Final Report

Reduced Parasitic Lasing in Ti: Sapphire Lasers. Removing a Bottleneck to New Ways of Acceleration

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to
Crystal Systems, Inc.
and
GT Solar Inc.

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Abstract

Powerful solid state lasers can be efficient sources of high energy electrons in a new concept of particle acceleration. One limitation to the power of these lasers is the phenomenon of parasitic lasing, in which oscillations transverse to the intended lasing direction can drain the stored energy and thus clamp the gain at some maximum value. This problem becomes more severe the larger the laser crystal becomes.

The source of the problem is the internal reflections in the larger sized gain material. This program was aimed at eliminating this bottleneck to more powerful lasers, to allow more intense high energy beams to be generated by this technique. The basic approach was to eliminate the internal reflections by making an absorbing layer on the outside of the crystal that had the same refractive index as the crystal. Thus, any transverse light would be absorbed before it reached the edge of the crystal and had a chance to reflect back into the interior.

During the first year, furnaces were renovated that and used to produce evaluation crystals. The first approach was to oxidize the outside surface of the titanium doped sapphire laser gain crystal. This oxidation is known to make the crystal be absorbing in the lasing wavelengths. A number of tests showed some modest absorption.

After the first year, 2 parallel paths were pursued to improve on this initial results – overgrowth and oxidation and grooving with index matched thermoplastic. Experiments on one of these techniques progressed to near-operational status as a commercial coating process. A test Ti:sapphire rod with a commercial coating technique was also supplied to the subcontractor for baseline testing.

The final product of the Phase II was a process that was reduced to a commercialized operation involving the complex series of steps of fabricating the crystal, polishing, grooving, providing an AR coating, then producing the absorbing layer and packaging the crystal in a manner that would allow easy handling for installation and deinstallation.

Near the end of the contract period, Crystal Systems was purchased by GT Solar (now GT Advanced Technologies.) This disruption resulted in the delay in the final report.

Introduction

Powerful solid-state lasers can be efficient sources of high energy electrons in a new concept in particle acceleration. Producing such novel sources of accelerated particle beams could have wide implications from fundamental high-energy physics to chemistry and to medicine. As an alternative to RF linear accelerators (linacs), laser driven plasma based accelerators are being studied at various universities and laboratories around the world. In analogy with RF linacs, the plasma based accelerator consists of an injector, an accelerating structure with longitudinal electric fields which provides mode control for the accelerating fields as well as guiding for the laser, and a suitable laser driver. The most significant differences between laser driven accelerators and RF accelerators are the two to three orders of magnitude larger longitudinal gradient and equally smaller wavelength of the accelerating fields. Accelerating gradients up to

100 GV/m have been demonstrated experimentally. Wavelengths on the order of 10-100 μm are used in laser driven accelerators compared to several centimeters in RF linacs. The high gradient offers the possibility to build compact high-energy accelerators. The short wavelength offers the possibility to produce intense ultra-short electron bunches. In addition, compact high intensity short pulse lasers are anticipated to have wide applications in medicine and chemistry, and fundamental research in materials science.

Experiments have demonstrated acceleration of electrons with energies up to 1 GeV by using 40 TW laser pulses and cm-scale plasma waveguides (1,2). The next major milestone is to achieve 10 GeV energy particle bunches containing on the order of 0.1 to 1 nC. From energy conservation arguments and simulations, reaching this energy level will require a laser system capable of delivering on the order of at least 40 J in pulse durations ranging from 30 to 100 fs. To achieve such laser pulses will require the development of a Petawatt Ti:Sapphire based chirped pulse amplification laser with a main amplifier that is pumped by at least 160 J of green pump beam and will require crystal amplifier sizes larger than 10 cm. For such large diameter Ti:Sapphire crystals a fundamental limitation in the amount of extractable energy can occur from clamping of the gain due to parasitic lasing.

Parasitic lasing in these applications is lasing due to scattered rays that propagate transverse to main pump beam. The scattered beams propagate towards the sides of the crystal and can reflect back in. Due to the long transverse path length in these large diameter crystals, they are efficiently absorbed and can cause spontaneous emissions. Only a small fraction of spontaneously emitted light needs to be reflected at the crystal surface in order to start parasitic oscillations along such paths. Parasitic oscillations reflect off of various surfaces inside the gain medium achieving long path lengths with low losses, resulting in substantial amplification and depletion of the stored energy. The net result is that gain is clamped at a low level, and the full amplification potential of the gain medium cannot be realized.

