ABSORPTION, LUMINESCENCE, AND INDUCED EMISSION OF YVO$_{4}$–Nd^{3+} CRYSTALS

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ABSORPTION, LUMINESCENCE, AND INDUCED EMISSION OF YVO_4–Nd^{3+} CRYSTALS

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The spectroscopic properties of yttrium orthovanadate crystals (YVO_4) activated by trivalent rare-earth elements have practically not been studied (apart from several papers on YVO_4 with Eu^{3+} and Er^{3+} ([1])). However, knowledge of these characteristics is now necessary, since induced emission has already been obtained in some of them (Tu^{3+}, Nd^{3+} ([2,3])).* In the present work we report some results of an investigation of the absorption, luminescence, and stimulated-emission spectra, as well as an analysis of optical centers and measurements of the lifetime (τ_{lum}) of the metastable term $^4F_{3/2}$ of Nd^{3+} ions in YVO_4 crystals synthesized at the Institute of Crystallography of the Academy of Sciences of the USSR.

The experimental technique for the study of absorption, luminescence, and measurement of τ_{lum} did not differ from that used in earlier works ([4,5]). The investigations were carried out at the temperatures of liquid helium, liquid nitrogen, and 300° K. Figure 1 shows the absorption spectra corresponding to the transition $^4I_{9/2} \rightarrow {}^4F_{3/2}$, obtained at 77 and 300° K with a DFS-12 spectrometer. As can be seen, they are distinguished by narrow, intense lines with a clearly resolved Stark structure. Figure 2 gives the luminescence spectra of a YVO_4—Nd^{3+} crystal (~ 2 at.%), corresponding to transitions from the levels of the term $^4F_{3/2}$ to the Stark components of the terms $^4I_{9/2}$ and $^4I_{11/2}$, obtained at 77 and 300° K. The spectra presented in Fig. 2a (transition $^4F_{3/2} \rightarrow {}^4I_{9/2}$) were recorded in such a way that, simultaneously with the luminescence, the resonance transitions in absorption were visible; they are especially well distinguished at 77° K. Figure 2b demonstrates the luminescence spectra corresponding to the transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$; the spectra consist of

[Figure 1 graph omitted]

Fig. 1. Absorption spectra of a YVO_4—Nd^{3+} crystal (~ 2 at.%), transition $^4I_{9/2} \rightarrow {}^4F_{3/2}$: *a*–300° K, *b*–77° K.

Figure 2

Figure 1: Figure 2

Fig. 3

Figure 2: Fig. 3

* In paper ([6]) the authors refer to a private communication by Johnson and Thomas on obtaining generation in YVO_4—Tu^{3+} crystals.

very narrow lines. For each temperature, for convenience in comparing the results obtained in the regions of weak lines, we presented additional spectra obtained at a higher sensitivity of the recording apparatus. In work (2) it was indicated that the luminescence spectrum of Nd^{3+} ions in YVO_4 crystals, corresponding to the transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$, contains half as many lines as the analogous spectrum of the $Y_3Al_5O_{12} - Nd^{3+}$ crystal (see, for example, (4)). Our data, and this is clearly seen from Fig. 2b, show that in the case of the YVO_4 crystal, at 300°K 10 lines are observed, and at 77°K 11 lines.

In addition to the experimental data listed above, we carried out an analysis of the Stark structure of the absorption and luminescence spectra associated with terms lying

Fig. 2. Spectra of unpolarized luminescence of a $YVO_4 - Nd^{3+}$ crystal (~ 2 at.%) at 77 and 300°K: *a* —transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$, *b* —transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$. The numbering of the lines is tied to the corresponding transitions in the energy-level scheme (see Fig. 3)

in the transparency range of the YVO_4 crystal (up to 30000 cm^{-1}) at 4.2, 77, and 300°K. Spectroscopic studies showed that in the YVO_4 crystal, with a content of Nd^{3+} ions of about 2 at.%, there exist at least two types of optical centers; we have designated them I and II. In the optical spectra, centers of type I are manifested most fully.

