PROCEDURE FOR THE INVESTIGATION OF AUTO-RESONANT ENERGY TRANSFER IN LASER ACTIVE MEDIA

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We describe two methods that permit a direct study of auto-resonant energy transfer processes in active media characterized by a variety of optical centers under stimulated emission conditions. One method employs a laser with a combined active medium (CAM) or with introduced selective amplification, and the other uses a multiple excess over threshold during lasing. Some spectroscopic and energy characteristics of CAM lasers with the crystals $CaF_2:YF_3:Nd^{3+}$ + $Y_3Al_5O_{12}:Nd^{3+}$ and $CaF_2:YF_3:Nd^{3+}$ + $CaWO_4:Nd^{3+}$ and the glasses $Y_3Al_5O_{12}$ + LGS-1 and $CaW_2O_4:Nd^{3+}$ with LGS-1 and LGS-6. Some new stimulated transitions could be detected in the CAM lasers investigated.

INTRODUCTION

INTEREST in mixed fluoride crystals characterized by a variety of optical centers (OC) has greatly increased recently[1]. On the one hand, they are promising active media for lasers and possess a number of unique properties (for example, high working temperature, constant efficiency in a wide temperature interval[2], high activator concentration), and on the other hand, by virtue of their special physical properties (when activated with TR, the mixed systems occupy with respect to their spectral characteristics a position intermediate between simple crystals and glasses, and at the same time they have a structure of typical crystals[3]) they are interesting objects for different physical investigations. Numerous brands of glasses activated with TR ions, particularly Nd^{3+}, are at the present time, alongside with ruby, the most widely used laser working media. Compared with fluoride mixed systems, they have been more thoroughly investigated[4]. Also included among the active media characterized by a variety of OC are several mixed crystals based on compounds with complex oxygen-containing anions[5] and inorganic liquids[6]. The variety of OC of these media is reflected in their spectral properties also in processes that occur in the stimulated-emission (SE) mode. The most interesting here, in our opinion, is the phenomenon of migration of energy between OC.

Worthy of special attention, in this connection, is auto-resonant transfer[1], i.e., the processes of energy transfer between OC having different structures but containing like ions. The importance of the investigation of such processes is indicated by problems connected with the generation of ultrashort pulses and high monochromaticity, without energy loss in Q-switched lasers, with selection of corresponding co-activators in media used in lasers to obtain high emission energies, etc. However, there are practically no published data[7] reporting studies of this phenomenon. The apparent cause is the lack of an appropriate procedure.

We describe in the present paper two procedures that make it possible to investigate directly, in the stimulated-emission mode, processes of auto-resonant energy transfer in active media characterized by a variety of OC. This is the method of a laser with combined active medium (CAM) or with introduced selective gain, and the method of multiple excess over threshold in the lasing mode.

ACTIVE MEDIA, EXPERIMENTAL APPARATUS, AND RESEARCH METHODS

We used in the experiments simple crystals ($CaWO_4$, $Y_3Al_5O_{12}$) and mixed fluoride systems ($CaF_2:YF_3$, α-$NaCaYF_6$, $LaF_3:SrF_2$) activated with Nd^{3+} ions, and also several brands of commercial glasses of the LGS type[1)]. The experiments were performed with samples in the form of cylindrical rods 25–40 mm long and ~ 5 mm in diameter with plane-parallel ends (~ 10″).

The luminescence and excitation spectra were investigated with a DFS-12 diffraction spectrometer with gratings of 600 lines/mm. The selective excitation source was a quartz monochromator ZMR-3 and a tungsten-iodine lamp type LNR. The lifetime of the excited $^4F_{3/2}$ state of the Nd^{3+} ion was measured by a procedure described in[8]. The SE spectra were photographed on I-1070 film with a DFS-8 spectrograph with dispersion ~ 5.9 Å/mm. The reference was the emission spectrum of a hollow-cathode (Fe) lamp in third order. The microphotometry of the spectra was with a modified MF-4 instrument. The threshold excitation energies E_{thr} for the individual generation lines were measured in analogy with[9]. The output energy of the laser E_g was registered by a spherical calorimeter (diameter 150 mm), the inner surface of which was coated with a diffusely reflecting paint having a constant reflection coefficient in a wide range of wavelengths. The radiation detector in the calorimeter was a thermo-

[1)] The crystals $CaWO_4$, $CaF_2:YF_3$, and $LaF_3:SrF_2$ were synthesized at the Physics Institute of the USSR Academy of Sciences under the guidance of V. V. Osiko, and the crystals $Y_3Al_5O_{12}$ and α-$NaCaYF_6$ at the Institute of Crystallography of the USSR Academy of Sciences under the guidance of Kh. S. Bagdasarov.

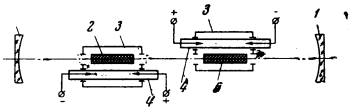

FIG. 1. Block diagram of lasers with CAM: 1–spherical mirrors, 2–active medium with variety of OC, 3–illuminating chambers of elliptic cross section, 4–type IFP-400 flash lamps, 5–simple crystal.

pile with a multirange high-resistance microvoltmeter F-116/2. The threshold sensitivity of calorimeter was 10^{-3} J [2].

Method of multiple excess over threshold. This method is based on the investigation of the distribution of the energy over the generation spectrum lines at different levels of the excitation energy E_{exc}. The method can be used to study active media that characterized by a variety of OC and are capable of emitting at least two lines within the limits of one transition. Such properties are possessed so far by four mixed fluoride crystals activated with Nd^{3+}, namely $CaF_2:YF_3$ [1], α-$NaCaYF_6$ [10], $CaF_2:SrF_2:BaF_2:YF_3:LaF_3$ [11] and $LaF_3:SrF_2$ [12]. Their emission lines are connected with the transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and lie in the wavelength range near 1.06 μ.

We used an ordinary laser consisting of a cylindrical illuminating system of elliptical cross section with an optical efficiency ~ 0.5 [10], an IFP-400 xenon flash lamp and an optical resonator made up of two dielectric-coated spherical mirrors (R = 550 mm) mounted confocally. The transmission of the output mirror at $\lambda = 1.06$ μ was $\sim 3\%$.

