

ENERGY TRANSFER BETWEEN NEODYMIUM IONS IN GLASS

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The shape and kinetics of filing up of the spectral hole induced by a giant pulse in the inhomogeneously broadened Nd^{3+} luminescence line in glass are investigated. For this purpose, the lateral luminescence of a laser was observed^[6,8]. Excitation energy transfer within an inhomogeneously broadened line of the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition of Nd^{3+} , due to nonradiative transfer by the ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition is analyzed theoretically. It is demonstrated that in the general case the transfer rate may depend on the frequency or the time. A method for the experimental determination of the transfer rate is proposed, based on a detailed analysis of the time variation of the Nd^{3+} luminescence after the giant pulse. For KGSS-3 silicate glass, the experimental values of the transfer rate lie in the range $(1-0.6) \times 10^4 \text{ sec}^{-1}$.

INTRODUCTION

THE investigation of the processes of excitation-transfer in laser materials is of considerable interest, since these processes influence greatly the energy, temporal, and spectral characteristics of lasers. In particular, since one of the most widely used laser materials is neodymium glass, many efforts have been directed at the study of excitation transfer in this material. The transfer of excitation between Nd^{3+} ions is the subject of a number of papers^[1-4], in which the transfer effect was investigated indirectly by determining the laser generation characteristics. The transfer rates obtained in these investigations fluctuate between 10^4 and 10^6 sec^{-1} .

An attempt was made in^[5] to observe directly the spectral hole by comparing the luminescence spectrum of laser alkali-silicate glass with Nd^{3+} before and after the generation of a giant pulse in the glass. It was observed that the width of the spectral hole greatly exceeds the width of the spectrum of the laser giant pulse producing this hole. This fact is sometimes interpreted as the result of rapid excitation transfer between the ions (of the order of $10^7-10^8 \text{ sec}^{-1}$).

In view of the contradictory character of the previously obtained data on the transfer rates, we have investigated excitation transfer between Nd^{3+} ions in glass by analyzing the kinetics of lateral luminescence of a neodymium laser operating in the giant pulse regime.

EXPERIMENTAL SETUP

A block diagram of the experiment aimed at observing lateral laser luminescence is shown in Fig. 1^[6].

A photomultiplier (FEU-22) provided with a light attenuator, a silicon filter $\sim 0.8 \text{ mm}$ thick, and a standard infrared filter (IKS-3) is used to observe the integral luminescence of neodymium in the region $\lambda \sim 1.06 \mu$ (transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$). Simultaneously, a diffraction monochromator (SFD-2) and a photomultiplier (FEU-62) make it possible to observe the luminescence in a narrow spectral interval ($\sim 8-10 \text{ \AA}$) both via the transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ and via the transition ${}^4F_{3/2}$

$\rightarrow {}^4I_{9/2}$ ($\lambda \sim 0.9 \mu$). The luminescence observation angle is $\sim 45^\circ$ to the laser axis.

The investigations were performed on a Q-switched laser using standard KGS-3 silicate neodymium glass (Nd_2O_3 concentration 2% by weight) of 10 mm diameter and 130 mm length, with the side surface ground dull. The pumping was with an IFP-800 lamp in a cylindrical illuminator of $\sim 60 \text{ mm}$ diameter, filled with distilled water. The distance between the axes of the flash lamp and the active rod was $\sim 23 \text{ mm}$. A $500 \mu\text{F}$ capacitor bank was used. The pump duration was $\sim 500 \mu\text{sec}$ at the 0.1 level. The Q-switching prism was rotated at 25000 rpm. The output mirror reflection coefficient was 0.85. The energy radiated in the giant pulse remained constant in all the experiments at $\sim 0.3 \text{ J}$ at a giant-pulse spectrum half-width $\sim 30 \text{ \AA}$.

Typical oscillograms of the observed luminescence are shown in Fig. 2. It is seen from these oscillograms that both the relative increase of the lateral luminescence resulting from the action of the giant pulse and its subsequent time variation depend strongly on the wavelength at which the observation is carried out. It is obvious that when the times of excitation transfer between the ions greatly exceed the giant-pulse duration ($\sim 10^{-8}-10^{-7} \text{ sec}$), the ratio of the populations at the start of the giant pulse to the population at the end of the pulse, as a function of the frequency, characterizes the form of the spectral hole that results from the action of the relatively narrow-band laser signal on the in-

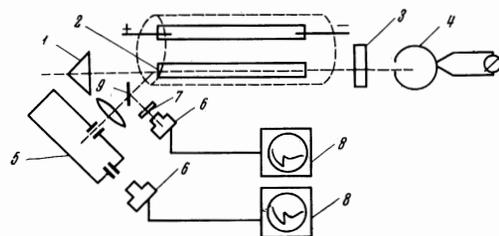


FIG. 1. Block diagram of the experiment: 1—rotating Q-switched prism, 2—laser rod, 3—output mirror, 4—calorimeter, 5—monochromator, 6—photomultipliers, 7—filter, 8—oscilloscope, 9—beam-splitting plate.

