INVESTIGATIONS OF NEW CRYSTALS FOR Q-SWITCHED LASERS

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The results are reported of spectroscopic investigations of the emission at 300°K, under free generation and Q-switching conditions, of lasers made of CaF_2-Nd^{3+} type I and type II, CaF_2-YF_3-Nd^{3+}, CaF_2-CeF_3-Nd^{3+}, BaF_2-LaF_3-Nd^{3+}, SrF_2-LaF_3-Nd^{3+}, $CaWO_4$-Nd^{3+} and $Y_3Al_5O_{12}$-Nd^{3+} crystals, and of KGSS-7 glass.

INTRODUCTION

Q-switched lasers [1-3] are usually made of activated crystals or glasses, capable of operation at room temperature. Unfortunately, the list of such working substances is short: mainly it is ruby (Al_2O_3-Cr^{3+}) and glasses activated with Nd^{3+} ions, less frequently scheelite ($CaWO_4$) and garnet ($Y_2Al_5O_{12}$) crystals. Yet the great variety of physical experiments in which such lasers are used call for the use of lasers of varying types, differing in their generation frequency, spectral composition, etc. Therefore, the search for new active media giving satisfactory laser action with instantaneous regeneration switching is a very urgent problem.

The aim of the present investigation was twofold. First, we wanted to investigate the possibility of construction Q-switched lasers, operational at 300°K, using a series of crystals in which only free generation has been obtained earlier. These are the mixed fluoride systems: CaF_2-YF_3-Nd^{3+}, [4-6] CaF_2-CeF_3-Nd^{3+}, [7] BaF_2-LaF_3-Nd^{3+} [8] and SrF_2-LaF_3-Nd^{3+}, [9] as well as type I and II CaF_2-Nd^{3+} crystals. [10-13] The second problem was to determine the relationship between the many optical centers [6,14] present in these mixed fluoride crystals, under the conditions of free and Q-switched generation.

INVESTIGATED CRYSTALS

Mixed fluoride crystals and CaF_2-Nd^{3+} crystals of type I were synthesized using a method described in [4,8,16]. In contrast to type I fluorite crystals, optical activator centers in type II CaF-Nd^{3+} crystals contain oxygen ions. In addition to the substances mentioned above, we investigated also garnet ($Y_3Al_5O_{12}$) and scheelite ($CaWO_4$) crystals, as well as glasses activated with Nd^{3+} ions. The grown single crystals were used to make samples in the form of polished circular rods with plane-parallel ends. The main properties of the active media employed and the geometrical dimensions are summarized in the table.

APPARATUS

The block diagram in Fig. 1 of the apparatus employed is shown. A conventional, highly efficient, elliptical chamber illumination system was used. The pumping sources were IFP-800 and IFP-400 xenon pulse-discharge lamps. The optical resonator consisted of a single spherical multilayer dielectric mirror, whose transmission at 1.06 μ was ≈0.5%, and a 90° total-internal reflec-

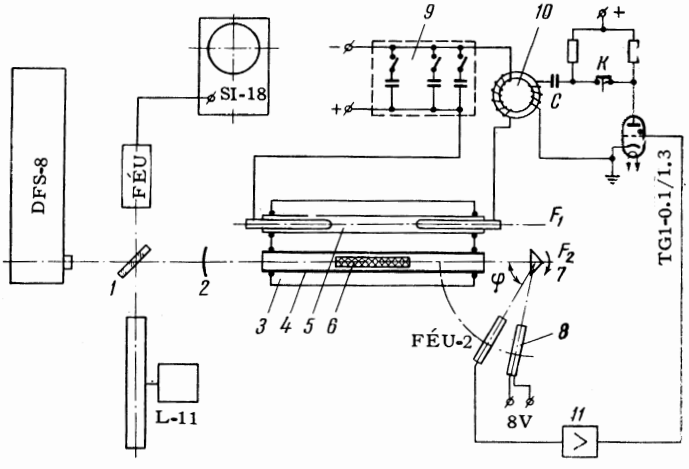

FIG. 1. Block diagram of apparatus: 1) plane-parallel glass plate; 2) spherical dielectric mirror; 3) cylindrical illumination chamber of elliptical cross section; 4) tubular filter of BS or ZhS-17 glass; 5) IFP-800 xenon pulse-discharge lamp, 6) working crystal; 7) 90° total-internal-reflection glass prism; 8) light source; 9) bank of IM-5-150 pulse-discharge capacitors; 10) hf pulse transformer; 11) pulse amplifier.

