

A new method of laser-spectroscopy investigation of solids with inhomogeneously broadened spectra

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A new method of selective laser excitation, which is used to investigate the Stark structure of inhomogeneously broadened absorption bands ${}^4I_{9/2} \rightarrow {}^2G_{7/2}$, ${}^4G_{5/2}$ of the Nd^{3+} ions in silicate glass, is described. A diagram of the location of the Stark levels of the ${}^2G_{7/2}$ and ${}^4G_{5/2}$ (Nd^{3+}) states in the glass is constructed for the first time. New possibilities of studying the energy migration along the spectra of laser excitation of the luminescence of Nd^{3+} with time and spectrum selection are proposed.

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In this paper we describe a new method of spectral investigation using selective laser excitation (SLE), which enabled us to obtain information on the structure of complex, inhomogeneously broadened absorption spectra of rare-earth (RE) ions in crystals and glass. It is based on recording the spectra of monochromatic excitation of luminescence using a tunable narrow-band ($\Delta\lambda_r < 1\text{\AA}$) laser spectrometer (λ_r -vary) and selective recording of radiation with respect to time (t_s) and with respect to the spectrum (λ_L) $I(t_s; \lambda_L) = f(\lambda_E)$ (the experimental setup is described in Ref. 1).

Let us examine the new capabilities of this method by investigating the structure of the absorption transition ${}^4I_{9/2} \rightarrow {}^2G_{7/2}$, ${}^4G_{5/2}$ of Nd^{3+} ions in commercial laser glass LGS-28, which has a large inhomogeneous spectral broadening attributable to the fact that Nd^{3+} ions are situated in structurally nonequivalent base positions. Figure 1a shows the luminescence spectra of the main transitions ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ and ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ of the Nd^{3+} ion, which were obtained as a result of nonselective excitation at $T = 77\text{ K}$. Using a monochromator we separated from these complex, low-structure spectra, nar-

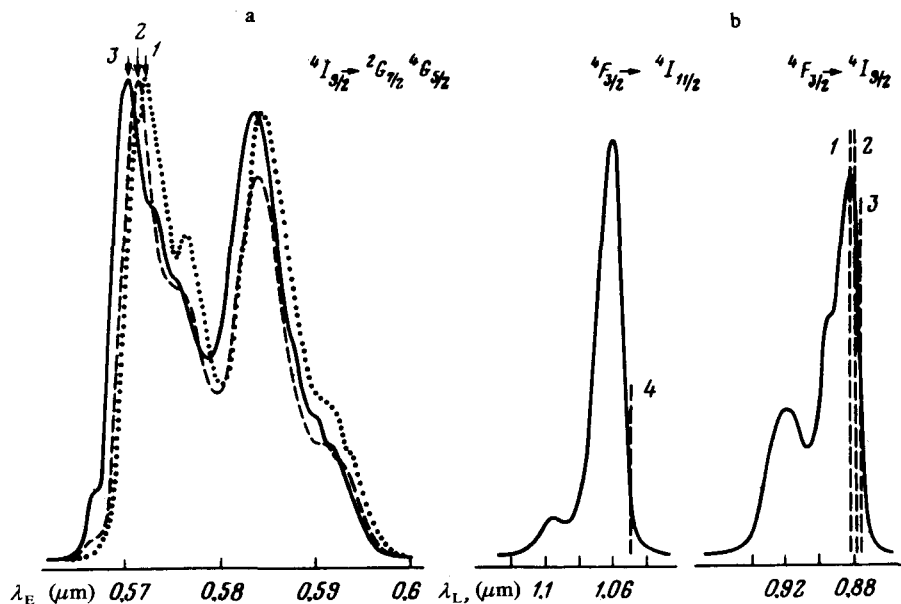


FIG. 1. a) Luminescence spectra of Nd^{3+} ions in LGS-28; b) spectra for monochromatic excitation of luminescence in the transition ${}^4I_{9/2} \rightarrow {}^2G_{7/2}$; ${}^4G_{5/2}$ for selective recording of luminescence in the transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$. Curves 1, 2, and 3 correspond to the wavelengths denoted by broken lines 1, 2, and 3 in Fig. 1a.

row spectral regions of recorded luminescence $[\lambda_L \pm (\Delta\lambda_L/2)]$ (marked by broken lines 1, 2, and 3 in Fig. 1a) and recorded the excitation spectrum of this luminescence by scanning the frequency of the tunable laser on the band corresponding to the investigated transition. These measurements were conducted at a low concentration of Nd_2O_3 (0.3 wt%) and a short delay time of the recording of luminescence relative to the moment of its excitation. The three excitation spectra in the transition band ${}^4I_{9/2} \rightarrow {}^2G_{7/2}$; ${}^4G_{5/2}$ for three different wavelengths of luminescence recording are represented by curves 1, 2, and 3 in Fig. 1b. It can be seen that the excitation spectra, which have a well-defined structure, depend greatly on the recording wavelength (λ_L). The number of spectral components (6–7) is much greater than that usually observed in the absorption spectrum (4) and approaches the maximum number of the Stark levels $3 \times ({}^4G_{5/2}) + 4({}^2G_{7/2})$.

