LIGHT ABSORPTION IN A CaWO\(_4\):Nd\(^{3+}\) CRYSTAL WITH Na\(^+\) COMPENSATION IN THE 1.06 \(\mu\) REGION

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Abstract

Full Text

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PHYSICS

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LIGHT ABSORPTION IN A $CaWO_4$:Nd^{3+} CRYSTAL WITH Na^+ COMPENSATION IN THE 1.06 μ REGION

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Johnson proposed[(1)] a level scheme for Nd^{3+} in a calcium tungstate matrix for the transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$ (see Fig. 1). In the absorption spectrum of the Nd^{3+} ion there is observed a group of bands, which are denoted in [(2)] by $R, S, A, B, C, \ldots$. In the scheme of Fig. 1, group R is the transition from the $^4I_{9/2}$ level to the $^4F_{3/2}$ level. The transition $^4I_{11/2} \rightarrow {}^4F_{3/2}$ is not observed in the absorption spectrum at room temperature because of the small population of the $^4I_{11/2}$ level (in the luminescence spectrum in the region $\lambda = 1.06\,\mu$, luminescence lines corresponding to the transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$ are observed).

Fig. 1. Energy-level scheme corresponding to the multiplets $^4F_{3/2}$ and $^4I_{11/2}$ in $CaWO_4$:Nd^{3+} (1). Arrows indicate transitions observed in luminescence.

In studying the absorption of light in a $CaWO_4$:Nd^{3+} crystal (the 1.06 μ region), we set ourselves the following tasks: a) to measure the population of the $^4I_{11/2}$ level as the temperature is raised; b) to measure the effective oscillator strength corresponding to the transition $^4I_{11/2} \rightarrow {}^4F_{3/2}$.

A $CaWO_4$ crystal with Nd^{3+} admixture crystallizes in the tetragonal system. Two crystals were studied, in the form of cylindrical rods of circular cross section, 49 and 33 mm long. When it was necessary to increase the absorption, both cylinders were placed one after the other (giving a length of 82 mm). The ends of the cylinders were polished plane-parallel. The tetragonal axis of the crystal was perpendicular to the axis of the cylinder. The crystals were illuminated

Fig. 2

Figure 2: Fig. 2

with polarized light. An Ahrens prism from a polarizing microscope served as the polarizer. By rotating the Ahrens prism, it was possible to obtain the σ- and π-components of the absorption spectrum. Absorption was observed at high temperatures (500–900°K). To heat the crystal, a tube furnace was used, consisting of a quartz tube 120 mm long with an internal diameter of 6 mm, uniformly wound with nichrome wire. On the outside the furnace was covered with a heat-insulating material. The diameters of the crystals were 5 mm. The crystal, wrapped in a sheet of aluminum (about 0.25 mm thick), fit tightly into the tube. To measure the temperature, the furnace had been previously calibrated as a function of the current. The error in measuring the temperature did not exceed $\pm 5^{\circ}$.

For illumination, a three-lens illuminating system was used, which made it possible to obtain uniform illumination of the slit. The absorption spectrum was studied on a home-made spectrograph with a plane diffraction-

grating, which had 600 lines per 1 mm; the dispersion in the region $\lambda = 1.06\,\mu$ was 27 Å/mm. Photography was carried out on I-1030 film.

Absorption at different temperatures was determined by the method of photographic photometry ([3]), using a platinum 9-step attenuator. The results obtained by the step-attenuator method were checked by the spectrograph-slit widening method ([3]). The scale of the absorption curve was determined with an accuracy of $\pm 15\%$. The dependences of the absorption coefficient χ on ν are presented in Fig. 2.

Fig. 2. Absorption spectra (π-component) of the transition ${}^4I_{11/2} \rightarrow {}^4F_{3/2}$ at crystal temperatures: *a*–580°K, *b*–640°K, *v*–850°K.

Experimental results. For our samples, absorption in the region 9500–9180 cm^{-1} becomes observable beginning at temperatures near 500°K. Figure 2 shows the absorption spectra for the π-component at various temperatures. For comparison, Fig. 3 gives the luminescence spectrum at 300°K (π-component).

In addition, absorption spectra χ were recorded for temperatures of 740 and 790°K. The measurements made at these temperatures are given below.

The lowest-temperature spectra are shown in Fig. 3–luminescence at 300°K– and in Fig. 2a–absorption (π-component) at 580°K. From both figures it is seen that both the absorption spectrum and the luminescence spectrum consist of four bands. The positions of the maxima of these bands are given in Table 1.

