

Comprehensive investigations of the spectral and lasing characteristics of the LuAlO_3 crystal doped with Nd^{3+}

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Combined experimental and theoretical investigations of the spectral and lasing properties of the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystal were carried out. The Stark-component energies of the lowest ${}^4F_{3/2}$ and ${}^4I_{9/2-15/2}$ multiplets were determined with high precision. Induced transitions between the ${}^4F_{3/2}$ and ${}^4I_{11/2,13/2}$ states were recorded and identified. The spectral composition of the stimulated emission, and the changes of the luminescence lifetime of the metastable ${}^4F_{3/2}$ state and of the luminescence line width with changing temperature were investigated. The spectroscopic quality coefficient of the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystal and its intermultiplet and Stark luminescence branching coefficients were measured. The latter were measured for the three principal crystallographic directions in $\text{LuAlO}_3:\text{Nd}^{3+}$ at 77 and 300 K. Orientation cross sections for all transitions between the ${}^4F_{3/2}$ and ${}^4I_{11/2,13/2}$ states were determined. The dependences of the excitation thresholds for generation lines *A* and *B* in the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystal ($F \approx \parallel [112]$) were studied and their theoretical interpretation presented. From the analysis one can deduce the main physical causes of the temperature dependence of these generation lines. The behavior of lasing lines *A* and *B* for an ordinary laser or laser with a combined active medium were studied in a broad temperature range (77–550 K). Some data are presented on the lasing properties of the YAlO_3 crystal for three transitions at 300 K.

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INTRODUCTION

Among the numerous activated crystalline media capable of stimulated emission (SE), a prominent place is occupied by the crystals $\text{Y}_3\text{Al}_5\text{O}_{12}$ and YAlO_3 doped with trivalent rare-earth (TR^{3+}) ions.^[1] These compounds belong to one binary chemical system $\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$ and are produced in it at sesquioxide ratios 3:5 and 1:5. Despite the difference in the crystal structure, they combine successfully satisfactory spectral and lasing properties with a number of such remarkable qualities as high mechanical strength, hardness, appreciable thermal conductivity, and transparency in a broad spectral interval. Research of recent years has shown^[1-3] that the same physical and chemical capabilities is possessed also by a number of other binary systems with general formula $(\text{TR})_2\text{O}_3-\text{Al}_2\text{O}_3$, stable compounds of which are of great interest for the physics and spectroscopy of solids.^[1-7] Some of these crystals are of definite practical interest. These include the recently obtained compounds of LuAlO_3 with Nd^{3+} ions.^[7] This crystal (just like YAlO_3) has a distorted perovskite structure (space group $D_{2h}^{16}-\text{Pbnm}$) and is produced in the $\text{U}_2\text{O}_3-\text{Al}_2\text{O}_3$ system at a component ratio 1:1. Another crystalline compound of this system with garnet structure (3:5) ($\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Nd}^{3+}$) was recently investigated in detail in^{[8], 1)}

The present paper is devoted to an experimental and theoretical investigation of the main spectral characteristics of the crystal $\text{LuAlO}_3:\text{Nd}^{3+}$, which cast the most light on the possibility of using this compound to obtain SE in different regimes, on the lines of its principal (${}^4F_{3/2} - {}^4I_{11/2}$) and supplementary (${}^4F_{3/2} - {}^4I_{13/2}$) emission channels and also help trace intelligently the SE parameters both when the crystal is varied and when other experimental conditions are changed. Among the primary tasks of our comprehensive investigations were

the following: establishment of the energy position of the Stark components of the Nd^{3+} ions, the transitions between which correspond to the optical transparency band of LuAlO_3 , and identification of the induced transitions (IT); determination of the intermultiplet ($\beta_{J, iE}$) and Stark (β_{ij}) branching coefficients of the luminescence from the levels of the metastable state ${}^4F_{3/2}$; measurement of the cross sections ($\sigma_{e, ij}$) of the transitions between the Stark levels of the multiplets ${}^4F_{3/2}$ and ${}^4I_{11/2,13/2}$ for the three crystallographic directions *a*, *b*, and *c*; study of the temperature dependence of the luminescence line width ($\Delta\nu_{\text{lum}}$) and of the excitation threshold (E_{thr}) of certain SI lines of the principal lasing channel.

CRYSTALS, EXPERIMENTAL TECHNIQUE, AND INVESTIGATION METHODS

In our comprehensive investigations we used $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals grown by the Czochralski method, in a commercial "Donets-3" iridium-crucible installation in an atmosphere of an inert gas (argon or nitrogen). The initial charge was a stoichiometric mixture of the sesquioxides Lu_2O_3 and Al_2O_3 in a ratio 1:1. The appropriate fraction of neodymium was introduced into the charge in oxide form. A distinguishing feature of the manufacture of $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals is the need for conditions that exclude the possibility of crystallization of the phase $\text{Lu}_3\text{AlO}_{12}$ or Lu_2O_3 on the primer during the priming process. The drawing rates employed range from 2 to 5 mm/hr at a rotation speed 20–45 rpm. Under our experimental conditions, when priming on an iridium wire, the natural and preferable growth direction was [112], along which most crystals were grown. To investigate the SE characteristics we prepared laser elements of length ≈ 18 and diameter ≈ 5 mm with plane-parallel end faces ($\approx 10'$). In the latter, the activator

concentration was about 1 at.%. Some of the laser elements had small twins over the radial cross section (internal losses $\approx 0.1 \text{ cm}^{-1}$). For precision luminescence measurements we used a single-block rectangular $\text{LuAlO}_3:\text{Nd}^{3+}$ sample of high optical homogeneity (with $\Delta n \leq 2 \times 10^{-5}$), with dimensions $4.5 \times 5 \times 8.2 \text{ mm}$, the sides of which were perpendicular (accurate to $\leq 2^\circ$) to the crystallographic directions a , b , and c .

Preliminary x-ray structure²⁾ and spectroscopic^[7] investigations of the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystal have shown that it has orthorhombic symmetry with a space group $D_{2h}^{16}-\text{Pbnm}$. The calculated x-ray density was 8.32 g/cm^3 , in good agreement with the density 8.339 g/cm^3 measured by hydrostatic suspension in toluene. The unit cell parameters of the LuAlO_3 crystal at 300°K were $a_0 = 5.100 \pm 0.003$, $b_0 = 5.324 \pm 0.002$, and $c_0 = 7.294 \pm 0.001 \text{ \AA}$. The microhardness measured along the three principal crystal directions was (in the Knoop scale) $\theta_a = 1293$, $\theta_b = 1460$, and $\theta_c = 2211 \text{ kg/mm}^2$. The specific heat of the LuAlO_3 crystal was $\approx 0.106 \text{ cal/g-deg}$. The refractive indices were estimated with an ellipsometer at the wavelength 6328 \AA and amounted to $n_a = 1.910$, $n_b = 1.934$, and $n_c = 1.885$.

All the spectroscopic measurements connected with the determination of the energies of the Stark levels and with the identification of the transitions between them were carried out by the traditional experimental methods of absorption and luminescence analysis supplemented by methods of SE spectroscopy.^[10] In these measurements we used the high-resolution spectral instruments DFS-8, DFS-12, SDL-1, UR-20, and SPM-2 (with a 300 line/mm grating). To excite the luminescence in a wide temperature interval we used effective illuminating systems employing quartz tungsten-iodine lamps of the KGM 12-100 and LIN-4 type. The line intensities of the luminescence of the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals were determined by the Judd-Ofelt method as modified by Kaminskii and Li,^[11,12] using an SF-8 spectrophotometer; the spectra were mathematically reduced with a 220-M computer. The excitation and investigation of the SE of the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals in the temperature interval from 77 to 600°K were carried out with experimental apparatus described in detail in^[13,14]. At 77°K , the laser elements were in direct contact with the liquid nitrogen. To excite the SI on low-intensity luminescence lines (${}^4F_{3/2} \rightarrow {}^4I_{11/2}$) we used a method that employed a laser with a combined active medium (CAM), developed by Kaminskii.^[15,16] In the latter case, the CAM were the laser crystals CaSc_2O_4 and Y_2SiO_5 with Nd^{3+} ions, and LGC-2 neodymium silicate glass. In all the SE experiments we used an external optical resonator with spherical interference mirrors arranged in a confocal scheme ($R \approx 600 \text{ mm}$) and having an approximate transmission 1% at the generation wavelength (λ_g).