The experiment was performed as follows. We first measured E_{thr} of each generation line, i.e., we determined the minimum pump energy at which a given to crystal begin to emit. We then photographed its SE spectrum $E_{exc} = mE_{thr}$. Before each generation act, the crystal temperature was 300°K. The energy distribution among the lines of the SE spectrum was determined from the density of the negative [13]. Because the energy (photon density) per generation line was proportional not only to the density at its center but also to its width $\Delta\nu_g$, we obtained the "integral density" over the entire line. We then plotted the percentage composition Π of the generation energy against mE_{thr}.

Method using a laser with combined active medium [14]. Figure 1 shows a block diagram of a CAM laser. We see that it consists of two illuminating chambers 3 with excitation sources 4 and a common optical resonator 1. Unlike an ordinary laser, its active medium is a combination of two or more substances, one of which is characterized by a variety of OC and is under investigation. The second medium should be able to generate narrow lines. Such media include simple crystals activated with TR [3]. We are interested in crystals containing only Nd^{3+} ions and capable of lasing at 300°K. The list of such media is now small—this is $Y_3Al_5O_{12}$ (10641 Å), $CaWO_4$ (10582 and 10652 Å), CaF_2 type I (10461 and 10630 Å), CaF_2 type II (10885 Å), SrF_2 type I (10371 and 10446 Å), Gd_2O_3 (10741 and 10789 Å), $SrWO_4$ (10630 Å), $CaMoO_4$ (10610 and 10673 Å) and $SrMoO_4$ (10576 and 10645 Å) [1], LaF_3 (10407 and 10633 Å) and CeF_3 (10410 and 10638 Å) [15], and YVO_4 (10641 and 10664 Å) [16] and $LiNbO_3$ (10846 Å) [17].

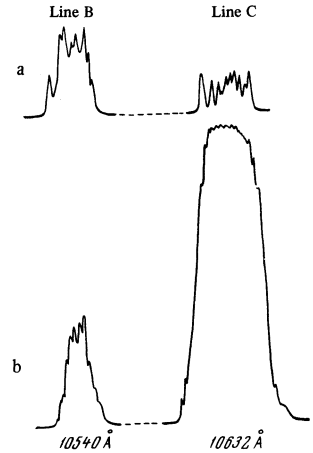

FIG. 2. Microphotograms of SE spectra of $CaF_2:YF_3$ (5%): Nd^{3+} (~2%) crystal at 300°K: a–$E_{exc} = 2E_{thr}$ (B), b–$E_{exc} = 20E_{thr}$ (B). The spectra were obtained from single generation pulses.

Inasmuch as CAM lasers, unlike ordinary lasers, have a controllable introduced selective gain determined by the spectroscopic properties of the simple crystals, we can excite at will in the mixed crystal or glass, over a wide frequency range (over the luminescent spectrum of the active medium with variety of OC), sections of the luminescence spectrum that coincide with the generation wavelengths λ_g or with the intense luminescence lines of the simple crystals. By performing spectroscopic, energy, temperature, and time-dependent investigations of the SE in different regimes (free generation, Q-switched generation) of such a laser as a function of the compositions of the media and the activator concentrations, we can obtain information on the kinetics of the processes that occur in active media with a variety of OC. The same pertains to the method of multiple excess over threshold.

We present below certain results of an investigation of the auto-resonant energy transfer in the crystals and glasses mentioned at the beginning of the present section. In our experiments, the CAM laser consisted of two identical illuminating chambers with optical efficiency ~ 0.5 with IFP-400 lamps. The spherical mirrors of the optical resonator (R = 550 mm) were placed 650 mm apart.

EXPERIMENTAL RESULTS

Method of multiple excess over threshold. Figure 2 shows microphotograms of the generation spectra (T = 300°K) of the crystal $CaF_2:YF_3(5\%):Nd^{3+}(2\%)$ [4] at E_{exc} equal to $2E_{thr}$(B) and $20E_{thr}$(C). We see that in the case of a large over-excitation a redistribution of the generation energy among the lines takes place. Figure 3 shows plots of $\Pi(n = E_{exc}/E_{thr})$ for two emission lines of yttrofluorite crystals, B (10540 Å) and C (10632 Å), at different concentrations of YF_3 and of the Nd^{3+} ions. We see that in all cases $E_{thr}(B) < E < E_{thr}(C)$. The difference in the values of E_{thr} increa-

[2] The author takes the opportunity to thank V. N. Gardash'yan and V. V. D'yachenko for supplying the calorimeter for the experiments.

[3] It is possible to use also other substances, for example gases.

[4] The concentration here and throughout is indicated in per cent by weight.

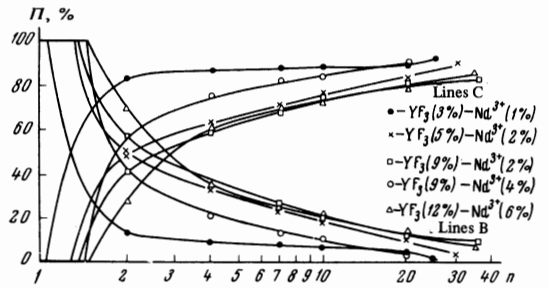

FIG. 3. Percentage distribution of the output energy over the generation lines as a function of the coefficient of over-excitation $n = E_{exc}/E_{thr}$ of yttrofluorite crystals.

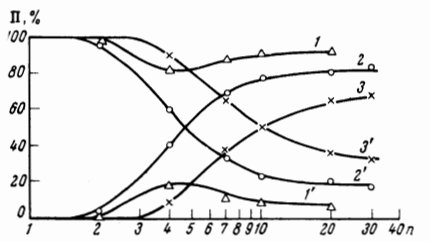

FIG. 4. Plot of $\Pi(n = E_{exc}/E_{thr})$ of hexagonal LaF3: SrF_2 (4%) + Nd^{3+} (~2%) crystal (curve 1) and α-$NaCaYF_6$:Nd^{3+} (curves 2 and 3 correspond to neodymium concentrations 1% and 4% respectively). The plots 1′–3′ pertain to the short-wave components of the generation spectra.

ses with increasing total concentration of the ions Y^{3+} and Nd^{3+}. Thus, at YF_3 and NdF_3 contents of 3 and 1%, respectively, we get $E_{thr}(C) \approx 1.05\, E_{thr}(B)$, and for crystals with respective concentrations 12 and 6% we have $E_{thr}(C) \approx 1.5\, E_{thr}(B)$. Nonetheless, with increasing E_{thr} the generation energy is radically redistributed towards the long-wave line C. As seen from Fig. 3, at $E_{exc} = 10E_{thr}$ practically the entire energy is in the line C.