By analyzing the energy state of the Stark components of the ${}^2G_{7/2}$ and ${}^4G_{5/2}$ levels as a function of the recording wavelength λ_L , we were able to construct for the first time an energy-level diagram for the continuous set of Nd^{3+} optical centers in the glass (Fig. 2). The energy of the luminescent transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ is plotted along the X axis and the energies of the Stark components of the ${}^2G_{7/2}$ and ${}^4G_{5/2}$ levels are plotted along the Y axis. A fragment of the luminescence spectrum of the ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition is given in the lower part of the diagram, where the shaded parts show the regions of the spectrum $\lambda_L \pm (\Delta\lambda_L/2)$ which were successively isolated by the monochromator.

By recording the spectra of monochromatic excitation and weak luminescence with the excited Stark component of the metastable ${}^4F_{3/2}$ level, which is weakly popu-

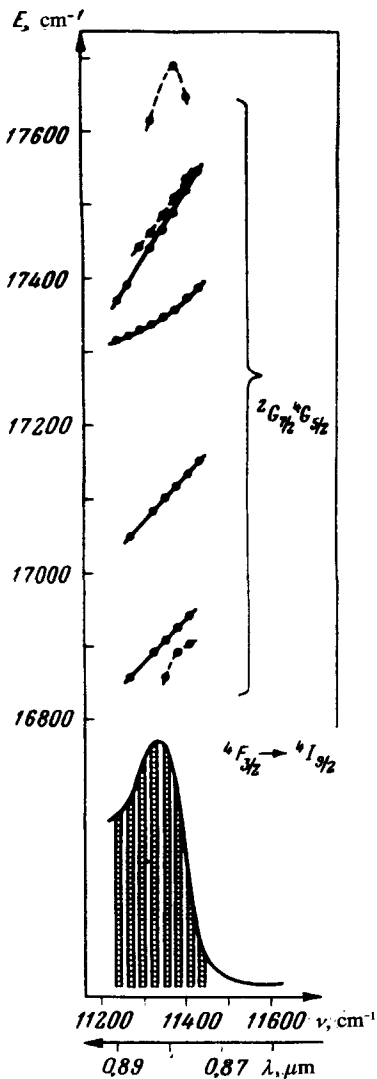


FIG. 2. A diagram for the position of the Stark components of the levels ${}^2G_{7/2}$ and $G_{5/2}$ for different optical centers of Nd^{3+} in the glass.

lated at $T = 77$ K, we were able to determine for the first time its energy state and the variation of the Stark splitting of the ${}^4F_{3/2}$ level for one Nd^{3+} center to another in the glass ΔE (${}^4F_{3/2}$). It turned out that ΔE increases smoothly on going from the low-energy centers $\Delta E = 85$ cm^{-1} to the high-energy centers $\Delta E = 160$ cm^{-1} .

Thus, the spectroscopic method of monochromatic excitation with the selection of time and spectrum luminescence enabled us to show for the first time the fine structure of the inhomogeneously broadened collective absorption spectra ${}^4I_{9/2} \rightarrow {}^4G_{5/2}$ and ${}^2G_{7/2}$ of the Nd^{3+} ions in the glass. This made it possible to obtain new information on the location of the Stark components of the high-lying energy levels ${}^4G_{5/2}$, ${}^2G_{7/2}$, and ${}^4F_{3/2}$ and on the variation of the Stark splitting from one Nd^{3+} center to another, which could not be obtained by conventional spectroscopic methods.

Moreover, this method affords new opportunities to observe experimentally the energy migration along the metastable levels of the spectrally nonequivalent optical centers.

