the ratio of the spot diameter on the screen to the distance from the screen to the sample, was approximately 4°. Processing of the photographs of the radiating face of the sample has shown that the generation was produced by a layer of thickness up to 1 mm. Thus, just as in the case of excitation of GaAs by the Stokes component of Raman scattering of light of a ruby laser in liquid nitrogen [1], a relatively large volume of the semiconductor participates in the generation. The generation efficiency, determined from the ratio of the GaAs emission energy to the energy of the pump optical pulse was approximately 0.1%.

Work is now in progress on the clarification of the physical picture of the excitation of GaAs. Attempts to observe a second harmonic of the pump frequency under the conditions described in [2] have not yet led to positive results.

[1] Basov, Grasyuk, and Katulin, DAN SSSR 161, No. 6 (1965)

[2] N. Bloembergen. International School of Physics "Enrico Fermi," Course XXXI, Varenna, 1963.

SELECTIVE EXCITATION OF RARE-EARTH ION CENTERS IN CRYSTALS

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Most solid lasers use presently as the active medium crystals doped with rare-earth ions (TR). As is well known, several types of optical centers, differing in structure, can exist simultaneously in these crystals. The relative concentrations of these centers depend on the over-all TR concentration and on several technological conditions. Each type of center has in this crystal specific properties and manifests itself separately in the lasing action. Therefore, in order to synthesize crystals with desirable types of TR centers, it is necessary to have a method for analyzing these centers. Such well-known methods as electron paramagnetic resonance, the Zeeman effect, and piezospectroscopic analysis make it possible to establish in many cases the symmetry of the crystalline field and yield some information on the energy level scheme of the given ion. In our papers delivered at the Symposium on Spectroscopy of Crystals Containing TR and Elements of the Iron Group (Moscow State University, 3 - 6 February 1965), we developed and employed a method for analyzing TR^{3+} centers in CaF_2 crystals (type I) from the concentration dependences of the optical spectra - the method of concentration series. This method makes it possible to separate from the total absorption spectrum the spectra belonging to different types of centers. However, this method does not make it possible to carry out an exhaustive analysis of the luminescence spectra.

In this communication we describe a method for the analysis of the Stark structure of luminescence spectra of crystals with TR ion impurity by selective excitation of the luminescence of the individual types of the centers. To carry out such an analysis, the excitation

of the centers must be carried out in individual Stark components of their absorption spectrum. This method was used to investigate CaF_2-Er^{3+} crystals (type I). They have strong absorption and luminescence in the wavelength intervals that are convenient for operation, with a well resolved Stark structure of the spectra.

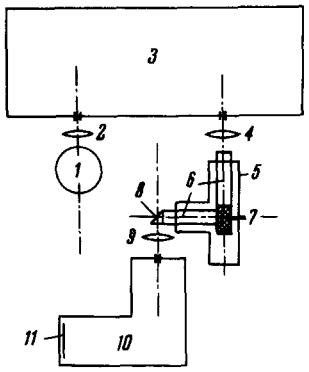

Fig. 1. Optical diagram of experimental setup: 1 - VRSh-1000 mercury lamp; 2, 4, 9 - lenses; 3 - monochromator of DFS-12 spectrometer; 5 - glass tubular Dewar; 6 - quartz light pipes; 7 - investigated crystal; 8 - right-angle prism; 10 - ISP-51 spectrograph with F-270 camera; 11 - casette.

Figure 1 shows a diagram of the apparatus for selective excitation of the centers. Its principal element is the monochromator of the DFS-12 spectrometer, which cuts out bands with half-width ~ 3 Å from the continuous spectrum of the excitation source. The luminescence is registered with an ISP-51 spectrograph. The excitation can be effected in two ways: statically, with the excitation produced in a preselected absorption line, and dynamically. The latter method provides for scanning the wavelength of the exciting lines with simultaneous displacement of the ISP-51 casette. The second method makes it also possible to obtain the excitation spectra of arbitrary individual luminescence lines with a resolved Stark structure.

Figure 2 shows the luminescence spectrum of the CaF_2 crystal (type I) with Er^{3+} concentration 0.3%, obtained in the dynamic mode at 77°K. The luminescence corresponds to the transition $^4S_{3/2} \rightarrow {}^4I_{15/2}$. For clarity we show simultaneously in Fig. 2 on a suitable scale, the absorption spectrum of the level $^2H_{11/2}$ (λ_{center} = 5170 Å), the Stark components of which were used for the excitation. The presence of a continuous inclined line in the right side of the figure is due to the scattered light of the excitation spectrum. The photograph shows clearly that the luminescence spectrum varies in the course of scanning of the exciting band.