Figure 4 shows analogous relations for the crystals α-$NaCaYF_6$ and LaF_3:SrF_2 with Nd^{3+} ions. Even in spite of the fact that $E_{thr}(C) \approx 3E_{thr}(B)$, the character of the generation-energy redistribution in the α-gagarinite crystals is close to the case considered above, i.e., the generation energy in the line C (~ 10628 Å) at $E_{exc} = 10E_{thr}$ prevails over the generation energy in the B line (~ 10541 Å). The Π(n) plot for the SE lines of the hexagonal crystal LaF_3:SrF_2 (4%) : Nd^{3+}(2%) behaves somewhat differently (see Fig. 4). This is due to the fact that E_{thr} of the long-wave line (~ 10635 Å) is half as large (~ 5.5 J) than that of the short-wave line (~ 10486 Å). The accuracy of the energy measurements in our experiments was 10–15%.

Method of laser with combined active medium. We investigated five pairs of active substances: CaF_2: YF_3 (9%) : Nd^{3+}(4%) + $Y_3Al_5O_{12}$: Nd^{3+}(1%), CaF_2: YF_3 (9%) : Nd^{3+}(4%) + $CaWO_4$: Nd^{3+}(~ 2%)[5], LGS-1 + $Y_3Al_5O_{12}$: Nd^{3+}(1%), LGS-1 + $CaWO_4$: Nd^{3+}(~ 2%), and LGS-6 + $CaWO_4$: Nd^{3+}(~ 2%). Figure 5 shows the luminescence spectra (transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$) and the diagrams of the arbitrarily separated energy levels of the Nd^{3+} ions in the CaF_2: YF_3 and $Y_3Al_5O_{12}$ crystals. The yttrofluorite is characterized by a variety of OC, while the garnet contains essentially one type of center, which determines its lasing parameter[18]. Whereas for $Y_3Al_5O_{12}$: Nd^{3+} we know the positions of the Stark components of the $^4F_{3/2}$ and $^4I_{11/2}$ terms, between which stimulated transitions can occur, it is practically impossible to construct such a level scheme for each OC in the case of CaF_2: YF_3: Nd^{3+}. The luminescence spectrum corresponding to the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition in the $Y_3Al_5O_{12}$: Nd^{3+} crystal has a distinct Stark structure, and that in yttrofluorite has an inhomogeneously broadened band constituting a superposition of the spectra of different centers. The generation spectrum of a garnet in an ordinary laser at 300°K contains one narrow line with $\Delta\nu_g = 0.9\ cm^{-1}$ at λ = 10641 Å, and in yttrofluorite it consists of two broad bands B and C (see above).

The condition for the onset of generation in a CAM laser at a frequency $h\nu_i$ can be written[14] in the form

$$\exp\{-2[(\alpha_{abs}^1 - \alpha_{amp}^1)l_1 + (\alpha_{abs}^2 - \alpha_{amp}^2)l_2]_{h\nu_i}\}R_1R_2 \approx 1,$$

where α_{abs}^1 and α_{abs}^2 are the absorption coefficient per unit length of the active media of the simple and mixed types, α_{amp}^1 and α_{amp}^2 are the corresponding amplification coefficients, l_1 and l_2 are the crystal lengths, and R_1 and R_2 are the reflection coefficients of the mirrors of the optical resonator at the frequency $h\nu_i$. We see that the emission is produced first at that frequency at which the effective amplification coefficient is the largest, provided, of course, the losses are the same in the frequency interval of their luminescence spectra.

The construction of the CAM laser has made it possible to study the lasing parameters of the crystals under both separate and simultaneous excitation. In all cases, both investigated active media were in the optical resonator. In the case of separate excitation of the $Y_3Al_5O_{12}$: Nd^{3+} in the CAM laser, its threshold energy was $E_{thr} = 11 \pm 0.3$ J, and in the case of the CaF_2: YF_3: Nd^{3+} it was approximately 23 J. The generation spectrum of the garnet, in the case of separate pumping at an energy of $55E_{thr}$ (~ 480 J) fed to the IFP-400 lamps (which were connected in series), consists of one line. It is shown in Fig. 6 together with the

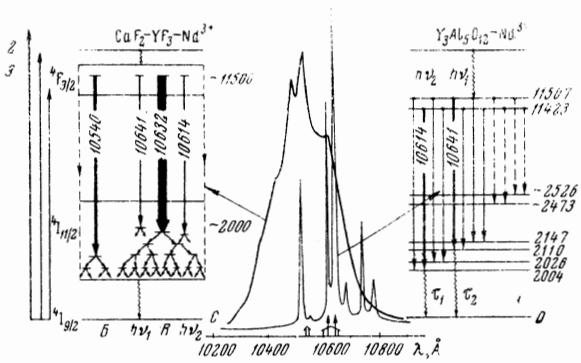

FIG. 5. Luminescence spectra of the crystals CaF_2 : YF_3 (9%): Nd^{3+} (4%) and $Y_3Al_5O_{12}$: Nd^{3+} (1%) at 300°K, transition $^4F_{3/2} \rightarrow {}^4I_{11/}$, and diagrams of conditionally separated levels. The level positions are indicated in cm^{-1}, and those of the transitions between them in Å. The thick arrows denote stimulated transitions and the wavy lines non radiative ones.

[5] The optical axis c coincides with the geometrical axis of the crystal.

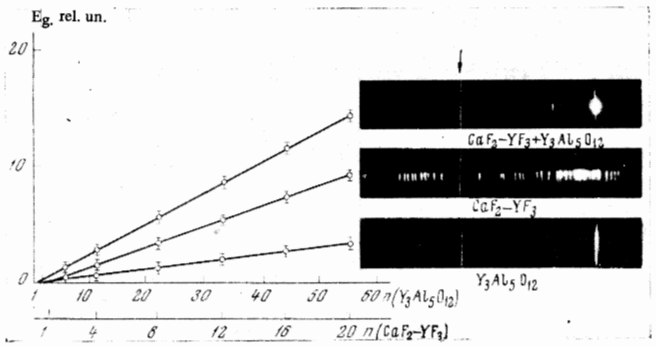

FIG. 6. Plot of $E_g(E_{exc})$ and generation spectra of $Y_3Al_5O_{12}$, $CaF_2:YF_3$, and $CaF_2:YF_3 + Y_3Al_5O_{12}$. The arrow denotes the reference line.