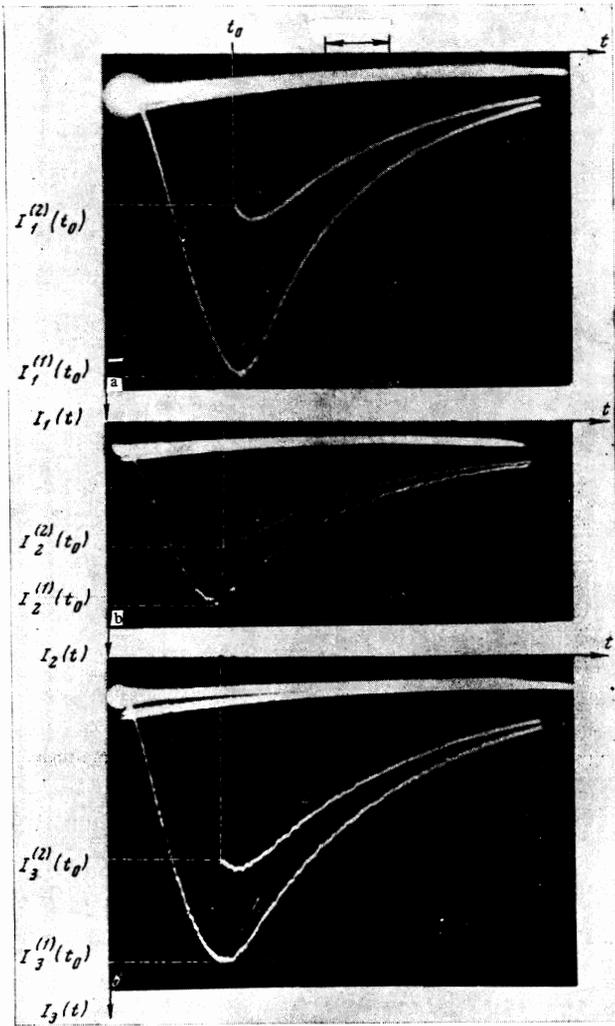
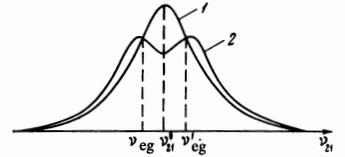


FIG. 2. Intensity I of the lateral luminescence of Nd^{3+} in a Q-switched laser (transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$): 1—luminescence in the absence of generation, 2—luminescence in the presence of generation. a—luminescence of Nd^{3+} , observed through a monochromator, $\lambda = 1.06\mu$, $I_1(t) = K_1 n_2(\nu_{21}, t)$, $I_1^{(1)}(t_0) = K_1 n_2^{(1)}(\nu_{21}, t_0)$, $I_1^{(2)}(t_0) = K_1^{(2)} n_2^{(2)}(\nu_{21}, t_0)$; b—luminescence of Nd^{3+} , observed through a monochromator, $\lambda = 1.095\mu$, $I_2(t) = K_2 n_2(\nu_{21}, t)$, $I_2^{(1)}(t_0) = K_2 n_2^{(1)}(\nu_{21}, t_0)$, $I_2^{(2)}(t_0) = K_2 n_2^{(2)}(\nu_{21}, t_0)$; c—integral luminescence of Nd^{3+} , observed without a monochromator $I_3(t) = K_3 N_2(t)$, $I_3^{(1)}(t_0) = K_3 N_2^{(1)}(t_0)$, $I_3^{(2)}(t_0) = K_3 N_2^{(2)}(t_0)$. Here K_1 , K_2 , and K_3 are the coefficients determined by the geometry and gain of the indication system. The time scale (designated by arrows) corresponds to 200 μsec .

homogeneously broadened neodymium luminescence line. The time variation of this luminescence can contain information on the transfer of excitation between the ions, which leads to the contraction of the indicated spectral hole.

Let us establish the connection between the shape of the spectral hole and the values of the relative luminescence drops observed through a monochromator and without a monochromator, without allowance for the Stark broadening of the working levels. For convenience we shall assign the numbers 0, 1, and 2 to the levels ${}^4I_{9/2}$, ${}^4I_{11/2}$, and ${}^4F_{3/2}$, respectively, and characterize each ion by two frequencies ν_{21} and ν_{20} , corresponding to the positions of its energy levels.

FIG. 3. Frequency distributions of the excited ions: 1—equilibrium— $g(\nu_{21})$ (prior to generation), 2—non-equilibrium— $g'(\nu_{21}, t_0)$ after generation.



