small amounts of impurities in gases but, for the purposes of certification of gas purity, it will be necessary to develop special methods because published information shows a lack of calibration techniques for the determination of impurities at the level of sensitivity and rapidity that we have attained.

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Selective-laser-excitation study of the structure of inhomogeneously broadened spectra of Nd^{3+} ions in glass

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The selective-laser-excitation technique was used to examine the structure of the inhomogeneously broadened bands of the Nd^{3+} ion in silicate glass. A survey diagram of the Stark splittings of the $^4I_{9/2}$ level in various optical centers in the glass is presented for the first time. The luminescence decay kinetics and the radiative transition probabilities A for various centers in neodymium glass are investigated. It is shown that the variations of the mean lifetime of the $^4F_{3/2}$ level under selective excitation of various optical centers in the glass correlate with the variations of A for the centers.

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Lasers with fixed wavelength, but especially tunable lasers, have recently been finding wider and wider application as excitation sources for spectroscopic studies. This is due to their unique capability of concentrating luminous energy in space, in time, and in spectrum. The use of laser sources to excite luminescence together with stroboscopic photorecorders that allow one to fix the emission (record the spectrum) instantaneously at selected times after the excitation enable one to approach the study of the Stark structure of inhomogeneously broadened spectra and the relaxation processes hidden by the inhomogeneous broadening at a qualitatively new level.[1,2]

Inhomogeneous broadening of spectra is a manifestation of the disorder of the structure of the activated

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medium. In media that are so widely used in quantum electronics as mixed crystals and glasses, the inhomogeneous broadening of the spectra of rare-earth ions is so great that it entirely smooths out the spectral characteristics of individual optical centers: the Stark splitting, radiative transition probabilities, lifetimes, and the magnitudes of the homogeneous broadening.

The lack of information on the structure of the inhomogeneously broadened bands and on the rates of energy exchange within the inhomogeneous contour substantially hinders the calculation of specific laser systems as well as the analysis and further study of the processes taking place in an excited activated medium. The selective laser excitation technique makes it possible to fill these gaps. This technique is based on the narrowing of nonuniformly broadened luminescence lines incident to narrow-band selective excitation of individual centers. This makes it possible to reveal the structure of the Stark splitting and other properties of individual optical centers. In this technique one must distinguish two schemes for observing the luminescence, depending on whether the luminescence to be observed is or is not in resonance with the transition used for selective excitation of the ions (Fig. 1). The greatest narrowing of the luminescence lines is seen when the luminescence is recorded at the same transition as is used for selective excitation—the resonance scheme (Fig. 1a)—since the inhomogeneous component of the luminescence-line broadening narrows to the width of the laser line and can be made very narrow.[1] When one observes nonresonance luminescence (Fig. 1b) the inhomogeneously broadened spectrum also appears narrower, but the narrowing may be less marked since the correlation between the energies of the radiative transition and the transition on which the laser photon was absorbed may be incomplete.[1]

The decrease of the inhomogeneous component of the broadening of the spectrum lines makes it possible to determine the following important characteristics of individual centers[1-5]:

1) The magnitude of the homogeneous broadening, its variation from center to center in the glass, and the temperature dependences of the spectrum-line broadenings,

2) The Stark-splitting energies for various optical centers and the correlation of the energies of transitions from center to center,

3) The lifetimes and radiative relaxation rates of transitions,

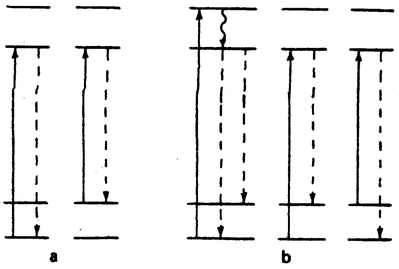

FIG. 1. Luminescence excitation and emission schemes: *a*—resonant, *b*—nonresonant.

4) The spectral migration of energy on the inhomogeneously broadened contour from laser-excited centers to spectrally distinct unexcited centers.