Simply stated, if the parasitic lasing problem was fully resolved, then the novel accelerator systems using these compact high intensity lasers would operate with significantly improved operational envelope in terms of power and intensity.

It was proposed during the Phase I that one way to achieve a reduction in the parasitic lasing would be to produce a Ti:sapphire crystal that had an outer region along the barrel with very high absorption of 800 nm radiation. These crystals are cylinders and the laser light is meant to move down the cylinder's axis. Light that reflects from side to side across the cylinder contributes to parasitic lasing. If the crystal had strong absorption of the scattered beam along the barrel of the crystal, this would minimize parasitic lasing. If the absorbing layer was an overgrowth of Ti:sapphire that had high absorption for 800 nm light, this crystal would take advantage of the high strength and high heat transfer properties of sapphire as well as its chemical inertness to virtually any coolant. By avoiding interfaces, this would avoid the Fresnel reflections that occur at the surface. Ti:Sapphire crystals of large diameter are routinely produced at Crystal Systems and used for laser applications. The main difference between currently produced crystals and that proposed is to increase the parasitic absorption of the main laser beam in select regions of the crystal which are not part of the clear aperture of the crystal. Proof of this concept was established during the SBIR Phase I program, where the capability was developed to process a rod for parasitic lasing, and still use it in a high power laser system. In the first year of the contract this concept was pursued, and another one was developed, as well. This second concept

involves using an index matched thermoplastic in which is mixed an absorbing species (as has been done before) but with machined light-trapping features in the barrel of the sapphire to enhance the effectiveness of this absorbing coating.

Anticipated Public Benefits

In addition to applications in HEP, the elimination of parasitic lasing would remove severe limitations in high power lasing in areas of medical, communications, and other fundamental areas of research. The dramatic rise in power densities achievable with high power, ultrafast lasers are only now becoming a commonplace tool, and already many potential applications are being pursued in biology, physics, and materials science.

If the problem of parasitic lasing can be solved, the gain can be increased to a point where the total amplified output energy will be limited only by the saturation fluence of the Ti:Sapphire material. This could also help to keep the full size of the optical amplifiers within a reasonable (table-top) range. Such high efficiency, high power density lasers developed as a result of this work can power simple, table top systems with tunable wavelengths, which can be used for medical applications such as laser accelerated proton therapy for cancer treatment, to cite one example.

Phase I Approach and Results

Overview

Scoping experiments from Phase I showed several clear paths to processing crystals to reduce parasitic lasing. The overall project structure has been to pursue these techniques for reducing parasitic lasing, then to test the effects of the treatments. The main way of testing these effects was to be to produce a test laser crystal with the best treatment and to send this crystal to LBNL to be tested in their laser system. By comparing the results to untreated crystals, it would be possible to evaluate the effectiveness of the treatment. Because of the effort needed to change laser crystals in the large LOASIS laser system at LBNL, it was also decided to analyze the effects of these treatments using ancillary measurements such as reflectivity. Also, modeling of the optical effects of the treatments was agreed to be performed at LBNL by an optical engineer who would be hired partially for this purpose. These activities were pursued in Phase II and a special jig was constructed to perform the reflectivity tests.

Technical Objectives for Phase II

The scoping experiments from Phase I were used to define the technical objectives of Phase II. The overall objectives of the Phase II project are to develop and scale up techniques for post growth processing of Ti:Sapphire rods that will result in substantial reduction or elimination of parasitic lasing by treating the cylindrical crystal in such a way that any light that scattered from the main path to the sides of the crystal did not reflect back into the crystal, but is absorbed, instead.

One technique that showed great promise from Phase I is a physical scoring of the barrel of the crystal to produce light trapping features. This technique has been evaluated, in conjunction with a variety of techniques for light trapping in the periphery to further reduce unwanted transverse reflections. One of the light trapping techniques is to use a thermoplastic that is a close match in refractive index to the sapphire with appropriate light trapping materials added. These have included carbon black, as discussed further below. Another technique is to add a fine powder made from finely ground heavily doped Ti:sapphire that has been optimally oxidized to maximize its absorbing characteristics. This sapphire has the added benefits of making the absorbing layer a composite material that more closely matches the Ti:sapphire in coefficient of thermal expansion and its thermal conductivity.

The parasitic lasing tests will be performed by the LOASIS Laser Facility at Lawrence Berkeley National Laboratories (LBNL), as was done for the Phase I. This is done by treating a suitable amplifier crystal which is then incorporated into a well instrumented high power laser at LBNL. The amplifier crystal is then pumped with increasing levels of energy until the gain is clamped by the parasitic lasing. This level is compared to the gain clamping of the same or a similar untreated rod to evaluate the effect of the treatment on the parasitic lasing.

