

⁵V. R. Regel', V. G. Govorkov, and A. F. Zakatov, *Zavodsk. laboratoriya*, No. 2, 243 (1960) [*Industrial Laboratory*].
⁶P. P. Perstnev and G. V. Berezhkova, *Izv. AN SSSR, Ser. Fiz.* **47**, 1133 (1983) [*Bull. USSR Acad. Sci., Phys. Ser.*].
⁷G. V. Berezhkova, N. P. Skvortsova, P. P. Perstnev, and V. R. Regel', *Fiz. Tverd. Tela (Leningrad)* **26**, 1074 (1984) [*Sov. Phys. Solid State* **26**, 654 (1984)].
⁸N. L. Sizova, V. D. Natzik, Y. Y. Gorina, and V. R. Regel, *Phys. Stat. Sol.* **a76**, 447 (1983).
⁹M. Sh. Akhchurin, V. G. Galstyan, V. R. Regel', and V. N. Rozhanskii,

Poverkhnost'. Fizika, khimiya mekhanika (Surfaces. Physics, Chemistry, and Mechanics), No. 3, 119 (1983).
¹⁰E. A. Stepanov and V. R. Regel', in: *Segnetoelektricheskie kristally pri prilozhenii razlichnykh polei (Ferroelectric Crystals with Application of Various Fields)*, Nauka, Leningrad, 1981, p. 17.
¹¹L. L. Regel, V. R. Regel, S. E. Boriskin, G. G. Knab, A. A. Urusovskaya, L. I. Alekseeva, and V. V. Klechkovskaya, *Phys. Stat. Sol.* **a73**, 255 (1982).
¹²V. R. Regel', V. A. Al'pert, D. Senesh, and Yu. A. Fadin, *Zh. Tekh. Fiz.* **51**, 134 (1981) [*Sov. Tech. Phys.* **26**, 76 (1981)].

A. A. Kaminskiĭ. *Physics and spectroscopy of laser crystals.* In little over a year we will observe the 25th anniversary of the creation of the first laser. As is well known, this was a quantum oscillator employing a ruby crystal ($\text{Al}_2\text{O}_3\text{-Cr}^{3+}$). Today laser crystals represent a complex scientific and practical problem which combines a number of aspects of solid-state physics, spectroscopy, quantum electronics, physical chemistry, and technology. The fundamental aspect of this problem involves study of the properties of activated crystals and physical phenomena occurring in them when they are excited, which result in the generation of stimulated radiation (SR). In solution of the applied problems in this area an activity of the first importance is the exploratory research which consists mainly of the development of new operational schemes and principles of excitation of SR of crystalline lasers, the search for new laser compounds and generation centers, discovery of new SR channels and means of increasing the efficiency of both new and already known laser compounds, and so forth,^{1,2}

The present report is devoted to some results in this area obtained in the Laboratory of Laser Crystal Physics at the Institute of Crystallography.

2. About 180 crystalline dielectric matrices are known, in which laser properties are provided by divalent and trivalent rare earth ions ($\text{Ln}^{2+}\text{-Sm}^{2+}$, Dy^{2+} , Tm^{2+} and $\text{Ln}^{3+}\text{-Pr}^{3+}$, Nd^{3+} , Sm^{3+} , Eu^{3+} , Tb^{3+} , Dy^{3+} , Ho^{3+} , Er^{3+} , Tm^{3+} , Yb^{3+}) and ions of the iron group (V^{2+} , Co^{2+} , Ni^{2+} and Cr^{3+} , Ti^{3+}) and U^{3+} ; these crystals are mainly fluorides and oxygen-containing compounds. Their principal generating activators are Ln^{3+} ions, in which SR is excited in 47 ($f\text{-}f$)

and 3 ($d\text{-}f$) intermultiplet transitions in the range from $\sim 0.17\ \mu\text{m}$ to $\sim 5.15\ \mu\text{m}$. Distinctive among them in the number of laser channels are the following ions: Ho^{3+} (12 channels), Er^{3+} (11 channels), and Pr^{3+} (10 channels), and distinguished in utilization is Nd^{3+} , after which we have the ions Er^{3+} , Ho^{3+} , Tm^{3+} , and Pr^{3+} . For Ln^{3+} activators several working laser schemes have been developed, of which the most used are four-level and sensitized schemes, and in recent years cascade schemes have also begun to be used actively.