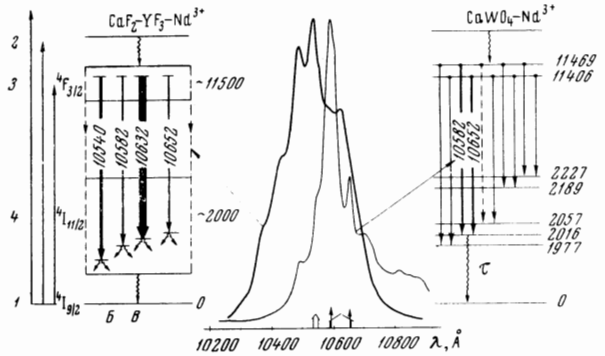

FIG. 7. Luminescence spectra of crystals $CaF_2:YF_3$ (9%):Nd^{3+} (4%) and $CaWO_4:Nd^{3+}$ (~2%) at 300°K, transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$; diagrams of conditionally separated levels. The notation is the same as in Fig. 5.

$E_g(E_{exc})$ plot. The same figure shows the SE spectrum of yttrofluorite, also obtained with separate excitation. Both were photographed at the same excitation energy. The latter, as can be readily seen, consists of two broad lines. Its $E_g(E_{exc})$ plot is sharper, since its efficiency is higher. It must be noted here that all the presented values of E_{thr} and E_{exc} correspond to the case when one of the lamp "idles." Such an excitation was technically realized by placing in the illuminating chamber a metallic shutter between the crystal and the lamp. If the $Y_3Al_5O_{12}:Nd^{3+}$ and $CaF_2:YF_3:Nd^{3+}$ are excited simultaneously, then this combined medium begins to generate at the garnet line $h\nu_1$ (10641 Å), with $E_{thr} = 8.7 \pm 7.3$ J. When E_{exc} is further increased to a value corresponding to E_{thr} ($CaF_2:YF_2$), no generation occurs at the yttrofluorite lines B and C. However, the results of the measurements of E_g have shown that the yttrofluorite takes part in the stimulated emission and generates at the $h\nu_1$ line of the garnet. The SE spectrum of the crystals $CaF_2:YF_3:Nd^{3+}$ and $Y_3Al_5O_{12}:Nd^{3+}$ when the two are simultaneously excited is also shown in Fig. 6, alongside the $E_g(E_{exc})$ plot. We see that besides the line $h\nu_1$ (10641 Å), a new narrow line $h\nu_2$ was produced with wavelength 10614 Å and $E_{thr} \approx 40$ J; this line is missing from the generation spectra of $Y_3Al_5O_{12}:Nd^{3+}$ and $CaF_2:YF_3:Nd^{3+}$ at 300°K. If we plot the total dependence of $E_g(E_{exc})CaF_2:YF_3 + E_g(E_{exc})\ Y_3Al_5O_{12}$, then it will not coincide with the obtained plot of $E_g(E_{exc})\ CaF_2:YF_3 + Y_3Al_5O_{12}$.

Figures 7 and 8 show the results of analogous investigations of CAM lasers on $CaF_2:YF_3:Nd^{3+}$ and $CaWO_4:Nd^{3+}$. Here, in the case of separate excitation, the value of $E_{thr}(CaWO_4)$ of the line with $\lambda = 10582$ Å is 6.7 ± 0.3 J. Simultaneous pumping of these media leads to a decrease of the threshold at this wavelength to 5.9 ± 0.3 J. It is seen from the generation spectrum that in this case there also arises a "new" line with $\lambda = 10652$ Å and $E_{thr} \approx 15$ J. The quotation marks emphasize here that this line is actually new for a $CaWO_4:Nd^{3+}$ crystal with c axis oriented parallel to the geometric axis F. The line with $\lambda = 10652$ Å is usually present in the emission spectrum of scheelite when $c \perp F$ or when c makes an appreciable angle with F. Just as in the preceding case, $E_g(E_{exc})$ with simultaneous pumping of the crystals is larger than $E_g(E_{exc})\ CaF_2:YN_3 + E_g(E_{exc})\ CaWO_4$.

Before we proceed to other media, we note that the lines B and C of yttrofluorite are not excited in CAM lasers based on $CaF_2:YF_3$, $Y_3Al_5O_{12}$, and $CaWO_4$ at limiting values of E_{exc} larger by 2–3 times than those indicated in Figs. 6 and 8.

Figures 9–12 demonstrate the results of an investigation of lasers with CAM based on the glasses LGS-1 and LGS-6 in conjunction with $CaWO_4$-Nd^{3+}, and also LGS-1 with $Y_3Al_5O_{12}:Nd^{3+}$. For the system LGS-1 + $CaWO_4:Nd^{3+}$ with separate excitation we have

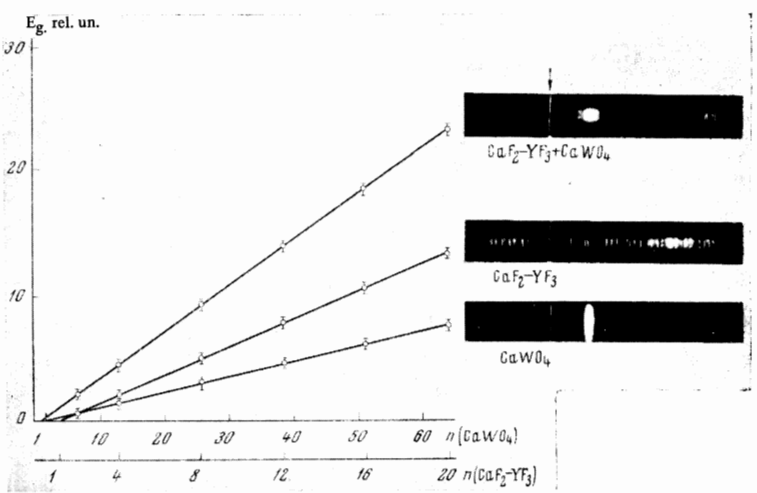

FIG. 8. Plots of $E_g(E_{exc})$ and SE spectra of the crystals $CaF_2:YF_3$ (9%): Nd^{3+} (4%) and $CaWO_4$ Nd^{3+} (~2%).

