A new generation of solid-state lasers

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A discussion is presented of recent advances in research and development relating to high-efficiency lasers made from activated ionic crystals (specifically, gadolinium-scandium-gallium garnet activated by Nd^{3+} and Cr^{3+} and yttrium aluminum garnet activated by Er^{3+}) and multicomponent glasses activated by Nd^{3+}, Er^{3+} and Yb^{3+}, along with tunable color-center lasers made from lithium fluoride. The physical processes, which occur in these active crystals and which lead to enhanced population inversion associated with the laser transition, are discussed, including energy transfer from sensitizer ions to active ions and cross-relaxation between the level systems of neighboring active ions. Processes that give rise to increased efficiency in tunable color-center lasers based on LiF crystals are analyzed; also mentioned is work relating to the fabrication of passive Q switches using LiF crystals with F_2^- color centers.

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1. INTRODUCTION

This year, scientists in every country of the developed world took note of the 25th anniversary of the discovery of the laser. In the course of the last 25 years, lasers of various types have undergone continual improvements, which have confirmed their ability to address a host of problems relating to practical applications: in communication engineering and medicine, technology and navigation, science, and ecology. In all fields of endeavor, use of the laser has led to revolutionary changes.

Let us first note that in the last quarter-century, progress in the laser field was marked by keen competition between various types of lasers, so that in various time periods the preference was now for one type, now for another type of laser. In spite of this strong competition, solid-state lasers, i.e., those made of crystals and glasses, were most often used in practical applications. For example, according to the statistics presented in Ref. 1, 42% of the total sales of lasers in the world market (excluding socialist countries) were for solid-state lasers. More than half the volume of sales of solid-state lasers was for lasers based on yttrium aluminum garnet with neodymium (Nd–YAG). These data reflected the state of affairs in 1982; in the past three years, the share of solid-state lasers in the world market has tended to increase even more.

The reason that solid-state lasers have become so widely disseminated lies in the fact that, although they do not exhibit record-breaking values for some specific laser parameters (e.g., average radiated power or efficiency), the overall performance of these lasers with respect to all the parameters taken together makes them unquestioned leaders in the field. The fundamental advantage of solid-state lasers is judged to be the combination of their high energy-generation parameters (up to 10 J per pulse, with an average generated power of up to 700 watts) and their ability to operate in various regimes—from ultrashort (picosecond length) pulses to continuous operation (CW)—along with high power, exceptional reliability and longevity; furthermore, these lasers are extremely compact.

In the last year, great advances have been made in understanding the physics of activated crystals and glasses. The most important regularities have been investigated and understood with regard to the formation of active centers in laser hosts,[2,3] transport processes and energy transfer of electronic excitations between active impurity ions,[4,5–8] and dependence of spectral-luminescence and laser properties on composition and structure of laser hosts.[4–6,8–11] As a result, a new generation of active media has emerged—crystals and glasses that can be used to make lasers that tune smoothly with wavelength over wide regions of the electromagnetic spectrum. The use of these media has abruptly increased the efficiency of the solid-state laser. Using these materials, lasers have been made which emit in formerly unexplored wavelength bands, e.g., the 1.5- and 3-micron bands. Among the new "neodymium" lasers, which emit at $\lambda = 1.06$, especially noteworthy are lasers:

—made from concentrated neodymium phosphate glasses (CNPG)[11];

—made from athermal phosphate glasses[12];

—made from gadolinium-scandium-gallium garnet with chromium and neodymium (GSGG-Cr-Nd)[10,13–18];

—made from yttrium aluminate with neodymium[19];

—made from potassium-gadolinium tungstate with neodymium (KGT–Nd).[20]

Lasers made of GSGG-Cr-Nd can deliver efficient emission at both 1.06- and 0.9-micron wavelengths.[21] In any list of "sesquimicron" lasers one should include lasers made of complex glasses with chromium, ytterbium and erbium.[22] The erbium laser has unique parameters, radiating at $\lambda = 2.94$ microns in crystals of yttrium-erbium-aluminum garnet.[23]

Compact and reliable tunable lasers whose output overlaps the visible and near infrared region of the spectrum and which work at room temperature are made from crystals of lithium fluoride with color centers,[24-26] as well as crystals of $BeAl_2O_4$–Cr^{3+},[27] GSGG–Cr^{3+},[28,15,18] and Al_2O_3–Ti^{3+}.[29,30] The new developments enumerated above have not only greatly enhanced the competitive edge that solid state lasers have in comparison to other types of lasers, but also have provided new solutions to an array of important practical problems.

2. LASERS BASED ON GALLIUM GARNET CRYSTALS CONTAINING CHROMIUM

Despite their multiple virtues, even the most commonly used lasers made from crystals and glasses have an important drawback-low emission efficiency. For example, the efficiency of the well-known neodymium garnet laser does not exceed 1–2%, either in pulsed or continuous operation.[31] The fundamental cause of this deficiency is the poor match between the CW spectrum of the flashlamp pump and the weak, band-like absorption spectrum of neodymium in the active element. The principle ways of increasing the efficiency of a solid-state laser are well-known: One is to increase sharply the concentration of active impurities (e.g., neodymium ions), thereby increasing the optical density of the active medium. Another is to use a doubly-activated medium which contains supplementary impurities that strongly absorb the flashlamp pump radiation and then effectively transfer this energy to the active impurities. However, we encounter fundamental obstacles to realizing either of these solutions. Thus, a simple increase of active impurity concentration in most cases does not lead to a positive result, because of the well-known phenomenon of concentration quenching of the luminescence. It is only in the last year that we have come to understand the mechanism of concentration quenching in laser hosts.[32-41]

This fact has prompted investigators to look for crystals and glasses with neodymium where the concentration quenching is anomalously weak. The best-known of these crystals and glasses are the "concentrated neodymium phosphate glasses" (CNPGs)[11]; outstanding among these glasses are Q-100[42] and LHG-8,[43] along with some others noted in the experimental literature. The efficiencies of lasers based on these glasses far exceed (by 2 or 3 times) those of their well-known predecessors, the neodymium garnet and neodymium glass lasers.

The crystal GSGG-Cr-Nd[10] has only recently achieved a level of recognition among researchers; its emergence has led to considerable progress in the fields of solid-state laser physics and engineering. In these doubly-activated crystals, ions of Cr^{3+}, which have broad, strong absorption bands, effectively absorb the pump light and are put in excited states; they then transfer the excitations nonradiatively by the multiple-resonance mechanism to the highest working level of the neodymium ion.

The first attempt to use this "sensitizer effect" to increase the efficiency of neodymium lasers was undertaken in 1964,[44] when a laser was made from YAG-Cr-Nd crystals in which the Cr^{3+} ion played the role of sensitizer. The researchers who made this laser reported something close to a doubling of the emission efficiency of the laser in CW operation. However, there was no subsequent follow-on research based on this result, and in a host of practical applications people continued to use YAG crystals with neodymium alone. It was necessary for some 10 years to pass before the mechanism which gives rise to Cr^{3+}–Nd^{3+} pair interaction was investigated. In 1978 the authors of Ref. 45 gave a description of certain complex chromium-neodymium-aluminoborate crystals in which rapid energy transfer from Cr^{3+} to Nd^{3+}—of the order of less than 100 nanoseconds—was observed for the first time. The power necessary to cause a population inversion in the $^4F_{3/2}$ level of Nd^{3+} was found to be down by an order of magnitude compared to crystals without chromium. The authors explained the high efficiency of sensitization on the basis of the high Nd concentration in the crystals they studied, which implied a rather small separation between Cr^{3+} and Nd^{3+}. An important step in understanding the mechanism of energy transfer between Cr^{3+} and Nd^{3+} was taken in Refs. 5, 6, 46–48. In these references it was shown that a decisive role was played in the energy transfer by the energy gap ΔE between the ^2E and $^4J_2$ energy levels of the Cr^{3+} ion (Fig. 1). Rapid energy transfer from Cr^{3+} to Nd^{3+} (i.e., rapid compared to the rate of deactivation of the upper level $^4F_{3/2}$ of Nd^{3+}) was possible only in the case where $\Delta E \leqslant kT$ (1). In particular, it was shown that the Cr^{3+} level scheme in the YAG crystals did not satisfy this condition, and so the introduction of chromium in Nd–YAG did not lead to significant improvement under pulsed excitation. Subsequent searches for crystals and glasses with efficient excitation energy transfer from chromium to neodymium led to the synthesis of a whole
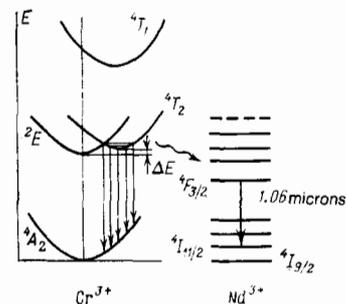
FIG. 1. Arrangement of low-lying energy states of the Cr^{3+} and Nd^{3+} ions.[5]
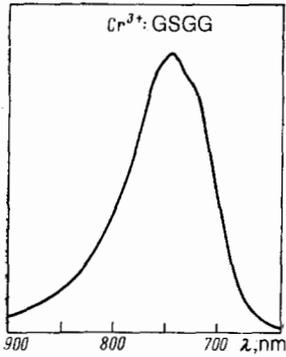
FIG. 2. Room temperature (300 K) luminescence spectrum for Cr^{3+} in GSGG crystals.[13]

range of crystals with the garnet structure, activated with Cr^{3+} and Nd^{3+} and satisfying condition (1). These materials were in essence substitutional compounds related to the gallium garnet group, among which the properties obtained in the crystal GSGG-Cr^{3+}-Nd^{3+} were found to be outstanding. This crystal is a complex gallium garnet in composition, into whose chemical makeup go, besides gallium, the elements gadolinium, scandium, chromium and neodymium.

