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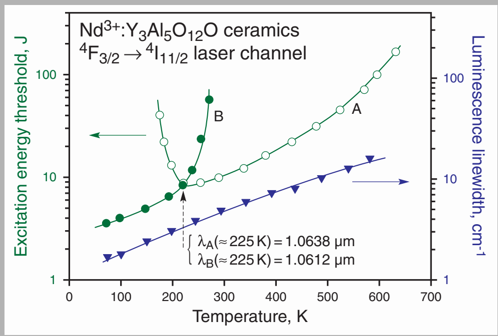
Abstract: Stimulated-emission spectroscopy methods were applied to investigate the temperature behaviour of main laser properties of the "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$. Obtained results clearly show that this novel form of the garnet gain material is an excellent alternative to the Nd^{3+}:$Y_3Al_5O_{12}$ single crystals.



Temperature dependences of threshold excitation energy for the A- and B-lasing lines, as well as luminescence linewidth for the A inter-Stark transition of fine-grained "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$

Stimulated-emission spectroscopy of fine-grained "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$ in a wide temperature range between 77 and 650 K

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1. Introduction

Numerous versatile investigations of past decade indicated conclusively that fine-grained "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$ and its single crystal analog have practically non-distinguished main spectroscopic and stimulated-emission (SE) properties (see, e.g. [1,2]). The Nd^{3+}-doped $Y_3Al_5O_{12}$ ceramics appears to have thermal and some physical characteristics comparable to melt-growing single-crystal counterpart.

However, the ceramic form has significant functional laser potential since it can be fabricated with non-limited strain-free dimensions and with homogeneous distribution lasants (e.g., Nd^{3+}). With these novel crystalline materials many remarkable laser advances have been made already, among them the Nd^{3+}:$Y_3Al_5O_{12}$ ceramic quasi-CW laser

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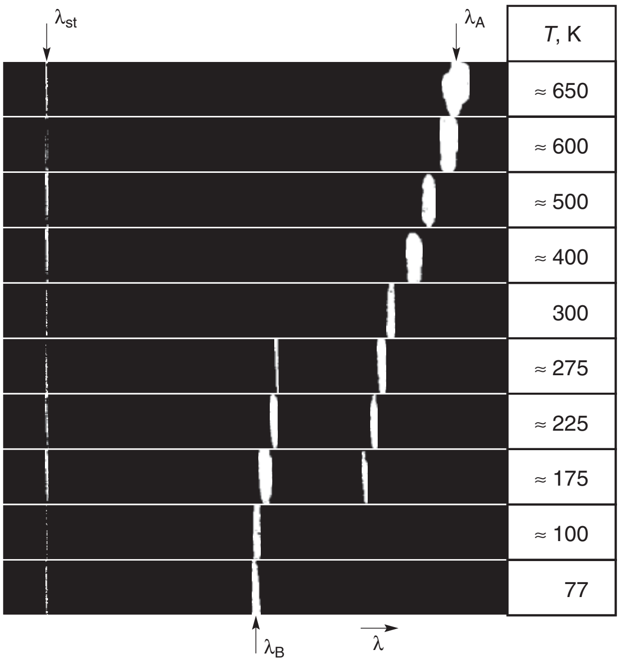

Figure 1 Stimulated-emission spectra ($^4F_{3/2} \to {}^4I_{11/2}$ laser channel) of fine-grained "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$ recorded in a wide temperature range. Wavelength of standard line $\lambda_{st} = 1.0561$ μm