We denote the running spectral densities of the number of excited ions at the upper laser level ${}^4F_{3/2}$ (level 2), corresponding to the time variation of the luminescence of the frequency ν_{21} with and without generation (Fig. 2a, b), by $n_2^{(2)}(\nu_{21}, t)$ and $n_2^{(1)}(\nu_{21}, t)$ ¹. By definition

$$n_2^{(1)}(\nu_{21}, t) = g(\nu_{21}, t) N_2^{(1)}(t), \quad N_2^{(1)}(t) = \int_0^\infty n_2^{(1)}(\nu_{21}, t) d\nu_{21}, \quad (1)$$

$$n_2^{(2)}(\nu_{21}, t) = g'(\nu_{21}, t) N_2^{(2)}(t), \quad N_2^{(2)}(t) = \int_0^\infty n_2^{(2)}(\nu_{21}, t) d\nu_{21},$$

where $g(\nu_{21})$ is the initial equilibrium frequency distribution (assuming broad band pumping) of the excited ions, $g'(\nu_{21}, t)$ is the non-equilibrium excited-ion distribution resulting from the action of the giant pulse, $N_2^{(2)}(t)$ and $N_2^{(1)}(t)$ are the total running numbers of ions at the level ${}^4F_{3/2}$ (per cm^3), corresponding to the luminescence curves with and without generation, observed without a monochromator (Fig. 2).

The relative luminescence dips are introduced in the following manner (assuming uniform generation over the cross section of the active element):

$$a_0 = N_2^{(1)}(t) / N_2^{(2)}(t) \quad (2a) \quad (2a)$$

is the luminescence dip observed without a monochromator, and

$$\alpha(\nu_{21}) = n_2^{(1)}(\nu_{21}, t) / n_2(\nu_{21}, t) \quad (2b)$$

is the luminescence dip observed through a monochromator. Here $t = t_0$ is the instant of emission of a giant pulse whose finite duration is not taken into account here, since it equals $\sim 10^{-8}$ – 10^{-7} sec; therefore the start and the end of the generation are practically indistinguishable in Figs. 2a and b.

From (2), taking (1) into account, we obtain the following relation for the determination of $g'(\nu_{21}, t)$ (Fig. 3) from the experimental data:

$$g'(\nu_{21}, t_0) = \frac{a_0}{\alpha(\nu_{21})} g(\nu_{21}). \quad (3)$$

The concrete form of the function $\alpha(\nu_{21})$, and consequently also $g'(\nu_{21}, t)$ depends on the character of the broadening of the spectral line and the degree of its inhomogeneity, as well as on the central frequency and on the spectral composition of the giant pulse. In particular, for a homogeneously broadened line we have

$$\alpha(\nu_{21}) \equiv a_0, \quad g'(\nu_{21}, t_0) \equiv g(\nu_{21}).$$

It is seen from Fig. 3 that the $g(\nu_{21})$ and $g'(\nu_{21}, t_0)$ curves intersect at the points $\nu_{21} = \nu_{\text{eq}}$ and $\nu_{21} = \nu'_{\text{eq}}$ where equilibrium population $n_2^{(2)}(\nu_{21}, t)$ obtains, i.e.,

$$\alpha(\nu_{\text{eq}}) = \alpha(\nu'_{\text{eq}}) = a_0. \quad (4)$$

¹Here and throughout we assume that the luminescence intensities observed through a monochromator and without a monochromator are proportional to $n_2(\nu_{21}, t)$ and $N_2(t)$, respectively.

ANALYSIS OF THE PROCESS OF EXCITATION TRANSFER WITHIN THE LIMITS OF THE INHOMOGENEOUSLY BROADENED LINE OF THE TRANSITION ${}^4F_{3/2} - {}^4I_{11/2}$ OF THE Nd^{3+} ION

After the end of the giant pulse there begins a contraction of the spectral hole, i.e., a nonequilibrium shape of the $g'(\nu_{21}, t)$ line, the initial shape of which is given by formula (3). This occurs at a certain rate determined by the character of the cross relaxation processes in the system and tends to an equilibrium line shape $g(\nu_{21})$. In particular, at the shape of the spectral hole shown in Fig. 3, restoration of the equilibrium calls for the presence of excitation transfer from the wings of the line ($\nu_{21} < \nu_{eq}$, $\nu_{21} > \nu'_{eq}$) to its center ($\nu_{eq} < \nu_{21} < \nu'_{eq}$). Let us see how such a process can occur for the case of Nd^{3+} ions in glass. The working transition is ${}^4F_{3/2} - {}^4I_{11/2}$, ($2 - 1$), so that it is of interest to consider the transfer of energy between ions having different values of the working frequency ν_{21} .

Since the energy gap between the lower working level ${}^4I_{11/2}$ and the ground level ${}^4I_{9/2}$ is $\sim 2000 \text{ cm}^{-1}$, the population of the ${}^4I_{11/2}$ level at room temperature is smaller by about four orders of magnitude than the population of the ${}^4I_{9/2}$ level. This allows us to assume that energy transfer from the ion excited at the ${}^4F_{3/2}$ level to the non-excited ion occurs mainly as the result of the transition $2 \rightarrow 0$ for the excited ion and $0 \rightarrow 2$ for the non-excited ion.