The luminescence spectrum of Cr^{3+} in GSGG-Cr-Nd at 300K consists of a broad electron-vibronic band with a maximum at 750 nanometers (Fig. 2) (the transition $^4T_2 \rightarrow ^4A_2$ in the scheme of Fig. 1). It is important that the luminescence from the ^2E level corresponding to the R-luminescence line of Cr^{3+} is completely absent at 300 K, which shows as a consequence that the gap ΔE between ^2E and $^4T_2$ is close to zero. Detailed study of the decay kinetics of the excited states of Cr^{3+} and Nd^{3+}, completed in Refs. 13, 49, showed that the decay of the Cr^{3+} level in the absence of Nd^{3+} was exponential with a decay constant equal to 120 microseconds, and that its value did not depend on the chromium concentration up to a value of 6×10^{20} cm^{-3} for the latter (Fig. 3). This was an important fact; it showed that in GSGG-Cr-Nd, concentration quenching of Cr^{3+} did not take place. When Nd^{3+} was introduced into the crystal, the decay time of the $^4T_2$ level of Cr^{3+} was sharply decreased; furthermore, the decay became nonexponential in character,

and could be described by the following expression:

$$I(t) = I_0 \exp\left[-\left(\frac{t}{\tau_{Cr}} + \gamma t^{1/2} + \bar{w}t\right)\right], \quad (1)$$

where I_0 is the normalized luminescence intensity at time t_0,

$$\bar{\gamma} = \frac{4}{3}\pi^{3/2}n_{Nd}c_{DA}^{1/2} \quad (2)$$

is the static energy transfer parameter for $Cr^{3+} \rightarrow Nd^{3+}$, n_{Nd} the neodymium concentration, c_{DA} the microscopic interaction parameter, and $\bar{w}$ the migration-induced relaxation probability of the $^4T_2$ state.

In this way it was shown that γ grows linearly with the Nd^{3+} concentration and does not depend [in agreement with (2)] on the Cr^{3+} concentration; for $n_{Nd} = 2 \times 10^{20}$ cm^{-3}, it equals 210 ± 20 sec^{-1}, which leads to $c_{DA} = 2 \times 10^{-38}$ cm^6/sec. A comparison of the latter quantity with c_{DA} for YAG-Cr-Nd, calculated in Ref. 47, shows that in the case of GSGG-Cr-Nd, the value of c_{DA} is 20 times larger than that of YAG. Thus, in crystals of GSGG-Cr-Nd the rate of energy transfer from Cr^{3+} to Nd^{3+} can be much larger than the deactivation rate for the upper laser level $^4F_{3/2}$ of Nd^{3+}.

Thus, the results of these investigations of the spectral-luminescence properties of GSGG-Cr-Nd clearly attest to the efficiency of sensitization of the radiating ion Nd^{3+} by the ion Cr^{3+}.

In Refs. 10, 47, 49, relations were obtained which connect the emission and luminescence properties of GSGG-Cr-Nd crystals. It was shown that the enhancement of the population inversion of the upper laser level $^4F_{3/2}$ of the Nd^{3+} ion due to introduction of Cr^{3+} ions is given in steady state by

$$q = 1 + \frac{N_0^{(Cr)}}{N_0^{(Nd)}} \frac{\gamma(\pi\tau_{Cr})^{1/2}}{2} \exp\left(\frac{\gamma^2\pi\tau_{Cr}}{4}\right)\left[1 - \Phi\left(\frac{\gamma\tau_{Cr}^{1/2}}{2}\right)\right], \quad (3)$$

FIG. 3. Decay curves for the excited state $^4T_2$ of the Cr^{3+} ion in GSGG–Cr^{3+} crystals (1), and in GSGG–Cr^{3+}–Nd^{3+} crystals containing: (2) 1×10^{20} cm^{-3} Cr^{3+} along with 2×10^{20} cm^{-3} Nd^{3+}, (3) 1×10^{20} cm^{-3} Cr^{3+} along with 3×10^{20} cm^{-3} Nd^{3+} and (4) 1×10^{20} cm^{-3} Cr^{3+} along with 6×10^{20} cm^{-3} Nd^{3+}.[5]
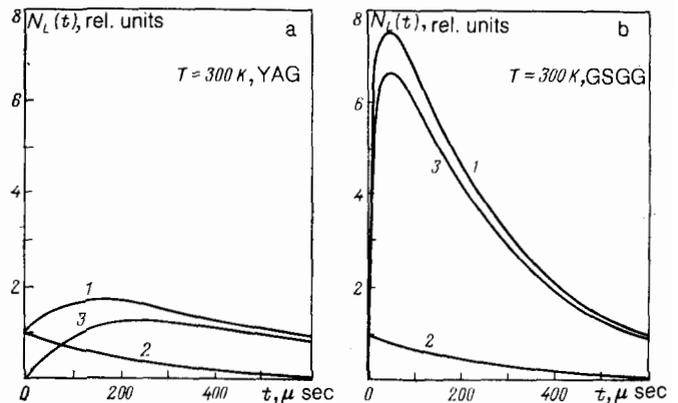
FIG. 4. Time dependences of excited-state populations $N_L(t)$ of the Nd^{3+} ion at 300 K in crystals co-activated with Cr^{3+} for excitation by δ function radiation pulses whose spectral content corresponds to the spectral content of light from a ISP-1000 lamp.[49] a) A YAG crystal with a neodymium concentration of 1×10^{20} cm^{-3} and a chromium concentration of 1.5×10^{20} cm^{-3}; b) A GSGG crystal with a neodymium concentration of 2×10^{20} cm^{-3} and a chromium concentration of 3×10^{20} cm^{-3}. Curves 1—total $N_L(t)$; curves 2—$N_L(t)$ for direct excitation of neodymium ions in their intrinsic absorption bands; curves 3—$N_L(t)$ for excitation of chromium ions, which then transfer their excitation to the upper level of neodymium ions.

TABLE I.

Characteristic	Value
Structure	cubic
Space group	$O_h^{10} - Ia3d$
Lattice constant, Å	12.545
Density, gm $-$ cm^{-3}	6.50
Melting temperature, °C	1850
Thermal conductivity, W-(Ohm^{-1}°K^{-1}	0.07
Specific heat, J-(gm-°K)$^{-1}$	0.448
Index of refraction	1.943 ($\lambda = 1060$ nm)
	1.955 ($\lambda = 750$ nm)
Elastic constants, 10^{11} Pascals	
c_{11}	2.69
c_{22}	1.02
c_{44}	0.774
Photoelastic constants	
p_{11}	-0.097
p_{22}	-0.040
p_{44}	-0.066
Modulus of hydrostatic stress, 10^{11} Pascals	1.58
Shear modulus, 10^{11} Pascals	0.8
Young's modulus, 10^{11} Pascals	2.05
Poisson ratio	0.28
Diffusion coefficient, Nd^{3+}	0.75
Diffusion coefficient, Cr^{3+}	1
Radiative lifetime, Nd^{3+} (microseconds)	280
Radiative lifetime, Cr^{3+} (microseconds)	120
Luminescent quantum efficiency for Nd^{3+}	1
Luminescent quantum efficiency for Cr^{3+}	1
Luminescent linewidth for laser transition of Nd^{3+} in the 1.06 micron region, Å	14
Maximum in the luminescence band of Cr^{3+}, nanometers	760
Half-width for luminescence band of Cr^{3+}, nanometers	130
Cross-section for laser transition of Nd^{3+} in the 1.06 micron region, 10^{-19} cm^2	1.5
Cross-section for laser transition of Nd^{3+} in the 0.9 micron region, 10^{-20} cm^2	4
Effective cross-section for laser transition of Cr for $T = 300$ K, 10^{-20} cm^2	0.9

where $N_0^{(Cr)}$ and $N_0^{(Nd)}$ are the densities of ions directly excited by the pump radiation—for Cr^{3+} and Nd^{3+} respectively—while γ and τ_{Cr} are determined in Eqs. (2, 3), and

$$\Phi(x) = \frac{2}{\pi^{1/2}} \int_0^x e^{-z^2}\, dz$$

is the error function.

