Fabrication and optical properties of single-crystal YAG fiber optics

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ABSTRACT

Single-crystal (SC) fiber optics have been grown for many years for use as passive fibers for the delivery of IR laser radiation and as active fibers useful as minirod lasers. By analogy with doped-YAG, bulk laser crystals it is expected that pure YAG SC fibers would be capable of transmitting extremely high laser energies. In this study we report on the growth of SC yttrium aluminum garnet, Y$_3$Al$_5$O$_{12}$ (YAG), fibers from undoped SC YAG source rods using the Laser Heated Pedestal Growth (LHPG) technique. The YAG transmits IR wavelengths up to approximately 4 $\mu$m which is a little beyond the transmission range for SC sapphire fibers. The garnet family of crystals is one of the most commonly used oxide crystal hosts for lasing ions in high power solid-state lasers, with the most commercially common laser host being YAG. The optical losses for 400- $\mu$m diameter YAG fibers have been measured to be about 1 dB/m at 2.94 $\mu$m. The longest length of YAG fiber grown has been about 65 cm.

Keywords: Infrared fiber optics, single-crystal fibers, oxide crystal fibers

INTRODUCTION

SC fibers have been grown since the early 1980s with most of the work concentrating on passive (pure) SC sapphire fibers. There has also been some limited work on growing doped and undoped YAG and other garnet SC fibers. Essentially all of these fibers have been grown by the LHPG technique. In this method a CO$_2$ laser is used to melt the tip of a crystalline source rod and a fiber is pulled upward from the molten tip. The SC fibers have been unclad in the sense that there is no true fiber cladding as is commonly associated with the core/clad structures of glass fiber. The most common SC fiber studied to date has been sapphire or Al$_2$O$_3$. Sapphire is an insoluble, uniaxial crystal (trigonal system, hexagonal class). It is an extremely hard and robust material with a usable fiber transmission from about 0.5 to 3.2 $\mu$m. Sapphire and other oxide crystals have outstanding physical characteristics, which rival even those of silica. For instance, the Young’s modulus of sapphire is approximately 6 times greater than silica; the melting point is over 2,000 $^\circ$C; and it is extremely hard. It is very inert and it can be grown and stored in an ambient atmosphere. These properties of sapphire and similar properties for the other oxide crystals such as YAG make them good SC fiber candidates for applications from 2 up to 5 $\mu$m.

The key physical properties of sapphire, YAG, and other oxide crystal fiber candidates are given in Table 1. The properties of these oxide crystals clearly indicate the outstanding physical characteristics of these materials and how well they rival even those of silica.

Table 1 – Properties of some oxide crystals used for fabrication of SC fiber optics

<table>
<thead>
<tr>
<th>Material</th>
<th>Symbol</th>
<th>Structure</th>
<th>$M_p$, °C</th>
<th>$n$ @ 3 $\mu$m</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sapphire</td>
<td>Al$_2$O$_3$</td>
<td>Hexagonal – uniaxial</td>
<td>2040</td>
<td>1.709</td>
</tr>
<tr>
<td>YAG</td>
<td>Y$_3$Al$<em>5$O$</em>{12}$</td>
<td>Garnet – cubic</td>
<td>1940</td>
<td>1.788</td>
</tr>
<tr>
<td>GGG</td>
<td>Gd$_3$Ga$<em>5$O$</em>{12}$</td>
<td>Garnet – cubic</td>
<td>2098</td>
<td>1.915</td>
</tr>
<tr>
<td>Spinel</td>
<td>MgAl$_2$O$_4$</td>
<td>Octahedral – cubic</td>
<td>2135</td>
<td>1.667</td>
</tr>
</tbody>
</table>

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FABRICATION OF SC FIBERS

There are basically two methods for fabricating oxide crystal fibers. In both cases a seed fiber or wire is used to pull a fiber from a molten reservoir. In one method a high temperature crucible usually made of tungsten is used to contain the melt. The SC fiber is most often pulled up from the melt as pioneered by LaBelle or pulled down through a small hole in the crucible. The method of LaBelle and Saphikon, Inc. (now Photran) is called the edge-defined, film-fed growth (EFG) technique. The alternative is a crucibleless method, called laser heated pedestal growth (LHPG). This method was first developed by Haggerty at MIT and then further refined by a number of researchers at Stanford University, Bell Labs, Rutgers University, University of South Florida, and others.