As is well known, the inhomogeneous broadening of the spectral lines of Nd^{3+} in glass is the result of variations of the surrounding of the neodymium ions, which for this reason may differ from each other in the positions of the energy levels 0, 1, and 2 of interest to us. Earlier, in considering the shape of the spectral hole in the luminescence line for the transition $2 - 1$, we have described the excited ions by a set of frequencies ν_{21} corresponding to the transitions $2 \rightleftharpoons 1$. However, since the contraction of the initial spectral dip should occur as the result of the transitions $2 \rightleftharpoons 0$, it will be convenient in what follows to characterize each ion, as noted above, by two frequencies ν_{21} and ν_{20} .

On the basis of the foregoing, we introduce the spectral density $n_0(\nu_{21}, \nu_{20})$ of the number of ions in the ground state, having specified values of the frequencies ν_{21} and ν_{20} , assuming time-independent weak pumping, with

$$\iint n_0(\nu_{21}, \nu_{20}) d\nu_{21} d\nu_{20} = N_0, \quad (5)$$

$$\int n_0(\nu_{21}, \nu_{20}) d\nu_{20} = n_0(\nu_{21}), \quad \int n_0(\nu_{21}, \nu_{20}) d\nu_{21} = n_0(\nu_{20}),$$

where N_0 is the total number of ions in the ground state.

We introduce analogously the running spectral density of the number of excited ions $n_2(\nu_{21}, \nu_{20}, t)$ in such a way that (see (1))

$$\iint n_2(\nu_{21}, \nu_{20}, t) d\nu_{21} d\nu_{20} = N_2(t), \quad (6)$$

$$\int n_2(\nu_{21}, \nu_{20}, t) d\nu_{20} = n_2(\nu_{21}, t), \quad \int n_2(\nu_{21}, \nu_{20}, t) d\nu_{21} = n_2(\nu_{20}, t).$$

In the period following the termination of the giant pulse, the balance equation for the spectral density of the number of excited ions $n_2^{(2)}(\nu_{21}, t)$ with allowance for excitation transfer due to the transitions $2 \rightleftharpoons 0$ can be written in the following form:

$$\frac{dn_2^{(2)}(\nu_{21}, t)}{dt} = W(t)n_0(\nu_{21}) - \frac{n_2^{(2)}(\nu_{21}, t)}{\tau} \quad (7)$$

$$+ \int n_2^{(2)}(\nu_{20}, t) d\nu_{20} \int \mathcal{F}(\nu_{20}, \nu_{20}') n_0(\nu_{21}, \nu_{20}') d\nu_{20}' - \int n_2^{(2)}(\nu_{21}, \nu_{20}, t) d\nu_{20} \int \mathcal{F}(\nu_{20}, \nu_{20}') n_0(\nu_{20}') d\nu_{20}',$$

where $W(t)$ is the probability of pumping the particles to the level 2, τ is the lifetime of the excited ions at the level 2, measured in the absence of the spectral hole, $\mathcal{F}(\nu_{20}, \nu_{20}')$ is the elementary rate of excitation from the excited ion, characterized by the frequency ν_{20} to the non-excited ion, characterized by the frequency ν_{20}' .

The first term in (7) is due to the pumping and the second to the spontaneous radiative and nonradiative transitions from the level ${}^4F_{3/2}$ to all the lower levels ${}^4I_{9/2, 11/2, 13/2, 15/2}$, the third to the energy transfer to the non-excited ions, characterized by a frequency ν_{21} , from all other ions, and the fourth to the energy transfer from the ions characterized by the frequency ν_{21} to all others.

In a regular crystal matrix, where the surrounding of all the impurity ions is practically the same and therefore the parameters of the crystal field are constant, the connection between ν_{21} and ν_{20} is firm. In glass, owing to random variations of the surrounding, the parameters of the crystal field vary randomly, and apparently to a considerable degree independently. The independent variations of these parameters cause the ions characterized by the same frequency ν_{21} to have different values of ν_{20} , and vice-versa. As the result, even resonant energy transfer in accordance with the transition $2 \rightleftharpoons 0$ should lead to a change in the number of the excited ions, characterized by a specified frequency ν_{21} .

In Eq. (7) we shall be interested primarily in the third and fourth terms, describing the excitation transfer between the working ions. For convenience in further transformations we introduce, in addition to (5) and (6), the form factors $g'(\nu_{21}, \nu_{20}, t)$ and $g(\nu_{21}, \nu_{20})$ in such a way that

$$n_2^{(2)}(\nu_{21}, \nu_{20}, t) = g'(\nu_{21}, \nu_{20}, t) N_2^{(2)}(t), \quad (8)$$

$$n_0(\nu_{21}, \nu_{20}) = g(\nu_{21}, \nu_{20}) N_0$$

under the following normalization conditions:

$$\begin{aligned} \iint g'(\nu_{21}, \nu_{20}, t) d\nu_{21} d\nu_{20} &= 1, & \iint g(\nu_{21}, \nu_{20}) d\nu_{21} d\nu_{20} &= 1, \\ \int g'(\nu_{21}, \nu_{20}, t) d\nu_{20} &= g'(\nu_{21}, t), & \int g(\nu_{21}, \nu_{20}) d\nu_{20} &= g(\nu_{21}), \\ \int g'(\nu_{21}, \nu_{20}, t) d\nu_{21} &= g'(\nu_{20}, t), & \int g(\nu_{21}, \nu_{20}) d\nu_{21} &= g(\nu_{20}). \end{aligned} \quad (9)$$

