# INFORMATIONAL CONTENT OF LUMINESCENCE OF THE ACTIVE MEDIUM OF A FREE-RUNNING LASER

# V. V. GRIGOR'YANTS

Institute of Radio Engineering and Electronics, USSR Academy of Sciences

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An analytic relation between the luminescence drops due to stimulated emission and effective radiation cross section, excitation transfer rate between the active ions, photon flux, and other laser parameters is established on the basis of kinetic equations for the integral and spectral populations of an inhomogeneously broadened metastable level of the active laser medium. Experimental data are obtained for Nd<sup>3+</sup> ions in various matrices by observing luminescence drops directly in the laser or in a small external active specimen irradiated by auxiliary laser.

# INTRODUCTION

THE behavior of luminescence from an active material exposed to its own or to an external coherent radiation has been the object of steadily increasing attention during the recent years [1-5]. This is first of all due to the fact that the nature of the luminescence drop, i.e., of the luminescence intensity decrease caused by a portion of the excited particles participating in stimulated emission, depends on such important laser parameters as effective cross section of stimulated emission, type of spectral line broadening, rate of excitation transfer between active particles, radiation energy, losses, etc., and it is possible to determine the role played by each such factor through suitable experimental arrangement. However the majority of the noted research efforts either deal with individual aspects of the problem or are qualitative in nature. On the other hand, as we show below, the kinetic equations describing population changes in the laser allow us to obtain the relation between the luminescence drops and the majority of the above laser parameters in a convenient analytical form, and to select and design suitable experiments.

Figure 1 shows the basic diagram of the experimental setup for the investigation of luminescence drops in lasers using neodymium glass as the active medium. Figure 2 shows typical oscillographic traces of neodymium ion luminescence in the absence and presence of free-running operation. The fundamental assumption used throughout this work is that the luminescence intensity observed in the setup shown in Fig. 1 is proportional to the population of the  ${}^{4}F_{3/2}$  metastable level of Nd<sup>3+</sup>. The measures necessary to accomplish this as well as the experimental details and results are given in the appropriate section of this paper.

# BEHAVIOR OF WORKING-LEVEL POPULATION DURING FREE-RUNNING OPERATION OF A LASER

Our investigation begins with analysing the behavior of the spectral and integral populations of an inhomogeneously broadened working level of a laser in free-



FIG. 1. Diagram of setup for the observation of luminescence from an active laser medium. 1-nontransparent mirror,  $r_1 = 1$ ; 2-output mirror,  $r_2 = 0.7$ ; 3-laser reflector; 4-diaphragm d = 4 mm; 5-beam splitter plate; 6-calorimeter; 7-blocking filter for  $\lambda = 1.06\mu$ ; 8-lens; 9-diaphragm d  $\approx$  1 mm, placed in the focus of lens 8; 10-DMR-3 monochromator; 11-photomultiplier; 12-oscilloscope; the input channel in the upper part of the diagram is used to measure F in the presence of a frequency selector in the laser resonator.

running operation according to a four-level scheme. These populations are denoted respectively by  $n^{(1)}(\nu, t)$  and  $N^{(1)}(t)$  in the absence of generation and by  $n^{(2)}(\nu, t)$  and  $N^{(2)}(t)$  in the presence of generation.

The equation describing the spectral density behavior of the population  $n^{(2)}(\nu, t)$ , assuming weak pumping and zero population of the lower working level, has the form<sup>[6,7]</sup>:

$$\frac{dn^{(2)}(\mathbf{v},t)}{dt} = W(t)N_0g(\mathbf{v},\mathbf{v}_0) - A_0n^{(2)}(\mathbf{v},t)$$
(1)  
$$^{(2)}(\mathbf{v},t) \quad \sigma_i f(\mathbf{v},\mathbf{v}')I(\mathbf{v}',t)d\mathbf{v}' + F[n_p \quad (\mathbf{v},t) - n^{(2)}(\mathbf{v},t)],$$

where  $\sigma_i$  is the stimulated emission cross section of the luminescence center,  $f(\nu, \nu')$  is the luminescence line shape of this center with a natural frequency  $\nu = \nu'$ ,  $I(\nu', t)$  is the spectral density of photon flux in the laser active medium, W(t) is the probability of ions being pumped to the working level,  $N_0$  is the concentration of active ions in the laser material,  $A_0$  $= 1/\tau_0$  is the probability of decay of the working level,  $\tau_0$  is the lifetime of excited ions at this level,  $g(\nu, \nu_0)$ is the equilibrium form of inhomogeneously broadened spectral line

$$\int_{0}^{\infty} g(v, v_0) \, dv = 1$$

 $\nu_0$  is the central frequency, F is the excitation trans-



FIG. 2. Curves of integral luminescence of the laser active material ( $\lambda \approx 0.88\mu$ ). 1–luminescence in the absence of generation, 1000 J pump level (200 J generation threshold); 2–luminescence in the presence of generation at the same pump level,  $E_{rad} \approx 1.9$  J,  $r_1 =$ 0.62; 3–luminescence in the absence of generation, 180 J pump level.