Figure 3 shows the spectra of monochromatic excitation of the Nd^{3+} ions

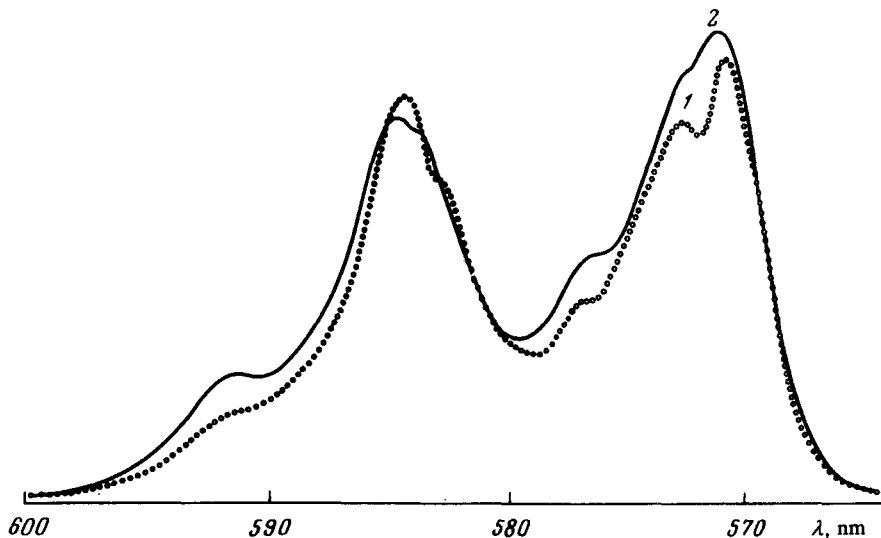


FIG. 3. Time evolution of spectra of monochromatic excitation for selective recording of luminescence $\lambda_L = 1.05 \mu\text{m}$ (broken line 4 in Fig. 1a). Curve 1 - $t_3 = 50 \mu\text{sec}$; curve 2 - $t_3 = 400 \mu\text{sec}$. $C = 2 \text{ wt}\%$. $T = 77 \text{ K}$.

(${}^4I_{9/2} \rightarrow {}^2G_{7/2}$; ${}^4G_{5/2}$ transition) obtained by selectively recording the luminescence at the wavelength $\lambda_L = 1.05 \mu\text{m}$ (broken line 4 Fig. 1a) and by varying the delay time t_3 of the recording of luminescence relative to the moment of its excitation. The spectra were taken at 77 K using glass samples with Nd_2O_3 concentration of 2 wt%, at which the ion-ion interactions $\text{Nd}^{3+} \rightleftharpoons \text{Nd}^{3+}$ usually begin to occur. Curve 1 in Fig. 3 represents an excitation spectrum obtained after a short delay time $t_3 = 50 \mu\text{sec}$. The fine structure of this spectrum shows the location of the absorption transitions only for that group of Nd^{3+} ions whose luminescence is recorded immediately ($\lambda_L = 1.05 \mu\text{m}$). As the delay time increases to $t_3 = 400 \mu\text{sec}$ (curve 2 in Fig. 3), additional lines of a large number of different centers begin to appear in the excitation spectrum, which in the time $t_3 = 400 \mu\text{sec}$ transfer the excitation energy from its ${}^4F_{3/2}$ metastable level to the ${}^4F_{3/2}$ level of the center that was isolated by selective recording $\lambda_L = 1.05 \mu\text{m}$. The result of such energy transfer is the leveling of the fine structure and broadening of the lines in the excitation spectrum, which, with increasing t_3 , begins to resemble the collective absorption spectrum of the Nd^{3+} ions in the glass. The unique feature of the proposed method of investigating the spectral energy migration is the directivity of this process, i.e., the transfer of energy from the large number of spectrally nonequivalent centers to the single isolated center, which distinguishes it from the other method of observing the migration.¹²¹

A study of this mechanism of energy migration may prove to be important from a

practical point of view for lasers that generate narrow spectral lines in active media with a large, inhomogeneous spectral broadening. In such lasers the energy migration facilitates additional monochromatization of the luminous energy before its stimulated coherent emission.

¹O.K. Alimov, T.T. Basiev, Yu. K. Voron'ko, Yu. V. Gribkov, A. Ya. Karasik, V.V. Osiko, A.M. Prokhorov, and I.A. Shcherbakov, *Zh. Eksp. Teor. Fiz.* **74**, 57 (1978) [*Sov. Phys. JETP* **47**, 29 (1978)].

²O.K. Alimov, T.T. Basiev, Yu. K. Voron'ko, L.S. Gaiigerova, and A.V. Dmitryuk, *Zh. Eksp. Teor. Fiz.* **45**, 690 (1977) [*Sov. Phys. JETP* **72**, 1313 (1977)].