This facility is ideally suited for characterizing the effects of processing on the laser crystal because of their highly instrumented cryogenic (100 K) amplifier system. This instrumentation includes beam profile monitoring cameras, remote controlled alignment options, beam-pointing stabilization loops, and an array of pulse measurement tools. The crystal will be supplied to LBNL in a laser-ready package after characterization at CSI and after superpolishing and coating at vendors. Beam energy measurements will be the primary means of determining the increase in gain allowed as parasitic lasing is reduced. This data will be utilized for improving the parasitic lasing reduction procedures. Samples may also be provided to other laser users for evaluation in their setups or laser systems.

The specific technical objectives of the Phase II and the status of these objectives are discussed further below.

<u>Technical Objectives, and Year 1 + 4 month status.</u> *Italics comments are from the previous report.* ***Bold and italics are for this period***.

1. Perform large diameter growth runs in HEM Ti:Sapphire furnace to produce Ti:Sapphire material of size necessary for testing. *One run has been done. One Ti:sapphire candidate rod has been fabricated.* ***This rod has been coated with a commercial coating to produce baseline results. This coated rod has been sent to LBNL for evaluation of parasitic lasing.***
2. Retrofit crystal growth furnace for performing overgrowth runs. This will involve changes to the heat exchanger, as well as other parts. *Completed*. Perform overgrowth experiments in experimental furnace to quantify the parameters necessary for quality overgrowths, including anneals and cooldown steps. These experiments will be aimed at optimizing thickness, concentration, temperature, time, growth rates, and annealing and cooldown conditions. *Retrofits completed for both the crystal growth furnace and the experimental furnace. Shakedown run completed in the crystal growth furnace and first overgrowth test run completed at large scale.* ***X runs at the 20 mm scale performed in the experimental***

furnace, optimizing parameters for that scale. System scaled up to crystal growth furnace, and Y 100 mm runs have been performed. Progress made in overcoming crucible, parameter, and dopant control issues.

3. Specify and purchase controlled atmosphere furnace. This includes all safety, emissions permitting, engineering and facilities work. Install the furnace, commission it and operate it to perform controlled oxidations of Ti:Sapphire test material. *Furnace manufacturer identified. Manufacturer also performs experiments on a toll basis and this will be the most cost effective approach at this point to evaluate the furnaces ability to achieve the desired oxidation.* ***No further action taken at this time.***
4. Determine operating parameters to yield optimal controlled oxidation of crystal. This will involve thermodynamic and solution calculations as well as gas chemistry calculations to produce initial conditions. *Ideal solution calculations have been performed. If experiments confirm results no more sophisticated solution models will be needed.* Follow on work will involve experimental work with Ti:Sapphire material to confirm the oxidation state, crystal quality, and diffusion effects. This will include fabricating test materials, and fabricating and polishing samples for laser absorption and UV-Visible analysis, and performing diffusion analysis. Consultations with laser experts at LBNL and other institutions will ensure that contemplated treatments are consistent with the production of high quality laser crystals. *Consultations with laser experts at LBNL and LOA as well as other laser experts at commercial institutions have been useful in directing the development of scoring techniques as well as suitable absorbing coatings.* ***No further action taken at this time***
5. Identify crucible types necessary, spec and purchase based on results of the optimum relationship between crucible and crystal from Phase I, and step 2, above. *In process. W crucibles necessary. These have been spec'd and will soon be purchased.* ***W crucibles purchased and small scale runs completed with successful results. Larger scale crucibles purchased, and are soon to be run, now that parameters are better optimized.***
6. Grow doped overgrowths onto fabricated laser rods, varying dopant concentration and thickness as well as growth parameters to arrive at the optimum combination of thickness, dopant and growth parameters to achieve suitable absorption at 800 nm energy. Test for α_{514}, α_{800} and crystal quality. *TBD* ***Overgrowths performed on fabricated surrogate sapphire rods. Parameters being optimized to produce homogeneous, stress free overgrowths.***
7. Perform controlled oxidations on Ti:Sapphire rods with high alpha overgrowths, and consult with laser experts at LBNL on the optimal treatment. *First meeting held at LBNL to review progress, including 9, below.*
8. Process a treated 40 mm rod with the optimized treatment to produce a laser rod. This rod will then be tested at LOASIS at LBNL for measurements of parasitic lasing. *A 40 mm rod that might be a candidate for subsequent treatment has been fabricated.* ***The barrel of the rod has been treated by a vendor using a commercial coating for reducing parasitic lasing. This will serve as a baseline test for parasitic lasing reduction to judge further improvements against. This rod has been delivered to LBNL where it is awaiting testing.***
9. Explore alternate techniques for reducing parasitic lasing, such as using FAST to machine light trapping features in the outside edge. *Major progress has been made in this area and this is being actively pursued as well. A new approach is to use the in house capability of slicing many parallel grooves in sapphire to produce light trapping features to be used alone or in conjunction with the overgrowths. This is discussed further below.* ***This technique has***