fer rate between the ions, and  $n_p^{(2)}(\nu, t)$  is the equilibrium spectral density of population related by definition to  $N^{(2)}(t)$  by the expression

$$n_{\rm p}^{(2)}(v,t) = g(v,v_0) N^{(2)}(t).$$
<sup>(2)</sup>

The instantaneous spectral population density  $n^{(2)}(\nu, t)$  can be related to  $N^{(2)}(t)$  by an analogous expression

$$n^{(2)}(v, t) = g'(v, v_0, t) N^{(2)}(t),$$
(3)

where  $g'(\nu, \nu_0, t)$  is the nonequilibrium spectral line shape occurring during stimulated emission in which

$$\int_{0}^{\infty} g'(v, v_0, t) dv = 1.$$

In the case of homogeneous broadening of the spectral line the integral population of the working level in an active laser medium in free-running operation is determined by the self-excitation condition and remains practically constant during generation<sup>[8]</sup>. In the case of inhomogeneous broadening of the spectral line the behavior of  $N^{(2)}(t)$  in free-running generation depends to a considerable degree on the excitation transfer rate F and the width of the laser generation spectrum. In fact, we assume that the generation occurs at the central frequency  $\nu_0$ , and the width of the generated spectrum is significantly less than the homogeneous width of the elementary center line. It is obvious that under these conditions the spectral population density at the frequency  $\nu_0$  and in its vicinity of the order of  $f(\nu, \nu')$  line width should remain constant during generation, i.e., according to (3)

$$\frac{dn^{(2)}(\mathbf{v}_0,t)}{dt} = \frac{dg'(\mathbf{v}_0,\mathbf{v}_0,t)}{dt} N^{(2)}(t) + \frac{dN^{(2)}(t)}{dt}g'(\mathbf{v}_0,\mathbf{v}_0,t) = 0,$$
(4)

whence

$$\frac{dN^{(2)}(t)}{dt} = -\frac{N^{(2)}(t)}{g'(v_0, v_0, t)} \frac{dg'(v_0, v_0, t)}{dt}.$$
 (5)

It follows from (5) that in the general case for an inhomogeneously broadened line  $N^{(2)}(t)$  is a convex

function of time passing through a maximum at the point in time when the depth of the spectral dip is also maximum.

 $N^{(2)}(t)$  can be a constant only if  $dg'(\nu_0, \nu_0, t)/dt = 0$ . This is possible either when  $F \rightarrow \infty$  or when the laser generation spectrum is broad, of the order of a spectral line width. In both cases the presence of generation fails to cause any noticeable spectral dip.

In the papers<sup>[9,10]</sup> reporting on the measurement of the effective cross section  $\sigma_{eff}^{(1)}$  of stimulated emission from the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  transition of the Nd<sup>3+</sup> ions in silicate glass the integral population of the  ${}^{4}F_{3/2}$ metastable level was assumed time-independent during generation; strictly speaking, this is incorrect in view of the above considerations. However the good agreement of the measured values of  $\sigma_{eff}$  with data reported elsewhere<sup>[11-13]</sup> shows that the spectral line shape of Nd<sup>3+</sup> in silicate glass little differs from the equilibrium line even when the generated spectral width in freerunning operation is several tens of angstroms. This means that the integral population of the metastable level varies negligibly during generation under these conditions, a fact confirmed by direct observation of the behavior of N<sup>(2)</sup>(t) described below.

# RELATION OF LUMINESCENCE DROP TO THE EXCITATION TRANSFER RATE BETWEEN IONS

Although the time dependence of  $N^{(2)}$  in the course of generation carries information on the excitation transfer rate F between ions, it is difficult to obtain an analytic relation between  $N^{(2)}(t)$  and F. This is due to the fact that the behavior of  $N^{(2)}(t)$  also depends on the excess over threshold, the spectral width of the generated signal, losses in the medium, the nature of spectral line broadening, etc. The transfer rate can be more simply related to the luminescence drop of the

<sup>&</sup>lt;sup>1</sup>)With inhomogeneous broadening of the spectral line gain is determined by the quantity  $\sigma_{\text{eff}} = \sigma_i \delta_i / \Delta$ , where  $\delta_i$  and  $\Delta$  are the homogeneous and inhomogeneous widths of the spectral line respectively [<sup>6</sup>].

active medium resulting from the participation of a portion of the excited particles in stimulated emission.

