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Luminescence of Nd-doped epitaxial single-crystal garnet films grown on $Gd_3Ga_5O_{12}$ substrates

V V Randoshkin, M I Belovolov, N V Vasil'eva, K A Zykov-Myzin, A M Saletskii, N N Sysoev, A N Churkin

***Abstract*. Epitaxial films with the atomic concentration of neodymium varied in the range from 1 to 15 % were grown on $Gd_3Ga_5O_{12}$ substrates with the orientation (111) by the method of liquid-phase epitaxy from a $PbO-B_2O_3$ overcooled melt solution. The absorption spectra of the films are recorded and their luminescence spectra and luminescence lifetimes of active ions are measured upon diode laser pumping. The concentration dependence of the luminescence lifetime of Nd^{3+} ions is determined.**

Keywords: *luminescence, epitaxial films.*

In microlasers and waveguide lasers based on epitaxial single-crystal garnet films, $Y_3Al_5O_{12}$ (YAG) substrates are commonly used [1, 2]. The disadvantage of this single crystal is its low isomorphous capacity caused by a small parameter (1.2005 nm) of its crystal lattice. The Al^{3+} ions have the lowest size among the ions entering tetra- and octahedral sublattices in the garnet structure, while among the ions entering dodecahedral sublattice only Lu^{3+}, Yb^{3+}, Tm^{3+}, and Er^{3+} ions have smaller sizes than Y^{3+} [3]. This restricts the maximum concentration of large ions (for example, Nd^{3+}) that can be doped to an epitaxial film on the YAG substrate because the film can be produced only when the lattice parameters of the film and substrate are matched with an accuracy of ~ 0.01 %.

The aim of this paper is to study the spectral, luminescence, and kinetic properties of Nd-doped epitaxial single-crystal garnet films grown on $Gd_3Ga_5O_{12}$ (GGG) substrates with the orientation (111).

Epitaxial single-crystal garnet films, in which the atomic concentration C_{Nd} of neodymium was varied from 1 to 15 %, were grown, as in Ref. [4], by the method of liquid-phase epitaxy from a $PbO-B_2O_3$ overcooled melt solution. At $C_{Nd} \geqslant 0.9$ %, to match the lattice parameters of films and substrates, the films were doped with ytterbium. The mixture composition was characterised by the following molar ratios ($\sum[Ln_2O_3] = [Gd_2O_3] + [Nd_2O_3] + [Y_2O_3]$)

$$R_1 = \frac{[Ga_2O_3]}{\sum[Ln_2O_3]} \approx 14.4,$$

$$R_2 = \frac{[PbO]}{[B_2O_3]} \approx 16.0,$$

$$R_3 = \frac{\sum[Ln_2O_3] + [Ga_2O_3]}{\sum[Ln_2O_3] + [Ga_2O_3] + [PbO] + [B_2O_3]} \approx 0.08,$$

where square brackets give the concentration of oxide in the mixture in molar percent.

To produce a single-crystal garnet film of a specified composition, the ratio of concentration of rare-earth oxides in the mixture was determined taking into account the known distribution coefficients for rare-earth elements [5, 6]. The saturation temperature of the melt solution was determined from the temperature dependence of the substrate etching rate [7]. The films were grown upon overcooling, which provided the absence of an additional absorption related to intervalence pair transitions in impurity Pb^{2+} and Pb^{4+} ions [8].

By weighting a substrate before the epitaxial growth and a film on the substrate after the film growth, we measured the total thickness $2h$ of films on both sides of the substrate and the growth rate of the film f_g (see Table 1). The difference between the quantitative compositions of the film and substrate was neglected. The time t_g of the film growth was varied from 30 to 180 min.

Table 1. Parameters of epitaxial single-crystal (Gd, Nd)$_3$Ga$_5$O$_{12}$ and (Gd, Y, Nd)$_3$Ga$_5$O$_{12}$ films doped with neodymium at different concentrations. The absorption coefficient α corresponds to absorption at 808 nm by two films with the total thickness $2h$.

Sample number	C_{Nd} (%)	t_g/min	f_g/μm min^{-1}	$2h$/μm	α/dB
1 (crystal)	1.0	–	–	2000	12
2	0.3	120	0.4	105	~0.1
3	0.9	30	1.3	77	0.3
4	1.7	120	0.4	96	0.9
5	2.3	100	0.4	74	1.0
6	3.3	30	0.9	54	0.95
7	5.0	120	0.3	68	2.0
8	10	180	0.2	69	3.6
9	15	120	0.2	54	7.8

The luminescence spectra were detected by exciting films from the side by a diode laser at 0.808 μm. For comparison, we also studied a single crystal Nd^{3+}:GGG plate (C_{Nd} = 1 %) of thickness 1 mm.

