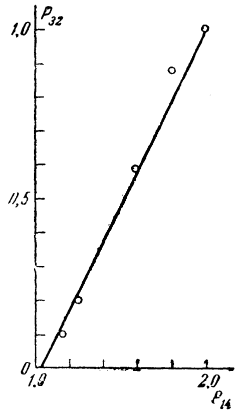


FIG. 3. Dependence of generated intensity on pump intensity ($CaF_2:U^{3+}$, T = 78° K).

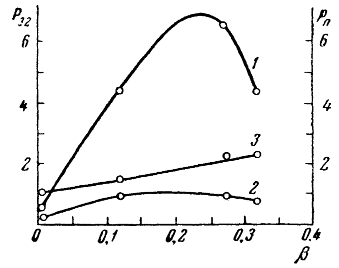


FIG. 4. Dependence of generated intensity P_{32} and threshold intensity P_n on the transmission coefficient of the mirror ($CaF_2:U^{3+}$, T = 78° K). Curves 1 and 2 are for P_{32} (1—voltage on pumping lamp, 1000 V; 2—700 V), and 3 for P_n.

Using the values obtained on the basis of luminescence and laser studies in $CaF_2:U^{3+}$ ($N_0 \approx 4.9 \times 10^{17}$ cm^{-3}, $k_0 \approx 3.6 \times 10^{16}$ cm^3, $A_{32}\Delta\nu_{31}/A_{31}\Delta\nu_{32} = 0.067$, $\sigma \approx 0.1$, $\Delta\nu_{32} = 1.3 \times 10^{12}$ sec^{-1}, $l = 2.5$ cm), we find from Eq. (10) that $R < 0.48$. Agreement with experiment is seen to be completely satisfactory.

Thus the results of experiments show that the elementary theory of steady-state generation developed above satisfactorily describes the principal features of a four-level laser.

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[2] Anan'ev, Gribkovskiĭ, Mak, and Stepanov, DAN SSSR, in press.

[3] Kaiser, Garrett, and Wood, Phys. Rev. **123**, 766 (1961).

Translated by L. M. Matarrese
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