For this purpose, using (1) and (2), we write the equations for  $n^{(1)}(\nu, t)$ ,  $n^{(2)}(\nu, t)$ , and  $n_{D}^{(2)}(\nu, t)$  on the assumption that the frequency  $\nu$  is sufficiently far from the stimulated emission frequency. This permits us to neglect stimulated transitions in describing the behavior of  $n^{(2)}(\nu, t)$  during generation. We have

$$\frac{dn^{(1)}(\mathbf{v},t)}{dt} = W(t)N_0g(\mathbf{v},\mathbf{v}_0) - A_0n^{(1)}(\mathbf{v},t),$$
(6)

$$\frac{dn^{(2)}(\mathbf{v},t)}{dt} = W(t)N_0g(\mathbf{v},\mathbf{v}_0) - A_0n^{(2)}(\mathbf{v},t) + F[n_p^{(2)}(\mathbf{v},t) - n^{(2)}(\mathbf{v},t)],$$
(7)
$$\frac{dn_p^{(2)}(\mathbf{v},t)}{dt} = W(t)N_0g(\mathbf{v},\mathbf{v}_0) - A_0n_p^{(2)}(\mathbf{v},t)$$

$$-g(\nu,\nu_0)\int n^{(2)}(\nu,t)\,d\nu\int\sigma_i f(\nu,\nu')I(\nu',t)\,d\nu'.$$
(8)

Subtracting (8) from (7) and from (6) and introducing the notation

$$\Delta n(\mathbf{v},t) = n^{(1)}(\mathbf{v},t) - n_{\rm p}^{(2)}(\mathbf{v},t), \ \Delta n^{*}(\mathbf{v},t) = n^{(2)}(\mathbf{v},t) - n_{\rm p}^{(2)}(\mathbf{v},t),$$
$$B = g(\mathbf{v},\mathbf{v}_{0}) \int n^{(2)}(\mathbf{v},t) d\mathbf{v} \int \sigma_{i} f(\mathbf{v}',t) I \, d\mathbf{v}',$$

we obtain

$$\frac{d\Delta n(\mathbf{v},t)}{dt} = -A_0 \Delta n(\mathbf{v},t) + B,$$
(9)

$$\frac{d\Delta n^*(\mathbf{v},t)}{dt} = -(A_0 + F)\Delta n^*(\mathbf{v},t) + B.$$
(10)

Integrating (9) and (10) over t within the limits from t = 0 to  $t = \infty$  and eliminating  $\int B dt$  we find an expression relating the luminescence drop due to generation to excitation transfer and spontaneous decay rates:

$$\frac{F + A_0}{A_0} = \frac{\Delta S}{\Delta S^*}; \qquad (11)$$
  
$$\Delta S \sim \int_{0}^{\infty} \Delta n(\mathbf{v}, t) dt, \quad \Delta S^* \sim \int_{0}^{\infty} \Delta n^*(\mathbf{v}, t) dt;$$

Here  $\Delta S$  in the area between curves  $n^{(1)}(\nu, t)$  and  $n_p^{(2)}(\nu, t)$  in arbitrary units, and  $\Delta S^*$  is the area be-tween curves  $n^{(2)}(\nu, t)$  and  $n_p^{(2)}(\nu, t)$  in the same units. The feasibility of experimental observation of areas

 $\Delta S$  and  $\Delta S^*$  is indicated by the relations

$$n^{(1)}(v,t) = g(v,v_0) N^{(1)}(t), \quad n_{\rm p}^{(2)}(v,t) = g(v,v_0) N^{(2)}(t),$$

It follows from these expressions that if the oscilloscopic trace  $N^{(1)}(t)$  is made to coincide with the trace  $n^{(1)}(\nu, t)$  by adjusting oscilloscope gain or graphically, the curve  $N^{(2)}(t)$  will be equivalent to curve  $n_p^{(2)}(\nu, t)$ after a similar scale adjustment<sup>[7]</sup>. Under these conditions  $\Delta S$  represents the area between curves  $N^{(1)}(t)$ and  $N^{(2)}(t)$ , and  $\Delta S^*$  the area between curves  $n^{(2)}(\nu, t)$  and  $N^{(2)}(t)$  (Fig. 3).

We now analyze (11). When  $F \rightarrow \infty$  the quantity  $\Delta S^* \rightarrow 0$ , i.e., there is no dip in the spectral line which behaves as a homogeneous line and, according to (5),  $N^{(2)}(t) = const.$  during generation.

When  $F \rightarrow 0$  the quantity  $\Delta S^* \rightarrow \Delta S$ . This means that curves  $n^{(2)}(\nu, t)$  and  $n^{(1)}(\nu, t)$  coincide, i.e., the generation at a frequency  $\nu_{gen}$  sufficiently distant from  $\nu$  fails to affect in any way the behavior of the spectral line wings; these are therefore regions of ordinary luminescence according to (6). In this case



FIG. 3. Superimposed curves of luminescence (external specimen).  $1-I_1 = k_1 n^{(1)}(\nu, t) = k_2 N^{(1)}(t); 2-I_2 = k_2 N^{(2)}(t) = k_1 n_p^{(2)}(\nu, t); 3 I_3 = k_1 n^{(2)}(\nu, t), k_1 = k_2 g(\nu_1, \nu_0).$ 

the integral population  $N^{(2)}(t)$  should vary significantly during generation<sup>[10]</sup>.