Thus, it is found that the sensitizing efficiency of the active medium of a broad-band-pumped laser is determined by the absorption spectra of the donor and acceptor ions along with the values of γ and τ_{Cr}. In Fig. 4(a) the time dependences are shown for the $^4F_{3/2}$ level of Nd^{3+} at 300 K in a YAG crystal with a Nd concentration of 1×10^{20} and a Cr concentration of 1.5×10^{20} cm^{-3}. The figure illustrates the time evolution of the excited neodymium, obtained either directly from the pump flashlamp or from the chromium ions, when an ISP-1000 pump flashlamp is used; the electrical power density discharged in the plasma column of the latter was 0.23 MW/cm^{-3}. Now, it is clear from Fig. 4(a) that introducing Cr into Nd-YAG leads to almost no enhancement of the upper-laser-level population of the Nd^{3+} in pulsed operation. A completely different picture emerges when we consider the GSGG-Cr-Nd crystal, however [Fig. 4(b)]. Here, the concentrations of chromium and neodymium are equal to 3×10^{20} cm^{-3} and 2×10^{20} cm^{-3} respec-

tively. Curves 2 and 3 show the contributions to the population due to energy directly absorbed by the neodymium atoms, along with the energy absorbed by the chromium and then transferred to the neodymium. Curve 1 shows the total population of the $^4F_{3/2}$ level for excitation in both the chromium and neodymium absorption bands. Clearly, in GSGG-Cr-Nd (as opposed to YAG crystals) sensitization leads to a significant increase in the upper-laser-level population of the Nd^{3+}. In Ref. 49, the optimal Nd^{3+} concentration was calculated for various Cr concentrations for optically thin smaples of GSGG-Cr-Nd. Table I lists those basic properties of the GSGG-Cr-Nd crystals that are relevant to its usefulness as a laser host.

The first results on emission from GSGG-Cr-Nd were obtained in Refs. 13–15 and 28. Subsequently, a whole series of lasers were made from these crystals, which exhibited record-breaking parameter values. Thus, in Ref. 17, a pulsed laser made from GSGG-Cr-Nd is described which operated over a pumping range from 1 to 3 J. The pumping was done by flashlamp; the light source was a pipe coated with a reflecting coating, whose external diameter was 7 mm. The dimensions of the active element, which had 2×10^{20} cm^{-3} and 3.5×10^{20} cm^{-3} concentrations of chromium and neodymium, respectively, were 3×50 mm^2. The absolute efficiency for free-running operation was 5.5%; it was 5.2% and 4% for pump energies of 3.2 J and 1 J, respectively, when the
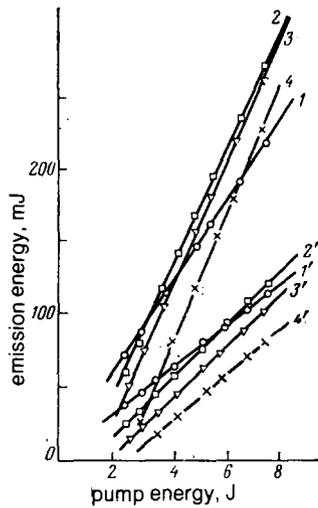
FIG. 5. Dependence of pulse energy for free-running lasers using the crystals GSGG–Cr–Nd and Nd–YAG with identical dimensions 5×50 mm^2 on pump energy for a pulse repetition rate of 20 sec^{-1} and various reflection coefficients R for the output mirror. Curves 1–4 are for GSGG crystals, 1'–4' for YAG crystals; $R = 0.87$ for 1, 1', $R = 0.66$ for 2, 2', $R = 0.43$ for 3, 3' and $R = 0.31$ for 4, 4'.

differential efficiency equaled 6.2%. When this laser was Q-switched with the help of a lithium niobate electro-optic modulator, the 3×50 mm^2 element with the concentrations of Cr and Nd described above exhibited an efficiency of 1.5% for a pump energy of 2 J. In this case, a silvered pipe was used as a light source.

An important enhancement in performance compared to lasers made from Nd-YAG crystals was also obtained for moderate pumping in the periodic-pulse regime. Figure 5 shows the dependence of the free-running output pulse energy on the energy per pump pulse for a repetition rate of 20 sec^{-1} and for various reflection coefficients of the output mirror. Stimulated emission was produced by a pulsed xenon flashlamp with a discharge gap of 3×45 mm^2 in a silvered quartz monoblock with an external diameter of 20 mm. In the figure, data are compared for GSGG-Cr-Nd and Nd-YAG elements with the same dimensions 5×50 mm^2. In the GSGG-Cr-Nd laser, the mean output power amounted to 5 watts for a pump power of 145 watts; for Q-switched operation using a lithium niobate electrooptic shutter cut at the Brewster angle, and for the excitation conditions described above, the mean emission power for the same pump power (145 watts) out of that element amounted to 2.9 watts, corresponding to a differential efficiency of 3%.[15]

Finally, much interest now centers on using GSGG-Cr-Nd to create more powerful solid-state lasers, using pulse-periodic pumping. In particular, for a GSGG-Cr-Nd active element with dimensions 5×88 mm^2, one obtains an average power in free-running operation for a pulse repetition rate of 50 sec^{-1} of about 40 watts for a pumping pulse energy of 18J (i.e., the absolute emission efficiency was 4.4%). For this active element, energies of 17 J in single-pulse operation were attained for pump energies of 400 J. Because of this sensitization phenomenon, we can use GSGG-Cr-Nd crystals to make efficient lasers operating on the transition of a
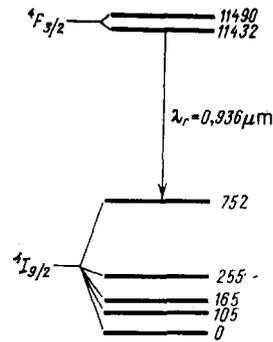
FIG. 6. Arrangement of low-lying energy states of the Nd^{3+} ion, including the laser transition $^4F_{3/2} \rightarrow ^4I_{9/2}$.[21]

Nd^{3+} ion to its $^4I_{9/2}$ ground state ($\lambda = 0.936$ micron; see Fig. 6). The concentrations of Cr^{3+} and Nd^{3+} in this laser are chosen in such a way that the resonant losses due to the absorption transition $^4I_{9/2}$ to $^4F_{3/2}$ of Nd^{3+} do not exceed tolerable values while the overall optical density of the active element remains high. Thus, for a Nd concentration of 5×10^{19} cm^{-3}, the magnitude of the coefficient of resonance absorption amounts to 0.02 cm^{-1}, which is comparable to typical values of losses in most active laser elements. For a chromium concentration of 2×10^{20} cm^{-3}, even for such a small content of neodymium, the optical density and efficient "booster pumping" of the upper working level $^4F_{3/2}$ by the chromium ions is preserved (Ref. 49). In the laser experiments performed in Ref. 21 with GSGG-Cr-Nd elements of dimensions 3×50 mm^2, and with chromium and neodymium contents of 2×10^{20} and 5×10^{19} cm^{-3} respectively, emission was obtained at $\lambda = 0.936$ micron for a threshold pump energy of 10 J with a differential efficiency of 0.4%.

In listing the unquestioned achievements of lasers using GSGG-Cr-Nd active elements, one should also note their resistance to damage by ultraviolet and gamma radiation. Figure 7 shows the dependence of the relative decrease in emitted energy for a GSGG-Cr-Nd and Nd-YAG elements, both of which were first irradiated by γ-rays from a ^{60}Co source at a rate of 600 Rad/sec, vs the received dose of radiation.[50,51] It is clear that the GSGG-Cr-Nd elements pre-
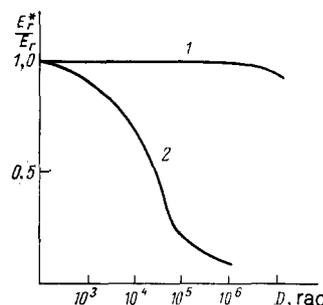
FIG. 7. Dependence of the relative emission energy decrement on γ ray irradiation dose for a dose rate of 600 Rad/sec from ^{60}Co for active elements made of GSGG–Cr–Nd (1) and Nd–YAG (2).[50]

serve their original output parameters right up to doses of 10^7 Rads. As was established in Ref. 52, the radiation hardness of GSGG-Cr-Nd for a caustic (?) of > 100 microns was 8×10^8 watts/cm^2 (i.e., the same as for Nd-YAG), while for smaller caustics it was 2×10^{11} watts/cm^2.

Recently it has been suggested that one might use the sensitization phenomenon to create lasers based not only on Nd^{3+} ions, but also on other rare-earth elements. In view of the broad-band luminescence of the Cr^{3+} ion from its excited state $^4T_2$, one should also expect efficient energy transfer to the ions Pr^{3+}, Sm^{3+}, Dy^{3+}, Ho^{3+}, Eu^{3+}, Tm^{3+}, and Yb^{3+}.[53]

3. LASERS BASED ON CONCENTRATED NEODYMIUM PHOSPHATE GLASS (CNPG)

In the previous section, we have already mentioned the fact that one of the principal means of increasing the efficiency of solid state lasers is to increase the concentration of active ions.

Figure 8 shows the dependence of the luminescent quantum efficiency of Nd^{3+} from the $^4F_{3/2}$ level in various laser crystals and glasses.[38] The sharp decrease in quantum efficiency for concentrations of Nd^{3+} above 1–3% is characteristic of most laser hosts. This "quenching"is caused by interactions among the transitions along the lines of those shown in Fig. 9, which becomes strong when the distance between neighboring Nd^{3+} ions falls below 3 Å.