Line [a]	Wavelength, μm	β_{ij} [b]	A_{ij}, s^{-1} [c]	$\Delta\nu_{lum}$, cm^{-1}	$\sigma^p_{e,ij}$, 10^{-19} cm^2 [d]
1	1.0521	$\approx$0.038	$\approx$370	$\approx$4.5	$\approx$0.94
3	1.0549	$\approx$0.002	$\approx$20	$\approx$4.5	$\approx$0.05
2(B)	1.0615	$\approx$0.080	$\approx$520	$\approx$3.6	$\approx$2.5
5(A)	1.06415	$\approx$0.1275	$\approx$1250	$\approx$5	$\approx$3.0
4(A′)	1.0644	$\approx$0.053	$\approx$345	$\approx$4.2	$\approx$1.44
7	1.0682	$\approx$0.034	$\approx$330	$\approx$6.5	$\approx$0.6
6	1.0737	$\approx$0.065	$\approx$425	$\approx$4.6	$\approx$1.64
8	1.0779	$\approx$0.046	$\approx$300	$\approx$7.0	$\approx$0.76
9	1.1055	$\approx$0.014	$\approx$135	$\approx$11	$\approx$0.15
11	1.1119	$\approx$0.029	$\approx$285	$\approx$10	$\approx$0.36
10	1.1158	$\approx$0.035	$\approx$230	$\approx$11	$\approx$0.41
12	1.1225	$\approx$0.032	$\approx$210	$\approx$10	$\approx$0.4

[a] The numeration line here and in Fig. 3 is the same.

[b] Measurement accuracy for strong lines is about 5% and for week and overlapping lines is less than 10%.

[c] Radiative transition probability was found from well known formulas $A_{ij} = 1/\tau^{rad}_{ij}$ and

$$\tau^{rad}_{ij} = \frac{\tau^{rad}(^4F_{3/2})}{\beta_{ij}} \frac{b_i}{\sum_i b_i},$$

here $\tau^{rad}(^4F_{3/2})$ is the radiative lifetime of the metastable $^4F_{3/2}$ state and $b_i = \exp[-\Delta E_{ij}(^4F_{3/2})/kT]$ is the Boltzmann factor, were ΔE_{ij} is the splitting of the $^4F_{3/2}$ state (at 300 K $\Delta E_{ij} = 84$ cm^{-1}, see crystal-field splitting scheme in Fig. 3) and $k = 1.38\times10^{-16}$ erg/K is the Boltzmann constant.

[d] For calculation of peak cross-sections we used formula (see, e.g. [5]): for homogeneously broadened luminescence lines

$$\sigma^p_{e,ij} = \frac{\lambda^2_{ij} A_{ij}}{4\pi^2 n^2 \Delta\nu_{lum}},$$

here n is the refractive index of ceramics studied at λ_{ij}, wavelength dispersion of which was measured recently in [11]. The effective peak cross-section $\sigma^{eff}_{e,ij}$ for the A-transition, which gives rise to SE at the wavelength λ_A=1.06415 μm at 300 K may be considered to be $(3.3\pm0.2)\times10^{-19}$ cm^2. This value results from a sum of corresponding parts of $\sigma^p_{e,ij}$ for two luminescence transitions A and A′, which have close wavelength (1.06415 and 1.0644 μm, respectively).

Table 1 Room-temperature inter-Stark luminescence branching ratios (β_{ij}), probability of radiative transitions (A_{ij}), as well as luminescence linewidth ($\Delta\nu_{lum}$) and emission peak cross-sections related to $i \to j$ transitions of Nd^{3+} ions in "garnet" ceramics $Y_3Al_5O_{12}$

with 100-kW level of output power [3]. As witness experience [4], for advantageous development of high power and high energy of solid-state lasers it is essential to know the spectroscopic behaviour of their gain materials in wide temperature interval. To regard of the Nd^{3+}:$Y_3Al_5O_{12}$ single crystals the comprehensive SE spectroscopic data for temperature range between 77 and $\approx$1100 K have been obtained already (see, e.g. [5]). In this work, using usual methods of SE spectroscopy (see, e.g. [6]) we expanded these temperature investigations upon the fine-grained "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$.