Equation (11) provides a convenient and simple method for the experimental determination of F which is one of the most interesting and important characteristics of laser material. However the structure of (11) shows that it may be appropriate for use if the assumed transfer rate F lies within the range 0.1  $A_0 \gtrsim F \gtrsim 10$ A<sub>0</sub>. Satisfactory accuracy of measurement is difficult outside this range where only qualitative data on the transfer rate are possible. The measurement accuracy can be improved, especially near the ends of this range, if the luminescence drop  $\Delta S$  amounts to a significant (not less than 20-30%) portion of the area under the luminescence curve in the absence of generation. It is obvious that  $\Delta S$  should grow with increasing total probability of stimulated emission in the investigated material; the latter is proportional to the photon flux intensity I(t) causing the luminescence drop and to the effective cross section  $\sigma_{eff}$  of stimulated emission.

# RELATION OF LUMINESCENCE DROP TO THE EF-FECTIVE CROSS SECTION OF STIMULATED EMISSION AND PHOTON FLUX THROUGH THE ACTIVE LASER MATERIAL

As we showed above the area  $\Delta S$  between curves  $n^{(1)}(\nu, t)$  and  $n_p^{(2)}(\nu, t)$  coincides with the area between curves  $N^{(1)}(t)$  and  $N^{(2)}(t)$ , provided curves  $n^{(1)}(\nu, t)$ and  $N^{(1)}(t)$  were brought into coincidence. Therefore the luminescence drop  $\Delta S$  can be evaluated with the aid of equations describing the behavior of integral populations  $N^{(1)}(t)$  and  $N^{(2)}(t)$  respectively:

$$\frac{dN^{(1)}(t)}{dt} = W(t)N_0 - A_0 N^{(1)}(t), \qquad (12)$$

$$\frac{dN^{(2)}(t)}{dt} = W(t)N_0 - A_0 N^{(2)}(t) - \sigma_{\rm eff} I(t) N^{(2)}(t).$$
(13)

Integrating these equations over t, from t = 0 to  $t = \infty$ , subtracting one result of integration from the other, and taking the theorem of the mean into account we obtain:

$$\frac{\int_{0}^{[N^{(1)}(t) - N^{(2)}(t)]dt}}{N^{(2)}(t_{av})} = \frac{\sigma_{eff} Q_{0}}{A_{0}}$$
(14)

where  $Q_0$  is the total photon flux passing through 1 cm<sup>2</sup> of the cross section of the active rod during a laser pulse:

$$Q_0 = \int_0^\infty I(t) \, dt$$

Considering that

$$\int_{0}^{\infty} \left[ N^{(1)}(t) - N^{(2)}(t) \right] dt \sim \Delta S,$$

i.e., areas between curves  $N^{(1)}(t)$  and  $N^{(2)}(t)$ , and  $N^{(2)}(t_m) \sim \overline{H}$  are equal to a mean amplitude of curve 2 during generation (Fig. 2), we find the relation between the experimentally observed relative luminescence drop and the values of effective cross section of stimulated emission and total photon flux:

$$\Delta S / \overline{H} = \sigma_{\text{eff}} Q_0 / A_0 a; \tag{15}$$

 $\Delta S$  and  $\overline{H}$  are measured directly from the oscillographic luminescence trace (Fig. 2) in arbitrary units (for example in cm<sup>2</sup> and cm respectively), and a is the scale of the axis of abscissas in sec/cm.

Since in deriving (11) and (15) we used only equations for the working level populations of the active material, both (11) and (15) can be also used in analyzing data on luminescence drops in a small excited specimen exposed to resonant coherent radiation from an auxiliary laser (Fig. 3)<sup>[13]</sup>. From the experimenter's point of view such a setup is in many cases more convenient and flexible. This is primarily due to the fact that the requirements of optical homogeneity and finish quality are relatively low when the investigated specimen is small and external to the laser; it is considerably easier to vary specimen temperature, the range of possible concentrations of the activator in the specimen is widened, and new matrix materials can be investigated that have not been previously known to generate with the given activator.

For an external small specimen ( $\sim 10 \times 10 \times 2$  mm), that permits us to neglect losses and gain, the total photon flux Q<sub>0</sub> in (15) causing the luminescence drop is expressed by

$$Q_0 = E_{\rm rad} / hvS_{\rm sp.},$$

where  $E_{rad}$  is the radiation energy of the auxiliary laser incident on the specimen, and  $S_{Sp}$  is the irradiated area of the specimen. If necessary, the total photon flux passing through the investigated specimen can be significantly increased by placing the specimen inside the resonator of the auxiliary laser.

We now return to the luminescence drop in the internal active medium of the laser. We find the relation between the total photon flux inside the laser resonator and the energy radiated through the semitransparent output window.