Combined medium	λ_g, μ	Threshold energies in separate excitation, J.					Threshold energies in simultaneous excitation in a laser with CAM, J.				
		CaF_2-YF_3	$Y_3Al_5O_{12}$	$CaWO_4$	LGS-1	LGS-6	CaF_2-YF_3 + $Y_3Al_5O_{12}$	CaF_2-YF_3 + $CaWO_4$	LGS-1 + $Y_3Al_5O_{12}$	LGS-1 + $CaWO_4$	LGS-6 + $CaWO_4$
$Y_3Al_5O_{12}$ +	1.0641		11				8.7				
	1.0614		—				40				
$+CaF_2-YF_3$	C 1.0540	33					—				
	B 1.0632	23					—				
$CaWO_4$ +	1.0582			6.7				5.9			
	1.0652			—				15			
$+CaF_2-YF_3$	C 1.0540	27						—			
	B 1.0632	19						—			
$Y_3Al_5O_{12}$ +	1.0641	10.4							7.5		
	1.0614	—							15		
+ LGS-1	1.06				17.5				70		
$CaWO_4$ +	1.0582		12								11
	1.0652		—								200
+ LGS-6	1.06					36					—
$CaWO_4$ +	1.0582		10							8	
	1.0652		—							75	
+ LGS-1	1.06				25						—

E_{thr}(LGS-1) = 25 J and $E_{thr}(CaWO_4)$ = 10 ± 0.3 J (λ = 10582 Å). In the case of the system LGS-6 + $CaWO_4$: Nd^{3+}, the values are E_{thr}(LGS-6) = 36 J and $E_{thr}(CaWO_4)$ = 12 ± 0.3 J (10582 Å). With simultaneous pumping, E_{thr}(LGS-1 + $CaWO_4$) for the line with λ = 10582 Å decreases to 8 ± 0.3 J, and for the combination LGS-6 + $CaWO_4$ it decreases to 11 ± 0.3 J. In the spectra of the considered active media, there also appears the aforementioned "new" line with λ = 10652 Å. For the first combination, its threshold was ~ 75 J, and for the second ~ 200 J.

In the CAM laser based on LGS-1 and $Y_3Al_5O_{12}$, the radiation of the LGS-1 glass with its characteristic broad line with λ = 1.06 μ is no longer registered. This is clearly seen from the spectrum of Fig. 12. In the case of separate excitation, the values are E_{thr}(LGS-1) = 17.5 J and $E_{thr}(Y_3Al_5O_{12})$ = 10.4 ± 0.3 J (λ = 10641 Å). In the case of simultaneous pumping of these media, E_{thr} of the 10641 Å line is already equal to 7.5 ± 0.3 J, and that of the second line with λ = 10614 Å, which also is produced in this case, is 15 J, while that of the generation line of glass is ~ 70 J. The threshold excitation energies of all the investigated CAM lasers are summarized in the table.

Besides these data, we point out one more circumstance: the centers of the generation lines of the LGS glasses do not coincide with the maxima of their luminescence bands. This is seen from Figs. 9–12. The discrepancy reaches 25–30 cm^{-1}. This holds also for KGSS glasses, which were not investigated in the present study. For yttrofluorite and other mixed systems, the SE also occurs most frequently at the long-wave maxima of the luminescence bands, which are not always the most intense in the spectra[2]. Thus, a general regularity is observed in active media characterized by a variety of OC, namely the tendency of shifting the generation wavelength towards the long-wave side relative to the center of the luminescence band.

DISCUSSION OF RESULTS

The existence of the process of migration of energy between different centers with like ions in active media characterized by a variety of OC was indirectly revealed on the basis of the results of numerous spectroscopic and generation investigations. The occurrence of auto-resonant cross-relaxation phenomena in activated glasses was discussed apparently for the first time in[19,20]. The existence of this phenomenon in mixed fluorite crystals was observed on the basis of the results of the very first experiments on SE[3,21]. This followed from the facts that lasers based on mixed fluorite crystals

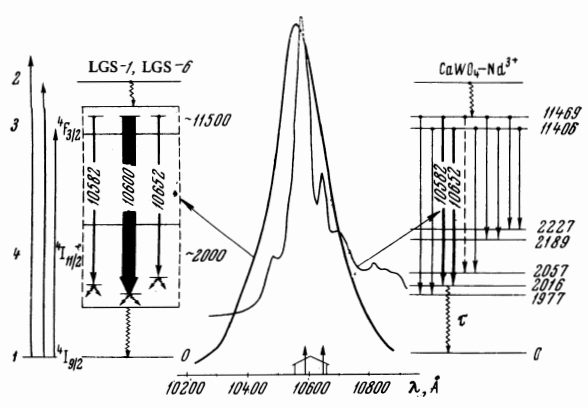

FIG. 9. Luminescence spectra of glasses LGS-1 and LGS-6 and of the crystal $CaWO_4$: Nd^{3+} (~2%) at 300°K, transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$, diagrams of conditionally separated levels. Notation the same as in Fig. 5.

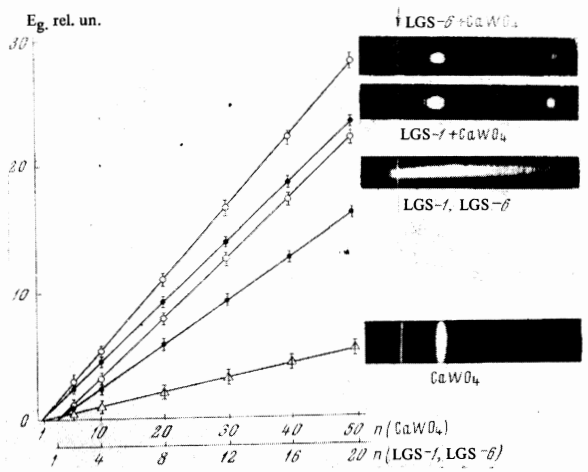

FIG. 10. Plots of $E_g(E_{exc})$ and of SE spectra of glasses LGS-1 and LGS-6 and of the crystal $CaWO_4$: Nd (~2%).