EXPERIMENTAL TECHNIQUE

To investigate the structure of inhomogeneously broadened spectra by the selective excitation method we developed a technique involving a narrow band tunable dye laser employing a rhodamine 6G solution[6] and operating in the periodic pulse regime ($\lambda_{gen} = 560$–630 nm, $\Delta\lambda = 0.1$ nm, $t_p = 10^{-8}$ sec, and $f = 12.5$–100 Hz), together with a photorecorder based on an FEU-83 photomultiplier and a strobe integrator (type PAR-162 boxcar integrator). The apparatus is diagramed schematically in Fig. 2.

The working dye (rhodamine 6G) was excited to generation in a three-prism dispersion resonator by the second harmonic radiation from a standard type LTIPCh-7 garnet laser containing neodymium. The wavelength of the emission from the tunable laser was monitored visually, using a Carl Zeiss type PGS-2 spectrograph. A type UTREKS cryostate (Cr) was used to control the temperature of the specimen; it enabled the temperature to be held at values from 4.2 to 300 °K. To register the spectra and isolate the luminescence lines we used a type MDR-2 high-transmission grating monochromator (MCh) having a dispersion of 4.0 nm/mm. The luminescence signal $I(t)$ was registered with an FEU-83 photomultiplier (PhM) and was fed into one of the channels of the strobe integrator. As was noted above, the strobe integrator is capable of recording delayed spectra $I(t_d; \lambda_L; \lambda_e)$ with a time resolution Δt down to 5 nsec.

The power output of the tunable dye laser varies as the generating wavelength λ_{gen} is altered. To compensate for these variations, part of the laser light was diverted to a bolometer (Bol) whose sensitivity is virtually constant over the entire spectral interval (we made special measurements to verify this). The reference signal from the bolometer was brought through an amplifier (Amp) to the second channel of the strobe integrator. The time instability of the laser intensity as well as the variations of the laser intensity incident

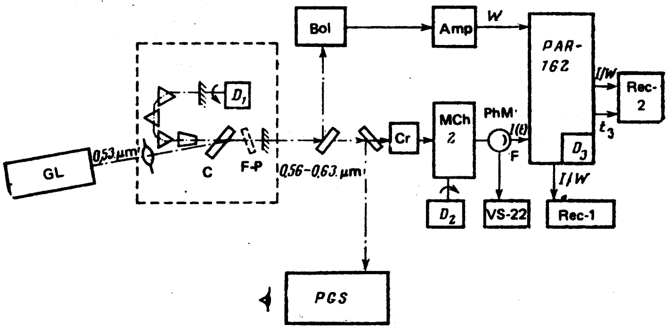

FIG. 2. Diagram of the apparatus: GL—type LTIPCh-7 garnet laser with neodymium, Bol—bolometer, Amp—amplifier, PGS—spectrograph, Cr—cryostat, MCh—type MDR-2 monochromator, PhM—type FEU-83 photomultiplier, PAR-162—boxcar integrator, Rec-1—type EPP-09 plotter, Rec-2—type PDS-021 recorder.

to varying λ_{gen} were taken into account by dividing the signal $I(t_d; \lambda_L; \lambda_e)$ by reference signal $W(\lambda_e)$ during the process of registering the entire spectrum. Then the signal I/W was recorded with an EPP-09 plotter (Rec-1) or a PDS-021 recorder (Rec-2).

Used in the time gating regime, this technique makes it possible to measure the luminescence decay curve $I_L(t)$ at fixed exciting and recording wavelengths λ_e and λ_L, the luminescence spectrum $I_L(\lambda_L)$ at fixed λ_e and fixed delay time t_d between the excitation and the beginning of recording, and the luminescence excitation spectrum $I_L(\lambda_e)$ with a resolution of 0.2–0.3 nm for fixed λ_L and $t_d(\Delta t)$. In the work reported here luminescence decay curves were recorded for various values of λ_e by photographing the screen of a TR-4401 oscilloscope when a 160-nm wide section of the spectrum in the vicinity of the maximum intensity of the $^4F_{3/2} \to {}^4I_{9/2}$ transition in Nd^{3+} was isolated.