On the basis of the results obtained by the concentration-series method for the absorption spectrum of the $^4I_{15/2} \rightarrow {}^2H_{11/2}$ group, it was possible to separate three systems of lines, belonging to three different Er^{3+} centers. It turned out that the excitation in all the lines of each of these systems gives identical luminescence spectra. By way of an example, Fig. 3 shows microphotograms of the luminescence spectra excited in the strongest absorption line of these systems. Excitation in the line of system I corresponds to a luminescence spectrum whose microphotograms are shown in Figs. 3a and b. In the lines of system 2 there is excited a second luminescence spectrum (Fig. 3, c, d). In addition to these spectra, a thorough analysis disclosed a third luminescence spectrum, which becomes particularly intense when the concentration of Er^{3+} is increased. Figure 3e shows a microphotogram of this spectrum for a CaF_2

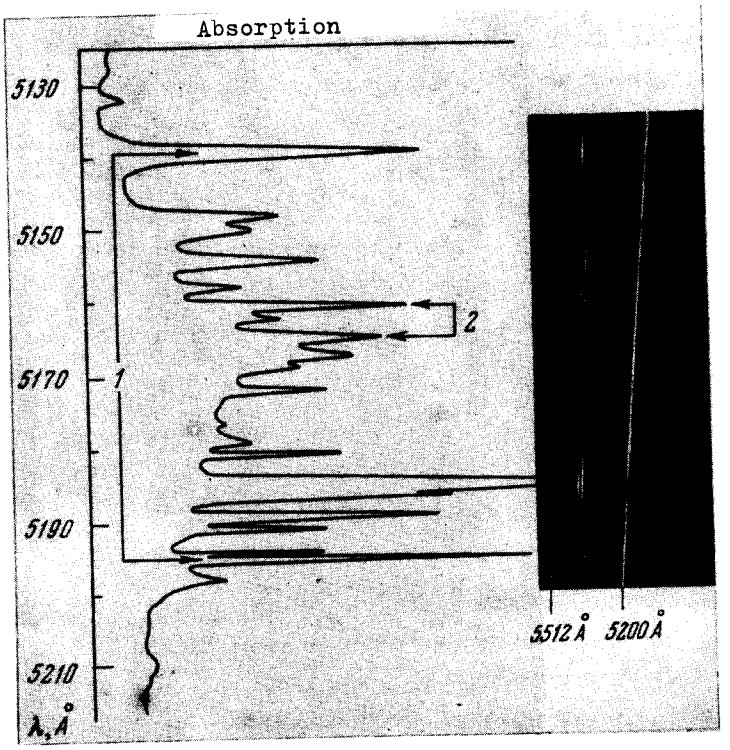

Fig. 2. Luminescence spectrum of the transition $^4S_{3/2} \rightarrow {}^4I_{15/2}$ of the CaF_2-ER^{3+} (0.3%) crystal. Left - absorption spectrum of the $^4I_{15/2} \rightarrow {}^2H_{11/2}$ group.

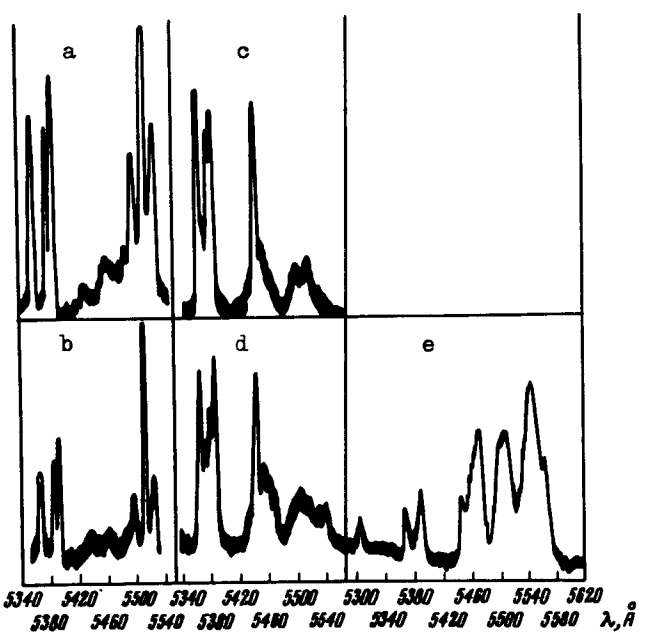

Fig. 3. Microphotograms of luminescence spectra of CaF_2-Er^{3+} (transition $^4S_{3/2} \rightarrow {}^4I_{15/2}$), obtained by exciting the transition $^4I_{15/2} \rightarrow {}^2H_{11/2}$ by individual Stark components. Spectra a, b (system I) and c, d (system II) were obtained by excitation with the absorption lines, noted by the arrows in Fig. 2.

containing approximately 1.0% of Er^{3+}.

Thus, we secceeded for the first time in analyzing experimentally, with the aid of the method of selective excitation, the total luminescence spectrum of a rare-earth ion in a crystal; that is to say, we were able to separate from the spectrum the Stark-structure lines belonging to different types of centers. The use of the method of selective excitation in conjunction with the method of concentration series makes it possible to carry out a detailed analysis of the Stark structure of optical spectra of rare-earth ions in crystals.

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