

Frequency selection of Nd^{3+} ions in glass excited by monochromatic laser radiation at the resonant transition

$${}^4I_{9/2} \rightarrow {}^4F_{3/2}$$

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A recently-developed repetitive-pulse tunable laser is first used to achieve resonant frequency selection of Nd^{3+} ions in a phosphate glass at the resonant transition ${}^4I_{9/2} \rightarrow {}^4F_{3/2}$. The stark splitting diagram of the ${}^4F_{3/2}$ and ${}^4I_{9/2}$ levels is developed for a set of spectrally-nonequivalent Nd^{3+} centers. Analysis of time variations in the fluorescence spectra due to instantaneous laser excitation shows the nature of Nd^{3+} ion coupling with phonons which leads to a spectral shift of energy to the longwave region.

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The method of selective laser excitation of fluorescence opens new possibilities in the study of optical media with non-homogeneous spectral broadening.⁽¹⁾ Until now, investigation of spectral-kinetic properties of fluorescence of Nd^{3+} ions excited by a resonant monochromatic radiation was impeded by the absence of suitable laser sources of light. In a very recent work,⁽²⁾ we reported on the development of a repetitive-pulse tunable laser based on a LiF crystal with F_2^+ centers. Featuring a selective resonator with a diffraction grating, the laser generates optical pulses with a 6×10^{-2} -

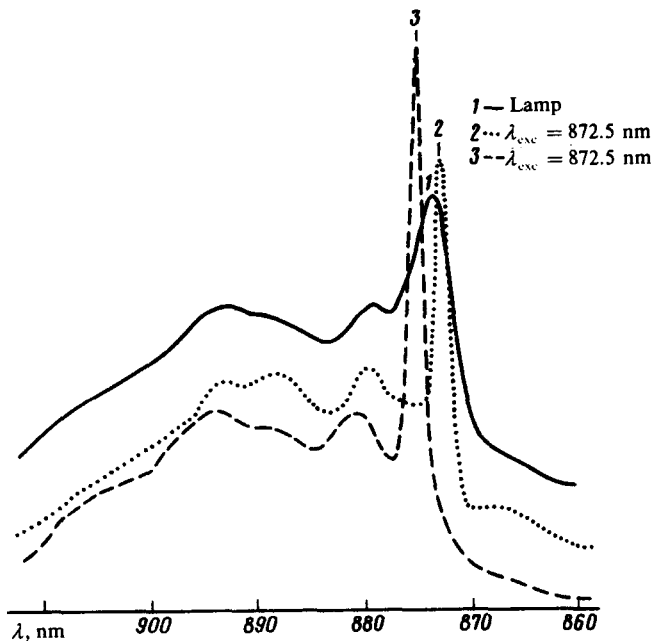


FIG. 1. ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ fluorescence spectra of Nd^{3+} ions in the case of broadband (curve 1) and resonant monochromatic excitation (curve 2— $\lambda_{\text{exc}} = 8725 \text{ \AA}$; curve 3— $\lambda_{\text{exc}} = 8752 \text{ \AA}$) $T = 77 \text{ K}$, $t_d = 40 \mu\text{sec}$, $n(\text{Nd}) = 2.7 \times 10^{20} \text{ cm}^{-3}$.

sec duration at a frequency of 50 Hz and a linewidth of 2 Å, which are tunable in a range 8700–9900 Å. The use of a three-prism dispersion-type resonator with a high Q -factor was responsible for extending the generation region to 8550–9990 Å but not without a certain broadening of the laser emission line. The development of such a laser makes it possible to conduct for the first time the frequency selection of the Nd^{3+} ions by a resonant monochromatic light.¹⁾

In this work we present the first results of investigation of the structure of inhomogeneously-broadened bands and migration of excitation electrons in the metastable ${}^4F_{3/2}$ levels of Nd^{3+} ions excited by resonant monochromatic radiation at the ${}^4I_{9/2} \rightarrow {}^4F_{3/2}$ transition.

As a specimen, we used Li-La-Nd phosphate glass that permits introduction of high Nd^{3+} concentrations and yielding smaller inhomogeneous broadening terms as compared to silicate glasses, which tends to impede investigation by the method of the nonresonant monochromatic excitation.⁽⁴⁾

Figure 1 shows the characteristic spectra of fluorescence ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ of Nd^{3+} ions excited by broadband and monochromatic resonant radiation $\lambda_{\text{exc}} = 8725 \text{ \AA}$ and $\lambda_{\text{exc}} = 8752 \text{ \AA}$ at $T = 77 \text{ K}$. Clearly, selective excitation leads to a sharp narrowing of the spectral fluorescence lines (especially resonant) whose position is essentially determined by the wavelength of the pumping radiation. This indicates a correlation of the energy states of a number of Stark sublevels of the electronic terms ${}^4F_{3/2}$ and ${}^4I_{9/2}$, and it permits construction of outline diagrams of the Stark splitting for a family of spectrally-nonequivalent Nd^{3+} centers in glass. The lower portion of this diagram in Fig. 2 shows an absorption spectrum for the ${}^4I_{9/2} \rightarrow {}^4F_{3/2}$ transition at which the selective