	Material	Ordinary generation conditions			Q-switched conditions			Concentration, wt. %	Rod length, mm	Rod diameter, mm	Plane-parallelism of ends, sec	Remarks
		Spectr. comp. $\lambda_{cent.}$, Å	Line width, cm^{-1}	Threshold, J	Spectr. comp. $\lambda_{cent.}$, Å	Line width, cm^{-1}	Threshold, J					
1	$CaF_2 - YF_3$	10540	9,5	100	—	—	—	3	75	6,5	6	YF_3 — 6%
		10632	~25	48	10632	7,8	96					
2	$CaF_2 - CeF_3$	10657	6,5	33	10657	3,0	450	0,5	45	5,0	20	CeF_3 — 3%
3	$BaF_2 - LaF_3$	10535	17	60	10535	0,4	90	0,7	75	6,0	18	LaF_3 — 30%
4	$SrF_2 - LaF_3$	10597	~25	75	10597	~25	120	1,0	45	5,5	25	LaF_3 — 30%
5	CaF_2 (type I)	10461	2,5	300	10461	2,4	800	0,5	75	6,5	22	
6	CaF_2 (type II)	10885	4,7	175	10885	3,6	400	0,5	45	6,5	18	
7	$Y_3Al_5O_{12}$	10641	0,6	5	10641	0,5	20	0,5	18	4,3	15	
8	$CaWO_4$	10652	~8	13	10652	1,0	35	3,0	40	4,5	10	
9	Стекло КГСС-7	10608	~75	50	10600	~60	85	6,0	38	4,0	10	

tion prism. The Q-switching was accomplished by rotating the prism at a rate of ≈370 rev/sec. The time lag ensuring population inversion of the $^4F_{3/2}$ level of the Nd^{3+} ions in the investigated active media, i.e., the interval between switching on the excitation lamp and regeneration, was achieved by means of an optomechanical delay chain, consisting of a directional source of light (an 8 V, 20 W incandescent lamp fitted with a lens), an FÉU-2 photomultiplier, and a rotating prism. At the rate of rotation of the prism mentioned above, we could vary this time lag from 250 to 450 μsec by varying the angle φ. The pumping lamp was triggered by a high-frequency (hf) discharge of a capacitor C through a TG1-0.1/1.3 thyratron and through the primary winding of an hf transformer. The lamp was switched by the simultaneous closure of the key K and of the optical system consisting of the source of light, the prism, and the photomultiplier.

The selection of an optical resonator in the form of a prism and a spherical mirror was governed by two circumstances: first, the investigated samples were not perfectly plane-parallel and some of the crystals (CaF_2-CeF_3) were of unsatisfactory optical quality; secondly, such a resonator could be tuned fairly simply by means of a gas laser (we used a laser of the OKG-11 type, with λ = 6328 Å).

The emitted light was recorded using FÉU-28 photomultipliers with oxygen-cesium photocathodes and a pulse oscillograph with a ≈25 Mc pass band. The spectral composition of the radiation was investigated by means of a DFS-8 diffraction spectrograph of ≈6 Å/mm (600 lines/mm) dispersion, whose working range was extended to 12 000 Å. The spectra were recorded photographically on I-1070 film. The spectrum of a lamp with an iron hollow cathode was used as a standard.

To reduce the undesirable effect of the photochemical reduction $Nd^{3+} \rightarrow Nd^{2+}$ ("aging"[12]) in fluoride crystals, caused by the action of strong light pulses from the excitation lamps,[16] we employed, in the generation experiments, tubular filters made of ZhS-17 or BS glass, which absorbed the ultraviolet part of the excitation spectrum.

Luminescence spectra were recorded with a DFS-12 diffraction spectrometer, using a method described earlier.[14]

LASER AND LUMINESCENCE SPECTRA

The spectral composition and the threshold values of the electrical excitation energy are listed in the table for all the investigated active media under free and Q-switched generation conditions. It is evident that the wavelengths generated by the various lasers overlap smoothly in a range ≈420 Å wide (from 10 461 to 10 885 Å).