Here $g'(\nu_{21}, t)$ and $g(\nu_{21})$ are respectively the non-equilibrium and equilibrium line shapes, and are connected at $t = t_0$ (the instant of termination of the giant pulse) by the relation (3).

Using (8) and (9) we can represent the last two terms of (7) in the form

$$\begin{aligned} N_0 N_2^{(2)}(t) \left\{ \int g'(\nu_{20}, t) d\nu_{20} \int \mathcal{F}(\nu_{20}, \nu_{20}') g(\nu_{21}, \nu_{20}') d\nu_{20}' \right. \\ \left. - \int g'(\nu_{21}, \nu_{20}, t) d\nu_{20} \int \mathcal{F}(\nu_{20}, \nu_{20}') g(\nu_{20}') d\nu_{20}' \right\}. \end{aligned} \quad (10)$$

Applying the theorem of the mean twice to (10), we obtain the term describing the excitation transfer between the ions

$$N_0 [F_1(\nu_{21}, t) g(\nu_{21}) N_2^{(2)}(t) - F_2(\nu_{21}, t) n_2^{(2)}(\nu_{21}, t)], \quad (11)$$

where²⁾

$${}^4F_1 = \mathcal{F}(\bar{\nu}_{20}(t), \bar{\nu}_{20}'(\nu_{21})), \quad F_2 = \mathcal{F}(\bar{\nu}_{20}(\nu_{21}, t), \bar{\nu}_{20}')$$

Expressions (10) and (11) are valid in the general case. After making certain assumptions concerning the character of the form factors, we can simplify these expressions.

In practice we are interested in the case when the stimulated emission in the form of a giant pulse occurs at the frequency ν_{21} (the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition for Nd^{3+}). As shown above, this leads to a distortion of the equilibrium line shape in accordance with (3). If it is assumed that a certain frequency ν_{21} corresponds to a number of frequencies in accordance with the transition $2 \rightarrow 0$, then the spectral hole occurring in generation at the transition $2 \rightarrow 1$, when luminescence is observed for the transition $2 \rightarrow 0$, becomes distributed over a set of frequencies ν_{20} corresponding to the fixed frequency ν_{21} . The distortion of $g(\nu_{20})$ will then be smoothed out compared with the distortion of $g(\nu_{21})$. The larger the set of frequencies ν_{20} corresponding to one frequency ν_{21} , the stronger this effect.

In the case of a sufficiently broad generation spectrum, with a width on the order of the magnitude of the inhomogeneous broadening, it can be assumed that the distortion of the distribution with respect to ν_{21} does not change the distribution with respect to ν_{20} , i.e.,

$$g'(\nu_{20}, t) = g(\nu_{20}) \quad (12)$$

(see (9)). Taking into account the fact that the term determining the excitation transfer vanishes at equilibrium, i.e.,

$$\int g(\nu_{20}) d\nu_{20} \int \mathcal{F}(\nu_{20}, \nu_{20}') g(\nu_{21}, \nu_{20}') d\nu_{20}' \\ = \int g(\nu_{21}, \nu_{20}) d\nu_{20} \int \mathcal{F}(\nu_{20}, \nu_{20}') g(\nu_{20}') d\nu_{20}',$$

we obtain in place of (10), with allowance for (12)

$$N_0 N_2^{(2)}(t) \int [g(\nu_{21}, \nu_{20}) - g'(\nu_{21}, \nu_{20}, t)] d\nu_{20} \int \mathcal{F}(\nu_{20}, \nu_{20}') g(\nu_{20}') d\nu_{20}'.$$

If we again employ the theorem of the mean, we obtain

$$N_0 F_3(\nu_{21}, t) \{n_2^{(2)}(\nu_{21}, t) - n_2^{(2)}(\nu_{21}, t_0)\}, \quad (13)$$

where

$$F_3 = \int \mathcal{F}(\bar{\nu}_{20}(\nu_{21}, t), \bar{\nu}_{20}') g(\nu_{20}') d\nu_{20}'.$$

