Comparative study of Yb (3+) doped LuYAG laser ceramic: optical, structure and lasing properties

V V Balashov¹, L Yu Zakharov¹, A Yu Kanaev², A B Kozlov³, S M Kozlova¹, A L Koromylsov⁴, K V Lopukhin¹, P P Faikov⁵, I M Tupitsyn⁶, E A Cheshev⁷

¹Kotelnikov’s IRE, RAS, Fryazino, Moscow region, Russia
²SLPG ‘Raduga’, FSE, Radyzhnyi, Vladimir region, Russia
³AO Stelmakh Polyus Res Inst, Moscow, Russia
⁴P.N.Lebedev Physical Institute of the Russian Academy of Sciences, LPI, Moscow, Russia
⁵D. Mendeleev University of Chemical Technology of Russia, Moscow, Russia

E-mail: intupitsyn@yandex.ru

Abstract. Materials with high thermo mechanical properties are required for high powered lasers. One of such materials is lutetium-aluminum garnet (LuAG) doped with ytterbium. In this work main properties of (LuₓY₁₋ₓ)AG:Yb³⁺ ceramics with x=0-1 are studied in order to determine optimal Lu/Y ratio for lasing purposes.

1. Introduction

Laser quality transparent ceramics, alongside with single crystals, are promising materials for development of either powerful or compact solid-state lasers working in continuous (CW) or q-switched modes. When constructing high average power laser, both spectral and thermal properties of lasing medium are very important. Optical quality of laser medium, possibility of fabrication elements of larger size and availability are also should be taken into account.

Early laser ceramics studies were focused on Nd³⁺-doped medium [1,2]. Subsequently, upon the emergence and wide availability of powerful semiconductor InGaAs lasers, purposeful research of Yb³⁺-doped lasers begins. Lasing wavelength of InGaAs lasers is 940 nm, and therefore they can be used for pumping Yb³⁺--doped media [3]. Yb³⁺ has some advantages over Nd³⁺ which was widespread at that point. First, the former dopant has small quantum defect than the latter, 9% and 6% for pumping wavelengths of 940 nm and 970 nm, respectively. This reduces the heat load of the active element [4]. Also, Yb³⁺ has simpler electron level structure, namely all absorption and emission transitions occur between ²F₇/₂ and ²F₅/₂ levels, which rules out possibility of up-conversion and cross-relaxation. Another advantage of Yb³⁺ is that it can be used in high concentration without concentration extinguishing taking place. Further, ytterbium has significantly wider absorption bandwidth than neodymium, which loosens requirements for diode pumping. And lastly, Yb³⁺ has longer life time of the upper level, the important advantage when constructing pulse lasers.

Spectral properties of the Yb³⁺ may vary depending on the choice of matrix, the feature that led to many studies on fabrication and properties of different ytterbium-doped laser media (crystals and ceramics). This includes studies of garnets (YAG, LuAG, YSAG, GGG) [5–7], sesquioxides (Lu₂O₃, Sc₂O₃, Y₂O₃, Lu₂SeO₃) [8], tungstates (KGW, KYW, KLuW) [9], borates (YAB, YCOB, LSB) [10] and fluorides (CaF₂, SrF₂) [11].
High-powered lasers require materials with high thermo mechanical properties. One of such materials is ytterbium-doped lutetium-aluminum garnet (LuAG). Its main advantage over yttrium-aluminum garnet is insignificant decrease of thermal conductivity when Yb$^{3+}$ concentration is increased [12]. This is due to the small difference of Yb$^{3+}$ and Lu$^{3+}$ ion radii, 1.008Å and 1.001Å, respectively. For comparison, ion radius of Y$^{3+}$ is 1.04Å [13]. In-between compounds (Lu$_x$Y$_{1-x}$)AG:Yb$^{3+}$ $x=0..1$ were initially investigated as an LuAG alternative with lower melting point and less expensive raw materials, as high purity lutetium oxide is particularly expensive. High melting point of LuAG makes growing crystal by methods, where crucibles are used, very challenging [14]. Afterwards it was discovered that intermediate compounds have other useful properties such as lasing wavelength shift to 2.02 µm for the Tm$^{3+}$ dopant [15] when $x=0.5$. This significantly reduces absorption by H$_2$O and CO$_2$ molecules [15,16].