The technique that we have used to grow YAG and other oxide crystal is the LHPG method. This crucibleless technique closely resembles the float-zone method of crystal growth. In the float-zone method, the molten zone is freely supported between the two ends of the crystal rod. LHPG is inherently the best technique for growing high optical-quality sapphire fibers because the molten zone is held in place by surface tension, eliminating the need for a crucible, which could be a possible source of contamination. Furthermore, a CO₂ laser beam, which provides a uniform, ultra-clean heat source, is used to melt the starting rod. Unlike the EFG technique, this growth method allows only one fiber to be grown at a time, so commercialization is difficult. LHPG, however, is the method used to produce the lowest loss sapphire fibers with losses approaching theoretical values at 3 μm.

In LHPG SC fiber growth, a CO₂ laser beam is focused onto the tip of a source rod creating a small molten bead of the oxide crystal source rod. A seed fiber is dipped into the molten region, shown schematically in Fig. 1, and slowly pulled upward forming the single-crystal fiber. The source rod, which may be single crystal, polycrystalline, sintered, or a pressed powder, is simultaneously fed upward to replenish the supply of molten material. The shape of the molten zone is a function of the laser power, the diameter reduction, and the material being grown. In general, the length of the maximum stable zone is approximately 3 times the fiber diameter. It is much more difficult to produce SC fibers with smooth surfaces than it is for a glass fiber. This is because the viscosity of a glass is very high during drawing whereas the viscosity of crystalline material at the molten zone is very low and, thus, sensitive to any minor perturbations in the system. Since the molten region is held in place simply by surface tension, any air currents, vibrations, laser-power fluctuations, etc. will have enormous effects on the stability of growth. For this reason it is necessary to use a very stable laser source; have a small molten zone; and grow with a source-to-fiber reduction ratio of 3:1.

The details of our LHPG apparatus used to grow our SC fibers have been previously published. A diagram of the key elements of the LHPG apparatus is shown in Fig. 2. In general, our LHPG system closely resembles the LHPG growth facilities at both Stanford University and the University of South Florida. Referring to Fig. 2, the key components are an amplitude stabilized, 50-W CO₂ laser beam, a laser micrometer for diameter control, and a reflaxicon optical system. Feedback from the laser micrometer is used to stabilize the laser power to ±0.5%. Approximately 10 to 20 W of laser power is needed to melt a 1-mm-diameter source rod. The growth rate is about 1 to 2 mm/min which translates to at least 8 hours to grow one meter of fiber. The seed fiber was either a YAG fiber or a Pt wire. For this work the final fiber diameters were about 400 μm when a 1-mm source was used. Smaller...
diameters can be obtained by regrowing these 400 μm fibers. The longest length of YAG fiber grown was 65 cm and all fibers were core only, i.e. unclad.

Figure 2 – Schematic diagram of LHPG apparatus showing the critical components.

OPTICAL PROPERTIES OF SCOXIDE FIBERS

IR absorption edge for oxide crystals

The IR absorption of most oxide crystals is well documented in the literature. In Fig. 3 we show the IR absorption edge for four different oxide crystals: sapphire, YAG, GGG, and yttria. The absorption at about 4.8 μm in YAG is due to multiphonon absorption and, therefore, an intrinsic property of the material. Yttria also has a weak multiphonon peak at 5 μm. As expected the IR edge is shifted toward longer wavelengths for oxides with the heaviest ions. For example, if we extrapolate from the IR absorption data the expected absorption for a fiber made from these materials, we would find that the loss for pure sapphire, YAG, GGG, and yttria is 18, 5.5, 0.5, and 0.15 dB/m at 4 μm, respectively. Based on these results it is clear that any fiber of a reasonable length, say greater than 1 m, would not transmit well beyond about 4 μm. The exception would be an SC yttria fiber. The difficulty with yttria is that it has a phase transition at about 100 °C below the melting point and thus it is hard to grow a single crystal of this material. In the past we tried to grow an SC yttria fiber starting with a polycrystalline (ceramic) source rod. In all cases we obtained small cracks in the outer diameter of the fiber indicative of the effects of the phase change in this material on cooling.