However, in 1967 (as reported in Ref. 54) the first apparent exception to this rule was found: in the crystal BaF_2–NdF_2, the luminescent quantum efficiency stayed high even at a Nd concentration of 4×10^{21} cm^3, i.e., almost 10 times larger than in the usual laser crystals. It was shown in this report that the cause of this weak quenching was that the $^4I_{15/2}$ and $^4I_{13/2}$ levels, through which the quenching occurred, were shifted compared to other hosts so that the radiative transition frequencies were smaller than the absorption frequencies: The transitions were nonresonant. During the 1970s, there were reports of another crystal with anomalously weak quenching: NdP_5O_{14}.[55] The Nd^{3+} concentration in this crystal was 4×10^{21} cm^{-3}; the authors explained the weak quenching by claiming that the crystal structure of
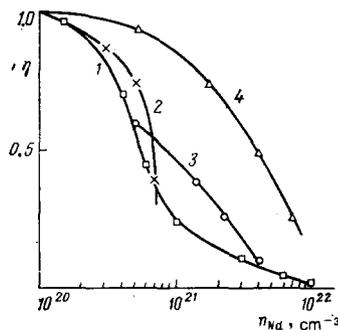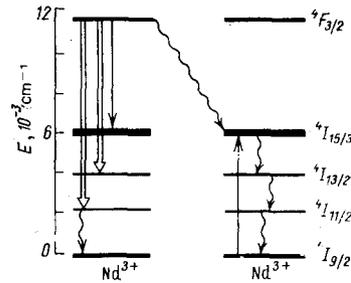
FIG. 9. Schematic arrangement of low-lying energy levels of the Nd^{3+} ion.

this compound was such that neighboring Nd^{3+} ions were separated by a distance of 5.2 Å (while in other hosts this distance was ~3Å). This disparity in ion separation turns out to be decisive, since the quenching probability due to dipole–dipole interactions is proportional to R^{-6}. In Refs. 56, 57, it was shown that in NdP_5O_{14}, in addition to the large spacing between the Nd^{3+} ions, the anomalous positions of the $^4I_{15/2}$ and $^4I_{13/2}$ levels also played an important role, just as in the case of BaF_2–NdF_2. From these results, the basic physical requirements for a laser medium with high active-ion concentration were identified: 1) a small value of the elementary quenching interaction parameter, and 2) a large value for the minimum separation between active ions allowed by the host crystal structure.

In various places throughout the world, researchers have recently synthesized about twenty or so crystals with high Nd concentrations. In essence, these crystals are all phosphates. Regrettably, these compounds are unstable and decompose upon heating; it is therefore difficult to obtain crystals whose size approaches the attainable for ruby or YAG laser crystals. For this reason, these crystals are used basically for making mini-lasers. A group of researchers at the General Physics Institute of the USSR Academy of Sciences has taken a second approach to these materials problems—that of fabricating high-concentration Nd glasses.[11,58–61] It is well known that using glasses allows one to make laser elements of large size, arbitrary shape and with high optical uniformity. It was shown by these workers that in order to make high-concentration glasses, one must be guided by the following principles: 1) the basic composition of the glasses must tolerate the high concentrations of Nd. Thus, if an oxide host is required, in place of the usual alkali or alkali-earth oxides one might use the sesqui-oxides such as Sc_2O_3, Y_2O_3, La_2O_3, Gd_2O_3. In this case, the introduction of large amounts of Nd_2O_3 into the glass does not lead to destruction of its structure, because the chemical properties and ionic dimensions of the Nd^{3+} and La^{3+}, as well as the ions they substitute for—Sc, Y, La, Gd—are very close. 2) The composition and structure of the glass must be such that the microscopic interaction-parameter must not exceed a value of $c_{DA} = (1–3) \times 10^{41}$ cm^{-6}/sec. 3) The distance between neighboring ions must be no smaller than 4.5 Å.

The glass Li-La-Nd phosphate (high concentration neodymium glass HCNPG) was synthesized by following these principles. In Fig 8, a curve is shown of the dependence

FIG. 8. Dependence of the luminescent quantum efficiency of neodymium on its concentration in various host materials.[11] Curve 1 is $La_{1-x}Nd_xF_3$; curve 2 is Nd–YAG; curve 3 is GLS-1 glass; curve 4 is HCNPG glass.

TABLE II. Some characteristics of HCNPG glasses.

Laser Properties	
Neodymium concentration	up to $3.0 \cdot 10^{21}$ cm^{-3}
Radiative luminescent lifetime of the metastable level of neodymium ($\lambda_{ex} = 0.53$ micron)	$330\,\mu$sec
Luminescent lifetime of the metastable level of neodymium in the presence of a 3×10^{21} cm^{-3} neodymium concentration.	$80\,\mu$sec
Emission cross-section for the neodymium transition.	$3.8 \cdot 10^{-20}$ cm^2
Optical Properties	
Emission wavelength	$1.0540\,\mu$sec
Index of refraction n	$1.3225\,\mu$sec
Nonlinear index of refraction	1.555
Thermooptical characteristics, 10^{-7} (deg)$^{-1}$	$1.35 \cdot 10^{-13}$ CGS units
w	31
p	26
Q	6
$\dfrac{dn}{dT}$	-13
Other Properties	
Chemical stability	0.2%
Density	2.85 gm/cm^3
Microhardness	340 kg/mm^2
Line broadening coefficient	$80(\pm 10) \cdot 10^{-7}$ deg^{-1}
Thermal resistance ΔT	$80(\pm 10)$ K

of the quantum efficiency of HCNPG glass on Nd^{3+} concentration. It is clear that the Nd^{3+} concentration can be reduced to 2.7×10^{21} cm^{-3} without a dramatic decrease in quantum efficiency. Table II describes the basic characteristics of HCNPG glasses.

The use of HCNPG glass to make active elements in lasers which operate in various regimes, i.e., from quasi-CW to single-pulse, in free-running or in Q-switched mode, allows one to obtain high energy parameters if the pump radiation is used efficiently.

3.1. Free-running operation

This mode of laser operation has been studied both for single and multiple pulses. In the monopulse regime, an absolute efficiency of $6.3 \pm 0.25\%$ and a differential efficiency of $8.4 \pm 0.35\%$ [62] were obtained from a HCNPG rod of length 70 mm and diameter 6.3 mm for pump energies up to 20 J. For a pulse repetition frequency of 5 pulses/sec, and a pump energy of 100 J, the mean output power exceeded 15 watts.[63] For a frequency of 25 pulses/sec and 10 J pump the output power came to 3.4 watts.[64]

3.2. *Q*-switched regime

Using the revolving-prism modulation technique, for a pump energy of 65 J the energy per pulse reached 0.64 J.[65] With a HCNPG electrooptic switch, an efficiency of 1% was reached even at 10 J pump energy, while for a pulse repetition frequency of 20 sec and the same pump energy the average output power came to 0.3 watts.

Recently, a passive element made of lithium fluoride with F color centers was used to Q-switch a HCNPG laser.[66,67] Using this element, emission in a single TEM$_{00}$ mode was achieved with an energy of 30 mJ for a pump energy of only 5.6 J (the dimensions of the HCNPG element were 2×60 mm).

3.3. Pulse-periodic operation

Quasi-CW emission [68] can be realized in a HCNPG laser by using laser pumping. A useful pump is the krypton laser; the active medium is made of HCNPG glass, activated either by Nd^{+3} alone (8×10^{20}cm^{-3}) or by Nd^{3+} (8×10^{20} cm^{-3}) and Cr^{3+} (8×10^{19} cm^{-3}) simultaneously. The pulse length of the pump equaled 0.5 microseconds with a 1:100 off-duty cycle. The pump operates either at the Nd^{3+} absorption line or within the Cr^{3+} band; in the first case, the efficiency reached 18%, in the second 8%.

The latter results show that the efficiencies of lasers made from HCNPG in the small-to-medium range of pump pulse energies, both in free-running and Q-switched regimes, are higher than those of lasers using well-known crystals and glasses. The essential advantage of HCNPG lasers lies in their high output parameters at low pumping levels. This makes it possible not only to make compact lasers but also to increase their reliability and lifetime significantly.