We can make an even firmer assumption, namely, that the ions characterized by a given frequency ν_{21} have the entire set of frequencies ν_{20} that it is possible in the given matrix and, conversely, the ions characterized by a given frequency ν_{20} have the entire possible set of frequencies ν_{21} . In this case

$$g'(\nu_{21}, \nu_{20}, t) = g'(\nu_{21}, t) g(\nu_{20}). \quad (14)$$

Taking (14) into account, we obtain in place of (11)

$$N_0 \int \mathcal{F}(\nu_{20}, \nu_{20}') g(\nu_{20}) g(\nu_{20}') d\nu_{20} d\nu_{20}' \{n_2^{(2)}(\nu_{21}, t) \\ - n_2^{(2)}(\nu_{21}, t_0)\} = N_0 C [n_2^{(2)}(\nu_{21}, t) - n_2^{(2)}(\nu_{21}, t_0)]. \quad (15)$$

We note that Denisov and Kizel^[7] proved the validity of the assumption (14) for the transitions ${}^5D_0 \rightarrow {}^7F_0$ and ${}^5D_0 \rightarrow {}^7F_1$ of the trivalent europium ion in glass. From

²⁾The choice of the average points $\bar{\nu}_{20}$ and $\bar{\nu}_{20}'$ depends on the parameters ν_{21} and t , which enter into the function that remains under the integral sign.

the theoretical point of view, for Nd^{3+} in glass, the validity of (14) is less obvious than for the indicated europium transitions, and calls for a special verification, which will be discussed later.

Thus, considering the excitation transfer within the limits of an inhomogeneously broadened line of Nd^{3+} due to the transitions $2 \rightarrow 0$, we have obtained a term describing the transfer in the form (11), (13), and (15), depending on the assumptions concerning the connection between the frequencies ν_{21} and ν_{20} . It is important that in all three indicated cases the total rate of excitation transfer is proportional to the quantity N_0 , which in the case of real pumping practically coincides with the concentration of the active ions in the matrix.

If we denote $N_0 C$ in (15) by F , then we obtain the transfer term in a form that coincides completely with the one introduced phenomenologically in^[11]. In the more general case (13), however, F is not a constant but depends on the frequency ν_{21} and on the time.

The foregoing analysis of the excitation-energy transfer process within the limits of an inhomogeneously broadened line of the transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ ($2 \rightarrow 1$) of neodymium in glass explains why the transfer term does not have a diffusion character. The point is that the energy transfer, according to our notions, proceeds actually in accordance with the transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ ($2 \rightarrow 0$), and there is no firm connection between the frequencies ν_{20} and ν_{21} . As a result, although the energy transfer via the ground state ($2 \rightarrow 0$) possibly represent a diffusion process, for the transition of interest to us ($2 \rightarrow 1$) the transfer is described by an equation that differs from the diffusion equation.

To determine the applicability of the particular form of the transfer term to our concrete case of excitation transfer between neodymium ions in KGSS-3 glass, we have used the experimental setup described above to measure the luminescence dips $\alpha(\nu)$ in a wide range of frequencies, both for the transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ and for the transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$. As already indicated, the energy and the spectral composition of the giant pulse remained unchanged in all the measurements.

In a real laser, the generation usually fills not the entire cross section of the rod, so that the signal reaching the receiver contains luminescence also from the nongenerating parts of the sample, leading to an error in the determination of $\alpha(\nu_{21})$ and α_0 ^[8]. The magnitude of this additional signal is the same for $n_2^{(1)}(\nu_{21}, t_0)$ and $n_2^{(2)}(\nu_{21}, t_0)$, and its relative contribution does not depend on ν_{21} . On this basis, it is more convenient to use in the reduction of the experimental data the quantities

$\delta n_2(\nu_{21}, t_0) = n_2^{(1)}(\nu_{21}, t_0) - n_2^{(2)}(\nu_{21}, t_0)$ and $\delta N_2(t_0) = N_2^{(1)}(t_0) - N_2^{(2)}(t_0)$, from which the influence of the non-uniformity of the generation is excluded.

In all cases, we took into account also the influence of the scattered light of the flash-lamp, which is incident on the photomultiplier together with the luminescence.

If $\delta n_2(\nu_{21}, t_0)$ and $\delta N_2(t_0)$ are determined from luminescence curves drawn to the same scale, then $\delta N_2(t_0)$ has in this case, as will be shown below, the meaning of the absolute value of the equilibrium dip. The results of the measurements in the form are the ratios

$$R_1 = \frac{\delta n_2(\nu_{21}, t_0)_{1.06}}{\delta N_2(t_0)}, \quad R_2 = \frac{\delta n_2(\nu_{20}, t_0)_{0.9}}{\delta N_2(t_0)}$$

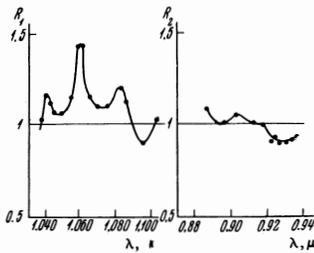


FIG. 4. Dependence of the drop of the luminescence on the frequency; $R_1 = \delta n_2(\nu_{21}, t_0)_{1,06} / \delta N_2(t_0)$, $R_2 = \delta n_2(\nu_{20}, t_0)_{0,9} / \delta N_2(t_0)$.

are shown in Fig. 4.