The threshold values of the excitation energy under free generation conditions, with the prism in a fixed position, were deduced from the appearance of a spiky laser pulse on the oscillograph screen. Unfortunately, the listed values of the threshold energies were not quite correct because the optical quality of the investigated crystals was not always satisfactory. The generation threshold under Q-switched conditions was also found oscillographically, using the appearance of a single strong pulse against a background

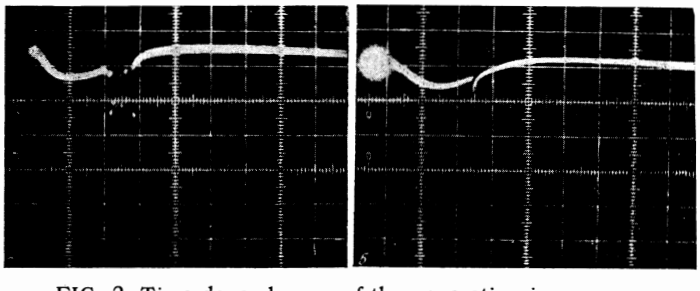

FIG. 2. Time dependences of the generation in a $Y_3Al_5O_{12}$-Nd^{3+} crystal at 300°K: a) free generation conditions; b) Q-switched conditions. Scale: 100 μsec per division.

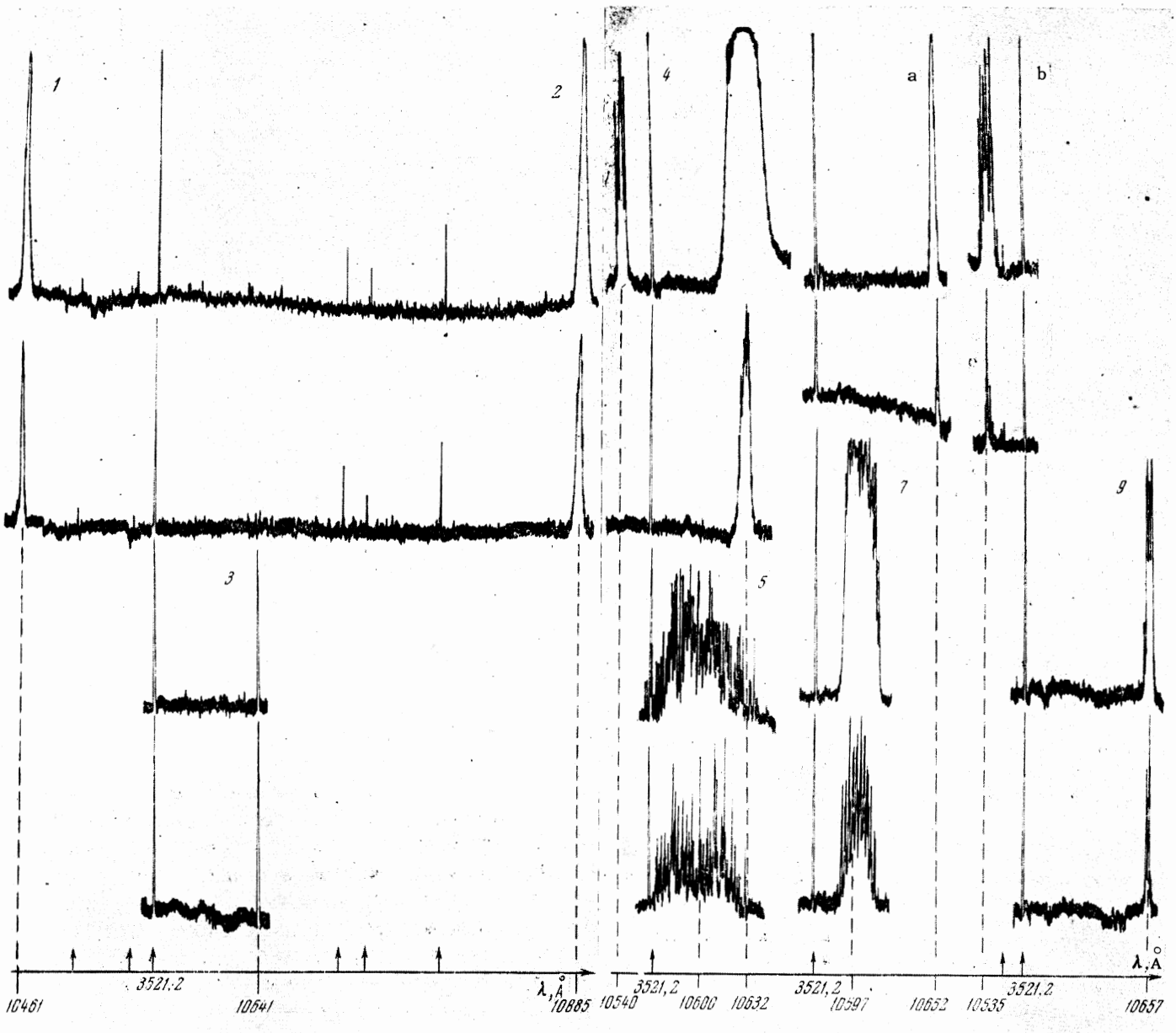

FIG. 3. Emission spectra at 300°K under free generation conditions (on the left) and under Q-switched conditions (on the right) of the following crystals, all containing Nd^{3+}: 1) CaF_2 type I; 2) CaF_2 type II; 3) $Y_3Al_5O_{12}$; 4) CaF_2-YF_3; 5) KGSS-7 glass; 6) $CaWO_4$; 7) SrF_2-LaF_3; 8) BaF_2-LaF_3; 9) CaF_2-CeF_3. The standard line at 3521.2 Å belongs to the third-order spectrum of iron (a hollow cathode lamp was used).

of the pulse of light from the pumping source.

Figure 2 shows oscillograms of the time dependence of the radiation emitted by a $Y_3Al_5O_{12}$-Nd^{3+} crystal under free generation conditions (Fig. 2a) and under Q-switched conditions (Fig. 2b). The optimum time lag between the pumping and generation pulses and the single-pulse conditions were selected by varying the angle φ (cf. Fig. 1).