To make it more general we consider that the photon flux varies along the laser rod, i.e., I = I(t, x). This is true if one mirror has a low reflection coefficient or in the case of a laser with a rotating-prism Q-switch. Since as a rule the prism Q-switch does not operate instantaneously, there may be a considerable light loss in the prism due to a generation time mismatch<sup>[14]</sup>. In this sense the prism is equivalent to a high-transmission mirror causing an uneven photon flux along the active rod in a Q-switched laser<sup>2</sup>).

We consider a laser in which the photon flux along the active rod is nonuniform due to one of the above reasons. We assume that the luminescence drop is observed sidewise at the end of the rod facing the output mirror with a reflection coefficient  $r_1(x = l)$  (Fig. 1). The photon flux passing through the indicated end of the active rod in both directions is

$$(l, t) (1 + r_1)$$

where  $I^{+}(l, t)$  is the photon flux toward the output mirror. Since

$$(1-r_1)\int_0^\infty I^+(l,t)\,dt = \frac{E_{\rm rad}}{hvS\,{\rm sp}}$$

the total photon flux passing through the rod end under consideration during the generation period is  $^{3)}$ 

$$Q_0 = (1+r_1) \int_{0}^{\infty} I^+(l,t) dt = \frac{1+r_1}{1-r_1} \frac{E_{\text{rad}}}{h_V S_{\text{sp}}}.$$
 (16)

Equation (16) is valid also when  $r_1 \approx r_2 \approx 1$  when the photon flux is practically constant along the rod. In this case the luminescence drop can be observed either sidewise at both ends of the rod or along its axis, using (16) in all cases to determine  $Q_0$ .

Substituting (16) into (15) we obtain a formula for the evaluation of the relative luminescence drop that can be obtained in the given laser:

$$\frac{\Delta S}{\overline{H}} = \frac{(1+r_1)\sigma_{\rm eff}E_{\rm rad}}{a(1-r_1)h_{\rm V}A_0S_{\rm sp}}$$
(17)

However the basic merit of (17) lies in its use to measure  $\sigma_{eff}$ , a quantity difficult to determine in fourlevel systems. The best accuracy of determination of  $\sigma_{eff}$  from (17) can be achieved with homogeneous broadening of the spectral line of the laser active medium, when  $N^{(2)}(t) = N_{th} = \text{const}$  during generation. However even in the case of inhomogeneous broadening, for example for Nd<sup>3+</sup> in silicate glass pumped with double or triple excess over threshold, the deviation of N<sup>(2)</sup>(t) from N<sup>(2)</sup>(t<sub>m</sub>) is small and as a rule amounts to about 20-30%. This means that the possible error in the experimental determination of  $\overline{H}$  from the oscilloscopic trace of luminescence drop (Fig. 2) will be still smaller.

We neglected in the above computations the effect of superradiance that shortens the lifetime of excited particles at the working level of the laser active material. Consequently (11), (15), and (17) that are valid from this viewpoint for small external specimens (Fig. 3) may require a suitable correction in the application to a laser, especially in the case of a high excess over pump threshold. For this purpose to correct (15), the value of  $A_0$  in (12) should be regarded as a function of population  $N^{(1)}(t)$  and consequently of time due to the superradiance effect. On the other hand the role of superradiance is negligible in (13) since the population  $N^{(2)}(t)$  is close to the threshold value during generation, ensuring a low total gain in the active material when the reflection coefficients of both laser mirrors are of the order of unity. The above considerations applied after the described operations on (12) and (13) lead to the following modification of (15)

$$\frac{\overline{AS^{(1)}-A_0S^{(2)}}}{\overline{H}} = \frac{\sigma_{\text{eff}}Q_0}{a}; \qquad (15a)$$

<sup>&</sup>lt;sup>2)</sup>These considerations and the subsequent derivation of (16) should be taken into account when using the results of  $[^{13,15,16}]$ .

<sup>&</sup>lt;sup>3)</sup>It is obvious that (16) also determines the total photon flux causing luminescence drop in a small active specimen when the latter is placed inside the auxiliary laser resonator next to the output mirror.

Here  $S^{(1)}$  and  $S^{(2)}$  are the areas under curves  $N^{(1)}(t)$  and  $N^{(2)}(t)$  respectively, and

$$\bar{A} = \int_{0}^{\infty} A(t) N^{(1)}(t) dt \Big/ \int_{0}^{\infty} N^{(1)}(t) dt$$

is a time-averaged probability of decay of the working level whose evaluation is described below. In the absence of superradiance  $A = A_0$  and (15a) converts into (15).

# DETERMINATION OF LIFETIME $\tau_0$ FROM LUMINESCENCE DROP

We must know the value of  $\tau_0 = 1/A_0$  both to determine mine the transfer rate F from (11) and to determine the luminescence drop or radiation cross section  $\sigma_{eff}$ from (17). The generally accepted method of measuring lifetime with a pulse taumeter<sup>[17]</sup> is inconvenient in this case because the lifetime of particles at the working level of the laser material can be significantly shortened by superradiance and is thus different from that measured by an ordinary taumeter. Furthermore such a taumeter is not always available to the experimenter.