Figure 3 – IR absorption edge for bulk oxide crystals. Note the excellent transmission of GGG and yttria at longer wavelengths.
Spectral loss for SC YAG preforms

The spectral loss for the starting YAG source rod or preform is shown in Fig. 4. The loss measurement was taken using a Bruker Tensor FTIR spectrometer. The source rod diameter was 1 mm and the length 9 cm. As mentioned above, the absorption near 4.8 μm is due to multiphonon absorption. The two small peaks around 3.5 μm are associated with OH defects. The two absorption lines at 3.45 and 3.54 μm are very near hydrogen-bond absorptions for the dodeca-octahedral and dodeca-tetrahedral bond lengths. These absorption bands are also observed for the SC YAG fibers. In addition, the absorption lines at 2.944 and 2.972 μm in both the YAG preform and fiber are well documented to be due to hydrogen-bond absorption.\(^{13,14}\)

![Graph showing spectral loss for SC YAG preforms](image)

Figure 4 – Spectral loss for SC YAG source rod or preform used to grow the SC fibers.

Spectral loss for SC YAG fiber

The spectral loss for the SC YAG fibers that we have grown is given in Fig. 5. The as-grown fibers were 400-μm in diameter and 65 cm long. For the spectral loss data shown in Fig. 6, a short 6-cm long fiber section was measured. Referring to the spectral data we again see the OH defect impurity bands near 3 and 3.5 μm as we saw in the starting source rod. From the data it appears that we have not introduced any appreciable increase in the impurity concentration over what was present in the starting material.

![Graph showing spectral loss for SC fiber](image)

Figure 5 – Spectral loss for SC fiber.
Loss at key laser wavelengths for SC YAG fibers

The loss for the SC YAG fibers, 400-μm in diameter and 65 cm long, was measured at four laser wavelengths. All lasers were DPSS lasers with excellent output mode quality and amplitude stability. The Er:YAG laser was made by Sheaumann Lasers (Marlborough, MA) and the other lasers were from Opto Engine (Midvale, UT). The data in the top part of the table shows the data for the as-grown fiber and below the dashed line the data after the fiber has been annealed for about 12 h at 1,000 °C in air. The losses for the fiber decrease significantly after the annealing process. This trend has been observed by Chang, et al. for SC sapphire fibers. It is not known precisely why the loss decreases in the visible region but it is suspected that annealing in air somehow decreases the oxygen-related impurities. We also see, as did Chang, et al., that there is virtually no change in the fiber loss at 2.94-μm after annealing.

<table>
<thead>
<tr>
<th>SC Fiber Material</th>
<th>Laser</th>
<th>Wavelength</th>
<th>Loss, dB/m</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-grown YAG fiber</td>
<td>Green</td>
<td>535 nm</td>
<td>4.4</td>
</tr>
<tr>
<td></td>
<td>Red</td>
<td>635 nm</td>
<td>3.6</td>
</tr>
<tr>
<td></td>
<td>Nd:YAG</td>
<td>1.06 μm</td>
<td>1.9</td>
</tr>
<tr>
<td></td>
<td>Er:YAG</td>
<td>2.94 μm</td>
<td>1.1</td>
</tr>
<tr>
<td>YAG fiber after anneal</td>
<td>Green</td>
<td>535 nm</td>
<td>1.7</td>
</tr>
<tr>
<td></td>
<td>Red</td>
<td>635 nm</td>
<td>1.8</td>
</tr>
<tr>
<td></td>
<td>Nd:YAG</td>
<td>1.06 μm</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>Er:YAG</td>
<td>2.94 μm</td>
<td>1.1</td>
</tr>
</tbody>
</table>