As mentioned earlier, the spectral composition of the emitted radiation was investigated photographically using the DFS-8 spectrograph. Each spectrum was the result of a single generation pulse and the energy supplied to the excitation lamp was ≈3 times as large as the threshold energy. Microphotograms of the spectra obtained are given in Fig. 3.

We can see that the spectral composition of the stimulated emission and the nature of this composition vary from crystal to crystal. For CaF_2-YF_3-Nd^{3+} under free generation conditions the spectrum has two lines, but under Q-switched conditions it has only one line (the one of longer wavelength). The line widths are slightly different for $Y_3Al_5O_{12}$ and CaF_2 type I and II crystals. In all these cases the lines generated contain many

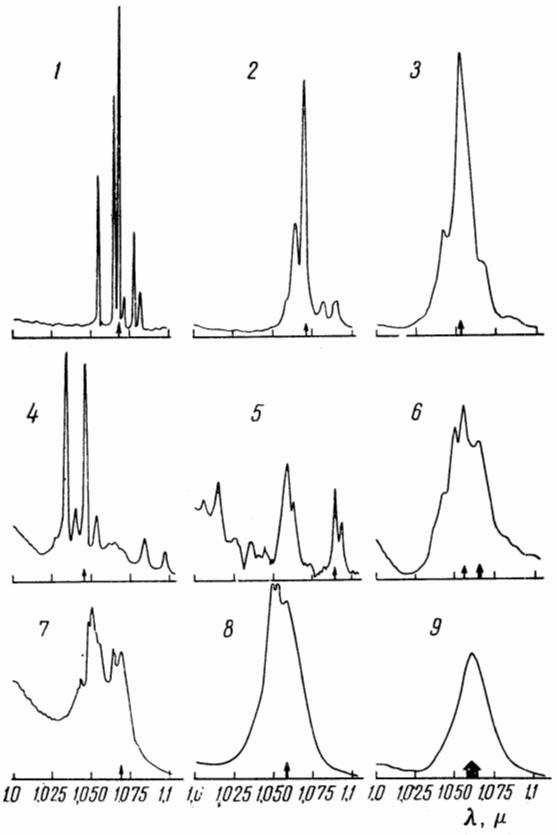

FIG. 4. Luminescence spectra of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition of the Nd^{3+} ion at 300°K in crystals: 1) $Y_3Al_5O_{12}$; 2) $CaWO_4$; 3) BaF_2-LaF_3; 4) CaF_2 type I; 5) CaF_2 type II; 6) CaF_2-YF_3; 7) CaF_2-CeF_3; 8) SrF_2-LaF_3; 9) KGSS-7 glass. Arrows indicate emission lines under free generation conditions and their relative widths.

very narrow components of various intensities and of widths of the order of 0.3—0.4 cm^{-1} (the limit of resolution of our instruments). In some cases these narrow lines were equally spaced (CaF_2-CeF_3 and SrF_2-LaF_3) but in the majority of the lasers they were distributed at random though fairly well reproducible from experiment to experiment (this was true of the positions of the strong components).