Another important approach that has been successfully pursued in the past year is the creation of "glass-like" lasers with high average radiated power. The so-called "athermal" neodymium phosphate glass is used to make the active elements in these lasers. It should be emphasized that Soviet scientists were pioneers in the creation of athermal laser glasses,[69,70] as well as these laser phosphate glasses.[71]

Investigation of phosphate laser glasses in the USSR had already begun in 1966. It was shown then that neodymium phosphate glasses provided narrower emission lines, lower thresholds and larger emission efficiencies than the well-known silicate glasses used earlier.[71-74] Using phosphate glasses leads to lasers which combine large cross-sec

tions for induced emission of Nd^{3+} with good thermal and optical properties, ensuring minimal dispersion of the laser radiation at large average emitted powers. The development of methods of determining the thermal and optical characteristics of such glasses[75-77] as well as the temperature dependence of these characteristics[73,74,77-79] allowed detailed investigations of their thermal and optical properties over a wide interval of temperature, leading to the manufacture of phosphate glasses whose compositions were suitable for fabricating lasers which combined good emission parameters with minimal angular dispersion of radiation.[74]

An important impediment to the use of the first generation of athermal phosphate glasses in lasers was the rather low thermal stability of active elements made from these glasses. Thus, an element of dimensions 8×100 mm^2 made from GLS-22 or GLS-26 glass can withstand a pump power of only 300–400 watts without damage.[80,81] Researchers *succeeded in increasing the thermal resistance considerably* by tempering the elements.[80-82] For example, tempered elements made from the glass LGS-I-3 (GLS-26) yielded a radiated energy of 0.6 J when subjected to a pump energy of 40 J per pulse at a pulse repetition rate of 10 sec^{-1}. The magnitude of angular dispersion corresponding to 0.8 of the total output energy amounted to $22'$.[80] However, much interest centered on an alternative way to increase the durability of these glasses—the fabrication of new phosphate glasses with increased intrinsic thermal resistance and thermal conductivity.[83,84] For an element of dimensions 8×130 mm^2 made *from such a glass as LGS-T (without tempering)*, an output radiated power in free-running operation higher than 40 watts was obtained for pump powers of 2 kW.[84] In the optimized case, the free-running efficiency from an element of dimensions 8×100 mm^2 made from LGS-T glass exceeded 3% (for a pulse repetition rate of 5–10 sec^{-1}) with angular dispersion of radiation smaller than for the glass LGS-I-3.[84] Active elements made from LGS-T glass reach free-running efficiencies of 0.7% for pulse repetition rates of 50–100 sec^{-1}.[83,84]

4. MID-IR LASERS USING $(Y, Er)_3Al_5O_{12}$ CRYSTALS AND ERBIUM GLASSES

The first high-power laser in the world to operate in the IR region of the spectrum ($\lambda = 2.36$ microns at $T = 77$ K) was made in 1966.[85,86] Using fluorite crystals activated by divalent dysprosium ions, lasers were fabricated whose output powers exceeded 100 watts in the continuous regime, corresponding to an efficiency of 1%. In the Q-switched regime, a gigantic pulse width of 20 nsec was achieved for pulse repetition rates from single pulse to 1000 sec^{-1}.[87] This was possible because of the availability of crystals of CaF_2-Ce-DY, which were thermally and optically stable and of high optical homogeneity, and because of the development of a successful system for cooling the crystals with circulating liquid nitrogen.

In recent years, the efforts of many investigators have been directed toward making efficient lasers in mid-IR band which work at room temperature with flashlamp pumping—extremely important preconditions to applying them both to scientific investigations and to practical goals. However, up to comparatively recent times this problem remained unsolved, despite the fact that much material had accumulated in the literature on investigations concerning various crystals, the theory and practice of making lasers, and methods of exciting crystals. The difficulty in propelling lasers into the long-wavelength regions of the spectrum is connected with large Stokes losses; furthermore, in this region of the spectrum, the working ion at which the emission effect is obtained often has a lower laser level whose lifetime exceeds that of the upper laser level (for certain ions ten times longer). Transitions to this kind of level are well-known in the literature to be self-saturating. Therefore, in the veiw of many authors, the way to increase the efficiency of such lasers is to find some way to depopulate the lower laser level[88]; however, work along these lines has not led to positive results.[89]

The use of crystals with high impurity concentrations, in which collective interactions between impurity atoms are at their strongest, opened up new possibilities for making efficient lasers in the mid-IR band. At these high concentrations, the relaxation of excited states is often determined not by the individual characteristics of the ion-activator, i.e., by its radiation probability or intracenter multiphonon interactions, but rather by processes of resonant nonradiative energy transfer due to ion–ion interactions (cross-relaxation). In addition to concentration quenching of the crystal luminescence, there are a whole series of other nonlinear effects caused by interactions between excited ions.[90] A large number of resonant transitions between rare-earth ions are possible in these high-concentration crystals; this allows one to envision various lasing schemes, many of which have larger energy efficiencies than the well-known three-or-four-level schemes despite their significant Stokes shifts. If one chooses to exploit these schemes, the most useful ion turns out to be Er^{3+}. Crystals and glasses containing erbium show comparatively intense absorption in the radiation bands of standard flashlamp pumps and radiative transitions in the near- to
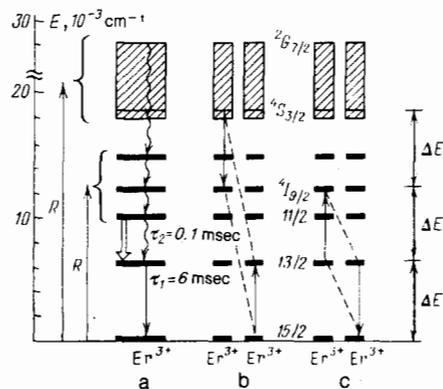
FIG. 10. Arrangement of energy levels of the Er^{3+} ion, and of relaxation processes for excited states of the Er^{3+} ion in $(Y, Er)_3Al_5O_{12}$.[96,97] a) Radiationless relaxation due to intracenter multiphonon transitions: $\rightarrow$ ——luminescent transitions; $— — \rightarrow$ —stimulated transitions ($\lambda = 2.94$ microns. b) Cross-relaxation mechanism for deactivating the state $^4S_{3/2}$ of Er^{3+}. c) Cross-relaxation transitions from the $^4I_{13/2}$ level caused by interaction between excited Er^{3+} ions.

mid-IR regions (Fig. 10). In Ref. 91, the first laser made from the mixed garnet crystal yttrium-erbium aluminate is described; this laser operated at room temperature at $\lambda = 2.94$ microns. In free-running operation this laser delivered an efficiency of $> 1\%$.[92] The high efficiency of the erbium laser was difficult to explain, because the lifetime of the upper laser level $^4I_{11/2}$ of Er^{3+} was one and a half orders of magnitude smaller than the lower laser level $^4I_{13/2}$, i.e., the transition should be self-saturating. An understanding of the mechanism by which such lasers operated was achieved only after it was first shown, in Refs. 93 and 94, that in high-concentration crystals ($n_{Er} > 10^{21}$ cm^{-3}) of Er^{3+}-YAG, the population inversion was achieved via cross-relaxation, thereby leading to important enhancement of the conversion efficiency of the flashlamp pump radiation into laser emission in the three-micron band. This enhancement made possible for the first time not only pulse but also CW emission on a "self-saturating" transition.

Once the physical processes that give rise to the population inversion and the kinetics of emission in the crystal were understood, it became possible to address all aspects of the problem of choosing a crystal composition and the conditions for its excitation which would result in an Er^{3+}-YAG laser with efficient cross-relaxation operating at a wavelength of $\lambda = 2.94$ microns, flashlamp-pumped at room temperature. In free-running operation, such a laser generated an average output power of up to 20 watts (for an efficiency of $> 1\%$).[91,92]

The level scheme and cross-relaxation transitions of Er^{3+} ions in YAG are displayed in Fig. 10. Luminescence from all other levels (except for $^4I_{13/2}$) is strongly quenched due to multiphonon transitions. The lifetime of the lower laser level $^4I_{13/2}$ is close to its radiative value, and amounts to 6.4 milliseconds, which significantly exceeds the lifetime of the upper level $^4I_{11/2}$ (100 microseconds). The high probability of multiphonon transitions leads to the efficient transfer of energy absorbed in various activator bands among the $^4S_{3/2}$, $^4I_{11/2}$, $^4I_{13/2}$ levels. For high concentrations of Er^{3+} (> 1 at.%) the deactivation of the state $^4S_{3/2}$ takes place because of the resonant cross-relaxation transitions $^4S_{3/2} \to {}^4I_{9/2}$, $^4I_{15/2} \to {}^4I_{13/2}$.[95–97]

For lower laser level populations of 10^{18}–10^{19} cm^{-3}, relaxation of an excitation from this level occurs due to interactions between excited ions (see Fig. 10c). In such an interaction one ion, excited to the energy state $^4I_{13/2}$, gives up its energy and falls to the ground state while the other ion, also excited to the energy level $^4I_{9/2}$, acquires this energy and goes to the $^4I_{11/2}$ level with subsequent relaxation to the upper laser level $^4I_{11/2}$. The interaction process between excited ions is intrinsically nonlinear, since its probability depends on the population of the $^4I_{13/2}$ level. The macroscopic rate constant for energy transfer due to the cross-relaxation transitions $^4I_{13/2} \to {}^4I_{9/2}$, $^4I_{13/2} \to {}^4I_{15/2}$ is determined from the kinetics of luminescence from the level $^4I_{11/2}$ measured at various excitation intensities.[98] For low intensities the build-up and decay of luminescence from the $^4I_{11/2}$ level occurs in a time which is characteristic of this level in isolation (100 microseconds). An increase of the excitation intensity leads
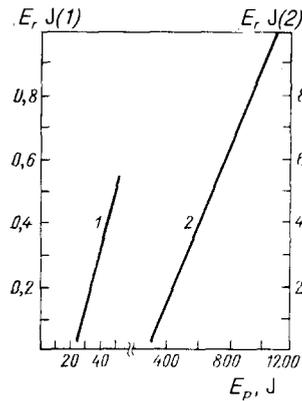
FIG. 11. Dependence of the output energy for a $(Y, Er)_3Al_5O_{12}$ laser on pump energy for various excitation time intervals T.[96,97] Curve 1: $T_{IMP} = 0.2$ msec; Curve 2: $T_{IMP} = 4$ msec.

to nonlinear growth of the luminescence intensity, and to a significant increase in its persistence time, thereby confirming the reality of energy transfer from the long-lived lower laser level to the upper laser level as a result of excited-ion interactions.