2. Temperature lasing measurement

The temperature effects observed in SE spectra of lasing crystalline materials doped with Ln^{3+} activator ions, in particular Nd^{3+}, usually are gradual change of emitted wavelengths, sudden switching of laser inter-Stark transitions, as well as considerable broadening of the generation lines. All of them basically govern by different manifestations of electron-phonon interaction and crystal-field parameters (splitting of Stark levels and transition probabilities between them), etc. The temperature effects with sudden switching of lasing wavelengths were recorded in many Nd^{3+}-doped crystals, for example, in temperature range above 300 K in CaF_2 [7], SrF_2 [8], and LaF_3 [9] at low temperatures in $Y_3Al_5O_{12}$ and $Lu_3Al_5O_{12}$ [6], as well as in $YAlO_3$ within both ranges above and below 300 K [10]. In present work, for spectroscopic temperature study of lasing "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$ we applied the same methods and equipments as in quoted above references [5–10].

In our temperature investigations we used samples (fabricated from commercial "Konoshima" $Y_3Al_5O_{12}$:Nd^{3+} ceramics, $C_{Nd} \approx 1$ at.%, and ≈ 5 μm average grain size) in the form of rods with plane-parallel ends ($l \approx 45$ mm, $\varnothing \approx 6$ mm) without antireflection coating for SE spectroscopy and plates of different thickness for absorption and luminescence measurements. Fig. 1 shows SE spectra ($^4F_{3/2} \to {}^4I_{11/2}$ laser channel) of "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$ recorded in restricted