Figure 4 shows the luminescence spectra of the Nd^{3+} ions in all the investigated media, corresponding to the transitions $^4F_{3/2} \rightarrow {}^4I_{11/2}$. The spectra were obtained at 300°K using the DFS-12 spectrometer. The accuracy of the measurements was ±3 Å. The arrows indicate the wavelengths at which stimulated emission was recorded. The thickness of the arrows represents roughly the widths of the stimulated lines under free conditions. It should be mentioned that the reported luminescence spectra were not corrected for the spectral sensitivity of the photomultiplier, although this can be done because the spectral sensitivity of the FÉU-28 photomultiplier is known.

DISCUSSION OF RESULTS

It is evident from the table that the threshold values of the excitation energy were relatively low under both investigated generation conditions, but they were somewhat higher under the Q-switched conditions. By way of example, we may mention that in a system with two spherical dielectric mirrors of ≈0.5% transmission, the threshold values of the excitation energy under free generation conditions were approximately five times smaller than under Q-switched conditions. The higher value of the Q-switched threshold in our experiments was due to the use of a prism, which was responsible for considerable losses in the optical resonator.

All the investigated active media could be divided into two groups of substances in respect of the variety of their optical activator centers. One group comprised crystals in which few types of (1—7) centers were formed at activator concentrations employed in lasers (0.5—5%): this group consisted of both type I and type II CaF_2-Nd^{3+},[12,13,17,18] $Y_3Al_5O_{12}$-Nd^{3+},[19,20] and $CaWO_4$-Nd^{3+}.[21] On the other hand, in mixed fluoride crystals[4-9,14] the number of types of optical center can be greater than 100. On this basis we assigned them to the second group of active media. In this respect, the fluoride crystals resembled glasses.[22]

It is evident from the table and from Figs. 3 and 4 that only one type of optical center took part in the 300°K generation in crystals of the first group. The fine structure of the lines generated in these media was evidently due to many modes of the multi-frequency optical resonator used in our experiments. The structure of the lines generated by these crystals was practically the same under Q-switched conditions ($CaWO_4$-Nd^{3+} was the only exception).

Voron'ko[21] showed that about five types of center were formed in scheelite crystals, even when the Nd^{3+} concentration was about 0.3%. Because of this, the observed luminescence line was nonuniformly broadened. This suggested an analogy with the mixed fluoride crystals in that the broad emission line observed under free generation conditions was the result of superposition of the emission lines of various centers. However, under Q-switched conditions, the radiation was emitted by only one type of center, for which the generation conditions were optimal.

As already mentioned, the mixed fluoride crystals contained many types of optical center and the introduced active impurity (Nd^{3+} in our

case) was distributed between these centers (cf.[4-9]). Because of this, independent generation by centers whose concentration was 0.01% or less was practically impossible in the presence of other centers whose total concentration was hundreds of times greater and which did not take part in the generation. It has been shown in [14] that, in such active media, energy may be transferred between the various centers or groups of centers. Our experimental results provided some support for this conclusion. Thus, some crystals (CaF_2-YF_3, CaF_2-CeF_3, BaF_2-LaF_3) exhibited narrowing of the spectrum on transition from free to Q-switched generation conditions. This could indicate that the time needed for energy transfer between individual centers was comparable with the duration of generation ($\approx 5 \times 10^{-7}$ sec in our case). In SrF_2-LaF_3 crystals energy evidently migrated in a shorter time interval.

Unfortunately, the nature of the relationship between optical centers under free and Q-switched generation conditions could not be determined from our experimental data. The radiation spectra of the neodymium glass KGSS-7 are given for comparison with the spectral composition of the emission by the investigated crystals.

CONCLUSIONS

1. Stimulated emission was obtained and investigated for the first time under Q-switched conditions at 300°K in six crystals containing Nd^{3+} as an impurity: CaF_2-YF_3,[1)] CaF_2-CeF_3, BaF_2-LaF_3, SrF_2-LaF_3, and both type I and type II CaF_2.

It was found that in crystals of the second group, characterized by a great variety of optical centers, energy was transferred between these centers; this was accompanied by a strong narrowing of the generation line.

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1)Generation under Q-switched conditions in CaF_2-YF_3-Nd^{3+} crystals was obtained also by G. P. Shipulo and V. V. Ragul'skii.

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14