An analysis of the kinetic equations, taking into account cross-relaxation transitions,[93,99] shows that the maximum laser efficiency is achieved under the condition that the cross-relaxation transition probability from the long-lived lower laser level exceeds the probability of spontaneous decay of this level. In this case, the laser level is not self-saturating and it is possible to set up a stationary population inversion. The duration of laser emission is determined by the duration of the pump pulse (Fig. 11), which significantly extends the energetic possibilities of the laser. Free-running operation is achieved experimentally for durations from 50 microseconds to 4 milliseconds with output energies per pulse up to 10 J (see Fig. 11).

The theoretical efficiency of the Er^{3+}-YAG laser significantly exceeds the efficiency determined by Stokes losses alone. Energy transfer of excitations from the $^4S_{3/2}$ and $^4I_{13/2}$ levels occurs because of cross-relaxation transitions; this process eliminates loss of pump energy to the transitions $^4S_{3/2} \to {}^4I_{9/2}$ and $^4I_{13/2} \to {}^4I_{15/2}$. In the steady-state emission regime, for each one-quantum absorption in the visible region of the spectrum the laser can radiate three quanta in the three-micron band.

Various techniques have been investigated for controlling the radiation from Er^{3+}-YAG lasers. To obtain radiation at the 2.94 micron wavelength, a passive shutter is made; by using this shutter, giant emission pulses can be generated. Associated liquids are used as materials for the passive shutter; these liquids contain hydroxyl groups, for instance water and ethanol. Energies of 20 mJ have been obtained in monopulses of length 20 nsec for a TEM_{00} mode of diameter 1.7 mm.[100] Because the relaxation time for self-transparency is long (~ 1 microsecond), it is not possible to use water-based passive shutters to produce self-mode locking of erbium lasers.

Q-switching with an electrooptic lithium niobate shut-

ter delivers giant emission pulses with output energies up to 0.1 J and durations of 65 nsec for single-mode operation; in this way it was established that Stark splitting of the laser levels $^4I_{11/2}$ and $^4I_{13/2}$ is essentially related to the energy and time parameters for laser emission.

Injection mode locking[102] was achieved by synchronous electro-optic modulation of the losses of the laser resonator, which contained partial polarizers[103] which ensured a modulation depth 2.6 times larger than modulation depths achievable with ideal polarizers. Under optimal conditions, a laser with a repetition frequency of 1 to $21.5\,\mathrm{sec}^{-1}$ generated a train of 25 ultra-short pulses with peak energies of 0.5 ± 0.015 mJ and an (average) length of around 80 picoseconds.[104] An investigation of the spectral-time characteristics of the radiation 4th harmonic and sum frequencies (from mixing with the neodymium laser radiation in the lithium niobate) showed that the probability of generating ultrashort pulses without substructure amounted to 1/3; the product of the pulse width of 4th harmonic and the width of its spectrum $\Delta\nu = 2$.

Under conditions of injection mode locking, the erbium laser operates in a new spectral region while possessing time and energetic characteristics typical of Nd–YAG lasers.

In recent years, a whole series of practical problems have surfaced whose solutions require lasers which emit in the 1.5-micron band. Among these are problems relating to navigation, meteorology and medicine—in particular, the requirements of ophthalmology are especially noteworthy: 1.5 micron lasers are needed for medical treatment of a whole series of serious illnesses.

Recently, efficient lasers have been fabricated from single-component erbium glasses. The active ions in these glasses are Er^{3+} ions, which radiate on the transition $^4I_{13/2}\rightarrow{}^4I_{15/2}$ ($\lambda = 1.536$ microns). One peculiar feature of this transition is that emission from it can be obtained only for rather low erbium concentrations, less than 1%. For these low concentrations, the weakly-absorbing transitions of the Er^{3+} ion cannot ensure high optical densities in the active elements; as a consequence, because of poor utilization of the flashlamp pump radiation, it is not possible to attain acceptable laser parameters in practice.

Under these circumstances, the only way to increase the efficiency of the 1.5 micron erbium laser is to use the sensitizer effect. A laser (described in Ref. 105) was made from erbium glass as early as 1965, in which the ion Yb^{3+} was used as a sensitizer. However, practical use of such lasers was rendered impossible by their high excitation thresholds for emission and low efficiencies. At the beginning of the 70s, the erbium glass LGS–E was made in the Soviet Union; lasers made from this glass exhibited high output parameters[106] when a powerful neodymium laser was used as a pumping source. By using laser pumping, it becomes possible to fabricate large-scale systems in which the lasers can emit pulses of $\lambda = 1.5$ micron radiation with energies of hundreds of joules, at a conversion efficiency of ~40% in free-running operation[107–109] and 10–15% when used to amplify nanosecond pulses.[110–113] These systems can provide an angular dispersion of output radiation which is close to the

diffraction limit. When elements made of the glass LGS–E are used as broad-aperture power amplifiers, gain coefficients of 0.4 cm^{-1} are attained along with values of stored excitation energy on the order of ~10 J/cm[110–113]; these parameters are significantly higher than those characteristic of disk amplifiers made of neodymium glasses.

Further progress in making 1.5 micron lasers was achieved and reported in Refs. 114–117. The efficiencies of flashlamp-pumped erbium glass lasers can be increased by 1.3–1.5 times by introducing still another sensitizer into the glass, which already contains erbium and ytterbium-chromium. The introduction of chromium can significantly increase the optical density of the glass.

To sum up, the efficiencies of flashlamp-pumped erbium lasers using active elements with dimensions 10×130 mm^2 can reach 3%, i.e., getting close to the efficiency of lasers made of standard neodymium glasses. At this time, the remaining inadequacy of these glasses is their tendency to have rather high emission thresholds of 70–100 J.[115–117]

5. TUNABLE LIF COLOR-CENTER LASERS

In 1965, Fritz and Menke[118] reported that they had obtained laser emission from color centers in the crystal KCl–Li by using a pulsed flashlamp pump at a temperature of 77 K. However, in the course of the next ten years this result was not pursued any further, and only in 1974 did the Mollenauer group in the USA report that they had obtained and investigated tunable color center lasers in KCl–Li and RbCl–Li with laser pumping.[119] Some time later, a group of investigators at the University of Hanover reported on a laser based on another kind of color center in the crystals KCl–Na and RbCl–Na,[120] while a group of Soviet authors at the Technological Institute (USSR Academy of Sciences, Siberian Division) reported emission on three kinds of color centers in crystals of LiF.[121,122] Since then, the number of publications in this area has grown rapidly. Comparatively recently, review articles have appeared which contain discussions of the physical processes by which color centers of various types are generated, and of how they can result in tunable laser emission.[24–26,123–125] From the very beginning, tunable color-center lasers were attractive to investigators because of their unique properties: broad overlapping tunable emission bands from 0.7 to 3.3 microns (in this region of wavelengths, lasers using mixed dyes are either unavailable or of low efficiencies), high frequency stability, small emission line widths, and the ability to work both in pulse-periodic and CW modes.

At the present time, emission has been obtained in a whole series of crystals: lithium fluoride, fluorides and chlorides of sodium and potassium, and rubidium chloride, as well as fluorides of calcium, strontium, magnesium and the complex fluoride $KMgF_3$.[126] In recent years, articles have appeared on emission from color centers in CaO[127] and Al_2O_3.[128] In their original state, all these crystals are colorless. Under the action of γ rays, high-energy electrons, X rays or upon heating in alkali-metal vapors (so-called "additive coloration"), intrinsic point defects appear in these
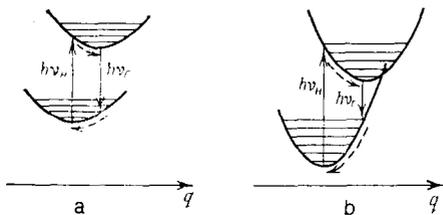
FIG. 12. Potential curves for ground and excited states of color centers in alkali-halide crystals. a) F_2, F_2^+ and F_2^- centers; b) F_A and F_B centers.

crystals, which localize electrons and holes at their lattice sites. The absorption bands of these charged defects give rise to the characteristic colors of these crystals. In crystals of LiF and alkali halides, the defect with the simplest structure is the F center, which is an anion vacancy which has captured one electron. The absorption bands of F centers in various crystals are located in the visible or ultraviolet parts of the spectrum; the centers on which emission is obtained are derived structurally from two or more F centers. Thus, the F_2 center consists of two adjacent F centers, on neighboring lattice sites: the F_2^+ paired center which has lost an electron, and the F_2^- paired center which has trapped three electrons. Sometimes there are cation impurities introduced into the crystal lattice; then the color centers (simple or paired), which lie on a line with the foreign cation (for example, Li^+ in place of K^+ in the KCl lattice), are labeled with the subscript A (for example, F_A). If two cation impurities are located on a line with an F center, then the center is denoted by F_B. Emission on color centers takes place according to a scheme analogous to that of dye lasers (Fig. 12): pump radiation is absorbed in a broad vibronic band, which subsequently relaxes via nonradiative processes to a minimum of the ("nuclear" or "lattice") potential curve for an excited state in a time 10^{-12}–10^{-13} sec. Thereupon, a radiative transition occurs with a time 10–100 nanoseconds to the lower state, which quickly relaxes to the potential minimum of the ground state.