INITIAL PREMISES AND PROBLEMS OF THE THEORY

1. *Optical spectra of the Nd^{3+} ion in the LuAlO_3 crystal.* The primary task in the spectroscopic study of activated crystals for lasers is to analyze the Stark structure of their spectra and to construct a detailed energy

level scheme for the activator. Without these data it is impossible to understand the nature of not only the SE but also of many other processes that take place in a laser crystal when it is excited.

The Nd^{3+} ions replace in the LuAlO_3 lattice Lu^{3+} ions having a position symmetry C_s and a twelve-fold oxygen surrounding. The total number of Lu^{3+} ions in LuAlO_3 , according to x-ray structure data is $2.017 \times 10^{22} \text{ cm}^{-3}$. Preliminary investigations have shown that at low concentrations the Nd^{3+} ions form in this compound one kind of activator center. The low symmetry of these centers causes the intracrystalline field to lift completely the orbital degeneracy of the multiplets. Certain luminescence data and information on the positions of the levels of the multiplets ${}^4F_{3/2}$ and ${}^4I_{9/2-13/2}$ of Nd^{3+} ions at 77°K in the LuAlO_3 crystal are given in a paper by Feofilov *et al.*^[7]

2. *Effective cross sections of radiative transitions.* For homogeneously broadened luminescence lines of isotropic media, connected with a singlet initial state, the formula for $\sigma_{e,ij}$ can be represented by

$$\sigma_{e,ij} = A_{ij} \lambda_{ij}^2 / 4\pi^2 n^2 \Delta \nu_{\text{lum}}, \quad (1)$$

where A_{ij} is the probability of the spontaneous transition $i \rightarrow j$ between the Stark levels i and j , and n is the refractive index at the wavelength λ_{ij} . For the case when the decay of the metastable level i proceeds only via radiative transitions ($1/\tau_{\text{lum}} = \sum_i A_{ij}$), the values of A_{ij} can be easily determined from the luminescence measurement data by means of the formula

$$A_{ij} = \beta_{ij} / \tau_{\text{lum}}. \quad (2)$$

For anisotropic crystals (of which LuAlO_3 is one) activated with Nd^{3+} , the experimental determination of A_{ij} is greatly complicated because of the redistribution of the excitation over the Stark levels of the metastable state ${}^4F_{3/2}$, and because of the different polarizations of the individual luminescent transitions. For this case, expression (2) takes the form

$$A_{ij} = \tau_{\text{lum}}^{-1} \beta_{ij}(\varphi, T) / b_i \Phi_{ij}(\varphi). \quad (3)$$

Here $\beta_{ij}(\varphi, T)$ is the spectral density of the luminescence for the line ij when observed at an angle φ to the chosen crystallographic direction of the crystal, and $\Phi_{ij}(\varphi)$ is a function that determines the dependence of the intensity of the ij line on the direction. The form of the function $\Phi_{ij}(\varphi)$ for crystals with various symmetries is given in a paper by Hellwege.^[17] In expression (3), b_i is the Boltzmann factor of the i -th Stark level of the state ${}^4F_{3/2}$ relative to its level ($i = 1$), and is determined by the relation

$$b_i = \exp(-\Delta E_{i1}/kT) / \sum_i \exp(-\Delta E_{i1}/kT), \quad (4)$$

where ΔE is the energy gap, k is the Boltzmann constant, and T is the temperature.

All the formulas describing the energy characteristics of the SI of the Nd^{3+} ions in anisotropic crystals contain

not the true probability of the spontaneous emission, which naturally, as follows from (3), is a constant for the transition $i-j$, but its value that characterizes the amplification properties of the crystal at the given temperature and orientation and at the given wavelength λ_{ij} . In this case, naturally, there is no need for selecting the function $\Phi_{ij}(\varphi)$ and we can use the arbitrary concept of orientational probability of spontaneous emission $A_{ij}^{or}(\varphi, T)$ and the orientational cross section $\sigma_{e,ij}^{or}(\varphi, T)$. For cubic crystals with Nd^{3+} ions (and other isotropic media) these characteristics will be only temperature-dependent. Then expression (1), taking the foregoing into account, is rewritten in the form

$$\sigma_{e,ij}^{or}(\varphi, T) = A_{ij}^{or}(\varphi, T) b_{ij}^2 / 4\pi^2 n^2 \Delta v_{lum}, \quad (5)$$

$$A_{ij}^{or}(\varphi, T) = \beta_{ij}(\varphi, T) / \tau_{lum} b_i = \beta_{j,iz} a_{j,iz}(\varphi, T) / \tau_{lum} b_i. \quad (6)$$

In formula (6), the parameter $a_{j,iz}(\varphi, T)$ describes the relative spectral density of the emission for the ij line of the given luminescence channel ${}^4F_{3/2} - {}^4I_J$. This characteristic can be determined from the orientational luminescence spectra, the line intensities of which are recalculated to take into account the spectral properties of the monochromators and the sensitivity of the photoreceivers.

3. Line intensities in the spectra of $\text{LuAlO}_3 : \text{Nd}^{3+}$.

The coefficient $\beta_{j,iz}$ in formula (6) is defined as

$$\beta_{j,iz} = A(i, J) / \sum_j A(i, J), \quad (7)$$

where $A(i, J)$ is the total probability of the spontaneous transitions from the initial levels (i) of the metastable states ${}^4F_{3/2}$ to all the Stark components of the final multiplets (J). Since the magnetic and quadrupole transitions between the states ${}^4F_{3/2}$ and 4I_J of the Nd^{3+} ion are forbidden ($|\Delta L| > 2$ and $|\Delta J| > 2$), the main contribution to the intensity of the (${}^4F_{3/2} - {}^4I_{9/2-15/2}$) luminescence will be made by electric dipole transitions induced by odd harmonics of the crystal field. Then the expression for $A(i, J)$, which enters in (7), takes the following form in the notation employed in^[18,19]:

$$A(aJ, bJ') = \frac{64\pi^4 e^2}{3h(2J+1)\bar{\lambda}_J^3} \frac{n(n^2+2)^2}{9} s(aJ, bJ'), \quad (8)$$

where

$$s(aJ, bJ') = \sum_{t=2,4,6} \Omega_i | \langle 4f^n [S, L] J \| U^{(t)} \| 4f^n [S', L'] J' \rangle |^2, \quad (9)$$

$\bar{\lambda}_J$ is the mean value of the wavelength of the intermultiplet transition $aJ - bJ'$, e is the elementary charge, h is Planck's constant, Ω_i is the Judd-Offelt theory^[18,19] are phenomenological parameters of the intensity, $\langle U^{(t)} \rangle$ is the matrix element of an irreducible tensor operator of rank t , which connects the initial and final states of the given transition, the wave functions of which are taken in the intermediate-coupling scheme in the form of linear combinations of Russell-Saunders states $|4f^n S, L\rangle$.