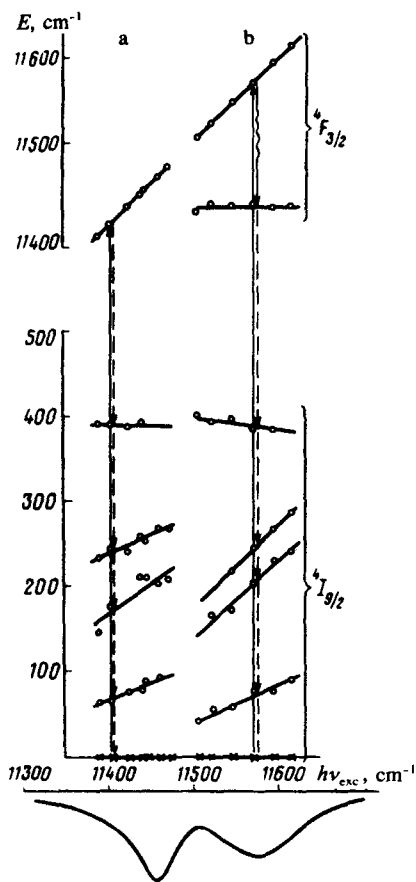


FIG. 2. Outline diagrams of the Stark splitting of ${}^4F_{3/2}$ and ${}^4I_{9/2}$ levels: a—excitation of the lower Stark component ${}^4F_{3/2}$; b—excitation of the upper Stark component ${}^4F_{3/2}$.

excitation of Nd^{3+} ions takes place. The abscissa represents the pumping radiation wavelength and the ordinate, the energy of the Stark sublevel states ${}^4I_{9/2}$ and ${}^4F_{3/2}$. A comparison of the two diagrams shows differences in the occurrence of inhomogeneous broadening of the transition spectra with the participation of the lower and upper components of the ${}^4F_{3/2}$ metastable level.

The significant factors requiring consideration when observing the frequency selection of Nd^{3+} ions in the excited states are concentration of the active ions in a specimen and registration of the onset of fluorescence after a rapid excitation of specimen. Thus, multipole coupling among the Nd^{3+} ions is enhanced at high dopant concentrations, which lead to nonradiative energy migration of electronic charges over the metastable levels ${}^4F_{3/2}$. This process may lead to a total loss of excitation selectivity in a family of the spectrally-nonequivalent centers.

Time gating of the fluorescence signal by means of the PAR-162 strobe-integrator permits observation of the frequency selection effect in the spectra even at high Nd^{3+} concentrations (in our case $n(\text{Nd}) = 2.7 \times 10^{20} \text{ cm}^{-3}$) and to study, from the time de-

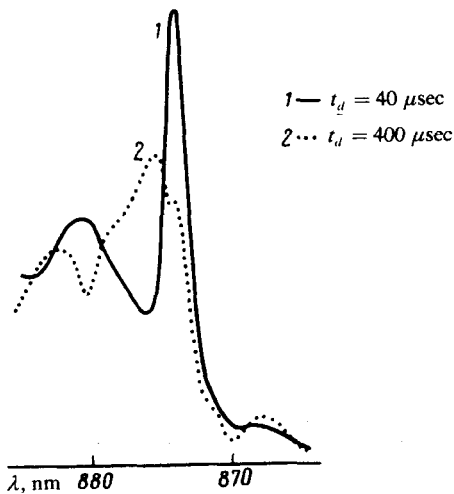


FIG. 3. Time dependence of fluorescence spectra in the case of resonant monochromatic excitation $\lambda_{exc} = 8737 \text{ \AA}$ and $T = 77 \text{ K}$: curve 1— $t_d = 40 \text{ \mu sec}$, curve 2— $t_d = 400 \text{ \mu sec}$.

pendence in the fluorescence spectra, energy transfer processes and coupling among the ions. Figure 3 shows sections of fluorescence spectra recorded at different times after excitation by resonant monochromatic radiation. Clearly, as t_d grows from 40 to 400 μsec , the spectra change substantially. The narrow spectral component broadens and shifts in the longwave direction. These changes depend on the migration coupling of the Nd^{3+} ions with a participation of the resonant transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ and they indicate a directed energy transfer from the high-energy centers with $\lambda_{fluo} = 8737 \text{ \AA}$ to centers with lower energies of the ${}^4F_{3/2}$ level $\lambda_{fluo} = 8753 \text{ \AA}$. This selective unidirectional transfer of electron excitation energy may be explained only in terms of nonresonant coupling of $\text{Nd}^{3+} \rightleftharpoons \text{Nd}^{3+}$ ions, where each act of nonradiative energy transfer is accompanied by a phonon emission. Moreover, energy transfer into the shortwave region of the spectrum requires that a certain portion of energy be absorbed from the phonon well. A low experimental temperature $T = 77 \text{ K}$ provides for a less effective process of up-migration with respect to the energy scale including phonon absorption, than down-migration accompanied by a phonon emission.

Subsequently, more detailed investigation of the temporal variations in the spectra with the resonant selection of Nd^{3+} ions permits us to determine the more important characteristics of inter-ion coupling, such as the multi-poleness parameter, the efficiency and its dependence on the frequency displacement.

¹¹Selective laser excitation of Nd^{3+} ions at the resonant transition was carried out concurrently with our work by means of a tunable semiconductor laser.¹¹

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