An analysis of luminescence behavior in active laser material in the absence and presence of generation shows that the lifetime  $\tau_0$ , just as F and  $\sigma_{eff}$ , can be determined from oscilloscopic luminescence traces of the type shown in Fig. 2. In fact the equations describing the behavior of integral populations  $N^{(1)}(t)$  and  $N^{(2)}(t)$  for times  $t \ge t_k$ , where  $t_k$  is the end point of free generation of the laser, are

$$\frac{dN^{(1)}(t)}{dt} = W(t)N_0 - \frac{N^{(1)}(t)}{\tau_0},$$
(18)

$$\frac{dN^{(2)}(t)}{dt} = W(t)N_0 - \frac{N^{(2)}(t)}{\tau_0}.$$
 (19)

(20)

Subtracting (19) from (18) and introducing the notation  $\Delta N(t) = N^{(1)}(t) - N^{(2)}(t)$  we obtain

 $\frac{d\Delta N(t)}{dt} = -\frac{\Delta N(t)}{\tau_0},$ 

whence

$$\Delta N(t) = \Delta N(t_0) \exp \{-(t - t_0) / \tau_0\}.$$
 (21)

Thus the lifetime  $\tau_0$  can be found from the time variation of the difference between the ordinates of luminescence curves of the active material in the absence and presence of generation. We should remember however that, as we noted above, the values of  $\tau_0$  in (18) and (19) can differ from one another and vary in time due to superradiance. This means that the lifetime determined from (21) is somewhat depressed in this case in comparison with the true  $\tau_0$  if  $t_0 \approx t_k$  is assumed in (21). If the luminescence signal is sufficiently high in relation to noise, the time  $t_0$  should be selected so as to have  $N^{(1)}(t_0) < N^{(2)}(t_k) \approx N^{(2)}_{th}$  i.e.,  $\tau_0$  should be measured in those regions of the luminescence curves in which superradiance is negligible.

When luminescence drop of small external specimens irradiated by an auxiliary laser is investigated, the above method of determining  $\tau_0$  yields results that coincide with the taumeter data since superradiance is practically nonexistent in small specimens several millimeters in size at real pump levels<sup>4)</sup>. To determine  $\overline{A} = 1/\overline{\tau}$  in (15a) we integrate (18) over time from zero to infinity, setting  $\tau_0 = \tau(t)$ :

$$N_0 \int_0^\infty W(t) dt = \int_0^\infty \frac{N^{(1)}(t)}{\tau(t)} dt = \frac{1}{\bar{\tau}} \int_0^\infty N^{(1)}(t) dt.$$
(22)

We further assume that we are observing luminescence of the laser active material in the absence of generation at two different pump levels: working pump power  $W_1(t)$  and subthreshold pump power  $W_2(t)$  with no superradiance, i.e.,  $\tau(t) \equiv \tau_0$ . We then obtain from (22)

$$\boldsymbol{\tau} = \frac{1}{\overline{A}} = \frac{\boldsymbol{\varkappa} S_1^{(1)}}{S_2^{(1)}} \boldsymbol{\tau}_0, \qquad (23)$$

where  $S_1^{(1)}$  and  $S_2^{(1)}$  are areas under curves  $N_1^{(1)}(t)$ and  $N_2^{(1)}(t)$  respectively (see Fig. 2, curves 1 and 3) in arbitrary units; and

$$\kappa = \int_{0}^{\infty} W_{2}(t) dt / \int_{0}^{\infty} W_{1}(t) dt = U_{2}^{2}/U_{1}^{2}$$

is the ratio of pump powers in both cases equal to the ratio of the corresponding squared voltages  $U_2$  and  $U_1$  at the capacitor bank of the laser.

# RELATION BETWEEN LUMINESCENCE DROP AND LOSSES IN THE ACTIVE MEDIUM OF THE LASER

We have shown above that the luminescence drop can be used to determine such parameters of the laser active material as transfer rate F, effective cross section of stimulated emission  $\sigma_{eff}$ , and lifetime of excited particles at the working level  $\tau_0$ . However this does not exhaust the information carried by the luminescence of the laser active material. It turns out that apart from these purely physical quantities the same method can also be used to determine some energy characteristics of lasers, such as total parasitic emission losses in the medium and resonator. To find these losses luminescence drops are compared for two different laser output mirrors having reflection coefficients  $r_1$  and  $r_2$ .