A remarkable consequence of the Judd-Offelt theory^[18,19] is that the parameter $s(aJ, bJ')$, which deter-

mines in (5) the integral intensity (it is sometimes called the line strength) of the luminescence, characterizes also the intensity of the absorption transitions of the given TR^{3+} ion. Therefore, by analyzing the absorption spectra we can obtain the value of the parameters Ω_i and calculated from them, knowing the matrix elements $\langle U^{(t)} \rangle$, the values of $A(aJ, bJ')$.

It follows from (8) and (9) that the values of $A(aJ, bJ')$ or $A(i, J)$ transitions from the levels of the state ${}^4F_{3/2}$ of the Nd^{3+} ion depend only on the parameters Ω_4 and Ω_6 , since the matrix elements of the second-rank operator between the multiplets ${}^4F_{3/2}$ and ${}^4I_{9/2-15/2}$ are equal to zero ($|\Delta J| > 2$). Starting from this fact, it has been shown in^[11,12] that the coefficients $\beta_{j,iz}$ are determined only by the ratio Ω_4/Ω_6 , which was called the spectroscopic quality parameter and designated by the symbol X . Its obvious consequence has made it possible to obtain in graphic form the analytic relations

$$\beta_{j,iz}(X) = \frac{(a_j X + b_j) / \bar{\lambda}_j^3}{\sum_j [(a_j X + b_j) / \bar{\lambda}_j^3]}, \quad (10)$$

where a_j and b_j are constants equal to the squares of the matrix elements of the irreducible tensor operator of fourth and sixth rank, respectively

$$a_j = | \langle {}^4F_{3/2} \| U^{(4)} \| {}^4I_J \rangle |^2, \\ b_j = | \langle {}^4F_{3/2} \| U^{(6)} \| {}^4I_J \rangle |^2,$$

and the numerical values of which were taken by Kaminskii and Li^[11,12] from Krupke's paper.^[20,13] Formula (10) leads to an extremely important conclusion—to each active medium with Nd^{3+} ions there corresponds a definite value of the parameter X , which determines uniquely the set of branching coefficients for its four luminescence channels ${}^4F_{3/2} - {}^4I_{9/2-15/2}$. The investigations of^[11,12] have also shown that the parameter X can be determined with sufficient accuracy by analyzing the integral absorption coefficient of only two absorption transitions: ${}^4I_{9/2} - {}^2P_{1/2}$ and ${}^4I_{9/2} - {}^4I_{15/2}$, a result of which was a simple expression for X , convenient for the experimenters:

$$X = 1.23s({}^4I_{9/2}, {}^2P_{1/2}) / s({}^4I_{9/2}, {}^4I_{15/2}). \quad (11)$$

Its convenience lies primarily in the fact that to determine X one needs not the absolute values of $s(aJ, bJ')$, but only their relative values. This makes it unnecessary to determine the concentration of the Nd^{3+} in the investigated medium, thus greatly simplifying the experiment.

4. *Temperature dependence of the lasing excitation threshold.* According to Vylegzhanin and Kaminskii,^[14,22] the $E_{thr}(T)$ dependence of the ij lasing line of the transition $i-j$ of the Nd^{3+} ions, in the case of quasi-stationary emission, can be described by the formula

$$E_{thr}(T) \sim m_1 A_3 [(1/\delta_{ij}^{or}) + \exp(-\Delta E_{zz}/kT)] \\ \times [1 + m_2 \exp(-\Delta E_{rs}/kT)]. \quad (12)$$

Here $m_{1,2}$ are proportionality coefficients that are practically independent of the temperature, with m_1 determining the ratio of the excitation rate and the excitation energy, and $m_2 = A_4/A_3$ (A_3 and A_4 are the combined

probabilities of the spontaneous emission from the levels of the states $R(^4F_{3/2})$ and $S(^4F_{5/2}, ^2H_{11/2})$ respectively; ΔE_{ZY} and ΔE_{RS} are the energy gaps between the nearest Stark components of the $^4I_{9/2} \rightarrow ^4I_{11/2}$ and $^4F_{3/2} \rightarrow ^4F_{5/2}$ multiplets, respectively; δ_{ij}^{or} is a dimensionless parameter characterizing the amplification properties of the lasing medium at the wavelength of the $i \rightarrow j$ transition in the given orientation under the condition that all the active ions are in the excited state,

$$\delta_{ij}^{or}(\varphi, T) = (N_0/\rho) (\lambda_{ij}^2/4\pi^2) (b_i A_{ij}^{or}(\varphi, T)/c \Delta v_{lum}), \quad (13)$$

where N_0 is the concentration of the ions in the active medium and ρ is the coefficient of the total losses. It follows from (12) and (13)⁴⁾ that the temperature dependence of E_{thr} is governed principally by the temperature dependence of the luminescence line width, by the distribution of the excitations over the Stark components of the metastable state $^4F_{3/2}$, and by the exponential terms in (12), which determine the populations of the levels of the multiplets $^4I_{11/2}$ and $(^4F_{5/2}, ^2H_{11/2})$.

If several transitions between equal-frequency Stark levels of a given pair of multiplets participate in the SE, it is necessary to substitute in (12) the quantity δ_{eff} , equal to (in the case of complete resonance) the sum of $\delta_{ij}^{or}(\varphi, T)$ for these transitions or their corresponding parts (in the case of partial overlap). When the temperature of the generating crystal is increased switching between the lasing lines may take place in the SE spectrum as a result of the shift and broadening of

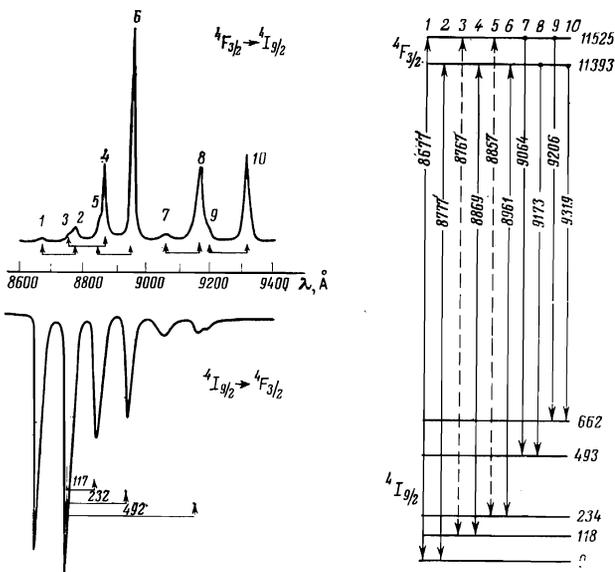


FIG. 1. Unpolarized luminescence spectra ($^4F_{3/2} \rightarrow ^4I_{9/2}$, 77 K), and absorption spectra ($^4I_{9/2} \rightarrow ^4F_{3/2}$, 300 K), and level scheme of crystal splitting of the multiplets of $^4F_{3/2}$ and $^4I_{9/2}$ of the Nd^{3+} ions in $LuAlO_3$ at 77°K. The brackets with the arrows on the spectra indicate the splitting of the metastable state $^4F_{3/2}$ and the positions of the levels of the principal $^4I_{9/2}$ multiplet. The dashed arrows on the level scheme correspond to insufficiently distinct lines on the spectra. The same numbering is used for the transitions on the scheme and the corresponding lines on the spectra. On the scheme, the level positions are indicated in cm^{-1} , and the transitions between them are given in Å.