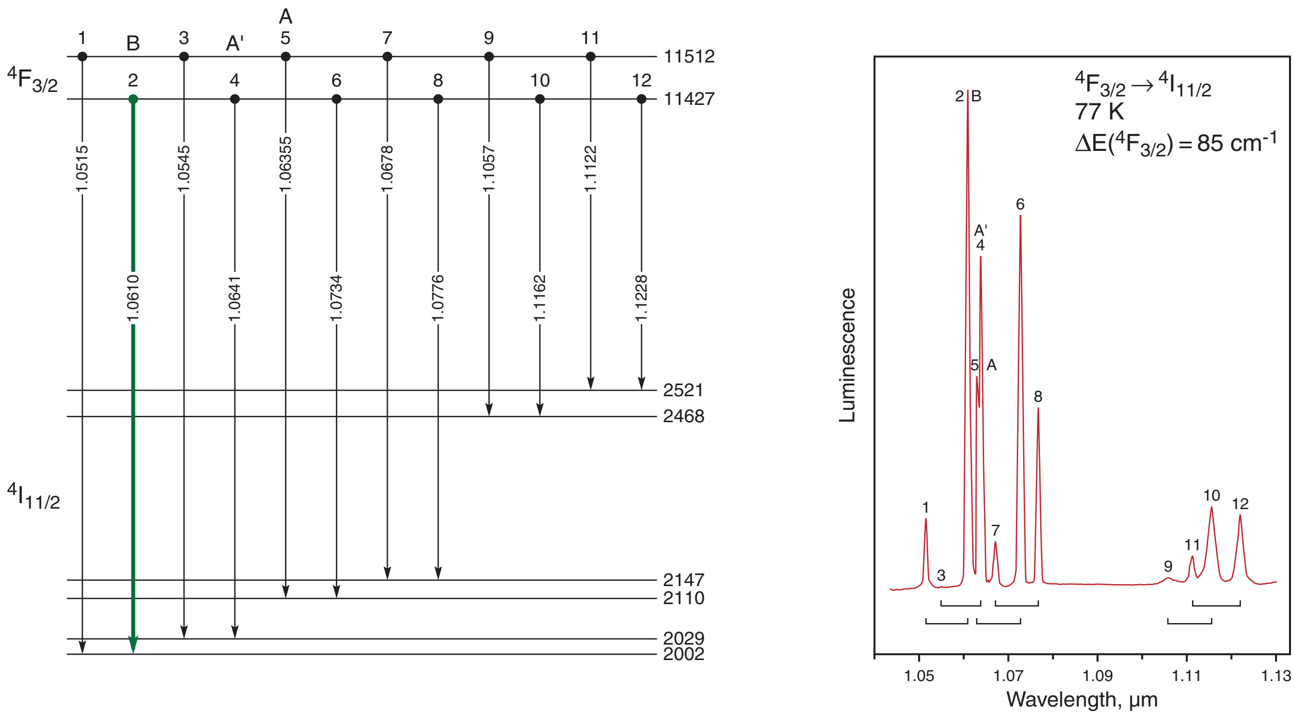

Figure 2 (online color at www.lphys.org) Luminescence spectrum ($^4F_{3/2} \rightarrow {}^4I_{11/2}$ inter-manifold transition) and crystal-field splitting scheme of the $^4F_{3/2}$ and $^4I_{11/2}$ manifolds of Nd^{3+} lasant ions in fine-grained “garnet” ceramics $Y_3Al_5O_{12}$ at 77 K. Stark level energy in the scheme given in cm^{-1} and the wavelengths between them are given in μm. Thick arrow indicates the B inter-Stark laser transition. The square brackets in the spectrum show the splitting of initial metastable $^4F_{3/2}$ state. Lines in the spectrum and corresponding transitions in the scheme are denoted by the same numeration

temperature rang between 77 K and $\approx$650 K It is evident from this figure that, along with a gradual wavelength change of SE on the A- and B-lines, switching from one to the other takes place at about 225 K. In Fig. 2 – Fig. 4 are given luminescence spectra and corresponding crystal-field splitting schemes for two manifolds $^4F_{3/2}$ and $^4I_{11/2}$ only for three measured temperatures 77 K, 300 K, and $\approx$650 K. As can be seen at 77 K, ceramics studied generates at the wavelength of the B-laser line (inter-Stark transition 11427 cm^{-1} $^4F_{3/2} \rightarrow {}^4I_{11/2}$ 2002 cm^{-1}, see Fig. 2), while at room temperature ceramics emits at the wavelength of A-laser line (11507 cm^{-1} $^4F_{3/2} \rightarrow {}^4I_{11/2}$ 2110 cm^{-1} transition, see Fig. 3). The laser inter-Stark A-transition is evidently associated with the upper Stark level of the metastable $^4F_{3/2}$ state, and the B-transition with the lower level. It is desirable now to gaze on Table 1, where listed all spectroscopic characteristics, which reflected SE potential of inter-Stark ij-transitions within main generation channel $^4F_{3/2} \rightarrow {}^4I_{11/2}$ of Nd^{3+} lasants in “garnet” ceramics $Y_3Al_5O_{12}$. For spontaneous (luminescence) A-transition (due to its higher radiative probability $A^A_{ij} \approx 1250$ cm^{-1} than that $A^B_{ij} \approx 520$ cm^{-1} for B-transition) above a certain temperature, according the Boltzmann thermal distribution, the population of the upper Stark level of the $^4F_{3/2}$ state increases and the A-luminescence line becomes stronger than that of the B-line. This is indeed just the case. The luminescence spectrum of Fig. 3 shows that at 300 K its A-line is already the strongest. To growing the intensity of the A-luminescence line ($\lambda_A = 1.06415$ μm) helps also the partly overlap of A′-line with close wavelength ($\lambda_{A'} = 1.0644$ μm). As result the effective peak cross-section at the wavelength of A-line to raise of about 10% (see note d) for Table 1).

Results of measurements of temperature dependences of threshold pump energies for A- and B-laser lines of 45-mm long Nd^{3+}:$Y_3Al_5O_{12}$ ceramic rods are shown in Fig. 5. They were obtained under the long-pulse ($\tau_{ext} \approx 250$ μs) excitation from of Xe-flashlamps (ISP-250) surrounded by quartz yellow filter with the use of two type of pumping chambers of elliptical cross-section. At cryogenic temperatures lasing rod placed in a glass tubular cryostat, was cooled either with liquid nitrogen or with its vapor. At room and elevated temperatures ceramic rod was heated by helical electric “furnace” made of platinum wire $\approx$0.3 mm in diameter. The temperature was monitored with a copper-constant thermocouple directly fastened to the ceramic rods. Spectral composition of SE was photographically studied with the use of grating spectrograph of a DFS-8 type and an I-1070-IR-film. Other exper-