If a change of the output mirror leaves other laser parameters unchanged, the balance of active particles can be written for these two cases (taking account of superradiance) in the following form:

$$\int_{0}^{\infty} \left[ \frac{N^{(1)}(t)}{\tau(t)} - \frac{N_{1}^{(2)}(t)}{\tau_{0}} \right] dt = \frac{E_{1 \operatorname{rad}}(\beta_{1 \operatorname{rad}} - \beta_{0})}{h_{v} V \beta_{1 \operatorname{rad}}},$$
(24)

$$\int_{0}^{\infty} \left[ \frac{N^{(1)}(t)}{\tau(t)} - \frac{N_{2}^{(2)}(t)}{\tau_{0}} \right] dt = \frac{E_{2rad} \left(\beta_{2rad} + \beta_{0}\right)}{h_{V} V \beta_{2rad}}$$
(25)

where  $\beta_0$  are parasitic losses in the active medium and resonator of the laser,

$$\beta_{2rad} = \frac{1}{2l} \ln \frac{1}{r_1}, \ \beta_{2rad} = \frac{1}{2l} \ln \frac{1}{r_2}$$

are losses in emission through the removable output mirror (the second mirror is considered non-transmitting),  $V = S_{Sp}l$  is the volume of the generating region of the active laser rod,  $S_{Sp}$  and l are the effective cross section and length of the rod respectively,  $E_{1}$  rad and  $E_{2}$  rad are emission energies for the reflection coefficients of the output mirror of  $r_{1}$  and  $r_{2}$ respectively, and  $N_{1}^{(2)}(t)$  and  $N_{2}^{(2)}(t)$  are instantaneous integral populations of the working level of the laser active material for  $r = r_{1}$  and  $r = r_{2}$  respectively.

<sup>&</sup>lt;sup>4)</sup>The above method of determining  $\tau_0$  can be also used in the case of luminescence drops due to a giant pulse [<sup>7,13</sup>].

After dividing (24) by (25) we obtain a formula to determine:

$$\frac{\beta_{0} + \beta_{2} \operatorname{rad}}{\beta_{0} + \beta_{1} \operatorname{rad}} \stackrel{\beta_{2} \operatorname{rad} \mathcal{E}_{1} \operatorname{rad}}{\beta_{1} \operatorname{rad} \mathcal{E}_{2} \operatorname{rad}} \frac{(\tau_{0} S^{(1)} - \bar{\tau} S_{2}^{(2)})}{(\tau_{0} S^{(1)} - \bar{\tau} S_{1}^{(2)})};$$
(26)

Here  $S^{(1)}$ ,  $S_1^{(2)}$ , and  $S_2^{(3)}$  are areas (in arbitrary units) under curves  $N^{(1)}(t)$ ,  $N_1^{(2)}(t)$ , and  $N_2^{(2)}(t)$ .

In deriving (26) we assumed that the volume of the generating region of the active laser rod does not depend on the reflection coefficient of the output mirror.

Since in the course of generation parasitic losses can vary in time, for example from heating of the active rod, (26) actually yields the magnitude of these losses averaged over the generation time. The change in  $\beta_0$  causes an analogous change in the laser excitation condition, i.e., a change in the threshold population of the working level. The change in the threshold population in the course of generation in turn affects the time-dependent behavior of  $N^{(2)}(t)$ . However the separation of the contribution due to the time-dependence of  $\beta_0$  from N<sup>(2)</sup>(t) is a fairly complex experimental problem especially with inhomogeneous broadening of the spectral line of the active medium. In fact as we pointed out above (see (5)),  $N^{(2)}(t)$  is in this case a function of a number of laser parameters whose individual effects can hardly be sorted out. Therefore the above method could be used to study loss kinetics primarily in lasers based on active media with homogeneously broadened spectral lines (such as ruby) where in the ideal case  $N^{(2)}(t) = N_{th}^{(2)} = const during generation. If the effect of nonuniform generation over$ the cross section of the active rod<sup>[16]</sup> has been eliminated in the observation of luminescence drop in such a laser, the time dependence of  $N^{(2)}(t)$  uniquely indicates an analogous time dependence of parasitic losses.

# EXPERIMENTAL INVESTIGATION OF LUMINES-CENCE DROPS OF NEODYMIUM IONS IN GLASS

It follows from (14), (21), (24), and (25) that to determine  $\sigma_{\rm eff}$ ,  $\tau$  and  $\beta_0$  we must investigate the behavior of integral populations  $N^{(1)}(t)$  and  $N^{(2)}(t)$  of the  ${}^{4}F_{3/2}$  metastable level. However it is difficult to observe the true behavior of integral luminescence during generation at the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  transition  $(\lambda \approx 1.06 \ \mu)$  because of the presence of a relatively strong diffusion of laser radiation in the active laser rod; it is practically impossible to eliminate such a diffusion. The transition  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{9/2} (\lambda \approx 0.88 \ \mu)$  is more convenient for this purpose. The recording of luminescence at this transition permits us also to prevent the superradiance signal from falling onto the photo-receiver and thus affords the opportunity to observe luminescence along the rod axis<sup>[4]</sup> in regions where generation is most uniform over the cross section (see Fig. 1).