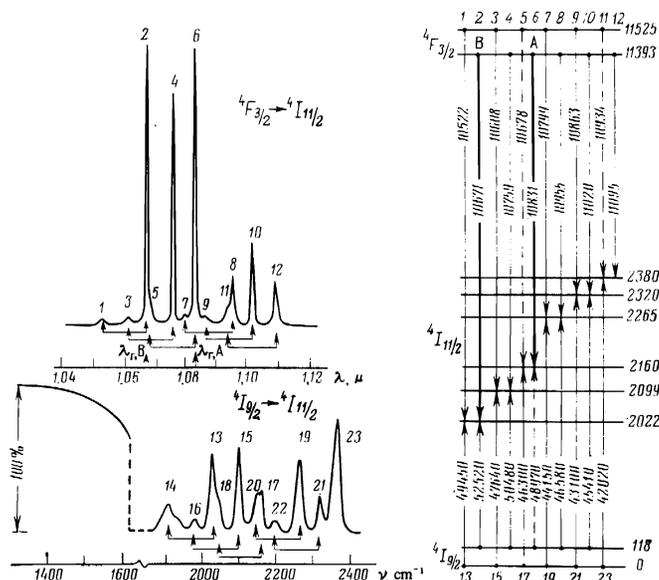


FIG. 2. Unpolarized luminescence spectra ($^4F_{3/2} \rightarrow ^4I_{11/2}$, 77 K), and absorption spectra ($^4I_{9/2} \rightarrow ^4I_{11/2}$, 90 K), and the scheme of the crystal splitting of the $^4F_{3/2}$ and $^4I_{11/2}$ multiplets of the Nd^{3+} ions in $LuAlO_3$ ($F \approx \parallel [112]$) at 77°K. The thick arrows mark the registered induced transitions. The brackets on the absorption spectra indicate the splitting up to the first excited Stark component of the ground state of $^4I_{9/2}$. The remaining symbols are the same as in Fig. 1.

the lines, and also as a result of the increased intensity of the luminescence lines due to transitions from the upper level of the $^4F_{3/2}$ state as a result of its population. For anisotropic crystals this phenomenon can also be enhanced by lasing-elements twins with inhomogeneous refractive-index patterns.

EXPERIMENTAL DATA AND THEIR DISCUSSION

1. *Determination of the Stark splittings of the Nd^{3+} ion in the $LuAlO_3$ crystal.* In our experiments we determined and refined the positions of the Stark components of most states of the Nd^{3+} ions in $LuAlO_3$, which extend up to $\approx 26\,000\,cm^{-1}$ ($\approx 3850\,Å$) at temperatures 77 and 300°K. Inasmuch as one of our tasks was a detailed study of the SE characteristics, greatest attention was paid in the spectroscopic measurements to the multiplet Stark levels between which induced transitions were possible. To increase the experimental accuracy these levels were investigated with the aid of the luminescence and absorption spectra. By way of example, Figs. 1 and 2 show the results of such an analysis for the transitions $^4F_{3/2} \rightarrow ^4I_{9/2}$ and $^4F_{3/2} \rightarrow ^4I_{11/2} \rightarrow ^4I_{9/2}$. In Table I we have also confined ourselves to data for the energies of the Stark components of the lowest $^4I_{9/2-15/2}$ and $^4F_{3/2}$ multiplets at 77°K. Comparison of the results with data on the positions of the Stark levels of the Nd^{3+} ions in other isostructural laser crystal matrices ($YAlO_3$,^[11] $GdAlO_3$ ^[25]) shows that the splitting of the multiplets of the lowest term 4I_j are practically identical. At the same time, as shown in^[7], the nepheloxetic shifts of the $^4F_{3/2}$ multiplets (relative to its position in the aqua-ion— $11\,580\,cm^{-1}$ ^[26]) is the largest in the $LuAlO_3$ crystal. According to our data, this effect

TABLE I. Splitting of the multiplets of the Nd³⁺ ion in the LuAlO₃ crystal 77°K.

Multiplets	Positions of Stark levels, cm ⁻¹	Number of components		ΔE, cm ⁻¹
		Theory	Experiment	
⁴ F _{3/2}	0, 118, 234, 493, 662	5	5	662
⁴ I _{11/2}	2022, 2099, 2160, 2265, 2320, 2380	6	6	358
⁴ I _{13/2}	3946, 4010, 4085, 4195, 4280, 4330, 4432	7	7	506
⁴ I _{9/2}	5760, 5895, 6010, 6240, 6300, 6405, 6685*, 6735	8	8	973
⁴ F _{5/2}	11393, 11525	2	2	132

manifests itself also to a considerable degree in the positions of the multiplets that lie higher. This means that in LuAlO₃ the activator-ligand bond is the most covalent of these three laser crystal matrices. The accuracy with which the ⁴F_{3/2} and ⁴I_{9/2-15/2} multiplet level positions are determined is not worse than 1-3 cm⁻¹. Since the lasing effect was observed in the LuAlO₃:Nd³⁺ crystal also at 300°K (⁴F_{3/2} - ⁴I_{11/2,13/2}), Figs. 3 and 4 show the corresponding luminescence spectra and the energy schemes with the results of the identification of the registered induced transitions.

2. Measurement of the cross sections of radiative transitions in the ⁴F_{3/2} - ⁴I_{11/2,13/2} channels. It follows from (5) and (6) that to determine σ_{e,ij}^{or}(φ, T) it is necessary to know the values of β_{ij}(φ, T), τ_{lum}, and Δν_{lum}. In turn, the Stark branching coefficients, according to (16), can be determined from the known values of β_{J, iE} and the luminescence spectra registered along the three crystallographic directions a, b, and c. The measurements of τ_{lum} of the metastable state of the Nd³⁺ ions in LuAlO₃ with small activator concentration (< 0.2 at. %) have shown that the lifetime, within the limits of the experimental accuracy, remains practically constant in the temperature interval 77 - ≈ 650°K and amounts to 160 ± 15 μsec. This indicates that the quantum yield of the luminescence from the levels of the state ⁴F_{3/2} is close to unity. Measurements of the intermultiplet coefficients β_{J, iE} of the Nd³⁺ ions in LuAlO₃ by the method described above, using formulas (10) and (11), have led to the results summarized in Table II. The obtained

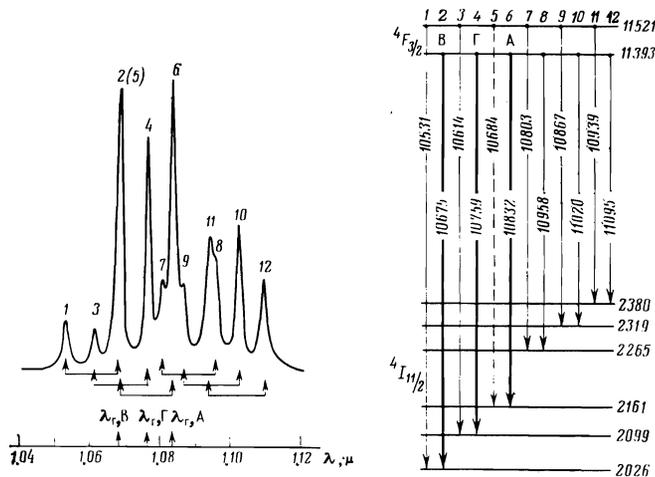


FIG. 3. Unpolarized luminescence spectrum (⁴F_{3/2} - ⁴I_{11/2}) and crystal splitting scheme of the states ⁴F_{3/2} and ⁴I_{11/2} of the Nd³⁺ ions in LuAlO₂ (F ≈ || [112]) at 300°K. The notation is the same as in Figs. 1 and 2.