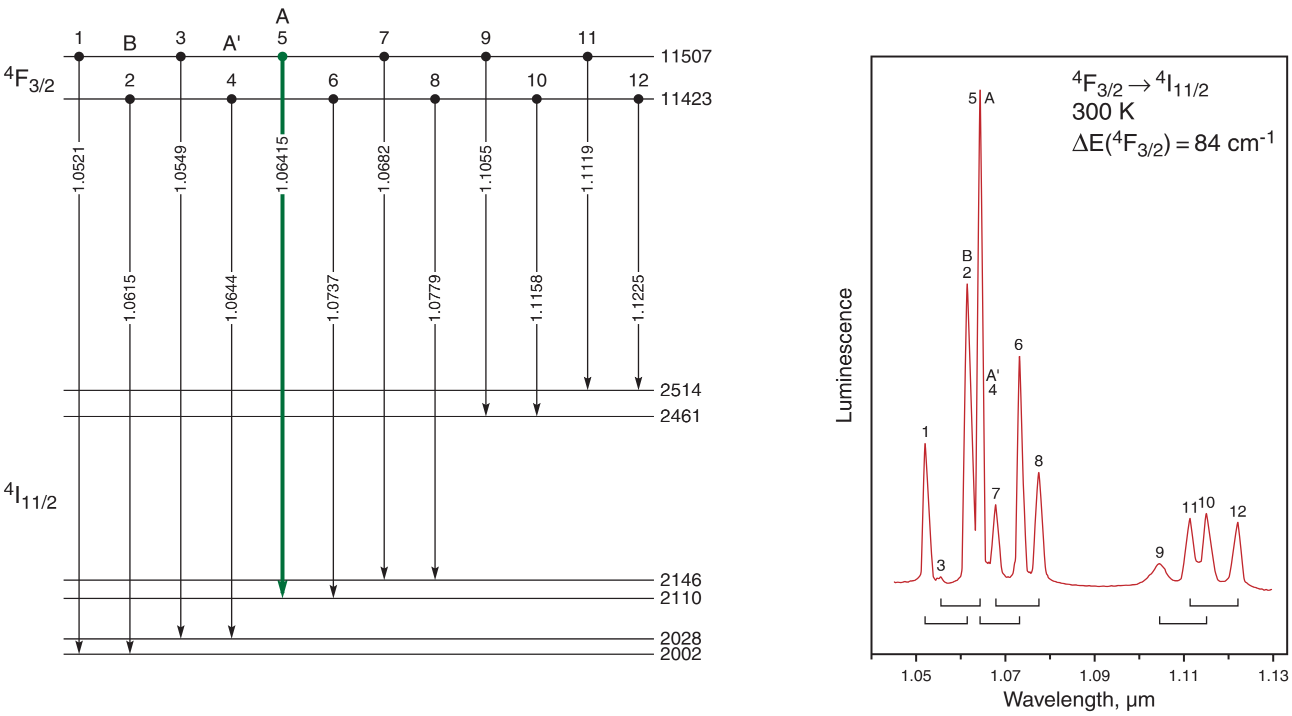

Figure 3 (online color at www.lphys.org) Room-temperature luminescence spectrum ($^4F_{3/2} \rightarrow {}^4I_{11/2}$ inter-manifold transition) and crystal-field splitting scheme of the $^4F_{3/2}$ and $^4I_{11/2}$ manifolds of Nd^{3+} lasant ions in fine-grained "garnet" ceramics $Y_3Al_5O_{12}$. Thick arrow indicates the A inter-Stark laser transition. Other notations are the same as in Fig. 2

imental components and details were similar to repeatable described in our papers (see, e.g. [6–10,12]).