2. Synthesis of ceramics
Precipitated Y$_2$O$_3$, Yb$_2$O$_3$ and commercially available Lu$_2$O$_3$ (Lanhit), Al$_2$O$_3$ (Sumitomo chemical) were used as starting materials. Mg(NO$_3$)$_2$, H$_3$BO$_3$, and TEOS were used as sintering aids with concentrations 0.04, 1.35, and 1 mol.% respectively. Mg(NO$_3$)$_2$, H$_3$BO$_3$ were used in the form of low concentration solutions in isopropyl alcohol in order to increase accuracy when weighting them. A series of samples with (Lu$_x$Y$_{1-x}$)AG:5%Yb$^{3+}$ ($x=0$-1) composition were fabricated with $x=0; 0.25; 0.5; 0.75; 1$. Their designations are Lu0-5, Lu25-5, Lu50-5, Lu75-5 and Lu100-5 respectively. Initial oxide powders were mixed in stoichiometric proportions and milled on a planetary mill with high purity alumina balls for 15 hours in isopropyl alcohol. After milling powders were dried for 48 hours at 50°C and sieved through a 200 mesh sieve, calcined at 1000°C for 6 hours to remove any volatile matter, and then compacted. The powders were pressed uniaxially at 100 MPa in stainless steel mold and then CIPed at 250 MPa for 5 minutes to obtain pellets $\approx$9 mm in diameter. These green bodies were calcined at 1000°C for 5 hours to remove residual organics and sintered at 1775°C for 8 hours in a furnace with tungsten heater under 10$^{-4}$ Pa vacuum to provide fully dense Yb:LuYAG ceramics. Then, sintered samples were annealed at 1300°C for 10 hours in air to remove oxygen vacancies and convert Yb$^{2+}$ to Yb$^{3+}$. Finally, all Yb:LuYAG ceramics was grinded and mirror polished on both sides using diamond slurries; the thickness of samples after mirror polishing was 1 mm.

3. Experimental
The microstructure of sintered Yb:LuYAG samples was investigated using scanning electron microscope Phenom ProX on thermally etched surfaces of polished samples. To evaluate the optical quality of ceramics transmission spectra of polished samples were measured using Specord UV-VIS spectrophotometer in 200-800nm wavelength range and custom made spectrophotometer based on POC 4 polychromator and Toshiba TCD1304AP CCD in 800-1050nm wavelength range.

4. Characterization of ceramics
The microstructure images of ceramics samples with different Lu/Y ratios sintered at 1775°C for 8 hours are presented in Figure 1(a-e). These are SEM microphotographs of thermally etched surfaces. Average grain size changes from 19.2µm for Lu0-5 sample to 29.5µm for Lu100-5 sample as shown on Figure 1(f). Images show no signs of abnormal grain growth and no inclusions of any extraneous phases. It can be seen from Figure 1(f) that with the increase of Lu/Y ratio average grain size also increases. If in future we’ll need to further decrease average grain size we’ll need to change sintering aids and/or use HIP treatment.
Figure 1. SEM micrographs of thermally etched surfaces of Yb:LuYAG ceramic samples (a)-Lu100-5, (b)-Lu75-5, (c)-Lu50-5, (d)-Lu25-5, (e)-Lu0-5 sintered at 1775°C for 8 hours (a-e). Grain size dependence on Lu/Y ratio for studied Yb:LuYAG ceramic samples (f).