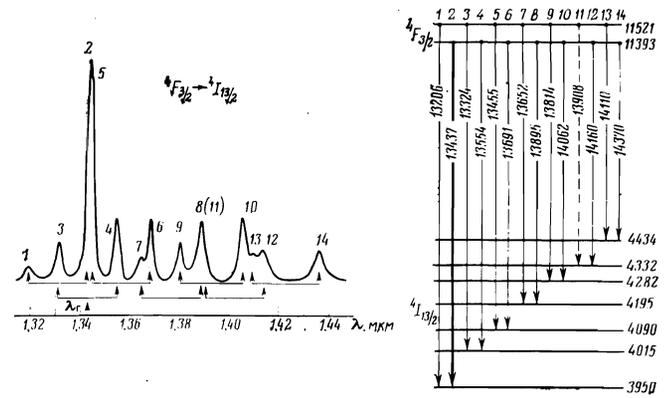


FIG. 4. Unpolarized luminescence spectrum (⁴F_{3/2} - ⁴I_{13/2}) and scheme of crystal splitting of the states ⁴F_{3/2} and ⁴I_{13/2} of the ions Nd³⁺ in LuAlO₃ (F ≈ || [112]) at 300°K. The notation is the same as in Figs. 1 and 2.

values of β_{J, iE} correspond to a spectroscopic quality parameter X = 0.265 ± 0.025. The values of the coefficients a_{J, iE}(φ, T), which enter in expression (6), were determined from the orientational luminescence spectra shown in Figs. 5-8. The experimentally obtained characteristics Δν_{lum} and β_{ij}(φ, T), as well as the calculated values of A_{ij}^{or}(φ, T) and σ_{e,ij}^{or}(φ, T) for the transitions of the principal (⁴F_{3/2} - ⁴I_{11/2}) and supplementary (⁴F_{3/2} - ⁴I_{13/2}) radiation channels are summarized in Tables III and IV.⁵⁾ The error in the determination of a_{J, iE}(φ, T), and consequently also β_{ij}(φ, T) do not exceed 10-15% for most lines. In these cases (especially for lines of low intensity), when the accuracy is low, the corresponding values of σ_{ij}(φ, T) in Tables III and IV are marked by an asterisk.

As already noted in item 4 of the preceding section, in the analysis of the E_{thr}(T) dependence it is necessary to take into account the amplification properties of the active medium at a wavelength λ_{ij}, due to resonance or to partial superposition of several emission lines (luminescence lines). This property, as shown by our investigations, are best characterized by a certain combined effective cross section σ_{e^{eff}}(φ, T), which consists of the corresponding parts with allowance for the polarization of the radiation) that take part in the resonance of the transitions. The measurements have shown that for certain SE lines of the LuAlO₃:Nd³⁺ crystal the resonance effect begins to influence substantially the behavior of E_{thr}(T) starting with temperatures ≥ 120°K. The numerical values of σ_{e^{eff}}(φ, T) for the most intense lines causing the stimulated emission on the transitions ⁴F_{3/2} - ⁴I_{11/2} and ⁴F_{3/2} - ⁴I_{13/2}, are given in Table V.

TABLE II. Intermultiplet luminescence branching coefficients β_{J, iE} from the levels of the metastable state ⁴F_{3/2} of Nd³⁺ ions in LuAlO₂ crystal.

Multiplet transition	Luminescence spectrum band, μ	β _{J, iE}
⁴ F _{3/2} → ⁴ I _{9/2}	0.868-0.932	0.27 ± 0.008
⁴ F _{3/2} → ⁴ I _{11/2}	1.052-1.109	0.585 ± 0.006
⁴ F _{3/2} → ⁴ I _{13/2}	1.320-1.438	0.138 ± 0.002
⁴ F _{3/2} → ⁴ I _{15/2}	1.735-2.149	≈ 0.006

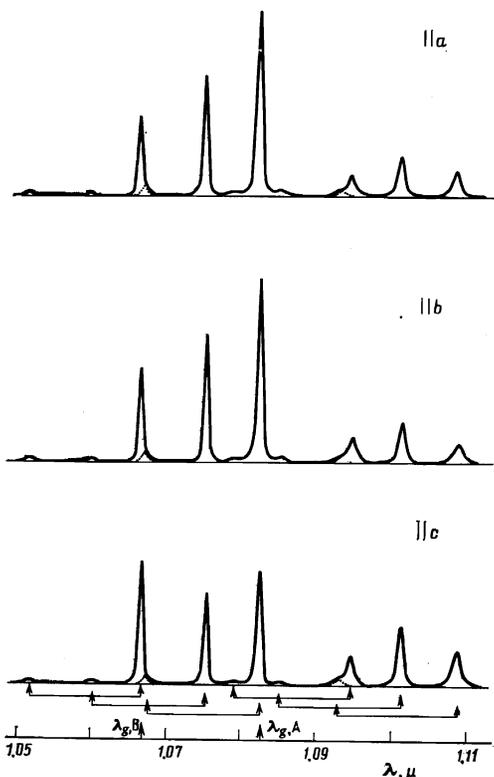


FIG. 5. Orientational luminescence spectra (${}^4F_{3/2} \rightarrow {}^4I_{11/2}$) of Nd^{3+} ions for three crystallographic directions of the LuAlO_3 crystal at 77°K. The dashed curves show the results of resolving the complex contours of the spectrum. The brackets with arrows indicate the splitting of the ${}^4F_{3/2}$ state. The line intensities are given in relative units.

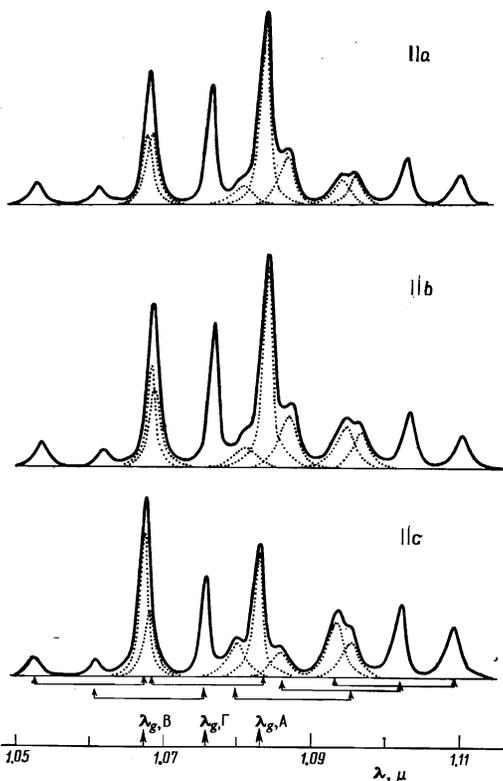


FIG. 6. Orientational luminescence spectra (${}^4F_{3/2} \rightarrow {}^4I_{11/2}$) of Nd^{3+} ions for three crystallographic directions of the crystal LuAlO_3 at 300°K. The notation is the same as in Fig. 5.

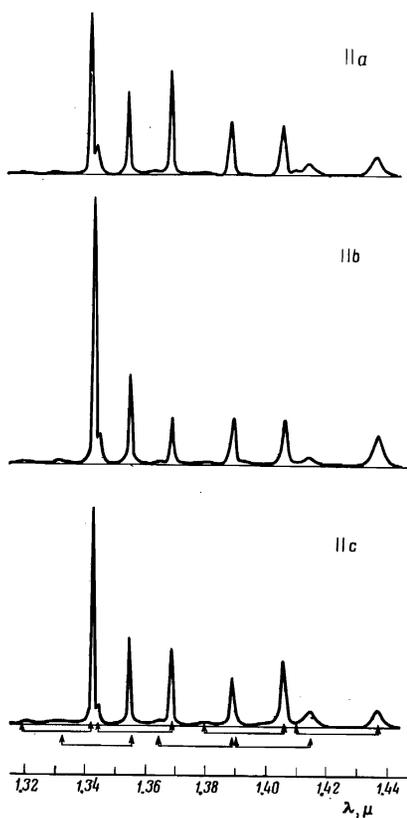


FIG. 7. Orientational luminescence spectra (${}^4F_{3/2} \rightarrow {}^4I_{13/2}$) of Nd^{3+} ions for three crystallographic directions of the LuAlO_3 crystal at 77°K. The notation is the same as in Fig. 5.