3. While in the 60s the number of lasing compounds discovered corresponded approximately to the number of lasers made on the basis of these compounds, in the following years, as can be seen from Fig. 1, the balance has shifted in favor of the latter. This progress is due to the methods which been developed for study of the spectroscopic properties of laser crystals and the understanding achieved of the nature of the processes which occur in them. Not all quantum generators employing activated crystals find application. The requirements of contemporary laser technology are satisfied only by those which lase with high efficiency at 300 K with lamp pumping and which have an extended period of operation. In solution of the problem of laser crystals, our contribution is shown by the hatched region of histogram 2 in Fig. 1.

4. Of the room-temperature crystalline lases developed by us we shall mention here only three—a laser employing the tetragonal fluoride $\text{LiYF}_4\text{-Pr}^{3+}$ which lases at a wavelength $\lambda_{\text{SR}} = 0.6395\ \mu\text{m}$ (the channel ${}^3P_0 \rightarrow {}^3F_2$) with a low excitation threshold ($E_{th} \approx 10\ \text{J}$) (Ref. 3); a laser employing

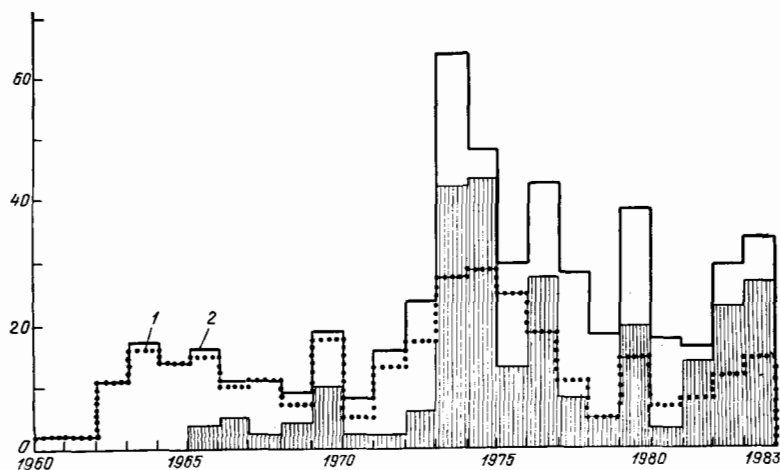


FIG. 1. Development of research on laser crystals from 1960 to 1983. 1—Number of compounds produced, 2—number of lasers constructed with these compounds.

the monoclinic crystal $\text{BaYb}_2\text{F}_8\text{-Er}^{3+}$ with $\lambda_{\text{SR}} = 1.9965 \mu\text{m}$ (the channel ${}^4F_{9/2} \rightarrow {}^4I_{11/2}$) and $E_{\text{th}} \approx 20 \text{ J}$ (Ref. 4); and a laser employing $\text{BaYb}_2\text{F}_8\text{-Ho}^{3+}$ with $\lambda_{\text{SR}} = 2.9054 \mu\text{m}$ (the channel ${}^5I_6 \rightarrow {}^5I_7$) and $E_{\text{th}} \approx 2.5 \text{ J}$.⁴ All these lasers are excited by the radiation of a xenon lamp and satisfy the requirement mentioned above.