EXPERIMENTAL RESULTS AND DISCUSSION

It is important to study the structure of the inhomogeneously broadened bands of the Nd^{3+} ion in glasses because of the widespread use of neodymium glass lasers. The commercial silicate glass LGS-28 is a typical example of a laser glass having considerable inhomogeneous broadening and, as a result, having almost structureless Nd^{3+} luminescence lines. The metastable $^4F_{3/2}$ level in Nd^{3+} lies lower than the corresponding level in the Eu^{3+} ion, which we investigated earlier,[1] so that for selective resonant excitation of the $^4I_{9/2} \to {}^4F_{3/2}$ transition one requires a pulsed periodic tunable laser whose λ_{gen} can be varied in the vicinity of 0.88 μm. No such lasers are available at present, so for our studies we chose the nonresonant scheme for selective excitation of the Nd^{3+} on the $^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ transition, using the radiation from a rhodamine 6G tunable laser.

We measured the luminescence and excitation spectra with time grating, as well as the kinetics of the luminescence decay, on specimens of LGS-28 glass containing 0.3 wt.% of Nd_2O_3 at temperatures T of 4.2 and 77 °K. Analysis of the luminescence spectra from the $^4F_{3/2} \to {}^4I_{9/2}$ and $^4F_{3/2} \to {}^4I_{11/2}$ transitions obtained by varying the wavelength of the laser light within the limits of the $^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ absorption band ($\lambda_e = 566$–600 nm) showed that these spectra do differ substantially from the spectra obtained by nonselective excitation (using an incandescent lamp with an SZS-21 filter) and, in addition, are strongly λ_e dependent. Thus, we were able to achieve the greatest narrowing of the luminescence spectra by exciting in the short wave and long wave wings of the absorption band. This can be understood, since the absorption is due to a transition from the ground level to individual Stark components of the $({}^4G_{5/2}, {}^2G_{7/2})$ state, the lowest and the highest.

Exciting in the center of the absorption band where the spectra of the Stark components of various centers pile up does not lead to narrowing of the lines, and in some cases one even finds the spectra to be broadened, owing, most likely, to the simultaneous preferential excitation of centers having the longest- and shortest-wavelength luminescence spectra. These facts indicate that there are correlations between the energies of the Stark levels of the $^4I_{9/2}, {}^4I_{11/2}, {}^4F_{3/2}, {}^4G_{5/2}$, and $^2G_{7/2}$ states, as is necessary if the structures of the bands are to be investigated by nonresonant selective excitation.

Figure 3 shows the luminescence spectra of the $^4F_{3/2} \to {}^4I_{9/2}$ and $^4F_{3/2} \to {}^4I_{11/2}$ transitions for excitation at two wavelengths in the short wave part of the absorption spectrum, namely at $\lambda_e = 575$ nm (Fig. 3.1) and $\lambda_e = 568$ nm (Fig. 3.2). It is evident that shifting the exciting light toward the shorter wavelenghts (to the wing of the absorption spectrum) leads to a large shift of the luminescence lines and brings out more clearly the structure of the $^4F_{3/2} \to {}^4I_{9/2}$ and $^4F_{3/2} \to {}^4I_{11/2}$ bands. Moreover, the fourth Stark component of the $^4I_{9/2}$ level is resolved, as well as the third (high-energy) Stark component of the $^4I_{11/2}$ level, which is not seen with unselective excitation and had not previously been observed. The changes in the luminescence spectra are such that the short wave components shift toward the shorter wavelengths and the long wave components, toward the longer wavelengths. This indicates that there are Nd^{3+} centers in this glass that have substantially different Stark splittings for the $^4I_{9/2}$ and $^4I_{11/2}$ levels. Thus, centers for which the energy of the $^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ transition is low ($\lambda_e = 575$ nm) have small Stark splittings (lie in a weak field of ligands), while for centers for which the $^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ transition energy is higher ($\lambda_e = 568$ nm) the Stark splitting is substantially greater (the ligand field is stronger).