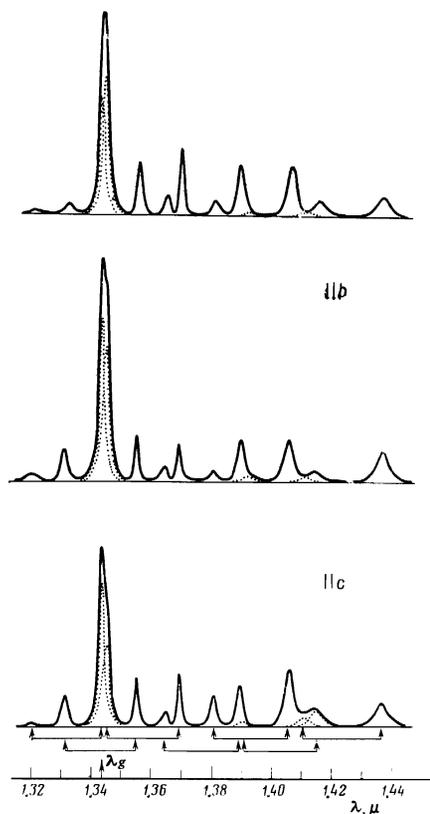


FIG. 8. Orientational luminescence spectra (${}^4F_{3/2} \rightarrow {}^4I_{13/2}$) of Nd^{3+} for three crystallographic directions of the crystal LuAlO_3 at 300°K. The notation is the same as in Fig. 5.

TABLE III. Spectral characteristics and orientational intensities of the luminescence lines of the transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ of Nd^{3+} ions in LuAlO_3 at 77 and 300°K.

Line, A	Luminescence line width, cm^{-1}	Crystallographic direction				
		a		b		c
		$\sigma_{e,ij}^{\text{or}} 10^{10}, \text{cm}^2$	$\sigma_{e,ij}^{\text{or}}, \text{cm}^2$	β_{if}^{or}	$A_{if}^{\text{or}}, \text{sec}^{-1}$	$\sigma_{e,ij}^{\text{or}} 10^{10}, \text{cm}^2$
T=77 K						
10 522	2.2±0.3	0.27	0.3	0.003	255	0.23
10 608	2.4±0.3	0.19	0.17	0.0027*	225	0.18
10 671 B	1.65±0.3	5.4	6.2	0.088	590	8.6
10 678	3.2±0.5	0.7	0.65	0.0102	485	0.52
10 759	1.9±0.3	8.2	8.3	0.071	480	6.2
10 799	4.5±0.5	0.17	0.14	0.0072*	590	0.27
10 831 A	2.8±0.3	12.4	12.1	0.123	830	7.4
10 863	5.9±0.5	0.34	0.29	0.006*	495	0.17
10 934	8.2±0.5	0.15	0.23	0.015*	1245	0.3
10 955	5.0±0.5	1.36	1.65	0.054	370	1.9
11 020	5.2±0.5	2.65	2.7	0.117	790	3.9
11 095	6.8±0.5	1.6	1.4	0.089	600	2.4
T=300 K						
10 531	11.5±1.0	0.24	0.26	0.014	250	0.18
10 614	12.0±1.0	0.17	0.16	0.013	230	0.17
10 675 B	7.5±1.0	0.83	0.95	0.063	600	1.36
10 680	12.5±2.5	0.84	0.75	0.047	845	0.62
10 759 Γ	8.5±1.0	1.34	1.36	0.049	470	0.95
10 803	15.5±2.5	0.23	0.2	0.034*	600	0.36
10 832 A	11.5±1.5	2.15	2.1	0.086	820	1.24
10 867	16.5±2.5	0.56	0.45	0.028*	495	0.28
10 939	20.0±2.5	0.29	0.43	0.070*	1245	0.6
10 958	14.5±2.5	0.33	0.4	0.038	370	0.45
11 020	16.5±1.5	0.6	0.58	0.082	790	0.87
11 095	18.5±1.5	0.41	0.37	0.062	600	0.6

For comparison the table shows also the cross sections of certain laser lines at 77 and 300°K, for which at the indicated temperature the influence of the resonance effect is negligible. The cross sections of these lines are marked by an asterisk in Table V.

3. *Results of the investigation of SE of $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals.* Figure 9 shows the temperature spectra of the lasing of $\text{LuAlO}_3:\text{Nd}^{3+}$ (the laser axis F of the element is oriented along the direction [112], which is close to the principal crystallographic direction c). Our investigations have shown that the emission of all the registered SE lines in the laser system has a degree of polarization close to 100%. As seen from the spectra, a laser based on $\text{LuAlO}_3:\text{Nd}^{3+}$ ($F \approx \parallel [112]$) at 77°K begins to generate on the line B, which is connected with the transition $11\,393\text{ cm}^{-1} {}^4F_{3/2} - {}^4I_{11/2}$ 2022 cm^{-1} (see Fig. 2). At a temperature $\approx 120^\circ\text{K}$, the line A due to the transition $11\,393\text{ cm}^{-1} {}^4F_{3/2} - {}^4I_{11/2}$ 2160 cm^{-1} also begins to be excited in the lasing. At $T \approx 160 \pm 5^\circ\text{K}$ temperature switching of the SE lines is observed, and at $T \geq 250^\circ\text{K}$ only the line A remains in the spectra. The behavior of the functions $E_{\text{thr}}(T)$ of lines A and B is shown in Fig. 10. An analysis of the line switching effect has shown that it is due to the increased value of $\sigma_e^{\text{eff}}(\varphi, T)$ of line A as a result of the superposition on this line of the neighboring lines 7 and 9 (see Figs. 2 and 3). These three SE lines have approximately the same radiation polarization.

At 300°K, the SE of the crystal $\text{LuAlO}_3:\text{Nd}^{3+}$ ($F \approx \parallel [112]$) was also excited at a wavelength $13\,437 \pm 10 \text{ \AA}$ with a relatively low pump threshold $\approx 15 \text{ J}$. The identification of this generation line (see Fig. 4) has shown that it is connected with the transition $11\,393\text{ cm}^{-1} {}^4F_{3/2} - {}^4I_{13/2}$ 3950 cm^{-1} , which is "aided" by the resonant transition $11\,521\text{ cm}^{-1} {}^4F_{3/2} - {}^4I_{13/2}$ 4090 cm^{-1} . The use of the method of stimulated emission spectroscopy with harmonic generation^[27] will make it possible to refine the spectral parameters of this emission line.

TABLE IV. Spectral characteristics and orientational intensities of the luminescence lines of the transition ${}^4F_{3/2} \rightarrow {}^4I_{13/2}$ of the Nd^{3+} ions in LuAlO_3 crystal at 77 and 300°K.

