Growth and characterization of Yb:Ho:YAG single crystal fiber

Yilun Yang a,b, Lihua Ye c, Renjie Bao c, Shanming Li a,b, Peixiong Zhang d, Min Xu a, Yin Hang a,*

a Key Laboratory of High Power Laser Materials, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China
b University of Chinese Academy of Sciences, Beijing 100039, China
c Department of Physics, Zhejiang University, Hangzhou 310027, China
d Department of Optoelectronic Engineering, Jinan University, Guangzhou 510632, China

HIGHLIGHTS
- The first study to grow Yb:Ho:YAG single crystal fibers by the LHPG method.
- The grown high-quality SCF possesses the same structure as the crystal bulk.
- Presently reported the minimum known diameter fluctuations of YAG single crystal fiber.

ABSTRACT
High quality Yb and Ho co-doped Y 3 Al 5 O 12 single crystal fibers have been successfully grown by the laser heated pedestal growth method of up to 124 mm in length and 450 μm in diameter for the first time. The results of inductively coupled plasma-atomic emission spectrometry analysis, X-ray diffraction and Raman spectroscopy reveal that the lattice structure and doping concentrations of the SCF are the same as that of the bulk. Scanning electron microscopy microphotographs shows that the fibers only have minor diameter fluctuations within 0.5%.

In recent years, fiber lasers with good thermal properties have been rapid development and have become a topic of intense research focus. However, the use of conventional silica fibers pose many problems, such as nonlinear effects which reduce spot quality, as well as low thermal conductivity [7–9]. Because of this, single crystal fiber (SCF) have become a promising candidate for the direction of high power fiber lasers. Now the Ho:YAG SCF grown by laser heated pedestal growth method has been reported in the literature, and achieved 23.5 W output power at 2.09 μm [10]. This means Ho 3+ ion doped SCF can be applied to fiber lasers.

The laser heated pedestal growth (LHPG) method is a new and efficient technique for the growth of single crystal fiber. It was first invented by Burrs in 1972, then rapidly became the best way to grow SCFs [11]. The LHPG method is a crucible-free, clean, and quick way to produce high-quality SCFs. Optical Ho:YAG SCFs have been made by this method [10,12,13]. In this paper, we have successfully grown Yb:Ho:YAG SCF using the LHPG method for the first time, to the best of our knowledge. Through the follow-up experiment we confirmed this SCF has the same structure as the YAG crystal bulk, with less than 0.5% diameter fluctuation.
2. Growth and experiments

2.1. Fiber growth

The 15 at.% \(\text{Yb}^{3+}/2.5\text{ at.}\%\ \text{Ho}^{3+}\) co-doped YAG bulk source material was grown using the Czochralski method [14]. A crystal bulk of square rods 1 mm\(^2\) × 40 mm size was used as the source rod. Two CO\(_2\) laser beams were used to heat the top of the source rod, the laser power was in the 15–30 W range. Further description of growth principles can be found in earlier publications [15,16]. These fibers were grown along <1 1 1> directions using a pre-grown undoped YAG SCF as seed. The seed was kept in the central axis of the source rod to remain roughly constant in diameter. The pulling rates ranged between 0.6 and 0.8 mm/h, with a rotation rate of 10 r/min. The ratio of pulling and feeding rates was chosen to obtain the diameter of the fiber, a number that is usually on the order of 3:1. The schematic of the LHPG method is shown in Fig. 2. The molten zone shown in Fig. 3 consists of a convex lens and a projection screen. The shape of the molten zone is related to the laser power, the diameter reduction, and the material being grown. Since the molten zone is held simply by surface tension, system vibrations, laser power fluctuations, and source rod diameter all have an enormous effect on the stability of the molten zone. Maintaining this stability after contact with seed is the key to growing high-quality single crystal fibers. In order to maintain the stability, we have adopted the following measures:

1. Strengthening the growth system. The foundation of the LHPG furnace reaches three meters, this effectively reduces the mechanical vibration of the devices; 2. Controlling the heating power. Higher power results in higher melting zone temperatures, and the surface tension decreases with increasing temperature, which will further lead to instability of the melting zone; 3. Maintaining the diameter ratio during growth. A large difference in diameter between the source rod and SCF will lead to drastic changes in the melting zone.

2.2. Experiments

The absorption spectrum in the range of 400–2000 nm was recorded by a Perkin Elmer Lambda 950 spectrometer. The fluorescence spectrum in range of 1800–2200 nm was measured under 915 nm LD pumping. The concentrations of Yb and Ho ions were
determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES) analysis. The X-ray powder diffraction (XRD) was used to investigate the structural relationship between single crystal fibers and YAG crystals. The Raman spectra were measured under excitation by 785 nm LD and recorded by Renishaw in Via spectrometer. The structure of the fiber was observed by a JEOL JSM-6390LV scanning electron microscope (SEM).

3. Results and discussions

For the best case so far, we obtained Yb:Ho:YAG single crystal fibers of length 124 mm, 450 μm at diameter, and the length-to-diameter ratio is greater than 270:1, as shown in Fig. 4(a). When the fiber length exceeds 140 mm it is difficult to maintain the stability of the melting zone, the diameter began to fluctuate sharply. This may attributed to the square shape of the source rod, the melt zone tends to become rounded under the liquid surface tension which affects stability. We believe that processing the source rod into rounded bars can effectively solve this problem. Fig. 4(b) shows the optical microscope photograph of the SCF, it is clear that the end facet of the fiber is hexagonal due to the melt supercooling at the growth interface [17]. The concentration of rare-earth element ions Yb$^{3+}$ and Ho$^{3+}$ is 12.65% and 1.67% respectively, which is close to the source crystal. This indicates that SCF growth with a small change in the concentration of doping ions can be achieved by the LHPG method.