Reasoning in this way, we may regard the energy of the $^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ transition as an index (parameter) of the strength f of the ligand field and plot the Stark splitting of the $^4I_{9/2}$ level in various optical centers in the glassy matrix as a function of this parameter (much as was done earlier for Eu^{3+} centers in sodium borosilicate glass[1]). For this purpose we recorded and analyzed a series of luminescence spectra for the $^4F_{3/2} \to {}^4I_{9/2}$ transition, varying the exciting wavelength λ_e from 575 to 566 nm in 1 nm steps (these measurements were made at $T = 77$ °K; reducing the temperature to 4.2 °K had little effect on the spectra). The results of these measurements are shown diagramatically in Fig. 4. The dashed curve in the upper part of the figure

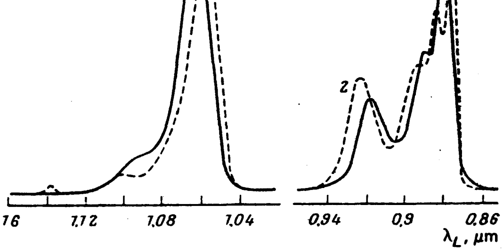

FIG. 3. Luminescence spectra of type LGS-28 silicate glass under selective laser excitation ($T = 77$°K); curve 1 (the full curve) is for $\lambda_e = 575$ nm, and curve 2 (the dashed curve), for $\lambda_e = 568$ nm.

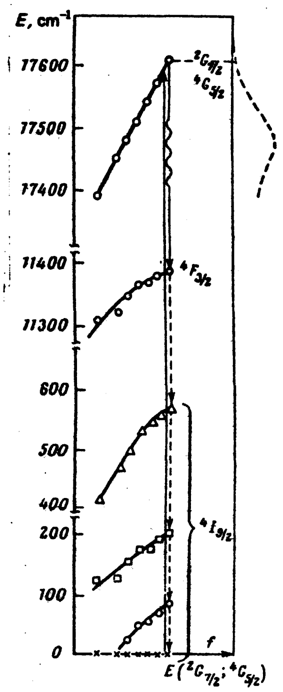

FIG. 4. Stark-level diagram for a continuous set of Nd^{3+} centers in LGS-28 glass.

shows the short wave edge of the absorption spectrum in which the Nd^{3+} ions were excited. The scheme for exciting and recording the luminescence is also shown. The laser light selectively excites only centers for which the $^4I_{9/2} \to (^4G_{5/2}, {}^2G_{7/2})$ transition (full arrow) has a definite energy. When the centers are excited they decay rapidly by a nonradiative transition to the metastable $^4F_{3/2}$ level (wavy line), and luminescence corresponding to transitions from the lower component of the $^4F_{3/2}$ level to levels of the $^4I_{9/2}$ multiplet is later emitted. The energies of the levels were determined from the positions of the four maxima in the luminescence spectrum as measured from the lower Stark component of the $^4I_{9/2}$ ground state (the fifth maximum corresponding to the transition to the fifth Stark level of the $^4I_{9/2}$ state was not observed).

The diagram shows the relation between the energies of the different Stark levels of the $(^4G_{5/2}, {}^2G_{7/2})$, $^4F_{3/2}$, and $^4I_{9/2}$ states, the correlated variations of these energies, and the quantitative values of the Stark splittings of the $^4I_{9/2}$ level for various optical centers in the glass. It reveals the complex structure of the inhomogeneously broadened collective spectrum of Nd^{3+} ions in such a highly disordered matrix as silicate glass.

The above demonstrated possibility of distinguishing various Nd^{3+} optical centers in glass by nonresonant laser excitation made it possible to investigate such important spectroscopic characteristics of the elementary emission centers in glass as the lifetime τ of the metastable level and the spontaneous radiative transition probability A.