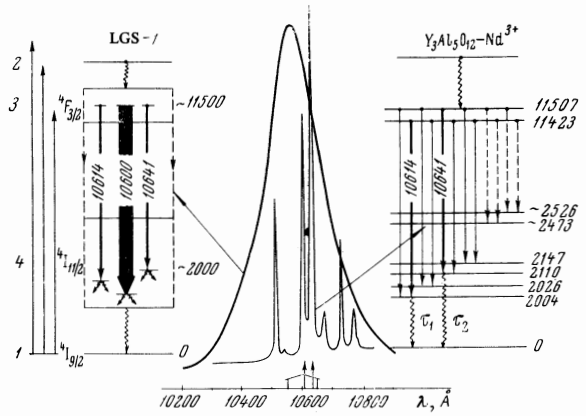

FIG. 11. Luminescence spectra of glass LGS-1 and of the crystal $Y_3Al_5O_{12}$:Nd^{3+} (1%) at 300°K, transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$; diagrams of conditionally separated levels. Notation the same as in Fig. 5.

generate appreciable energies in sufficiently narrow lines, several cm^{-1} wide, with low E_{thr} at very wide bands in the luminescence spectra, reaching several hundredths of cm^{-1}. Further experiments on the characteristics of generation in processes occurring in the excited state in a laser based on crystals with disordered structure under different regimes and conditions[2,22] have additionally confirmed that such acts of energy transfer play an important role in such media. By now we know of more than 15 mixed crystals (solid solutions) that can operate successfully in various types of lasers (see the reviews[1] and the bibliography in[2]). An estimate of the time of energy transfer between OC in mixed crystals and glasses, in accordance with the data of[4,7,19,29,22], leads to values from 10^{-4} to 10^{-8} sec.

An analysis of the foregoing results, and also of data obtained in similar investigations of other media, has shown that in each substance there exists such OC or groups of centers, between which the probability of auto-resonant transfer varies in a wide range. The lower limit, i.e., the longest time, is apparently close to the value of the radiative lifetime of the metastable term $^4F_{3/2}$ (for Nd^{3+} ions), and the shortest can be less than 10^{-8} sec, as indicated by experiments with lasers operating in the Q-switching regime[20,22]. The relative weight of the OC capable of exchanging energy within a definite time can be revealed with the aid of the considered method using a CAM laser, by selectively choosing the excitation along the luminescence line contour and measuring the energy in the individual generation lines with varying Q-switching time. At the present time, the production of SE pulses of duration 10^{-10}–10^{-13} sec, is quite realistic[23]. Such investigations make it possible to choose the optimal regime and lasing wavelength within the luminescence-line contour. Our experiments have shown that in this respect the yttrofluorite crystals have better parameters than LGS-1 and LGS-6 glasses, i.e., they can emit shorter pulses without an appreciable loss of efficiency.

Since the purpose of the present investigation was not a detailed study of the probabilities of auto-resonant transfer in mixed crystals and glasses, we stop to discuss only general questions.

The results reduce briefly to the following: 1) we have established by a direct method the existence of energy migration in mixed crystals and glasses activated with Nd^{3+}. 2) We have established that if an active medium characterized by a variety of OC is capable of generating at several lines within the limits of one transition, then at larger E_{exc} a redistribution of the radiation energy takes place in the direction of the long-wave component of the spectrum. 3) A long-wave shift of the centers of the SE lines relative to the maxima of the luminescence bands has been observed in glasses LGS and KGSS with activated Nd^{3+} ions. 4) It has been observed that in lasers with CAM the generation occurring in the lines of simple crystals stimulates the SE media characterized by a variety of OC, and the optical centers of these media, taking part in the emission in the ordinary-laser scheme, can be made to generate at the lines of the simple crystals. 5) We have observed in CAM lasers the occurrence of new stimulated transitions, which are not observed in ordinary lasers based on separate combining media. 6) We have established that the generation energy of a laser with CAM is larger than the sum of the generation energies in the case of separate excitation of the individual media under the same conditions. The slight increase in the efficiency is apparently connected with the decrease of the energy loss to luminescence processes preceding the SE.

The totality of the obtained data gives grounds for suggesting the possible existence in the active media with a variety of OC of two mechanisms of auto-resonant energy transfer. Figure 13 shows the arbitrarily separated energy levels of the terms $^4F_{3/2}$ and $^4I_{11/2}$. Let us consider these two possible variants as applied to the

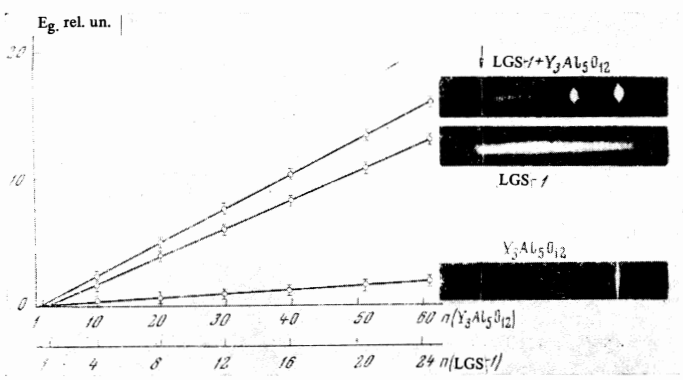

FIG. 12. Plots of $E_g(E_{exc})$ and SE spectra of LGS:1 glass and of crystal $Y_3Al_5O_{12}$:Nd^{3+}.

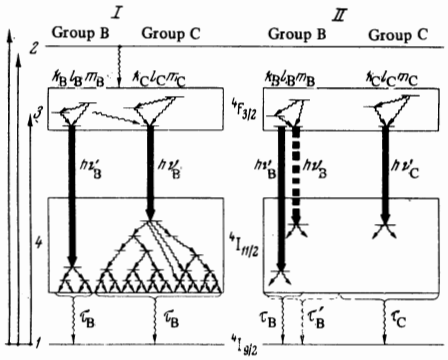

FIG. 13. Conditional schemes of the terms $^4F_{3/2}$ and $^4I_{11/2}$ of the Nd^{3+} ions in yttrofluorite crystals, with separated levels of certain centers of groups B and C under two assumptions. The thick arrows denote induced transitions, the wavy arrows denote nonradiative transitions.

results obtained by the method of many fold excess above threshold.