It follows from Fig. 4 that the spectral dependence of the luminescence dip in the ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition is considerably smoothed out compared with this dependence in the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition. This smoothing becomes manifest most distinctly at wavelengths $0.890 \mu \leq \lambda \leq 0.91 \mu$. When $\lambda < 0.890 \mu$ and $\lambda > 0.915 \mu$, the dips change much more abruptly. However, owing to the stronger influence of the scattered light of the flash lamp, which is difficult to take into account, on the edges of the spectral line of the ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition, the true cause of this phenomenon remains unclear. In our opinion, the data of Fig. 4 allow us to conclude that in the case of Nd^{3+} in glass the transfer term should in all probability have the form (13). However, the degrees of the dependence of the rate of excitation transfer F , which enters in (13), on the time and of the frequency, should be determined separately. We shall henceforth assume, as an approximation, that $F = \text{const}$, i.e., we shall use the transfer term in the form (15).

It is interesting to note that the maxima of the luminescence dips in the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition agree well with the known frequencies of the transitions between two Stark sublevels of the state ${}^4F_{3/2}$ and between the two groups of unresolved Stark sublevels of the state ${}^4I_{11/2}$ of the Nd^{3+} ion in glass^[5]. The width of each peak could yield information concerning the magnitude of the homogeneous broadening of the levels under the condition that the spectral width of the giant pulse of the investigated laser is narrowed down. Thus, the observation of a spectral dependence of the luminescence dip under the influence of the stimulated emission directly in the laser or in a small external sample^[10] is one method of investigating the structures of unresolved lines.

In analogous investigations of phosphate glass with neodymium, no frequency dependences of the luminescence dips was observed within the limits of the time resolution of our apparatus ($\sim 10^{-6}$ sec). This fact can be attributed to the presence in the phosphate glass of rapid excitation transfer between the Nd^{3+} ($> 10^6 \text{ sec}^{-1}$)^[9], or else to the fact that the homogeneous level broadening is much larger than in silicate glass.

DETERMINATION OF THE RATE OF EXCITATION TRANSFER BETWEEN NEODYMIUM IONS

To determine the rate of excitation transfer between neodymium ions, we use Eq. (7), in which we write the transfer term in the form (15). We have

$$\frac{dn_2^{(2)}(\nu_{21}, t)}{dt} = W(t)N_0g(\nu_{21}) - \frac{n_2^{(2)}(\nu_{21}, t)}{\tau} \quad (16)$$

$$+ F[n_2^{(2)}(\nu_{21}, t) - n_2^{(2)}(\nu_{21}, t)].$$

Since

$$n_2^{(2)}(\nu_{21}, t) = g(\nu_{21})N_2^{(2)}(t), \quad (17)$$

the kinetic equation for this quantity is analogous to the equation for $N_2(t)$:

$$\frac{dn_2^{(2)}(\nu_{21}, t)}{dt} = W(t)N_0g(\nu_{21}) - \frac{n_2^{(2)}(\nu_{21}, t)}{\tau}. \quad (18)$$

Subtracting (16) from (18), we get an equation describing the process of contraction of the spectral hole occurring as the result of the action of the giant pulse at the instant $t = t_0$:

$$\frac{d\Delta n_2(\nu_{21}, t)}{dt} = -\left(F + \frac{1}{\tau}\right)\Delta n_2(\nu_{21}, t), \quad (19)$$

where

$$\Delta n_2(\nu_{21}, t) = n_2^{(2)}(\nu_{21}, t) - n_2^{(2)}(\nu_{21}, t_0).$$

It follows from (19) that the contraction of the spectral hole follows an exponential law

$$\Delta n_2(\nu_{21}, t) = \Delta n_2(\nu_{21}, t_0) \exp\left\{-\left(F + \frac{1}{\tau}\right)(t - t_0)\right\}, \quad (20)$$

which can be used to determine F .

The initial deviation $\Delta n_2(\nu_{21}, t_0)$ of the spectral population density from the equilibrium density is actually determined by the form of the nonequilibrium spectral line $g'(\nu_{21}, t_0)$ (see (3)).

Let us see how we can trace the contraction of the spectral hole under the concrete conditions of our experiment.

If we align, either by choosing the gain of the oscilloscopes or graphically, the oscillograms of the integral luminescence of neodymium and of the luminescence at a certain frequency ν_{21} of the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition in the absence of generation, then it follows from (1) that the coefficients K_1 and K_3 (Figs. 2a, b) are chosen here in such a way as to satisfy the condition $K_3/K_1 = g(\nu_{21})$. Then, if the generation instants coincide, we can assume, taking (17) into account, that the integral luminescence curve in the presence of generation will coincide with the time variation of $n_2^{(2)}(\nu_{21}, t)$ at the given frequency. Thus, in this case the quantity $\Delta n_2(\nu_{21}, t)$ can be read directly from the oscillograms, as the difference between the curves of the integral luminescence and the luminescence observed through the monochromator at the frequency ν_{21} in the post-generation period (Fig. 5).