5. Extension of the spectral region of generation of crystalline lasers to the middle of the infrared range, as studies have shown, can be achieved only by use of the cascade principle of excitation of stimulated radiation.^{1,2} Confirming this are the first results on lasing in the 4- μm and 5- μm ranges of Ln^{3+} ions in dielectric crystals.⁵⁻⁷ For example, in our laboratory in a multiray employing $\text{BaF}_2\text{-LaF}_3$ we have detected the stimulated radiation of Nd^{3+} ions by the direct cascade scheme ${}^4F_{3/2} \rightarrow {}^4I_{13/2} (\lambda_{\text{SR}} = 1.328 \mu\text{m}) \rightarrow {}^4I_{11/2} (\lambda_{\text{SR}} = 5.15 \mu\text{m})$.⁶ In Ref. 7 with $\text{YAlO}_3\text{-Er}^{3+}$ lasing was obtained with the scheme ${}^4S_{3/2} \rightarrow {}^4I_{9/2} (\lambda_{\text{SR}} = 1.6632 \mu\text{m}) \rightarrow {}^4I_{11/2} (\lambda_{\text{SR}} \approx 4.75 \mu\text{m})$.

The cascade principle is also promising for experiments on excitation of induced microwave phonon transitions both in the intermultiplet channels and in the inter-Stark radiationless channels of Ln^{3+} activators in ionic crystals. Analysis of the kinetics of lasing of $\text{YAlO}_3\text{-Er}^{3+}$ according to the cascade scheme ${}^4S_{3/2} \rightarrow {}^4I_{9/2} (\lambda_{\text{SR}} = 1.6628 \mu\text{m}) \rightarrow {}^4I_{11/2} \rightarrow {}^4I_{13/2} (\lambda_{\text{SR}} \approx 2.73 \mu\text{m})$ at $\sim 110 \text{ K}$ has made it possible to conclude that generation of phonons in the terahertz range is excited in transition between the states ${}^4I_{9/2}$ and ${}^4I_{11/2}$.⁸ A certain applied interest is presented also by feed-flowing cross-cascade laser schemes. These utilize extra feed of the initial laser state of the generating ions from sensitizer ions and an outflow from the final working level of the residual excitation to de-activator ions. With the garnet $\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Yb}^{3+}, \text{Cr}^{3+}\text{-Er}^{3+}, \text{Tm}^{3+}$ and the schemes $(\text{Yb}^{3+})^2F_{5/2}, (\text{Cr}^{3+})^2E \sim (\text{Er}^{3+})^4I_{11/2} \rightarrow {}^4I_{13/2} (\lambda_{\text{SR}} = 2.697 \mu\text{m}) \sim (\text{Tm}^{3+})^3H_4 \rightarrow {}^3H_6 (\lambda_{\text{SR}} = 2.0205 \mu\text{m})$ results have been obtained which indicate the possibility of producing a 3- μm crystalline continuous laser.⁹

6. The problem of low-threshold neodymium laser crystals is interesting scientifically and also is important practically. Its solution is based on the search for compounds in which Nd^{3+} ions have a large peak effective cross section σ_e for laser transitions. Until recently the list of such active media was headed by the monoclinic potassium rare earth tungstates $\text{KY}(\text{WO}_4)_2, \text{KGd}(\text{WO}_4)_2,$ and $\text{KLu}(\text{WO}_4)_2$, stimulated radiation of which was observed early in the 1970's.¹⁰ Recently at our institute a new class of low-threshold laser crystals was found: $\text{NaGdGeO}_4, \text{NaLuGeO}_4,$ and so forth, in which the Nd^{3+} ions have record high values of σ_e both in transitions of the main channel ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ ($\sigma_e \approx 4 \cdot 10^{-19} \text{ cm}^2$) and in transitions ${}^4F_{3/2} \rightarrow {}^4I_{13/2}$ ($\sigma_e \approx 2 \cdot 10^{-19} \text{ cm}^2$).¹¹ This quality is due not to the eight-fold anion environment usual for Nd^{3+} ions, but to a six-fold environment.