The most important characteristic of a crystalline laser is the excitation threshold. It depends on almost all spectroscopic parameters of the gain medium (single crystal or crystalline ceramics), and exhibits a pronounced sensitivity to various peculiarities of the interaction of lasants ions with host-matrix. Of course, the energy (pumping) SE threshold depends on the operating regime, the excitation conditions, the optical quality of gain material, the optical resonator parameters, etc. (see, e.g. [4,13]). But, its temperature behaviour primarily governs by the nature of crystal-field splitting of Stark levels of the lasant ions (in our case, Nd^{3+}) and their electron-phonon interaction with vibration modes of host-medium. The comprehensive investigation of the interrelation between the temperature behaviour of SE parameters and electron-phonon interaction in lasing Nd^{3+}:$Y_3Al_5O_{12}$ single crystal was conducted in [6]. By virtue of the fact that carried out measurements have no revealed any perceptible difference in the temperature dependences of the excitation thresholds for lasing line A and B in Nd^{3+}:$Y_3Al_5O_{12}$ ceramics studied and single crystal, we shortly note here only their peculiar properties. In studied ceramics, there are two inter-Stark transitions between levels of the $^4F_{3/2}$ and $^4I_{11/2}$ state, which have nearly equal wavelengths. These transitions correspond to the A- and A′-lines in the low-temperature luminescence spectrum of Nd^{3+}:$Y_3Al_5O_{12}$ ceramics are slightly separated as shown in Fig. 2. When the temperature rise is above 77 K, the A- and A′-lines move closer to each other, due to shift of Stark levels that governed by temperature change of crystal-field and activity of electron-phonon processes. As evidenced spectroscopic analysis, the separation between these two lines for $T \geq 300$ K becomes less than their linewidths and A′-transition begins to participate in SE at $\lambda_A \geq 1.06415$ μm wavelengths (see, also note *d)* for Table 1). Analysis of the experimental dependence given in Fig. 5 shows that up to $T \approx 450$ K the main influence of the temperature behaviour of pumping threshold of laser A-line is the temperature change of luminescence linewidth of its transition. At higher temperatures, the threshold energy of A-laser transition is essentially influenced by the thermal increase of the part of population of the levels of the $^4F_{3/2} + {}^2H(2)_{9/2}$ higher lying manifolds above the metastable $^4F_{3/2}$ state, as well as by Boltzmann population of terminal third Stark laser level of the $^4I_{11/2}$ state. Latter indicates that our laser based on "garnet" ceramics Nd^{3+}:$Y_3Al_5O_{12}$, which generates on the A-line began to lose its "four-level" properties at elevated temperatures above 300 K. Of course, such temperature loss of "four-level" distinguishing features will be manifests for all neodymium solid-state lasers emitted at transitions of their "one-micron" $^4F_{3/2} \rightarrow {}^4I_{11/2}$ channel. It should be particularly added

