Nd\(^{3+}\):GGG and Cr\(^{4+}\):GGG Epitaxial Films for Neodymium Lasers

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Received November 18, 2008

Abstract—An efficient technology was developed for growing the gadolinium gallium garnet (GGG) single-crystal films doped with Nd\(^{3+}\) or Cr and Ca ions. The films with thickness up to 100 \(\mu\)m have been grown by liquid-phase epitaxy method on undoped GGG substrates of small and big sizes (5–8 mm and up to 76 mm in diameter, respectively). The dependence of absorption, luminescence spectra and optical losses at the wavelength of 1 \(\mu\)m on growth temperature and melt-solution composition was studied. We demonstrated that Cr\(^{4+}\) centers have been implemented in epitaxial films and these films may be used as passive Q-switches for laser systems.

PACS numbers: 42.55.Px, 42.60.Gd, 42.70.Hj, 81.15.Lm

DOI: 10.3103/S1541308X09020010

1. INTRODUCTION

Great progress has been made in the development of high-efficiency solid-state diode-pumped lasers during the last 20 years [1–6]. Commercial lasers of this type are widely used, for example, in laser rangefinders and lidars [7], labeling tools [8], and medicine [9]. This is due to a great extent to the progress in diode pump systems operating in a wide power range (from \(10^{-3}\) to \(10^{2}\) W). At the same time, the use of active elements of conventional shape meets serious difficulties, related to the formation of a pump beam of necessary geometry (strongly divergent pump radiation must be transformed into a long narrow beam). As a result, such systems become not only more complicated and expensive but also less efficient in many cases.

Lasers and amplifiers with optical (diode) pumping are most effective when the waveguide geometry is used, in which pump radiation is strongly overlapped with cavity modes, the volume of laser modes is minimized, and the product of the pump intensity with the interaction length may exceed the corresponding values for the bulk active elements by several orders of magnitude. This concept is implemented in erbium fiber amplifiers, ytterbium fiber lasers, etc. Currently, the average lasing power of a fiber laser with a core diameter of about 8 \(\mu\)m reaches 1.5 kW, and the lasing pulse energy is limited by several microjoules. The lasing power in planar elements is limited by several hundreds of watts, and the lasing energies in the Q-switched mode are at a level of 0.5 J. Another advantage of planar active elements over classical ones is smaller thermo-optical beam distortions [10, 11]. Therefore, the researchers all over the world pay much attention to the methods for fabricating both fiber and planar active elements.

The studies devoted to design of planar optical waveguides based on garnet crystals doped with rare earth ions have been under way since the 1970s [12, 13]. However, the films obtained by different methods in the first studies were of low crystalline and optical quality, and it was fairly difficult to obtain lasing even on neodymium active elements [14]. Moreover, the luminescence spectra of neodymium ions differed from those of Czochralski-grown bulk single crystals.

The methods of ion implantation [15, 16] and thermal diffusion [16], which have been thoroughly elaborated in electronic technology and production of electronic devices, make it possible to easily obtain waveguide structures in glasses and crystals of different types, including nonlinear. The thus
obtained structures have high optical characteristics but relatively large loss at a wavelength of 1 μm (~1 dB cm⁻¹). The neodymium- and erbium-activated structures, obtained by the above-mentioned methods, exhibited lasing at wavelengths of 1 and 3 μm. In particular, in a 20-μm Nd:YAG planar waveguide prepared by ion implantation, the lasing threshold was ~1 mW at 808-nm laser-diode pumping. The lasing efficiency was ~15%. The poor laser characteristics were explained by both the low pump efficiency and high loss at the lasing wavelength. The loss source was not exactly determined. However, the most likely reason was the damage of the substrate crystalline structure, induced by an ion beam.

The method for obtaining planar structures by laser sputtering with subsequent deposition on a substrate [14, 17] may be optimal for preparing multicomponent thin films due to the possibility of congruent target-to-substrate transport of material. In addition, this technique does not require to match the film and substrate lattice parameters. Moreover, single-crystal films can be deposited even on glass substrates [21]. The loss at a lasing wavelength of 1 μm in the structures fabricated in this way is also high (2–6 dB cm⁻¹). Nevertheless, lasing was obtained on neodymium planar structures produced by this method. However, the lasing efficiency did not exceed 20% of the absorbed pump power.

The methods of vapor-phase epitaxy (VPE) [18] and liquid-phase epitaxy (LPE) [19, 20] are more complex in comparison with those considered above, because growth of high-quality films requires careful matching of the substrate and film lattice parameters and choice of appropriate thermal growth conditions. However, specifically these methods make it possible to obtain planar structures of the highest quality with the least loss at the lasing wavelength. For example, samples of Yb:YAG planar structures with a loss less than 0.05 dB cm⁻¹ (i.e., about 0.12 cm⁻¹) at a wavelength of 1 μm have been implemented. This value exceeds the loss in commercial crystals grown by the Czochralski method by a factor of only two. In this case, the lasing efficiency under laser-diode pumping was about 77% of the diode power [22]. A drawback of VPE is the low growth rate and small film thickness.

The interest in garnet crystals activated with Cr⁴⁺ ions is caused by their wide practical application in Q-switches for 1-μm lasers and in tunable near-IR (1.2–1.5 μm) lasers. The growth technique of bulk Cr⁴⁺:YAG and Cr⁴⁺:GGG crystals has been developed for more than 15 years [23]. Currently, the studies aimed at improving the optical characteristics of Cr-containing garnets and developing a physical model to explain the mechanism of formation of Cr⁴⁺ ions in crystals are under way [24–26]. Compact laser systems are of particular interest: they must be based on effective (simple, reliable, and inexpensive) short-pulse lasers with stable single-mode lasing [27]. A possible design is a single block composed of a crystalline active element and an epitaxial single-crystal layer on its surface, operating as a Q-switch [28, 29]. This is a complex technological problem to be solved in several stages.

The purpose of this study was to search for the optimal conditions for obtaining high-quality homogeneous single-crystal gadolinium gallium garnet films, doped with neodymium and chromium ions, on substrates of large diameter (more than 70 mm), and determine the spectroscopic characteristics for their use as active elements and Q-switches in compact laser systems.

2. DEVELOPMENT OF THE METHODS FOR GROWING SINGLE-CRYSTAL EPITAXIAL FILMS WITH GARNET STRUCTURE AND OPTIMIZATION OF GROWTH CONDITIONS

An analysis of the data in the literature shows that VPE and LPE make it possible to obtain planar structures of the highest quality and least loss at a lasing wavelength. However, VPE barely makes it possible to grow films up to 100 μm thick. Therefore, we chose LPE to grow laser garnet films for planar active elements. Growth of epitaxial films from melt-solution allows one to vary the growth temperature in a certain range in order to optimize the film parameters. We used immersion in a supersaturated melt-solution with garnet components.

As a substrate and film material, we chose crystals with garnet structure (in particular, gadolinium gallium garnet (GGG)) for the following reasons:

(i) modern technology makes it possible to grow crystals up to 150 mm in diameter, due to which not only optical elements with a large aperture can be prepared but also elements of smaller sizes with more uniform properties;

(ii) chemical (thermodynamic) stability and mechanical strength of these crystals facilitate their use;

(iii) high thermal conductivity allows high-power applications;

(iv) the presence of ions of different size in the garnet composition facilitates doping with different elements.

In this study, the emphasis was on the growth of two epitaxial films of different composition, doped with Nd³⁺ ions or Ca²⁺, Cr³⁺, and Cr⁴⁺ ions, with minimization of optical loss at a wavelength of 1 μm.