After annealing in air at 1300°C for 10 hours all samples became colorless. Transmission spectra of annealed samples are shown on Figure 2. At 850nm transmission of annealed samples is ranging from 80.7% for sample Lu25-5 to 83.4% for the rest of them. At 400nm transmission of annealed samples is ranging from 75.3% for sample Lu25-5 to 79.5% for sample Lu50-5 and 80.6% for the rest of the samples.
5. Laser operation
For studying of lasing characteristics, disk laser elements similar to ones described in [17] were made. Thickness of active media after machining was 800 μm. This thickness was chosen to provide enough amplification in case of direct pumping from one source without multi-pass pumping. Besides that, in the case of thick disk, effects caused by thermal conductivity and amplification of spontaneous emission (ASE) will be more pronounced than in the case of thin disk (200-300 μm). On one side of samples MgF₂ antireflection coating for 1030nm wavelength was applied. On the other side of samples R>99,98% mirror for lasing wavelength was applied. Prepared samples were installed one at a time mirror side towards copper-tungsten heatsink by means of vacuum cold diffusion bonding with indium used as solder.

For comparative measurements of lasing characteristics of fabricated ceramic laser elements, a laser stand was assembled, schematically shown on Figure 3. Disk active element was located on the temperature-stabilized heat sink with possibility for temperature adjustment.

Semiconfocal resonator was formed by heat sink facing side of disk laser element (where mirror coating was applied) and spherical mirror with 200m curvature radius and 95% reflectivity for 1030nm wavelength. This mirror (M2) was installed on motorized platform. In all experiments pumping was performed by laser module with 100 μm fiber output, NA=0.22 and maximum power of 70W. Study of lasing characteristics was carried out in CW and quasi-continuous (QCW) modes. Latter mode was used to reduce thermal effects. Resonator length was 101mm. Central pumping wavelength was 938nm, such choice was made due to similar absorption coefficients of studied samples at this wavelength. Pump radiation was injected into samples at 13-14° angle forming spot 250 μm in size. Heat sink temperature was kept at 14°C during the experiments.
Figure 3. Experimental setup for disk laser element testing (top view). HS – heat sink, M₁, M₂ – resonator mirrors, AM – active element, L₁ – focusing system, X₁,₂ – focal lengths of the focusing system.

Laser generation was obtained on all studied samples in CW and QCW modes. The dependences of lasing power on absorbed pump power in CW and QCW modes are shown on Figure 4.

Figure 4. Dependence of lasing power on absorbed pump power for samples studied in CW and QCW modes of laser operation.

It can be seen from study of lasing characteristics that in case of LuYG matrix doped with 5% Yb³⁺ substituting up to 50% of Lu for Y does not lead to decrease in laser performance compared to pure LuAG. For samples doped with 5% Yb³⁺, difference in thermal conductivity doesn’t affect the output lasing characteristics significantly. In CW mode 3,2 to 4W of output power was obtained for samples Lu50 to Lu100, 6 to 7 W in QCW mode. For samples Lu0 and Lu25 obtained output power was 2.5W in CW mode and 5W in QCW mode. This difference in output characteristics is attributed to lower thermal load of active element in QCW mode.

We can compare slope efficiency of 30-45% measured in this work to results obtained in [6], where 60% slope efficiency was measured for similar configuration, namely for 5%Yb:LuAG with T=5,8% output mirror and 75% with optimal mirror with T=39.1%. In our case lower slope efficiency was attributed to inhomogeneity and inconsistency of pumping with the cavity axis, inherent to the angular pumping scheme, and also to the use of output mirror with T=5% which is not optimal. In our
experiments we had no goal of achieving high efficiency of laser system. But it can improved by selecting optimal transmission of output mirror.

6. Conclusion
Various optical, structural and lasing characteristics of obtained ceramic samples were studied. Carried out research has shown that fabricated LuYAG:5% Yb ceramics with different Lu/Y ratios are of the laser quality and substituting up to 50% of Lu for Y does not lead to a decrease in laser performance compared to pure LuAG.

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