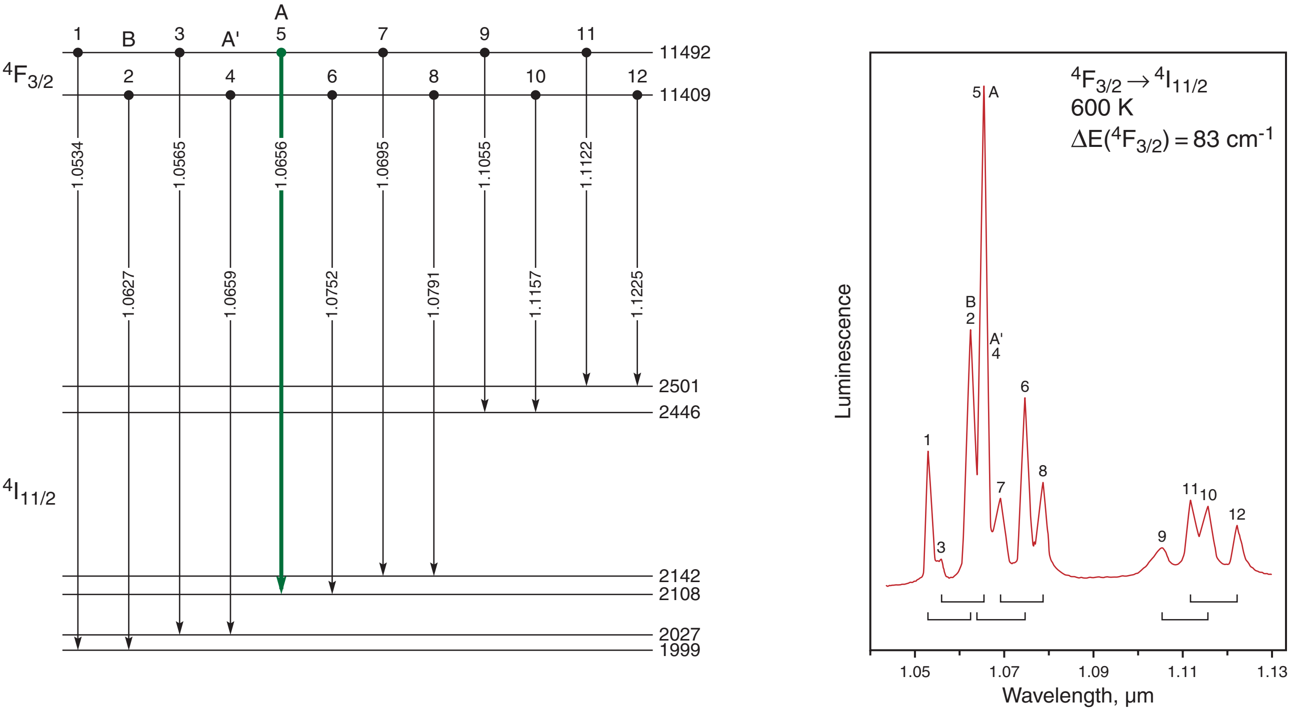

Figure 4 (online color at www.lphys.org) Luminescence spectrum ($^4F_{3/2} \rightarrow {}^4I_{11/2}$ inter-manifold transition) and crystal-field splitting scheme of the $^4F_{3/2}$ and $^4I_{11/2}$ manifolds of Nd^{3+} lasant ions in fine-grained “garnet” ceramics $Y_3Al_5O_{12}$ at 600 K. Thick arrow indicates the A inter-Stark laser transition. Other notations are the same as in Fig. 2

here that in temperature range between ≈ 250 and ≈ 650 K lasing wavelength if the A-line increases linearly with the rate $\approx 4.85 \times 10^{-2}$ Å/K.

3. Conclusion

In summary, results of carried out study of characteristics of the Xe-flashlamp pumped laser based on the fine-grained ceramics Nd^{3+}:$Y_3Al_5O_{12}$, namely the change of spectral composition of its “one-micron” SE of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ generation channel within temperature range from 77 to ≈ 650 K, as well as temperature behaviour of the excitation thresholds for two inter-Stark SE transitions, additionally confirmed natural likeness of main spectroscopic and stimulated-emission properties in both “garnet” gain materials the ceramics and the single crystals.

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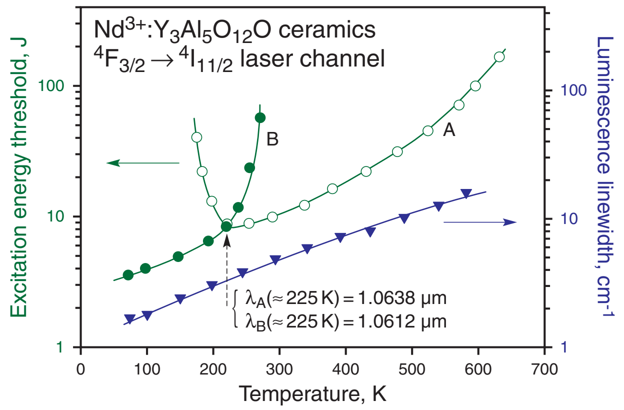

Figure 5 (online color at www.lphys.org) Temperature dependences of threshold excitation energy for the A- and B-lasing lines, as well as luminescence linewidth for the A inter-Stark transition of fine-grained “garnet” ceramics Nd^{3+}:$Y_3Al_5O_{12}$

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