The results obtained allowed us to construct schemes of the crystal-field splitting of the levels of Nd^{3+} ions in the crystal for optical centers I and II. In Fig. 3, certain parts of the schemes of the energy levels of Nd^{3+} ions (terms ${}^4F_{3/2}$, ${}^4I_{9/2}$, and ${}^4I_{11/2}$) are shown for three temperatures. As can be seen, the splitting of the term ${}^4F_{3/2}$ (ΔE) of optical centers of type I at room temperature is 14 ± 1 cm^{-1}, at 77°K $\Delta E = 18 \pm 1$ cm^{-1}, and

Fig. 3. Schemes of the crystal-field splitting of the terms ${}^4F_{3/2}$, ${}^4I_{9/2}$, and ${}^4I_{11/2}$ of Nd^{3+} ions in a YVO_4 crystal at 4.2, 77, and 300°K for optical centers of types I and II. The positions of the levels are given in cm^{-1}, and the transitions between them in Å. Induced transitions are indicated by bold arrows.

at 4.2°K $\Delta E = 19 \pm 1$ cm^{-1}, while for centers of type II it is 58, 56, and 55

cm^{-1}, respectively. It should be noted here that the splitting of the term $^4F_{3/2}$ of centers of type I is the smallest for Nd^{3+} ions among all the crystals known to us. The accuracy of measurement of the positions of the energy levels in our experiments was 1-2 cm^{-1}.

The lines belonging to optical centers of type II are clearly visible in the absorption spectra (Fig. 1) and luminescence spectra (Fig. 2a). In luminescence, the lines of centers I are mainly distinguished, but, as can be seen from Fig. 2b, at 77°K, alongside the most intense components (1-4), weak lines are present (in the spectrum they are denoted by the letter c— "satellites"). With greater sensitivity of the recording apparatus, the "satellites" are also visible alongside the lines of groups 5-7 and 8-11. Unfortunately, it is very difficult to identify them as belonging to centers of type II or to other centers.

In the experiments on stimulated emission, a cylindrical illumination system of elliptical cross section with a pulsed xenon lamp of type IFP-400 [4] was used. The optical resonator consisted of spherical mirrors ($\tau \cong 2\%$) with dielectric coatings, mounted confocally ($R = 576$ mm). Recording of the spectra of induced emission and of the threshold excitation energies (E_{th}) for individual lines was carried out according to the procedures described in [7]. At 300°K

Optical quantum generators based on $YVO_4 - Nd^{3+}$ crystals ($\sim$ 2 at. %) * generate at two wavelengths, with $\lambda = 10\,641$ Å (9398 cm^{-1})—line A, and with $\lambda = 10\,664$ Å (9377 cm^{-1})—line B. The induced transitions in the energy-level scheme (Fig. 3) are indicated by bold arrows. As can be seen, they connect the lower component of the term $^4F_{3/2}$ (11 362 cm^{-1}) with two levels (1964 and 1985 cm^{-1}) of the term $^4I_{11/2}$. The threshold excitation energy for line A at 300°K is $\sim$ 2 J, and for line B $\sim$ 60 J. At room temperature the widths of the generation lines are less than 1 cm^{-1}.

Measurements of luminescence decay showed that for YVO_4 crystals with a concentration of Nd^{3+} ions of about 2 at. %, τ_{lum} at 300°K is equal to $\sim$ 90 μs, and at 77°K to $\sim$ 75 μs (according to the data of work [2], $\tau_{lum} = 33$ μs at 77°K).

The results obtained by us, as well as the physicochemical properties of YVO_4 crystals, show that they may be promising media for continuous-wave optical quantum generators.

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* The optical axis makes a small angle with the geometrical axis.

Note: Figure translations are in progress. See original paper for figures.