Line, A	Luminescence line width, cm^{-1}	Crystallographic direction				
		a		b		c
		$\sigma_{e,ij}^{\text{or}} 10^{10}, \text{cm}^2$	$\sigma_{e,ij}^{\text{or}} 10^{10}, \text{cm}^2$	β_{if}^{or}	$A_{if}^{\text{or}}, \text{sec}^{-1}$	$\sigma_{e,ij}^{\text{or}} 10^{10}, \text{cm}^2$
T=77 K						
13 194	1.8±0.3	0.03	0.02	0.0003*	25	0.04
13 312	2.0±0.3	0.05	0.18	0.0015*	120	0.16
13 429	1.65±0.3	4.1	6.2	0.035	235	5.5
13 447	2.4±0.5	0.72	0.74	0.004	340	0.44
13 546	1.8±0.3	2.0	2.16	0.014	90	1.95
13 642	3.5±0.5	0.08	0.05	0.001*	75	0.07
13 685	2.5±0.3	2.47	1.1	0.017	120	1.85
13 803	5.0±0.5	0.05	0.03	0.002*	170	0.11
13 891	4.0±0.5	1.34	1.24	0.014	100	0.98
13 903	7.0±0.5	0.02	0.01	0.001*	65	0.03
14 059	4.5±0.5	1.4	1.23	0.025	175	1.55
14 097	9.0±1.0	0.2	0.01	0.0008*	65	0.02
14 160	6.5±0.5	0.3	0.22	0.001	50	0.32
14 365	7.0±0.5	0.5	0.77	0.012	0	0.51
T=300 K						
13 206	9.5±1.0	0.03	0.02	0.0015*	25	0.04
13 324	10.5±1.0	0.05	0.017	0.006	120	0.16
13 437	7.5±1.0	0.67	0.96	0.024	230	0.84
13 455	10.5±2.0	0.77	0.8	0.02	235	0.46
13 554	9.0±1.0	0.29	0.31	0.01	90	0.28
13 652	12.0±1.5	0.11	0.07	0.004*	75	0.09
13 691	11.0±1.5	0.38	0.18	0.012	120	0.3
13 814	14.0±1.5	0.09	0.06	0.01	170	0.18
13 895	13.0±2.0	0.3	0.27	0.01	95	0.22
13 908	19.0±2.5	0.03	0.03	0.04*	65	0.06
14 062	15.0±2.5	0.3	0.26	0.018	170	0.33
14 110	21.0±2.5	0.03	0.02	0.004*	65	0.05
14 160	18.0±2.5	0.01	0.06	0.005*	50	0.08
14 370	20.0±2.5	0.12	0.18	0.009	80	0.13

The $E_{\text{thr}}(T)$ dependence was interpreted only for the line A. The interpretation was based on formula (12) using the data of that part of the article where the objects of the investigations are described, and the data of Table III and Fig. 1, which show also the experimental plots of $\Delta\nu_{\text{lum}}(T)$. The theoretical curve 1 on Fig. 10 was obtained without allowance for the "leakage" of the excitation via the levels of the multiplets ${}^4F_{5/2}$ and ${}^2H_{11/2}$ on account of their thermal population by radiative transitions. In this case, as seen from Fig. 10, the $E_{\text{thr}}(T)$ curve accurate to an exponential coefficient b_i , follows the variation of $\Delta\nu_{\text{lum}}(T)$. The theoretical curve 2 was obtained with the second exponential term of (12) taken into account. Better agreement between theory and experiment was obtained at $A_4/A_3 = 3$. The influence of the first exponential term $\exp(-\Delta E_{2X}/kT)$ was insignificant in our case because of the large losses in the lasing crystal. The abrupt increase of E_{thr} at $T \geq 500^\circ\text{K}$ is due to causes not accounted for in the assumed model. The most important among them is con-

TABLE V. Effective cross sections of certain intense luminescence lines of the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystal.

Transition	Line, A	Temperature, °K	Crystallographic direction		
			a		c
			$\sigma_e^{\text{eff}} 10^{10}, \text{cm}^2$	$\sigma_e^{\text{eff}} 10^{10}, \text{cm}^2$	$\sigma_e^{\text{eff}} 10^{10}, \text{cm}^2$
${}^4F_{3/2} \rightarrow {}^4I_{11/2}$	10 671 B	77	5.4*	6.2*	8.6*
	10 759 ** D	77	8.2*	8.3*	6.2*
	10 831 A	77	12.4*	12.1*	7.4*
	10 675 B	300	0.83*	0.95*	1.36*
	10 684 ** E	300	0.84*	0.75*	0.62*
	10 759 D	300	1.34*	1.36*	0.95*
${}^4F_{3/2} \rightarrow {}^4I_{13/2}$	10 832 A	300	2.2	2.18	1.28
	13 429 **	77	4.1*	6.2*	5.5*
	13 437	300	≈ 0.9	≈ 1.2	≈ 1.0
	13 455 **	300	≈ 0.9	≈ 1.0	≈ 0.55

Note. No stimulated emission has been excited so far on the lines noted by two asterisks.

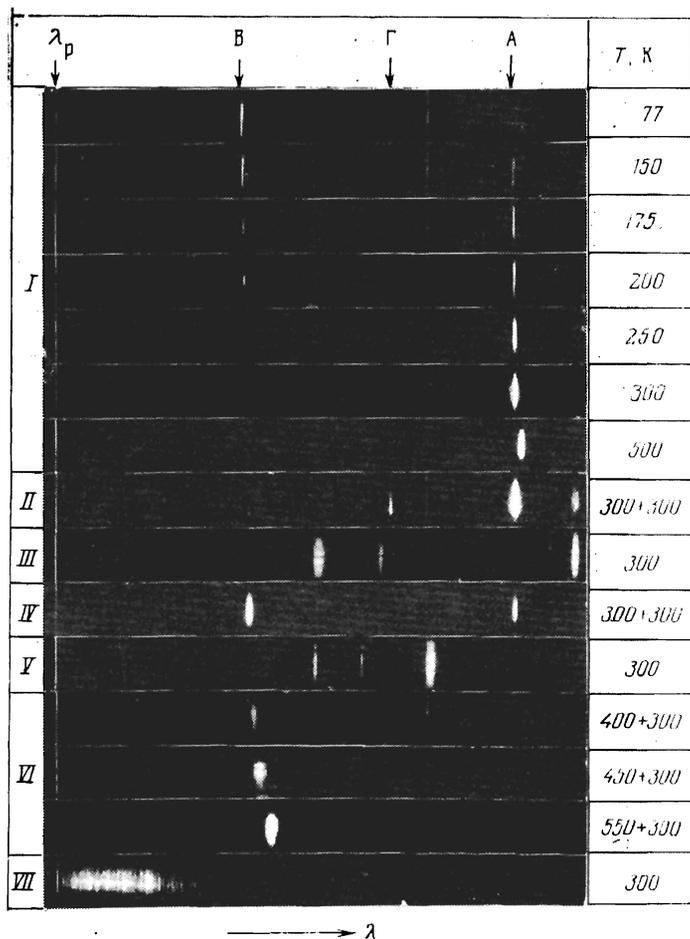


FIG. 9. Temperature spectra of SE of $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals ($F \approx [112]$) in the usual laser scheme and in the scheme of a laser with CAM. Reference line with $\lambda_p = 10561 \text{ \AA}$ is indicated by the arrow: I— $\text{LuAlO}_3:\text{Nd}^{3+}$ laser; II— $\text{LuAlO}_3:\text{Nd}^{3+} + \text{CaSc}_2\text{O}_4:\text{Nd}^{3+}$ laser with CAM; III— $\text{CaSc}_2\text{O}_4:\text{Nd}^{3+}$ laser; IV— $\text{LuAlO}_3:\text{Nd}^{3+} + \text{Y}_2\text{SiO}_5:\text{Nd}^{3+}$ laser with CAM; V— $\text{Y}_2\text{SiO}_5:\text{Nd}^{3+}$ laser; VI— $\text{LuAlO}_3:\text{Nd}^{3+} + \text{Nd glass (LGS-2)}$ laser with CAM; VII—Nd glass (LGS-2) laser.

nected with the presence of extraneous impurities in the crystal. In the absence of the yellow filter ZhS-17 surrounding the excitation lamp, the "turning off" of the SE begins to manifest itself at temperatures $\sim 400 \text{ K}$.^[28]

To excite new SE lines and to study the $\lambda_g(T)$ dependences of the lines A and B we used, as noted above, the method of SE spectroscopy using a laser with CAM. As a result we obtained lasing on the line (transition $11393 \text{ cm}^{-1} {}^4F_{3/2} - {}^4I_{11/2}$ 2099 cm^{-1}) at 300 K (see Figs. 3 and 9) and investigated the $\lambda_g(T)$ of line B up to T

$\approx 500 \text{ K}$. Plots of $\lambda_g(T)$ of generation lines A and B are shown in Fig. 11.