V V Randoshkin, N V Vasil'eva General Physics Institute, Russian Academy of Sciences, ul. Vavilova, 119991 Moscow 38, Russia
M I Belovolov, K A Zykov-Myzin Fiber Optics Research Center, General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia
A M Saletskii, N N Sysoev, A N Churkin Department of Physics, M V Lomonosov Moscow State University, Vorob'evy Gory, 119899 Moscow, Russia

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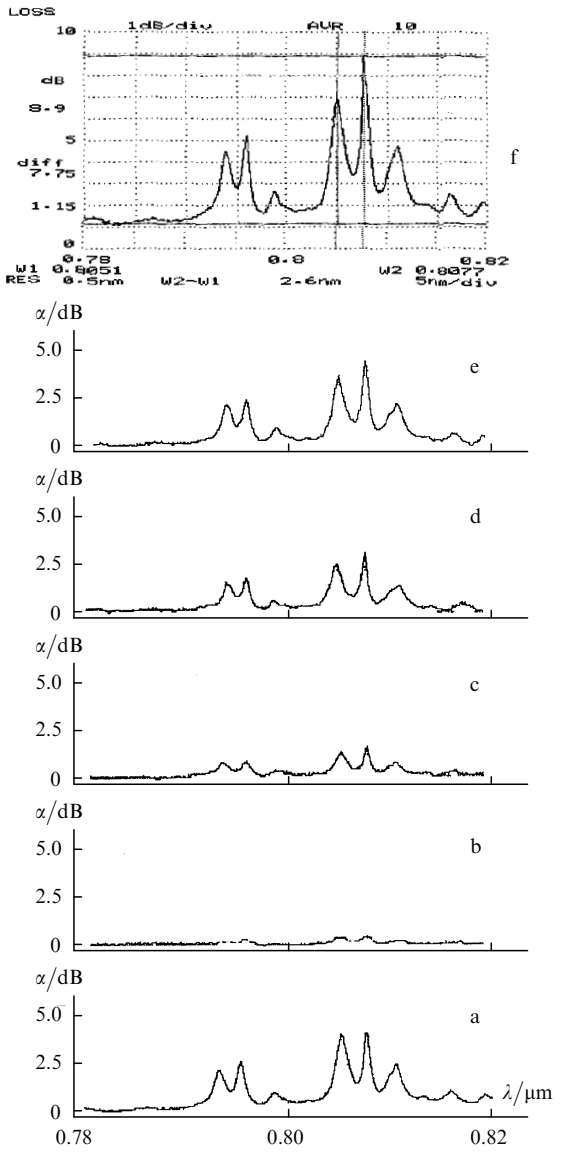

Figure 1. Absorption spectra of a single-crystal Nd^{3+}:GGG plate ($C_{Nd} = 1\,\%$) (a) and epitaxial single-crystal films with $C_{Nd} = 0.9$ (b), 1.7 (c), 5.0 (d), 10.0 (e), and 15.0 % (f).

Fig. 1 shows the absorption spectra of epitaxial films with different neodymium concentrations. One can see that, although absorption increases with increasing concentration of Nd^{3+} ions (Figs. 1b–f), the shape of the absorption spectrum almost does not change and coincides with that of the absorption spectrum of the single-crystal Nd^{3+}:GGG plate (Fig. 1a). The wavelengths 0.805 and 0.808 μm are optimal for pumping. Note that approximately 1/3 of the pump power is absorbed during the double passage of light at these wavelengths through the films with $C_{Nd} = 1.7\,\%$–2.3 %, which allows the use of these films in transversely-pumped lasers. At a lower concentration of Nd in films, either the film thickness should be increased or the films should be used in waveguide lasers.

Fig. 2 shows a typical luminescence spectrum of Nd-doped epitaxial films. The positions of the luminescence peaks of epitaxial single-crystal films at any concentration of Nd^{3+} ions studied coincide with those observed for the single-crystal plate. The 1.061-μm and 1.33-μm luminescence bands exhibit four characteristic peaks each, while the 1.105-μm luminescence band has three peaks. The maximum luminescence intensity was observed at 1.061 μm.

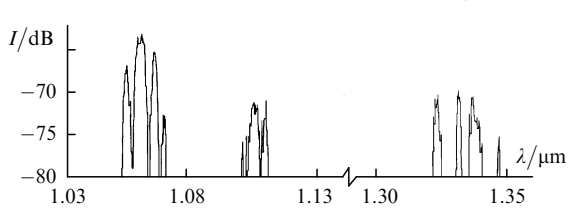

Figure 2. Luminescence spectrum of an epitaxial single-crystal film doped with neodymium at a concentration of 5.0 %.

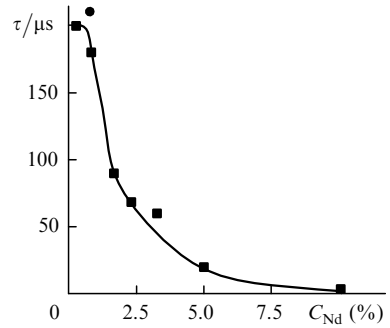

Figure 3. Dependence of the luminescence lifetime τ on the concentration C_{Nd} of Nd^{3+} ions in epitaxial single-crystal films (■) and a bulk sample (●).

Fig. 3 shows the concentration dependence of the luminescence lifetime τ. One can see that τ decreases with increasing concentration of Nd^{3+}. The films with C_{Nd} up to 3.3 %, for which the luminescence lifetime is 60 μs, are promising for lasing. At $C_{Nd} > 10\,\%$, the luminescence lifetime τ becomes shorter than 10 μs.

Thus, we have shown in this paper that Nd-doped single-crystal films grown by the method of liquid-phase epitaxy on the GGG substrates offer promise as materials for microlasers and waveguide lasers.

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