Fig. 3. Luminescence spectrum (π-component) of the transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ at 300°K.

Fig. 3

Figure 3: Fig. 3

As the temperature is raised, the spectral bands broaden, and maxima Nos. 2 and 4 become poorly resolved; this is seen in Figs. 2b and 2v. The presence of maximum No. 4 is clearly visible in the luminescence spectrum (Fig. 3). Its presence may also be suspected in Fig. 2 from the asymmetric shape of the band $\nu = 9432$ cm^{-1}, which is asymmetrically broadened toward larger ν.

The maxima of the spectral bands, with increasing temperature, apparently shift toward the region of low frequencies, as can be seen from Table 1.

Table 2 gives the half-widths of the absorption and luminescence bands; bands 3 and 4 are combined into one band.

Table 1

Position of the maxima of the absorption and luminescence bands (in cm^{-1}) at different temperatures

Maximum no.	1	2	3	4
Luminescence, 300°K	9250	9349	9450	9492
Absorption, 580°K	9255	9340	9440	–
640°K	9255	–	9436	–
740°K	9245	–	9436	–
790°K	9243	–	9432	–
850°K	9240	–	9432	–

Table 2

Half-width of the absorption and luminescence bands (in cm^{-1})

Band no.	1	2	3 and 4
Luminescence, 300°K	60	40	40
Absorption, 580°K	70	60	80
640°K	70	–	90
740°K	110	–	95
790°K	110	–	100
850°K	110	–	110

Determination of the intensities of the absorption bands

From the absorption curves one can obtain the oscillator strength corresponding to the transition ${}^4I_{11/2} \rightarrow {}^4F_{3/2}$. The oscillator strength is determined by the well-known formula [4]

$$f = \frac{4mc^2\nu_0}{e^2N}\,\mu \int \chi(\nu)\,d\nu, \tag{1}$$

where m is the electron mass, e is the electron charge, c is the speed of light, ν_0 is the transition frequency in cm^{-1}, N is the number of absorbing centers in a volume of 1 cm^3, μ is the refractive index of the medium, and χ is the absorption coefficient, which is determined from the formula

$$I = I_0 \cdot 10^{-\chi \frac{4\pi l}{\lambda} 0.434}, \tag{2}$$

l is the thickness of the absorbing layer. In our case this quantity has been estimated very roughly; however, until now it had not been estimated in any way.

The inaccuracies in determining the oscillator strength are as follows:

1) The oscillator strength is determined for the entire transition ${}^4I_{11/2} \rightarrow {}^4F_{3/2}$ in π-polarization, but the number of absorbing centers entering formula (1) is calculated only for the 2016 cm^{-1} level. The transition from this level to the upper sublevel of the ${}^4F_{3/2}$ term corresponds to the band having the greatest intensity.

2) The absorption band is broad and, in addition, consists of several maxima. However, for ν_0 the transition frequency corresponding to the principal maximum will be taken.

Table 3

Polarization	T, °K	$S = \int \chi(\nu)\,d\nu \cdot 10^4$, cm^{-1}	$N \cdot 10^{-17}$, from Boltzmann formula	$\frac{S}{N} \cdot 10^{22}$	$f \cdot 10^5$, referred to the whole band
π	580	0.89	5.0	1.78	4.6
π	640	1.3	8.3	1.57	4.1
π	740	2.0	14	1.43	3.7
π	790	2.4	18	1.34	3.5
π	850	3.4	23	1.48	3.8
σ	850	2.4	23	1.04	2.7

3) Some inaccuracy is also contained in the calculation itself of N for the 2016 cm^{-1} level. Namely, the calculation is carried out by the Boltzmann formula

$$N = N_0 e^{-h\nu c/kT}, \tag{3}$$

where N_0 is the number of ions on the very lowest (zero) level of the $^4I_{9/2}$ term. In determining N_0 it is assumed that at all temperatures below

At 850°K the total number of Nd^{3+} ions in 1 cm^3 is distributed only over the sublevels of the term $^4I_{9/2}$. Thus, we neglect the fact that the total number of ions in 1 cm^3 is distributed over all terms.

4) Owing to thermal expansion, the number of ions in 1 cm^3 is a function of temperature.

The result of the calculations and the processing of the curves is given in Table 3.

The systematic trend of f is noteworthy. Possibly this indicates that $\varkappa$ is a function of temperature.

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