Comparing the level schemes for F_A and F_B centers on the one hand and F_2, F_2^+ and F_2^- centers on the other, one finds important differences in the Stokes shift. Thus, for F_A and F_B centers the absorption transition lies in the 0.5–0.6-micron range, while the luminescence is in the 2–3 micron region; this corresponds to a Stokes shift of ~ 15000 cm, which in some cases exceeds the luminescence energy of the

transition. This leads to the following circumstance: the intersection point of the potential curves for the ground and excited states is found close to the ground-state energy minimum, resulting in large nonradiative losses, so that lasers with F_A and F_B centers operate only at temperatures substantially below room temperature (< 100 K). A way of making tunable lasers based on F_A and F_B centers which operate at temperatures near 100 K was pursued by scientists in the USA and the Federal Republic of Germany;[118-120,123,125] Soviet physicists have worked primarily on making lasers which operate at room temperature. As distinct from F_A and F_B centers, the F_2, F_2^+ and F_2^- centers have small Stokes shifts compared to their luminescent energies; this ensures that their quantum efficiencies will be high and only weakly temperature-dependent even at temperatures above room temperature. At the same time, the Stokes shift is sufficient to ensure a four-level emission scheme. The spectral positions of the broad vibronic absorption bands and the color center luminescence depend strongly on the color center type and fundamental parameters of the crystal lattice in which it is made. According to the Mollwo-Ivy relation, for a given type of center the wavelength position of maximum absorption in the bands varies according to a power-law dependence on the cubic crystal-lattice parameter d: $\lambda_{max} \sim d^x$ ($x = 1.5$–2.5). The luminescence bands for F_A (II), F_B (II) and F_2^+ color centers are shown in Fig. 13. It is clear that by choosing this or that crystal containing the same center, one can significantly change the emission band wavelengths over an overlapping near-continuous range from 2.2 to 3.3 microns for F_A (II) and F_B (II) centers, and from 0.82 to 2 microns for F_2^+ color centers.

Not long ago, a new group of crystals was introduced into the laser ranks, containing F_A type color centers which included in their compositions Tl^+ [129,130] or Ag^+.[130] The F_A (Tl^+) center [as opposed to F_A (II) and F_B (II) centers] are characterized by moderate values of the Stokes losses; crystals containing them make possible lasers which cover the important spectral region from 1.3 to 1.8 microns. The creation of complex quasimolecular centers of F_2 and F_2^+ type associated with monovalent and divalent metals (they are denoted as $(F_2)_A$ and $(F_2^+)_A$ centers) which are "doped" into the crystal matrix leads to significant shifts— in the thousands of inverse centimeters in the absorption and luminescence bands of these structures, thereby broadening the range of tunable emission still more. Furthermore, it is noteworthy that the direction of this shift in the case of mon-
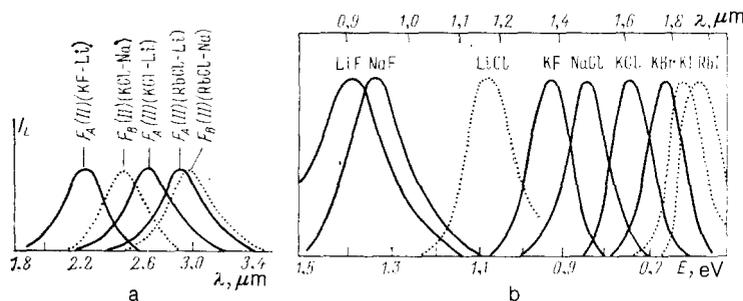
FIG. 13. Luminescence bands for F_A (II) and F_B (II) centers (a) and F_2^+ centers (b) as functions of the cubic crystal lattice parameter for alkali halides.

ovalent impurities (Li^+ in NaF, Na^+ in KCl, Na^+ in CaF_2 and SrF_2) is to longer wavelengths, while for divalent impurities (Mg^{2+}, Ni^{2+}, Co^{2+}) in LiF, the shift is to shorter wavelengths[24-26,124,125,130-133]; this allows one to control the optical properties of laser crystals with color centers.

At present, the most advanced color center lasers are those which make use of the crystal LiF. The F_2, F_2^+, and F_2^- centers in LiF possess the strongest oscillator strengths for vibronic transitions, along with broad luminescence bands and room-temperature four-level transition schemes. Besides, the LiF crystal is rather stable mechanically, only weakly soluble in water, and easy to grow.

The acceptor character of the F_2^+ center, along with peculiarities in its electronic structure, ensures that crystals containing them have a rather high resistance to damage by the pump radiation. A fundamental drawback of the F_2^+ center, however, is its low thermal stability: the half life of its decay at room temperature is only 12 hours. This low stability has up until now been a fundamental obstacle, preventing the manufacture of high-efficiency lasers using these centers, although emission was obtained from them even in 1977.[122] In Refs. 25 and 134, experiments were undertaken to stabilize the F_2^+ centers by introducing OH^- into LiF; a certain fraction of the F_2^+ centers did indeed acquire some thermal stability, but despite this, the basic emission characteristics of the crystal degraded as did the radiative stability. In Refs. 135, 136, a new method of building up ionized centers was proposed, which allowed the stability problem to be solved for the laser parameters in $LiF-F_2^+$. In essence, the idea was to combine the thermal stability of the neutral F_2 center with the photo-stability of the F_2^+. To this end a technology was developed which allowed one to obtain crystals of LiF which contained only F_2 centers. The spectrum of one such crystal is shown in Fig. 14. Under the action of a pump consisting of second harmonic radiation from a neodymium laser (0.53 micron) or the direct output of a rhoda-

FIG. 15. Kinetics of the buildup of F_2^+ color centers in the active region of a LiF crystal due to the action of $\lambda = 0.53$ micron radiation, for various power densities. The kinetics of the process $F_2 \rightarrow F_2^+$ tracks the change in absorption of the radiation from the He–Ne laser ($\lambda = 0.632$ micron).

mine 6G dye laser (0.55 to 0.58 micron), the F_2 centers were excited in the pulse-periodic emission mode. However, the strong photoinstability of the F_2 centers in the path of the pump beam leads to their ionization into F_2^+ centers. This leads to some decrease in the intensity of the F_2 absorption band and to the appearance of an F_2^+ band (see Fig. 14). Provided that the pump radiation can ionize an F_2 center and excite an F_2^+ center equally well, after ten or so pulses there appears in the pump beam a smooth replacement of F_2 emission by F_2^+ emission. The kinetics of F_2^+ buildup is shown in Fig. 15. It is clear that a short time after the pump is switched on the absorption in that region of the spectrum corresponding to the maximum absorption band of the ionized centers increases sharply and is thereby stabilized. Corresponding to this stabilization, F_2^+ centers are generated. Measurement of the kinetics of absorption in the crystal at $\lambda = 0.63$ micron for an initial rate of change of the F_2 color

FIG. 14. Absorption spectrum (continuous curves) and luminescence spectrum (dotted curves) for (a) a γ-irradiated LiF crystal with F_2 centers, and (b) the same crystal in the beam of a second-harmonic pump from a neodymium laser ($\lambda = 0.53$ micron).
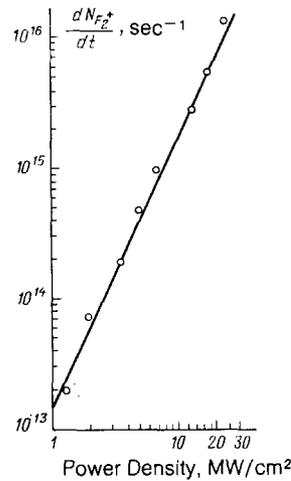
FIG. 16. Dependence of the rate of change of the F_2^+ color center population in the active region of a LiF crystal, averaged over a large number of pulses, on the power density of $\lambda = 0.53$ micron pump radiation.[137,138] The experimental values (circles) agree closely with a quadratic dependence for the rate of change of the color center population (continuous line).

center concentration shows a quadratic dependence of the rate of transformation of F_2^+ color centers on the pump power density (Fig. 16), which documents the two-step character of the phototransformation process of F_2^+ color centers into F_2 centers.[137,138] By applying the method described here, one can for the first time make use of a new active element in LiF lasers ($F_2 \rightarrow F_2^+$), with a working efficiency at room temperature. In a non-selective resonator, the energy efficiency of these elements is found to be 24–30% with a quantum efficiency of $>40\%$ (relative to the pump). An active LiF element $F_2 \rightarrow F_2^+$) can operate with a pulse repetition rate of $100\ \text{sec}^{-1}$ or larger without forced cooling, generating radiation whose angular dispersion is less than 10^{-3} rads, i.e., close to the diffraction limit.[135,136]

Below, various schemes will be discussed for obtaining tunable emission based on these new LiF ($F_2 \rightarrow F_2^+$) active elements. These schemes make use of dispersive resonators made with prisms and holographic selectors, Fabry-Perot etalons and grazing-incidence diffraction gratings with autocollimation. They allow us to make a whole gamut of nano- and picosecond lasers which are tunable over the 0.81–1.1 micron range with spectral widths of 0.07 to 3 cm^{-1}. These lasers are stable at room temperature or higher (up to 370 K both in pulsed and in quasicontinuous operation with peak power of 2 megawatts.[139–142]

The efficiency of the process of preionization of the $F_2 \rightarrow F_2^+$ described above, along with the high quality of the new active elements, confirms the aforementioned results as regards obtaining emission from these elements at low excitation thresholds for flashlamp pumping at room tempera-

ture.[143] For an active element $4 \times 8 \times 40\ \text{mm}^3$ and an energy of 100 J supplied to the pump flashlamp over a discharge time of 25 microseconds, wideband emission is obtained from the F_2^+ centers with $\lambda_{max} = 0.90$ micron and an output energy 14 mJ. The threshold pump energy amounted to 30–35 J.