We made use of this possibility to investigate the decay kinetics of the Nd^{3+} luminescence excited by monochromatic laser light at various wavelengths. The Nd^{3+} luminescence of the LGS-28 glass was found to decay in a nonexponential manner, as it had previously been found to do under broader-band excitation;[7] moreover, the luminescence decay rate was found to depend substantially on the exciting wavelength[1)] λ_e. Figure 5 shows a series of decay curves taken at 4.2°K and at several values of λ_e from 566 to 573 nm in the shortwave region of the absorption spectrum, for which we already have an analysis of the Stark structure of the $^4I_{9/2}$ state in various centers (see Fig. 4). It is evident from curves 1–3 in Fig. 5 that there is a certain decrease in the luminescence decay rate at all stages of the decay process as the exciting wavelength is increased from 566 nm to 567 and then to 568 nm. Comparison with the diagram (Fig. 4) shows that these changes correlate with the decrease in the Stark splitting (weakening of the ligand field strength) of the centers excited at $\lambda_e = 568$ nm as compared with those of the centers excited at $\lambda_e = 566$ nm. Such correlation between τ^{-1} and ΔE allows us to suppose that the changes in τ^{-1} are due to a change in the radiative transition probability A incident to a simultaneous decrease of both the even ligand field parameters (which are responsible for ΔE) and of the odd ones (which are responsible for A). However, such a correlated change in both the even and odd field strength parameters is not at all necessary, and in the general case it may not occur (for example, incident to a change in the symmetry of the optical center).

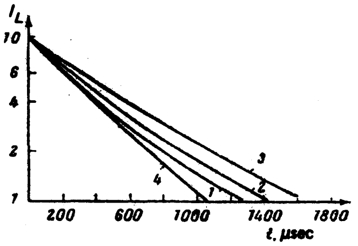

FIG. 5. Decay curves for the luminescence from the $^4F_{3/2}$ level of Nb^{3+} in LGS-28-0.3 glass under selective laser excitation ($T = 4.2$°K); curves 2—4 are for $\lambda_e = 466$, 567, 568, and 573 nm, respectively.

It is evident from Fig. 5 that as λ_e increases monotonically the lifetime τ first rises smoothly and then falls sharply (the decay rate rises) when λ_e reaches 573 nm (curve 4). This increase in τ^{-1} is no longer correlated at all with the small Stark splitting ΔE for these centers (Fig. 4), and there are two possible reasons for it: 1) an increase in the radiative transition probability A for the center concerned occasioned by an increase in the odd component of the crystal field (even though the even component decreases), as a result of which the electric dipole transitions $^4F_{3/2} \to {}^4I_J$ are no longer forbidden; and 2) and increase in the nonradiative losses from the $^4F_{3/2}$ level for this center.

It is very important to discover the causes for such sharp changes in the decay rate of the metastable state, since the same causes determine the relaxation channels for the energy stored in the laser, and this, in the end, affects the laser's efficiency. In an attempt to discover these causes we conducted an independent study of the relative variations of the radiative transition probabilitites from center to center as λ_e was varied and compared them with the corresponding change of the mean lifetime $\tau(\lambda_e)$ for the same centers. The relative variations of the radiative transition probability $A(\lambda_e)$

can be obtained by comparing the excitation spectra for the Nd^{3+} luminescence at the ${}^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ band with the absorption spectra at this same transition. The luminescence intensity I_L immediately after rapid excitation (at $t=0$) is of course given by

$$I_L(0, \lambda_e) = n^*(0, \lambda_e) A(\lambda_e) h\nu_L. \tag{1}$$

It is evident from this formula that the dependence of the luminescence intensity on the wavelength of the exciting light (the luminescence excitation spectrum) is determined by the λ_e dependences of two parameters: $n^*(0, \lambda_e)$, the number of excited Nd^{3+} ions, and $A(\lambda_e)$ ($h\nu_L$ is the luminescence photon energy). Under the assumption that there are no (or relatively few) Nd^{3+} ions that have zero quantum yield, i.e., that absorb photons but do not emit them, the number $n^*(0, \lambda_e)$ of Nd^{3+} ions excited to the metastable level will be equal to the number of photons absorbed in the specimen (active absorption):

$$n^*(0, \lambda_e = W_e(\lambda_e) D(\lambda_e)/h\nu_e, \tag{2}$$

where $W_e(\lambda_e)$ is the energy dispersion of the light from the tunable laser, $D(\lambda_e) = I_{abs}(\lambda_e)/I_0$ is the relative absorption of energy on the ${}^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ transition or the absorption spectrum in relative units for a specimen of low optical density, and $h\nu_e$ is the photon energy of the exciting radiation.