Variant I. An investigation of the dependences of E_{thr} of lines B and C of yttrofluorite crystals on the Nd^{3+} ion concentration has shown that these stimulated transitions belong to different groups of OC. In Fig. 13 we have separated these groups with levels k, l, and m. In connection with the fact that $E_{thr}(B) < E_{thr}(C)$ lasing at the 10540 Å line sets in at small E_{exc} and is connected, say, with center l_B, since it has the best conditions for the occurrence of SE in this group of OC. With increasing E_{exc}, the centers of group C enter into the lasing process. Up to a value $E_{exc} = (1.5-3)\,E_{thr}(B)$, these groups of centers apparently emit independently, as indicated by the linear increase of the values of their E_g. But when E_{exc} exceeds the indicated values, the output energy begins to become redistributed in the direction of group C. Such a redistribution can be attributed to the appearance of "burning out of the inversion" or to the occurrence of a phonon bottleneck due to the long time τ_B of the final levels.

We investigated the radiative lifetime of the metastable term $^4F_{3/2}$ at different frequencies of the contours of the luminescence lines corresponding to the transitions $^4F_{3/2} \rightarrow {}^4I_{9/2-13/2}$. As expected, these times were the same in all cases. We also investigated the luminescence line contours corresponding to the transitions $^4F_{3/2} \rightarrow {}^4I_{9/2-11/2}$ in the case of selective excitation in several absorption bands lying above 9000 cm^{-1}. The luminescence line contours always remained unchanged. Analogous results were obtained by studying different brands of glasses activated with Nd^{3+} ions, in [19]. All this gives grounds for concluding that the resonantly coupled levels of the $^4I_{11/2}$ term of the Nd^{3+} ions in such media are characterized by one nonradiative lifetime. Then the occurrence of the phonon bottleneck at the frequency of the lines B will differ from the ordinary case when the levels of the $^4I_{11/2}$ term do not overlap (for example, for simple crystals).

We propose that the following processes take place here. In the lasing regime, the depletion of the final levels of the centers of group B proceeds with lower levels of other centers taking part. Obviously, in the case of emission of photons of higher energy, a smaller number of levels will be affected. In our case (see Fig. 13), the energy is transferred to four centers. In other words, with the same time of nonradiative relaxation, the effective time will be one-fourth as long. But there may not be enough of these nonradiative-relaxation channels, and the effect of the phonon bottleneck may appear. On the other hand, in the case of radiation from centers C, this channel is already much broader, and the conditions for the occurrence of the phonon bottleneck are no longer produced, since $\tau_C \ll \tau_B$. The realization of such processes in lasing can explain to some degree the tendency of the generation lines of active media with a variety of OC to shift towards the long-wave side.

The possible appearance of the SE channel with the considered properties superimposes a condition on the time of auto-resonant energy transfer of the excitation between the OC. At high E_{exc}, the widths of the generation lines B and C, as shown by our experiments, increase insignificantly; this fact indicates that new centers are not "turned on" in the presence of generation in the $CaF_2:YF_2:Nd^{3+}$ crystals, and the centers that already started to radiate are "supplied" with energy stored by other centers. This mechanism calls for satisfaction of the ordinary conditions for the existence of auto-resonant transfer—coincidence of the transitions (plus and minus phonon additions).

Variant II. At small E_{exc}, emission at the line B begins just as in the preceding case. But here the field of the SE of the line C, which occurs at a higher pumping level, stimulates the generation of centers l_B at a frequency $h\nu_B$. This can take place with another like center l_B, lying above the energy level of the term $^4I_{11/2}$, or of another OC of group B, which is auto-resonantly coupled with the center l_B. The occurrence of generation of the centers l_B at the frequency $h\nu_C$ is impossible under ordinary conditions, since the probability of SE at the frequency $h\nu_C$ is larger. But if an "external" SE field appears at a frequency $h\nu_C$, then a jump across the generation channel is already possible. This circumstance can also be contributed to by the occurrence of the phonon bottleneck as a result of the large time τ_B. When generation switches over to the frequency $h\nu_C$, just as in the first case, the effective time is $\tau'_B \ll \tau_B$. We see that this mechanism does not call for satisfaction of the condition for the interaction between the OC, and the coupling factor in this case is the field of the SE itself. The real existence of such a process is demonstrated by the presence of an entire series of CAM lasers [14].

Thus, in the first case, a role is played in the process of stimulated emission only by acts of auto-resonant energy transfer both for "feeding" the generating centers and in processes of nonradiative relaxation from the final levels of the $^4I_{11/2}$ term. The second case is characterized by the fact that the produced initial field of the SE stimulates the generation of other OC at its own wavelength. In this case, even if auto-resonant transfer processes do occur during the radiation time, their role is not primary.

We note also that the method of multiple excess over threshold can be used to investigate auto-resonant migration of energy also in the case of active media with a variety of OC, which have one line in the SE spectrum (for example, glasses). It is possible to use for

this purpose a disperse optic resonator, for example one with a diffraction grating[24] or with prisms[25] tuned to two or more wavelengths within the luminescence band.

A few words concerning lasers with CAM. Results of their investigations have shown that they have low E_{thr} and narrow generation lines, qualities which they have acquired from the simple crystals, as well as a sufficiently high efficiency, which they acquired from the active media with a variety of OC. If we compare these qualities with the parameters of ordinary lasers, then we see an increased efficiency compared with lasers based on simple crystals, and an increase of spectral brightness and a lowering of E_{thr} compared with lasers based on mixed-type media or glasses[14].

CONCLUSION

1. Two procedures were described for direct investigations of the processes of energy migration in active media characterized by a variety of optical centers, in the stimulated-emission regime—the method of a laser with combined active medium, and the method of multiple excess over threshold.

2. The results of the investigations of the crystals $CaF_2:YF_3:Nd^{3+}$, α-$NaCaYF_6:Nd^{3+}$, and $LaF_3:SrF_2:Nd^{3+}$ by the method of multiple excess over threshold in ordinary lasers, and of CaF_2-$YF_3:Nd^{3+}$ and glasses of LGS type in conjunction with $Y_3Al_5O_5$-Nd^3 and $CaWO_4$-Nd^{3+} crystals, in a laser with CAM have made it possible to propose the existence of two mechanisms of auto-resonant migration of energy in media characterized by a variety of optical centers.