Measurement of  $\sigma_{eff}$  according to the setup of Fig. 1 showed that N<sup>(2)</sup>(t) varies negligibly during generation in ordinary lasers with plane mirrors even at pump levels equal to 1.2–1.3 of threshold. According to (5) this indicates a smoothing of the central dip in the line, a result of a comparatively broad generation spectrum (at the above pump level  $\Delta \lambda \approx 50-60$  Å). Therefore the generation spectrum width must be



FIG. 4. Diagram of a setup for the observation of luminescence of an external active specimen irradiated by an auxiliary laser. 1-auxiliary laser with a narrow generation spectrum; 2-collecting lens; 3-pump lamp for the investigated specimen; 4-external specimen protected by a jacket-diaphragm whose apertures admit pumping light and emission from the auxiliary laser; 5-calorimeter; 6-lens; 7-SFD-2 monochromator; 8-photomultiplier; 9-oscilloscope.

forcibly limited, as with a frequency selector, in order to measure the excitation transfer rate directly in the laser. In our experiments we measured F in a small external specimen of KGSS-3 glass (Fig. 4) irradiated by an auxiliary phosphate neodymium glass laser with a narrow generation spectrum ( $\Delta\lambda \approx 3-5$  Å). The resulting value was F  $\approx (0.5-1.0) \times 10^5$  sec<sup>-1</sup>.

The results of measuring  $\sigma_{eff}$  and the computation according to (15a) for the case of Nd<sup>3+</sup> in various glass matrices are given below:

Matrix	KGSS-3	LGS-5	LGS-28	Phosphate glass
$\sigma_{\rm eff} \times 10^{20}$ , cm <sup>2</sup>	2.0	2.2	1.7	3.6

The completed analysis and experimental investigation of the behavior of the metastable level population indicate a high informational content of the luminescence drop of the active medium due to the action of stimulated emission. The simple analytic relation of the luminescence drop to the basic laser parameters, and the relatively uncomplicated, clear, and standard measurement method applicable to both free-running and giant pulse generation<sup>[7,13-16]</sup> render the above method of investigating laser properties quite effective and universal.

In addition to the quantities measured above the observation of luminescence drops directly in the laser yields information on the field distribution in optical resonators of various types, on the effectiveness and true form of the pumping energy, and on the processes of generation establishment. For example, curve 2 in Fig. 2 plotted for a photomultiplier time constant of  $\sim 1 \times 10^{-6}$  sec clearly shows that the population at the beginning of spiking slightly exceeds the stationary population in contrast to the results predicted in<sup>[18]</sup>.

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<sup>&</sup>lt;sup>1</sup>M. D. Galanin and Z. A. Chizhikova, Opt. Spektrosk. 17, 402 (1964).

<sup>&</sup>lt;sup>2</sup>I. H. Parks and A. Javan, Phys. Rev. 139, A1351 (1965).

<sup>&</sup>lt;sup>3</sup>M. Mishon, IEEE J. Quant. Electr. QE-2, 612 (1966).

<sup>&</sup>lt;sup>4</sup>B. I. Stepanov, S. A. Mikhanov, and A. N. Rubinov, Zh. Prikl. Spectr. 4, 122 (1966).

<sup>&</sup>lt;sup>5</sup>B. Decomps and M. Dumont, France J. Phys. 29,

443 (1968).

<sup>6</sup>A. Y. Cabezas and R. P. Treat, J. Appl. Phys. 37, 3556 (1966).

<sup>7</sup>V. R. Belan, Ch. M. Briskina, V. V. Grigor'yants, and M. E. Zhabotinskii, Zh. Eksp. Teor. Fiz. 57, 1148

(1969) [Sov. Phys.-JETP 30, 627 (1970)].

<sup>8</sup>N. Toshizo, J. Appl. Phys. Japan 5, 689 (1966).

<sup>9</sup>J. G. Edwards, Brit. J. Appl. Phys. Ser. 21, 449 (1968).

<sup>10</sup> V. V. Grigor'yants, Radiotekh. i Élektr. 14, 176 (1969).

<sup>11</sup> P. Maurer, Appl. Optics 3, 433 (1964).

<sup>12</sup>A. Y. Kabezas, R. P. Treat, and McAllister, Ng. J. Appl. Phys. 38, 3487 (1967).

<sup>13</sup>V. R. Belan, V. V. Grigor'yants, and M. E.

Zhabotinskii, IEEE J. Quant. Electr. QE-3, 11, 425

(1967).

<sup>14</sup> V. R. Belan, V. V. Grigor'yants, and M. E. Zhabotinskii, Digest of Techn. Papers, Laser Conference, Washington, 1969.

<sup>15</sup> V. R. Belan, V. V. Grigor'yants, and M. E.

Zhabotinskii, ZhETF Pis. Red. 6, 721 (1967) [JETP Lett. 6, 200 (1967)].

<sup>16</sup> V. V. Grigor'yants, Radiotekh. i Élektr. 13, 2186 (1968).

<sup>17</sup> N. A. Tolstoĭ and A. M. Tkachuk, Opt. Spektrosk. 6, 1659 (1959). <sup>18</sup>R. Dusnmuir, Electronics and Control 10, 453

(1961).

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