On substituting expression (2) into formula (1) we obtain

$$\frac{I_L(0, \lambda_e)}{W_e(\lambda_e)} = A(\lambda_e) D(\lambda_e) \frac{h\nu_L}{h\nu_e} \ . \tag{3}$$

Our technique for registering the luminescence using the PAR-162 allows us simultaneously to record $I_L(t, \lambda_e)$ and $W_e(\lambda_e)$ and so to obtain the yield function in the form of the required ratio $I_L(t, \lambda_e)/W(\lambda_e)$. The form of the excitation spectrum at the short delay time[2)] $t_d = 10$ μsec (at the initial instant) as corrected in this manner is shown by curve 1 in Fig. 6. The excitation spectrum for the long delay $t_d = 3$ msec (curve 2) is also given, but only to show the long lived centers more prominently, since by that time the short lived centers will have died out. The absorption spectrum $D(\lambda_e)$ is shown by curve 3 (in relative units) for comparison. It will be seen that for centers excited at $\lambda_e < 573$ nm ($\lambda_e > 573$ nm) the excitation spectrum for $t_d = 10$ μsec (curve 1) runs below (above) the absorption spectrum (curve 3). As formula 3 shows, this can be explained only by an increase in the Einstein coefficient A with increasing excitation wavelength near $\lambda_e = 573$ nm ($h\nu_e$ changes little—by less than 10%—within the limits of the absorption band).

Dividing the excitation spectrum (curve 1) by the absorption spectrum (curve 3), we obtain A (in relative units) as a function of λ_e. The resulting $A(\lambda_e)$ curve is shown as curve 1 in Fig. 7. To compare this curve with the decay rates of the metastable ${}^4F_{3/2}$ state in different centers we determined the average lifetime τ_{av} for different λ_e values by integrating the corresponding nonexponential luminescence decay curves:

$$\tau_{av} = \frac{1}{I_L(0)} \int_0^\infty I_L(t)\,dt. \tag{4}$$

These values of τ_{av} are also plotted on Fig. 7 (curve 2). Comparison of the $A(\lambda_e)$ and $\tau_{av}(\lambda_e)$ curves shows that there is a sharp fall in τ_{av} as λ_e rises from 569 to 575 nm, corresponding to the rise in the radiative transition probability A on that same interval.

In the work reported here we attempted to obtain a quantitative correspondence between $A(\lambda_e)$ and $\tau_{av}(\lambda_e)$, so in determining the λ_e dependence of A we did not take all the Nd^{3+} luminescence lines into account, but only a region 160 nm wide cut from the peak of the ${}^4F_{3/2}-{}^4I_{9/2}$ luminescence spectrum with the aid of an MDR-2 monochromator. As a result of the redistribution of the transition probability A over the Stark levels or multiplets for the different centers, the λ_e dependence of the total transition probability $\Sigma A(\lambda_e)$ may differ somewhat from the dependence shown by curve 1 in Fig. 7.

We also examined the luminescence decay kinetics for various values of λ_e at $T = 77$°K in an Li-La-Nd-phosphate glass (containing 1 wt.% Nd_2O_3) that has been described elsewhere,[8,9] and which has substantially narrower spectrum lines than the LGS-28 glass. Unlike the LGS-28 glass, the Li-La-Nd-phosphate glass has a quantum yield of nearly unity for the luminescence from the ${}^4F_{3/2}$ level at the specified activator concentrations.[9,10] The variations of τ in the phosphate glass for different excitation wavelengths amounts to ~13% (from 330 to 380 μsec) while the luminescence decay curves turn out to be fairly well approximated by exponentials. On the basis of these facts we may conclude that the variations of the radiative transition probabilities for the different optical centers in the phosphate glass that we investigated are very small.

We note that only correlated variations of $\tau(\lambda_e)$ and

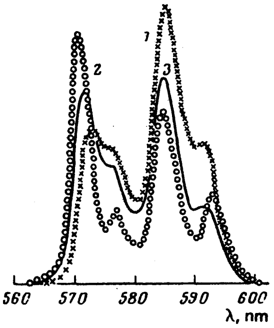

FIG. 6. Luminescence excitation spectrum for Nd^{3+} in LGS-28-0.3 glass at different delay times t_d (curve 1, $t_d = 10$ sec; curve 2, $t_d = 3$ msec) and the absorption spectrum at the ${}^4I_{9/2} \to ({}^4G_{5/2}, {}^2G_{7/2})$ transition (curve 3) at 77°K.