CHALLENGES IN THE GROWTH OF SC OXIDE FIBERS

Growing any high purity SC fibers from materials like sapphire, YAG, GGG, or yttria, presents some serious challenges both from the viewpoint of the purity of the material after fiber growth and in the growth process itself. The data for YAG shows absorptions due to OH related defects in both the starting source rod crystals and in the as-grown fibers. These impurities and defects are present as two absorption peaks near 3 and 3.5 μm. It is unclear just why these occur or what we might do to eliminate these impurity absorptions. Annealing in air has reduced the attenuation in the visible portion of the spectrum for the YAG as well as for SC sapphire fibers. In future work it will be interesting to experiment more with different atmospheres in the growth chamber to see if it is possible to reduce this impurity absorption. An interesting observation by Chang, et al. was that growing the fibers in a He atmosphere enabled them to grow sapphire fibers at ten times the growth rate compared to the growth rate used when growing in ambient air conditions.

One of the biggest difficulties faced in the growth of these oxide crystal fibers is control of the fiber diameter. The situation is quite different for glasses because fiber is drawn from glassy materials which are in a highly viscous state. The melt zone in a crystal has a viscosity close to that of water so any vibration or irregular motion during growth can lead to a diameter fluctuation. In our LHPG apparatus we control the laser power by virtue of a feedback signal from the laser itself. The source feed and fiber pulling motors are attached to belt drives so that the source and fiber are moved very smoothly. In addition, the source rod passes through a small bore tube to help guide the rod smoothly into the melt zone. The fiber rides in Teflon coated V-groove again to prevent sideways motion of the fiber. The motors, laser power, and laser micrometer are all controlled by a LabView program. Yet we still have diameter control issues that occur as the fiber is grown. In Fig. 6 we show two microphotographs of the same YAG fiber. The difference is that in one case there is poor diameter control (A) leading to a diameter variation of about ±8% and in the other (B) there is excellent diameter control of better than ±1%. The variation in fiber diameter...
shown in Fig. 6 occurs as a result of some movement of the fiber or source rod that is, for some reason, not well controlled. Sometimes this occurs when the fiber goes into or out of the tractor feed mechanism or, less often, if the source or fiber slips just a small amount in their respective feed mechanisms. We are working to improve our drive controls which seem to be the most significant source of diameter fluctuations.

![Figure 6(A) – Poor diameter control](image1)

![Figure 6(B) – Good diameter control](image2)

**SUMMARY AND CONCLUSIONS**

Oxide crystal fibers have some distinct advantages over other IR fibers in the 2 to 5 μm range. Their extremely high $M_p$ makes them suitable for applications involving temperatures above 1,200 °C. They are very inert and non-toxic. However, most of the SC fibers made to date lack a suitable cladding and therefore some applications such as SC fiber lasers have been slow in coming. The emphasis in this work has been on passive SC fibers for the delivery of high power laser radiation. In principle, these fibers should have very high laser damage thresholds. This is particularly true of YAG fiber which, based on Nd:YAG lasers, should be capable of withstanding energy densities produced by cw or pulsed high power lasers. The losses for our YAG SC fibers are much higher than the intrinsic loss by a factor of about four at 3 μm. That is, for YAG we measure a loss at 2.94 μm of 1 dB/m compared to a calculated value of about 0.25 dB/m at this Er:YAG laser wavelength. We attribute the higher measured losses to fiber diameter fluctuations and impurities. The YAG fiber that we have grown to date is not as uniform in diameter as the sapphire fiber that we have grown in the past. Work is underway to improve the diameter control and also to minimize the defects and impurities by using purer starting material; annealing the fiber in air; and growing the fiber in a controlled atmosphere such as He or O$_2$.

**ACKNOWLEDGEMENTS**

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**REFERENCES**