Thus, comprehensive spectral and lasing investigations of Nd^{3+} were carried out in a new laser crystal matrix LuAlO_3 in a wide range of temperatures. Spectroscopic information was obtained on the principal physical phenomena that govern the SE effect in this active medium. The orientational transverse cross sections of all the Stark transitions of the Nd^{3+} were determined, for the first time, for laser crystals, for two lasing channels of definite practical importance. The

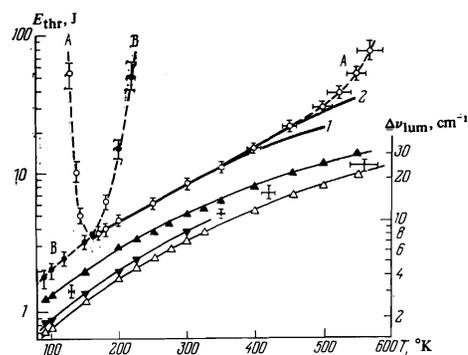


FIG. 10. Plots of $E_{\text{thr}}(T)$ or $\Delta\nu_{\text{lum}}(T)$ of certain emission lines of $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals ($F \approx [112]$). Curves 1 and 2—theoretical plots of $E_{\text{thr}}(T)$, explanation in the text. Plot of $\Delta\nu_{\text{lum}}(T)$: \blacktriangle —line A, \triangle —line Γ , \blacktriangledown line B.

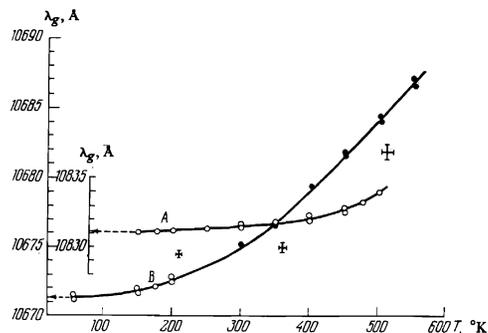


FIG. 11. Plots of $\lambda_g(T)$ for two SE lines of $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals ($F \approx [112]$) obtained in a scheme with an ordinary laser (o) and in a scheme with a laser with CAM (\bullet).

presented theoretical interpretation of the $E_{\text{thr}}(T)$ dependences of lasing lines A and B has revealed the main causes of their temperature dependence. The results offer evidence that LuAlO_3 doped with TR^{3+} can be used as effective media for solid-state lasers.

As shown by our subsequent investigations, among the promising media for "room-temperature" crystal lasers are also compounds produced in the binary systems $\text{Y}_2\text{O}_3\text{-Al}_2\text{O}_3$ and $\text{Lu}_2\text{Al}_3\text{-Al}_2\text{O}_3$ with Ho^{3+} and Er^{3+} ions. This is attested by our following data: at 300 °K, lasing of Ho^{3+} ions was obtained on the ${}^5I_6\text{-}{}^5I_7$ transition in three media: YAlO_3 with $\lambda_g = 29180 \text{ \AA}$ and $E_{\text{thr}} = 7.5 \text{ J}$; $\text{Y}_3\text{Al}_5\text{O}_{12}$ with $\lambda_g = 29403 \text{ \AA}$ and $E_{\text{thr}} = 30 \text{ J}$; $\text{Lu}_3\text{Al}_5\text{O}_{12}$ with $\lambda_g = 29460 \text{ \AA}$ and $E_{\text{thr}} = 60 \text{ J}$. Lasing of the Er^{3+} ions in YAlO_3 took place on three transitions: ${}^4S_{3/2}\text{-}{}^4I_{13/2}$ at wavelengths 8497.5 and 8594 Å with $E_{\text{thr}} \approx 100$ and 65 J; ${}^4S_{3/2}\text{-}{}^4I_{9/2}$ with $\lambda_g = 16632 \text{ \AA}$ and $E_{\text{thr}} = 20 \text{ J}$; ${}^4I_{11/2}\text{-}{}^4I_{13/2}$ with $\lambda_g = 27309 \text{ \AA}$ and $E_{\text{thr}} = 40 \text{ J}$. Similar results were obtained with $\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Er}^{3+}$, namely, ${}^4S_{3/2}\text{-}{}^4I_{13/2}$ with $\lambda_g = 8632.5 \text{ \AA}$ and $E_{\text{thr}} = 45 \text{ J}$; ${}^4S_{3/2}\text{-}{}^4I_{9/2}$ with $\lambda_g = 17762 \text{ \AA}$ and $E_{\text{thr}} = 30 \text{ J}$; ${}^4I_{11/2}\text{-}{}^4I_{13/2}$ with $\lambda_g = 29408 \text{ \AA}$ and $E_{\text{thr}} = 55 \text{ J}$. In the crystal $(\text{Lu}_2\text{Er})\text{Al}_5\text{O}_{12}:\text{Er}^{3+}$ ($\approx 35 \text{ at. \%}$), lasing was observed with $E_{\text{thr}} = 12 \text{ J}$ at a wavelength 23395 Å. The system $\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Ho}^{3+}$ ($\approx 3 \text{ at. \%}$), Tu^{3+} ($\approx 1 \text{ at. \%}$), Er^{3+} ($\approx 9 \text{ at. \%}$) generates with $\lambda_g = 26990 \text{ \AA}$ ($E_{\text{thr}} = 25 \text{ J}$). Finally, at 300 °K with $E_{\text{thr}} = 45 \text{ J}$, SE of Er^{3+} ions in $\text{Y}_3\text{Al}_5\text{O}_{12}$ was observed⁶⁾ at a wavelength 8627 Å (${}^4S_{3/2}\text{-}{}^4I_{13/2}$).

In conclusion, the authors thank corresponding member of the USSR Academy of Sciences P. P. Feofilov for interest in the research and for direction in the source of the first laser experiments with the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystals, Kh. S. Bagdasarov for support, D. N. Vylezhagin and G. A. Bogomolov for a discussion of the results and for help, G. V. Anan'ev for orientation and x-ray structure data, and I. V. Morov and V. I. Pshenitsyn for crystal-optical measurements.

¹⁾Information on the spectral and lasing characteristics of crystals with garnet structure ($O_h^{10}\text{-}Ia3d$), which are produced in $(\text{TR})_2\text{O}_3\text{-Al}_2\text{O}_3$ systems can be obtained from [1-3,9].

²⁾The features of the technology of obtaining $\text{LuAlO}_3:\text{TR}^{3+}$ single crystals and the details of the measurement of the x-ray-structure and physical properties will be described by G. V. Aran'eva, A. O. Ivanov, T. I. Mekulyaeva, and I. V. Molchanov.

³⁾The values of $\langle \|U^{(t)}\| \rangle$ for most transition from the ground states of TR^{3+} ions were calculated and tabulated by Carnall *et al.* [21] for the intermediate-coupling case.

⁴⁾Formula (13) has been written out for the case of a Lorentz contour of the luminescence line.

⁵⁾In order not to clutter up Tables III and IV the values of β_{ij}^{ex} and A_{ij}^{ex} are given only for one orientation ($\|c$) of the $\text{LuAlO}_3:\text{Nd}^{3+}$ crystal.

⁶⁾The investigations with lutecium garnet crystals with rare-earth ions were performed by A. G. Petrosyan and T. I.

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