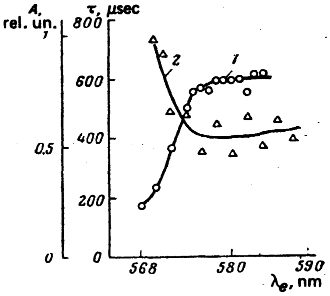

FIG. 7. Average delay time τ_{av} for the luminescence from the ${}^4F_{3/2}$ level of Nd^{3+} (curve 2) and the radiative transition probability $A({}^4F_{3/2} \to {}^4I_{9/2})$ (curve 1) vs exciting-light wavelength λ_e.

$A(\lambda_e)$ detected by two different methods make it possible to establish at least one of the reasons for the nonexponential decay of the Nd^{3+} luminescence in LGS-28 glass and for the λ_e dependence of τ_{av}—the reason associated with the variations of the radiative transition probabilities A for different optical centers in the glass. Just the fact that a λ_e dependence of τ has been observed, however, is not enough to permit a correct conclusion to be reached concerning the mechanism responsible for the changes in the τ values for the different centers.

Thus, our results show that by using nonresonant selective excitation one can obtain new information concerning the Stark structure, the variations of the decay kinetics, and the variations of the radiative transition probabilities A for various optical centers in a disordered activated medium, i.e., information that is usually quite hidden by large inhomogeneous broadening when traditional spectroscopic methods are used.

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[1] In an earlier study[7] it was not possible to detect the λ_e dependence of τ because of the higher temperature ($T = 300$°K) and broader-band excitation sources ($\Delta\lambda_{gen} = 1.0$–$1.5$ nm) that were used.

[2] The choice of a short delay time of 10^{-5} sec in recording the excitation spectra eliminates the effects of slow nonradiative deactivation processes such as intracenter relaxation and $Nd^{3+} \rightleftarrows Nd^{3+}$ cross relaxation, which, as our measurements show, cannot change the population of the $^4F_{3/2}$ level in so short a time as 10^{-5} sec when the Nd^{3+} concentration is as low as it was (0.3 wt.%).

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Investigation of the absorption spectrum of a two-level system subjected to intense two-photon excitation

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We investigate the absorption spectrum of weak emission from a two-level system (the Zeeman-splitting levels of ^{113}Cd atoms) under intense two-photon excitation. It is observed that, at sufficiently high probability of two-photon excitation, a change in the sign of the absorption coefficient (i.e., amplification) takes place without population inversion, analogous to the changes of the absorption contour in resonant (single-photon) excitation. A theory is constructed that describes the shape of the absorption line in the case of two-photon excitation of a two-level system; the results of the theory agree well with the experimental data. It is shown that reversal of the sign of the absorption coefficient can be used to convert laser emission frequency (to generate the third optical harmonic). Estimates of the efficiency of the frequency tripling of the 7784 Å emission in sodium vapor show that near the third-harmonic frequency it is possible to attain a gain of ~ 1 cm^{-1} at a sodium atom concentration $\sim 10^{16}$ cm^{-3} and at a pump intensity $\sim 10^7$ W/cm^2.

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1. INTRODUCTION

The changes in the absorption and emission line shapes of a two-level system under intense resonant excitation have by now been investigated in sufficient detail.[1-11] The usual Lorentz shape of the absorption and emission lines is the consequence of the exponential law governing the damping of the upper and lower states of the system. The change in the kinetics in the damping of the states of an atom subjected to intense excitation is the reason for the change in the atomic emission and absorption line shapes.

The most interesting phenomenon, first predicted theoretically,[1] is the essential difference between the changes in the line shapes of the spontaneous emission and absorption of auxiliary weak radiation. In the case of intense resonant excitation, the probability of the transition of an atom to an upper state comes close to the stationary value, oscillating about this value at a frequency $dE/\hbar$ (d is the matrix element of the dipole