The 400–2000 nm absorption spectrum of Yb:Ho:YAG are shown in Fig. 5(a). This means that through the transition of Yb$^{3+}$:$^{2}F_{7/2}$ → $^{2}F_{5/2}$ and energy transmission process from Yb$^{3+}$:$^{2}F_{5/2}$ to Ho$^{3+}$:$^{5}I_{6}$, the Yb:Ho:YAG crystal can be pumped by commercial 940 nm pumping sources. Fig. 5(b) shows the fluorescence spectrum of the Yb:Ho:YAG crystal bulk excited by 915 nm LD. It is clear to see that there are two strong fluorescence emission peaks, appearing at 2091 nm and 2122 nm.

The X-ray powder diffraction patterns Fig. 6 confirms that the structures of Yb:Ho:YAG SCF and YAG crystal bulk are almost exactly identical. The crystallographic data for the Yb:Ho:YAG SCF are a = 12.0679 Å, volume = 1.7575 nm$^3$, Z = 8, and p = 4.49 g/cm$^3$. 

Fig. 4. (a) Yb:Ho:YAG single crystal fiber. (b) Optical microscope photograph of end facet of Yb:Ho:YAG single crystal fiber.

Fig. 5. (a) Absorption spectra of the Yb:Ho:YAG crystal. (b) Fluorescence spectra of the Yb:Ho:YAG crystal excited by 915 nm LD.

Fig. 6. X-ray powder diffraction patterns of Yb:Ho:YAG single crystal fiber and YAG crystal.
which are close to the YAG crystal, indicating that the fibers grown by LHPG have a YAG crystal structure with only slight lattice distortion.

Fig. 7 shows the Raman spectra of the Yb:Ho:YAG SCF and bulk, the observed wavenumbers are given in Table 1. The main Raman peak of the Yb:Ho:YAG SCF is same as the crystal bulk and similar to the YAG. The peak position of the offset is also consistent with the conclusions found in literature, which mention that in the low-wavenumber region, the Raman peak shifts gradually to lower wavenumbers as the rare-earth ion content increases [18]. This shows that the structures of single crystal bulk and fiber are very similar, and the grown Yb:Ho:YAG SCF is of good quality.

In Fig. 8, the SEM microphotographs of Yb:Ho:YAG SCF shows, the diameter of fiber is very stable. The maximum diameter change shown in Fig. 8(c) falls within 0.5% range. This means that the LHPG method is suitable for the growth of morphologically stable SCF. In Fig. 8(d) the end face of the fiber presents a hexagonal shape, like we’ve seen in optical microscopes before. No grooves were found on the sides, which is common in the LHPG growth of the SCF [10].

![Fig. 7. Raman spectra of Yb:Ho:YAG SCF and bulk.](image)

![Fig. 8. SEM microphotographs of the Yb:Ho:YAG single crystal fiber. (a) overview, (b) and (c) side of fiber in different resolutions, (d) end facet.](image)

### Table 1
Wavenumbers (cm\(^{-1}\)) observed in the Raman spectra of Yb:Ho:YAG crystal bulk and SCF. (See below-mentioned references for further information.)

<table>
<thead>
<tr>
<th>YAG [19]</th>
<th>Yb:Ho:YAG crystal bulk</th>
<th>Yb:Ho:YAG SCF</th>
<th>Assignment</th>
</tr>
</thead>
<tbody>
<tr>
<td>139</td>
<td>130</td>
<td>130</td>
<td>Y or Yb translatory</td>
</tr>
<tr>
<td>220</td>
<td>217</td>
<td>218</td>
<td>Translatory + rotatory + (v_3(\text{AlO}_4))</td>
</tr>
<tr>
<td>261</td>
<td>260</td>
<td>260</td>
<td>(v_1=\text{v}_3(\text{AlO}_4))</td>
</tr>
<tr>
<td>298</td>
<td>311</td>
<td>312</td>
<td>(v_1=\text{v}_3(\text{AlO}_4))</td>
</tr>
<tr>
<td>373</td>
<td>370</td>
<td>370</td>
<td>(v_1=\text{v}_3(\text{AlO}_4))</td>
</tr>
<tr>
<td>401</td>
<td>399</td>
<td>400</td>
<td>(v_1=\text{v}_3(\text{AlO}_4))</td>
</tr>
<tr>
<td>715</td>
<td>719</td>
<td>718</td>
<td>(v_1=\text{v}_3(\text{AlO}_4))</td>
</tr>
<tr>
<td>783</td>
<td>783</td>
<td>783</td>
<td>(v_1=\text{v}_3(\text{AlO}_4))</td>
</tr>
</tbody>
</table>
4. Conclusion

In this paper, high-quality Yb:Ho:YAG single crystal fibers of length-to-diameter ratio exceeding 270:1 were successfully grown by the LHPG method. These fibers were found to be transparent and colorless with a smooth surface, and of 124 mm in length and 450 \( \mu \text{m} \) in diameter. The 0.5% diameter fluctuation shows that fibers grown by the LHPG method are of overall good quality. These fiber have the same structure and concentration of rare earth ions as the source crystal bulk, with only minor lattice distortion. All of the results indicate that the Yb:Ho:YAG single crystal fibers are very promising for laser applications. Detailed studies on growth and laser properties of Yb:Ho:YAG SCF have been carried out and reported.

Conflict of interest

The authors declared that there is no conflict of interest.

Acknowledgement

This research was supported by the National Key R&D Program of China (No. 2016YFB0701002).

References