7. Recently the attention of researchers has switched from self-activated neodymium laser crystals to highly concentrated holmium, erbium, and praseodymium laser crystals. In study of the compounds $\text{Er}_3\text{Al}_5\text{O}_{12}, \text{ErAlO}_3, \text{KEr}(\text{WO}_4)_2, \text{BaEr}_2\text{F}_8, \text{LiErF}_4, \text{CaF}_2\text{-ErF}_3,$ and $\text{SrF}_3\text{-ErF}_3$ with 100% content of Er^{3+} ions in our laboratory, we have

observed in them a number of new lasing properties associated with the ${}^4S_{3/2}$ and ${}^4F_{9/2}$ multiplets.^{4,12,13} These states are characterized by low quantum yields of the radiative transitions as a result of the bypassing of the latter by active radiationless transitions and the influence of cross-relaxation quenching channels. Analysis of the occurrence of unlike-wave lasing in these crystals (the channels ${}^4S_{3/2} \rightarrow {}^4I_{9/2-13/2}$ and ${}^4F_{9/2} \rightarrow {}^4I_{11/2}$) and its kinetics have shown that in the movement of energy between multiplets of Er^{3+} ions at low temperatures an appreciable role is played also by the imprisonment of electronic excitation.

8. Continuing the search for laser crystals with the garnet structure, we have recently discovered new possibilities for this class of active media. In the 4-cation garnet $\text{Ca}_3\text{Ga}_2\text{Ge}_3\text{O}_{12}$ with lamp pumping, lasing was excited in the near infrared region ($\lambda_{\text{SR}} \approx 1.28 \mu\text{m}$), which is due to stable defect centers created by choice of appropriate conditions during growth of the crystal.¹⁴ The production of stimulated radiation in defect centers opens a new field in the physics of ionic laser crystals. Previously we discovered the laser properties of more than twenty garnets, including several 4-cation garnets— $\text{Y}_3\text{Sc}_2\text{Al}_3\text{O}_{12}, \text{Y}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}, \text{Gd}_3\text{Sc}_2\text{Ga}_3\text{O}_{12},$ with Nd^{3+} ions, etc.^{2,15}

9. In solution of the problem of increasing the efficiency of solid-state lasers by creation of disordered compounds, we found a new series of oxygen-containing laser compounds with Nd^{3+} ions, among them $(\text{La}_{1-x}\text{Nd}_x)_3\text{Ga}_5\text{SiO}_{14}$ and $(\text{La}_{1-x}\text{Nd}_x)_3\text{Ga}_5\text{GeO}_{14}$ with the structure of trigonal calcium gallium germanate $\text{Ca}_3\text{Ga}_2\text{Ge}_4\text{O}_{14}$.^{16,17} These crystals have turned out to be promising not only for creation of efficient lasers, but also in connection with piezoelectric technology and acoustics.¹⁸

10. Analysis of the actual intensity of luminescence lines of Ln^{3+} ions in crystals requires data on the probability of spontaneous radiative transitions A_{ij} and radiationless transitions W_{ij} . Usually for determination of the former one first estimates the intermultiplet probabilities (on the basis of the Judd-Ofelt approximation),² and then precision mea-

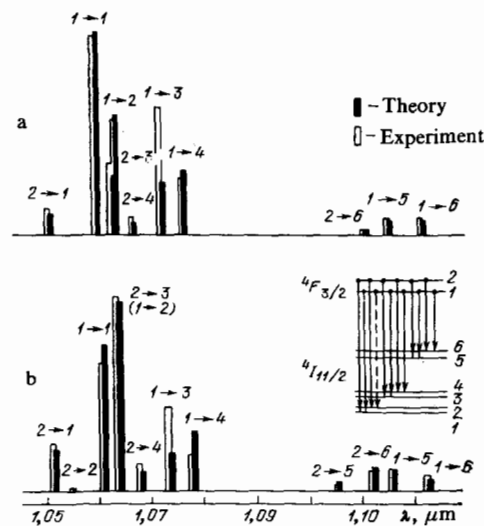


FIG. 2. Intensity of luminescence lines of Nd^{3+} ions (the channel ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$) in $\text{Y}_3\text{Al}_5\text{O}_{12}$. a) At 77 K; b) at 300 K.