It is obvious that the luminescence curves corre-

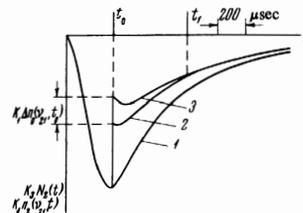


FIG. 5

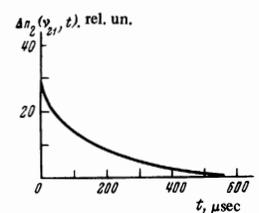


FIG. 6

Fig. 5. Aligned (graphically) luminescence curves of Nd^{3+} , observed through a monochromator ($\lambda = 1.06 \mu$) and without it: 1— $K_3 N_2^{(1)}(t) = K_1 n_2^{(1)}(\nu_{21}, t)$, 2— $K_3 N_2^{(2)}(t) = K_1 n_2^{(2)}(\nu_{21}, t)$, 3— $K_1 n_2^{(2)}(\nu_{21}, t)$.

Fig. 6. Experimental plot of contraction of the spectral dip ($\lambda = 1.06 \mu$).

sponding to $n_2^{(2)}(\nu_{\text{eq}}, t)$, $n_2^{(2)}(\nu'_{\text{eq}}, t)$ and $N_2^{(2)}(t)$ will coincide if one aligns the scales at the frequencies $\nu_{21} = \nu_{\text{eq}}$ and $\nu_{21} = \nu'_{\text{eq}}$ (Fig. 5), from which we have in accordance with (4) $\alpha(\nu_{\text{eq}}) = \alpha(\nu'_{\text{eq}}) = \alpha_0$.

On the other hand, if the luminescence curves without generation are aligned at frequencies different from ν_{eq} and ν'_{eq} , then $n_2^{(2)}(\nu_{21}, t)$ curves will lie higher than the $N_2^{(2)}(t)$ curve if $\nu_{21} < \nu_{\text{eq}}$ and $\nu_{21} > \nu'_{\text{eq}}$, and below this curve if $\nu_{\text{eq}} < \nu_{21} < \nu'_{\text{eq}}$.

By measuring the ratio $\Delta n_2(\nu_{21}, t)/\Delta n_2(\nu_{21}, t_0)$ from Fig. 5 for different instants of time, we can immediately determine the value of S. It is seen from Fig. 5 that the process of contraction of the spectral hole practically terminates at the instant $t = t_1$, when the curves $N_2^{(2)}(t)$ and $n_2^{(2)}(\nu_{21}, t)$ coalesce.

Even more illustrative is Fig. 6, which shows on a magnified scale the time dependence of $\Delta n_2(\nu_{21}, t)$ taken from Fig. 5. An analysis of plots of this type makes it possible to ascertain whether the transfer rate F depends on the time or on the frequency. However, for the experiments performed in accordance with the block diagram of Fig. 1, this is difficult to do because the lifetimes of the ions τ , which enter in Eqs. (16) and (18), can be different and can vary in time because of superluminescence, the contribution of which, in turn, depends on the frequency. More promising from this point of view, therefore, should be analogous investigations of the luminescence of a small sample with neodymium excited prior to the production of the populated inversion, to which a giant pulse is applied from an external Q-switched neodymium laser^{10,11}. The indicated variant of the experiment is convenient also because in this case it is simpler to decrease the harmful influence of the scattered light of the flash lamp (observation along the sample axis¹¹) and the inhomogeneity of the pumping of the sample and of the flux of photons from the external laser; requirements with respect to optical inhomogeneity of the surface finish of the samples are also less stringent. In addition, the possibility of carrying out the investigations at low temperatures becomes realistic.

As follows from Fig. 6, the rate of excitation transfer in KGSS-3 glass is $F = 6 \times 10^3 - 1 \times 10^4 \text{ sec}^{-1}$. If it is assumed that the excitation transfer in accordance with the ${}^4F_{3/2} \rightleftharpoons {}^4I_{9/2}$ transition is resonant and results from dipole-dipole interaction, then we can obtain in our case, by using the data of¹², the following tentative estimate of F:

$$F = 0.9 \cdot 10^{-15} N_0 / R_M^3 [\text{sec}^{-1}], \quad (21)$$

where N_0 is the number of neodymium ions per cm^3 , R_M is the minimal distance between the neodymium ions (in Angstroms) possible in the given matrix. Taking $N_0 = 3.5 \times 10^{20} \text{ cm}^{-3}$, we obtain for different values of R_M :

$R_M, \text{ \AA}:$	2	3	4
$10^{-15} F, \text{ sec}^{-1}:$	3.9	1.2	0.5

Comparison of the calculated and the experimental values of the transfer rate makes it possible to conclude that the contribution of the resonant dipole-dipole interaction to the excitation transfer of interest to us, between the neodymium ions in silicate glass at room temperature, can be quite appreciable.

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