3. At high E_{exc} in lasers based on glasses of type LGS and KGSS, a long-wave shift of the center of the generation line was observed relative to the maximum of the luminescence band.

4. We have described several energy and spectral parameters of lasers with CAM based on yttrofluorite, and glasses of LGS type with garnet and scheelite crystals. The presented experimental results offer evidence that lasers with CAM differ from ordinary lasers in that they have controllable selective gain in a wide frequency characteristic at fixed parameters of the optical resonator. In connection with the fact that lasers with CAM have low threshold excitation energies and narrow generation lines, and also a sufficiently high efficiency, they can also be used independently.

5. The study of CAM lasers has revealed new induced transitions that do not appear in ordinary lasers based on separate combining media. These are the lines with λ = 10614 Å of the crystal $Y_3Al_5O_{12}:Nd^{3+}$ and with λ = 10652 Å of the crystal $CaWO_4:Nd^{3+}$.

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[1] A. A. Kaminskiĭ and V. V. Osiko, Izv. AN SSSR, ser. Neorgan. materialy 1, 2049 (1965) and 3, 417 (1967).

[2] A. A. Kaminskiĭ, Zh. Eksp. Teor. Fiz. 54, 727 (1968) [Sov. Phys.-JETP 27, 368 (1968)].

[3] Yu. K. Voron'ko, A. A. Kaminskiĭ, V. V. Osiko, and A. M. Prokhorov, Izv. AN SSSR, ser. Neorgan. materialy 2, 1161 (1966).

[4] E. Snitzer, Appl. Optics 5, 1487 (1966); G. O. Karapetyan and A. L. Reĭshakhrit, Izv. AN SSSR ser. Neorgan. Materialy 3, 217 (1967).

[5] G. E. Peterson and P. M. Bridenbaugh, Appl. Phys. Lett. 4, 173 (1964); G. M. Zverev and G. Ya. Kolodnyĭ, Zh. Eksp. Teor. Fiz. 52, 337 (1967) [Sov. Phys.-JETP 25, 217 (1967)]; A. M. Morozov, P. P. Feofilov, M. N. Tolstoĭ, and V. N. Shapovalov, Opt. Spektr. 22, 414 (1967).

[6] A. Lampicki and A. Heller, Appl. Phys. Lett. 9, 108 (1966).

[7] V. S. Mashkevich and M. S. Soskin, ZhETF Pis. Red. 5, 456 (1967) [JETP Lett. 5, 369 (1967)]. N. S. Belokrinitskiĭ, A. D. Manuil'skiĭ, and M. S. Soskin, Ukr. fiz. zh. 12, 1720 (1967).

[8] A. A. Kaminskiĭ, L. S. Kornienko, and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. 48, 1262 (1965) [Sov. Phys.-JETP 21, 844 (1965)].

[9] A. A. Kaminskiĭ, L. S. Kornienko, and A. M. Prokhorov, ibid. 48, 476 (1965) [21, 318 (1965)].

[10] Kh. S. Bagdasarov, A. A. Kaminskiĭ, Ya. E. Lapsker, and B. P. Sobolev, ZhETF Pis. Red. 5, 220 (1967) [JETP Lett. 5, 175 (1967)].

[11] Yu. K. Voron'ko, A. A. Kaminskiĭ, and V. V. Osiko, Kristallografiya 13, 332 (1968) [Sov. Phys. Crystallogr. 13, 267 (1968)].

[12] M. V. Dmitruk, A. A. Kaminskiĭ, V. V. Osiko, and T. A. Tevonecjan, Phys. Stat. Sol. 25, 725 (1968).

[13] I. M. Kustanovich, Spektral'nyĭ analiz (Spectral Analysis), Vysshaya shkola, 1962.

[14] A. A. Kaminskiĭ, ZhETF Pis. Red. 7, 260 (1968) [JETP Lett. 7, 201 (1968)].

[15] M. V. Dmitruk, A. A. Kaminskiĭ, Zh. Eksp. Teor. Fiz. 53, 874 (1967) [Sov. Phys.-JETP 26, 531 (1968)]. M. V. Dmitruk, A. A. Kaminskiĭ, and I. A. Shcherbakov, ibid, 54, 1680 ((968) [this issue, p. 900].

[16] Kh. S. Bagdasarov, A. A. Kaminskiĭ, V. S. Krylov, and V. I. Popov, Phys. Stat. Sol. 27, 2 (1968).

[17] N. F. Evlanova, A. S. Kovalev, V. A. Koptsik, L. S. Kornienko, A. M. Prokhorov, and L. N. Rashkovich, ZhETF Pis. Red. 5, 351 (1966) [JETP Lett. 5, 291 (1966)].

[18] A. A. Kaminskiĭ, Zh. Eksp. Teor. Fiz. 51, 49 (1966) [Sov. Phys.-JETP 24, 33 (1967)].

[19] G. O. Karapetyan, Ya. E. Kariss, S. G. Lunter, and P. P. Feofilov, Zh. prikl. spektroskop. 1, 193 (1964).

[20] B. B. McFarland, R. H. Hoskins, and B. H. Soffer, Nature 207, 1180 (1965).

[21] Kh. S. Bagdasarov, Yu. K. Voron'ko, A. A. Kaminskiĭ, V. V. Osiko, and A. M. Prokhorov, Kristallgrafiya 10, 746 (1965) [Sov. Phys. Crystallogr. 10, 626 (196)].

[22] A. A. Kaminskiĭ and V. N. Shpakov, Zh. Eksp. Teor. Fiz. 52, 103 (1967) [Sov. Phys.-JETP 25, 67 (1967)].

[23] D. A. Stetser and A. J. De Maria, Appl. Phys. Lett. 9, 118 (1966).

[24] B. H. Soffer and B. B. McFarland, Appl. Phys. Lett. 10, 206 (1967).

[25] V. I. Kravchenko, M. S. Soskin, and V. V. Tarabrov, ZhETF Pis. Red. 5, 355 (1967) [JETP Lett. 5, 293 (1967)].

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