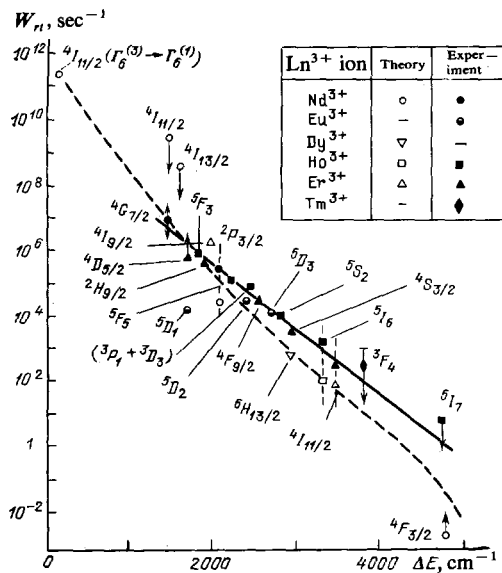


FIG. 3. Probability of radiationless transitions at $T = 0$ of Ln^{3+} ions in a YAlO_3 crystal as a function of the gap energy. The experimental points are from Ref. 23.

measurements are used to find the fraction of radiation associated with a given inter-Stark transition. Multiphonon radiationless transitions are most often studied on the basis of the dependence of their probability on the energy (the energy-gap law).¹⁹ The empirical nature of these methods does not bring out the relation of the measured characteristics to the microstructure of the Ln^{3+} center, knowledge of which is extremely important in the directed search for new laser media. For a number of years the theory of the questions enumerated above has been developed in many laboratories, and the first concrete calculations were made recently with our participation with application to the widely used laser crystals $\text{Y}_3\text{Al}_5\text{O}_{12}$ and YAlO_3 with Ln^{3+} ions.

In a medium with an isotropic refractive index $n(\nu_{ij})$ the probability A_{ij} of an inter-Stark transition is given by the formula

$$A_{ij} = \frac{32\pi^3 n(\nu_{ij})}{3hc^3} \nu_{ij}^3 \sum_{\nu} |\langle \nu | D'_{ij} | j \rangle|^2 \quad (1)$$

where D'_{ij} is the effective-dipole-moment operator. To determine this probability in Ref. 20 we calculated the local field at the Nd^{3+} ion and the dipole polarization of the $\text{Y}_3\text{Al}_5\text{O}_{12}$ lattice in the field of an electromagnetic wave and calculated the parameters of the static and dynamic crystal fields. In Fig. 2 the results of this calculation are compared with experiment. For W_{ij} between the levels of two multiplets the following expression was obtained in Ref. 21:

G. A. Gusev and I. M. Zheleznykh. *On the possibility of detection of neutrinos and muons on the basis of radio radiation of cascades in natural dielectric media (antarctic ice sheet and so forth).*

1) *Detectors of ultralarge size and problems of high-energy physics and astrophysics.* Detectors of extensive air showers (EAS) operating in our country and abroad have areas of several tens of km^2 (at Yakutsk, Havera Park, etc.),

$$W_{ij} = \sum_f A_{nf}^2 \int_{-\infty}^{\infty} e^{i\Omega_{if}t} D_f(t) dt, \quad (2)$$

where Ω_{if} is the Bohr frequency of the transition, A_{nf} is a factor describing the wave functions of the levels, and $D_f(t)$ is a parameter which takes into account the dynamic properties of the matrix-base in terms of which the phonon integral is written:

$$D_f(t) = \frac{\hbar}{6N S m_f} \int_0^{\max} \omega^{-1} \rho(\omega) \{ \bar{n}(\omega) + 1 \} e^{i\Omega t} + \bar{n}(\omega) e^{-i\Omega t} dt, \quad (3)$$

where N is the number of unit cells in the crystals, m_f is the mass of atom f in the cell ($f = 1, \dots, S$), $\bar{n}(\omega)$ is the Planck rms value of the phonon filling number, and $\rho(\omega)$ is the effective phonon density in the definition of Ref. 22. For $\text{YAlO}_3 - \text{Ln}^{3+}$ the results of the calculation are summarized in Fig. 3.

The coauthors of the publications cited in this report are our colleagues of the Institute of Physics Research of the Armenian Academy of Sciences, the Institute of Inorganic Chemistry of the Siberian Division, USSR Academy of Sciences, the Moscow, Kazan' and Kishinev State Universities and several laboratories of our institute.