The elucidation and investigation of the spectral-selective processes of photoionization and recombination that take place in the collective aggregate of color centers in LiF crystals, along with the nature of color-center coupling to the impurity composition of the crystal and to the pump radiation spectrum,[137,138] have led to an increase by two orders of magnitude in the pulse capabilities of active elements of LiF with F_2 centers, pushing it up to 10^5 pulses. Lasers with LiF–F_2 active elements overlap the 0.65–0.74 micron wavelength band with an efficiency of up to 10% at the maximum of their tuning curves.

Another type of color center encountered in LiF, which is of considerable interest to those who seek to make tunable lasers operating at room temperature, is the F_2^- center.[122,144] The half-life of these centers at room temperature is estimated to be many years; furthermore, they are stable in the presence of the infrared pump radiation. Here the fundamental problem lies in the difficulty in obtaining LiF crystals with a sufficiently high concentration of these color centers and at the same time with small passive losses in the emission region. Recently this problem has been successfully resolved; as a result, active elements have been made from LiF–F_2^- with high concentrations of F_2^- and low optical losses.[145] Lasers made from these elements have record-breaking

TABLE III. Color center lasers in LiF crystals, operating at room temperature.

Center type	Pump source	λ_p, microns	Working regime	Pulse duration	Emission power per pulse, MW	Emission energy per pulse, J	Tuning range, microns	Efficiency, %	Reference
F_2	Nd–YAG laser	0.53	Pulse-periodic, $f < 100$ Hz	10 nsec	0.02	$2 \cdot 10^{-4}$	0.65 0.74	10	138
F_2^+ (OH$^-$)	Nd–YAG laser,	0.53 0.69	the same	10 nsec	—	—	0.8 1.15	10–30	92
$F_2 \rightarrow F_2^+$	rhodamine, ruby the same	0.53 0.69	the same	10 nsec	2	0.02	0.81 1.1	30	26,135,136, 138,139
$F_2 \rightarrow F_2^+$	the same	0.53 0.69	Pulse-periodic, $f < 10$ Hz	10–30 psec	—	—	0.84 1.1		140
$F_2 \rightarrow F_2^+$	the same	0.53 0.69	Quasi-CW, $f < 30$ kHz	50 psec	0.03	$1.5 \cdot 10^{-6}$	0.85 1.09	7	141
$F_2 \rightarrow F_2^+$	IFP-1200 lamp	-0.6 0.7	Pulsed	25 μsec	—	0.014	0.91	0.014	143
F_2^-	Nd-YAG laser	1.06	Pulse-periodic, $f = 100$ Hz	10 nsec	12.5	0.125	1.08 1.25	43 (50)	80,81,77
F_2^-	the same	0.53 0.69	the same	100 μsec	—	$0.5 \cdot 10^{-3}$	1.14	0.5	80
F_2^-	the same	1.06	Pulsed	<0.7 psec	—	—	1.08 1.25	—	82
F_2^-	LGS-247-2 glass laser	1.055	Monopulse	100 nsec	10^3	100	1.12 1.16	15	85
F_2^-	4 IFP-1200 lamps	0.8–1	Pulsed	50 μsec		0.05	1.185	0.01	83,84

TABLE IV. Nonlinear frequency conversion for LiF color center lasers.

	Nonlinear Crystal	Emission Band, microns
ω (LiF-F$_2^-$)	—	1,08—1,25
ω (LiF-F$_2^+$)	—	0,84—1,1
ω (LiF-F$_2$)	—	0,65—0,74
2ω (LiF-F$_2^-$)	LiIO$_3$, LiNbO$_3$	0,54—0,62
ω (LiF-F$_2^-$)+ω (1,06)	LiIO$_3$, LiNbO$_3$	0,535—0,57
2ω (LiF-F$_2^+$)	LiIO$_3$	0,42—0,55
ω (LiF-F$_2^+$)+ω (1,06)	LiIO$_3$	0,47—0,54
ω (LiF-F$_2^+$)−ω (1,06)	GaSe	5,5—16,5

emission parameters; they can operate with no loss of stability at room temperature with pulse repetition rates in excess of 100 sec^{-1}. When a pump consisting of the fundamental emission line from a nanosecond-pulse neodymium garnet laser was used, the efficiency relative to this pump was found to be 40%, with a differential efficiency of $\sim$50%. The emission band had a width of 1000 cm^{-1} with a maximum at 1.17 microns. A tunable laser made from LiF-F$_2^-$ had an emission linewidth of < 5 Å and a tuning range of 1.08-1.25 microns. The efficiency relative to the pump at the maximum of the tuning curve ($\lambda_{max} = 1.17$ microns) equaled 25%.[125,145,146] Emission on the F$_2^-$ centers was obtained with an efficiency of $\sim$0.5% as well when the element was pumped by quasi-CW laser pulses; the laser operated in free-running mode for a duration of 10^{-4} sec.[145]

The use of a neodymium glass laser as a pumping source operating in the mode-locked regime has led for the first time to the construction of a tunable LiF-F$_2^-$ laser whose output consists of subpicosecond pulses of widths < 0.7 psec.

Laser emission was obtained for the first time from LiF-F$_2^-$ active elements of dimensions 8.5 × 90 mm^2 under flashlamp pumping at room temperature. The output energy emitted amounted to 50 mJ for a stored energy of 800 J in a capacitor and a discharge time to the flashlamp of 50 microseconds.[148,149]

Recently, much success has been achieved in large-scale fabrication of active elements made from lithium fluoride with F$_2^-$ centers. Lasers are made from elements with dimensions of 220 × 80 × 40 mm^3 that work at room temperature with peak powers of 10^3 megawatts (which are record values for tunable lasers) for emission energies of 100 J and pulse widths of 100 nanoseconds.[150] A monopulse laser made of neodymium glass usually serves as a pump, putting out an energy of 700 J. The LiF color center laser parameters are diplayed in Table III.

In recent years, LiF-F$_2^-$ crystals have found yet another application in the field of solid state lasers. As reported in Ref. 151, nonlinear absorption due to F$_2^-$ centers was detected for the first time at $\lambda = 1.06$ microns; this result allowed a whole gamut of neodymium lasers to be made using passive Q switches made with LiF-F$_2^-$ crystals.[152] As an example, we mention small-scale pulse lasers made of HCNPG (2 × 50mm^2) with passive laser Q switches which initially have 64% transmission at $\lambda = 1.06$ microns; these

lasers have an output energy from 8 to 30 mJ for 3 to 5.6 J pump energies.[66,67] A second example is a pulse-periodic laser made from YAG with neodymium (6.3 × 80 mm^2) with a LiF-F$_2^-$ Brewster passive Q switch whose initial transmission was 17%; this laser had an output energy of 0.1 to 0.2 J for a pump energy of 16 to 25 J and average radiated power greater than 2 watts.[153] Finally, not long ago a powerful compact radiator was made out of the neodymium glass LGS-247-2 (a slab 40 × 240 × 720 mm^3) with a passive Q switch made from LiF-F$_2^-$ of dimensions 40 × 180 × 40 mm^3 with 70% transmission, having output energies of 150 J in a pulse of 150 nanosecond duration when pumped with an energy of 7.5 × 10^4 J.[147] We remark that this was the first time that anyone had demonstrated the feasibility of constructing such simple and at the same time reliable and efficient systems.

Tunable LiF lasers using F$_2$, F$_2^+$ and F$_2^-$ color centers allow us to cover the important 0.65-to 1.25-micron band of the optical spectrum (which is badly served by dye lasers), using only a single pumping source: the neodymium laser. An important extension of the range of frequencies which LiF color center lasers can cover can be achieved with the help of harmonic generation and frequency mixing in nonlinear crystals (Table IV).

Clearly, then, LiF color center lasers are basic to the practice of wide-band spectrometry in the visible and infrared bands.[26,142,146]

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