- ¹A. A. Kaminskiĭ, *Lazernye kristally (Laser Crystals)*, Nauka, Moscow, 1975.
- ²A. A. Kaminskiĭ, *Laser Crystals, Their Physics and Properties*, Berlin, Heidelberg, New York: Springer-Verlag, 1981.
- ³A. A. Kaminskiĭ, *Dokl. Akad. Nauk SSSR* **271**, 1357 (1983) [*Sov. Phys. Doklady* **28**, 668 (1983)].
- ⁴A. A. Kaminskiĭ *et al.*, *Neorganicheskie materialy* **18**, 482 (1982).
- ⁵L. Esterowitz *et al.*, *Appl. Phys. Lett.* **35**, 236 (1979).
- ⁶A. A. Kaminskiĭ, in: *Proc. of Intern. Conf. on Lasers 80*, McLean: STS Press, 1981, p. 328.
- ⁷A. A. Kaminskiĭ, *Izv. AN SSR, Ser. fiz.* **45**, 348 (1981) [*Bull. USSR Acad. Sci., Phys. Ser.*].
- ⁸A. A. Kaminskiĭ, *Dokl. Akad. Nauk SSSR* **267**, 106 (1982) [*Sov. Phys. Doklady* **27**, 1039 (1982)].
- ⁹A. A. Kaminskiĭ *et al.*, *Dokl. Akad. Nauk SSSR* **257**, 79 (1981) [*Sov. Phys. Doklady* **26**, 309 (1981)].
- ¹⁰A. A. Kaminskiĭ *et al.*, *Neorganicheskie materialy* **8**, 2153 (1972).
- ¹¹A. A. Kaminskiĭ *et al.*, *Kristallografiya* **27**, 522 (1982) [*Sov. Phys. Crystallogr.* **27**, 316 (1982)].
- ¹²A. A. Kaminskiĭ *et al.*, *Neorganicheskie materialy* **18**, 1910 (1982).
- ¹³A. A. Kaminskiĭ *et al.*, *Neorganicheskie materialy* **17**, 1121 (1981).
- ¹⁴A. A. Kaminskiĭ *et al.*, *Neorganicheskie materialy* **19**, 2056 (1983).
- ¹⁵A. A. Kaminskiĭ *et al.*, *Phys. Stat. Sol.* **a34**, K109 (1976).
- ¹⁶A. A. Kaminskiĭ *et al.*, *Dokl. Adad. Nauk SSSR* **264**, 93 (1982) [*Sov. Phys. Doklady* **27**, 403 (1982)].
- ¹⁷A. A. Kaminskiĭ *et al.*, *Neorganicheskie materialy* **19**, 1762 (1983).
- ¹⁸A. A. Kaminskiĭ *et al.*, *Phys. Stat. Sol.* **a80**, 607 (1983).
- ¹⁹L. A. Riseberg and M. J. Weber, in: *Progress in Optics*, Amsterdam North-Holland, 1976, p. 91.
- ²⁰A. A. Kaminskiĭ *et al.*, *Izv. AN SSSR, Ser. fiz.* **46**, 979 (1982) [*Bull. USSR, Acad. Sci., Phys. Ser.*].
- ²¹Yu. E. Perlin *et al.*, *Fiz. Tverd. Tela (Leningrad)* **24**, 685 (1982) [*Sov. Phys. Solid State* **24**, 386 (1982)].
- ²²M. G. Blazha *et al.*, *Izv. AN SSSR, Ser. fiz.* **40**, 1851 (1976) [*Bull. USSR Acad. Sci., Phys. Ser.*].
- ²³M. J. Weber, *Phys. Rev.* **B8**, 54 (1973).

and the volumes of underground detectors exceed 10^3 m^3 (the Baksan scintillation telescope, the IMB detector, and so forth). However, the study of elementary-particle physics of ultrahigh energies, the search for new phenomena and particles predicted by contemporary theories (superheavy magnetic monopole, supersymmetric particles, and so forth), and the problems of astrophysics require the construction of surface, underground, and deep underwater detectors of much