been continually improved, and matured towards a commercially viable method to treat rods. Thirteen rods have been fabricated and used, in many cases multiple times to develop the operation of coating and . Issues dealt with include geometry of the light trapping features, temperature of the coating process, optimizing the absorbent content, incorporating the process into the workflow of producing a finished Ti:sapphire rod, and protecting the super-polished faces and the AR coating throughout the process. Other issues being investigated include finish packaging for a laser-ready rod.

Progress Results

The overall goal of the Phase II will be to produce a 40 mm rod with demonstrably improved parasitic lasing, and a technique that can be applied to larger crystals, as well. The technique will be demonstrated by processing a rod with the developed technique, and measuring the output energy vs. pump energy to determine the effect on parasitic lasing. The end result of the Phase II will be a reliable technique for post growth processing of laser rods to eliminate parasitic lasing. This will be pursued in Phase III as a service or as an integral part of the CSI product line.

The progress to date for individual tasks as well as new findings are discussed briefly, below.

YEAR 1 PHASE II RESULTS

Perform Overgrowths

Overgrowth tests have been conducted both in the R&D furnace used in Phase I, as well as in a retrofitted larger production furnace. Much of the work of the overgrowths will involve proprietary parameter optimization. The overgrowths are essentially small scale crystal growth with highly doped systems. During these experiments, both furnaces required a rebuild due to broken elements which cost a run and further delayed the start of the overgrowth experiments.

The overgrowth approach consists of optimizing growth parameters of time, temperature, pressure, crucible size, dopant concentration and annealing conditions to produce highly doped overgrowths of high optical quality. The results from these runs are shown below.

Gh-26

This was an attempt to produce an overgrowth of Ti sapphire on a small cylinder of sapphire 12.3 mm diam by 61mm long using much heavier Ti dopant than had been tried in the past. The run parameters used were based on results from the Phase I. After the run was over, the cylinder was sectioned. The top 2/3 of the rod was well coated, however, the bottom third the overgrowth did not adhere.

Fig. 2. Top 2/3 of the rod from overgrowth run Gh-26. The irregular dark specks are accumulations of very late stage opaque material on the outermost surface.

Fig 3. The bottom 2/3 of the Gh-26 rod showed poor adherence and fracturing.

Gh-27

Based on the results from Gh-27, This was an attempt to produce an overgrowth of Ti sapphire on a cylinder of sapphire (12.2 mm diam by 61.5 mm long) using the same heavy Ti dopant, but using modified parameters to produce better results at the base of the rod.

After the run, the rod was recovered and sectioned, similarly to Gh-26. This rod was in one piece for the whole length, and displayed good adherence of the Ti:Sa overgrowth across the entire length of the rod. Although some gas bubbles at the lower end prevented Ti:sapphire material from filling in at the bottom, the improved adherence and lack of fracturing except at the very bottom were indications of success.

Fig. 4. Run Gh-27 post run. The adherence to lower levels was improved over Gh-26, as well as the interface quality.

Based on these results, another run was planned, however, it was discovered that the element on the R&D furnace had broken. While it was being replaced, it was decided to try a larger scale run on a production scale furnace. This furnace was refurbished, and an overgrowth experiment was tried on a large scale (78 mm diam by 56 mm high) sapphire crystal. This is actually a larger scale than will be initially tried with a Ti:sapphire crystal. This crystal was recovered and sectioned for examination after the run.

As can be seen in Figure 5, there was some fracturing of the overgrowth on this large scale run. (Fractures inside the crystal are a function of residual stress, as they generally happen after the cylinder is sectioned. This shows the need for more time for an in-situ anneal.). However, as a first attempt at an overgrowth at this scale it was considered encouraging, and good adherence to the rod from top to bottom was achieved.

Fig. 5. Result of large scale overgrowth run. The two cracks inside the crystal occurred after slicing and are no doubt due to residual stresses. This indicates the need for a longer in situ anneal.

Controlled Oxidation of Outer Rind

Results from the Phase I indicate the requirement for a gas mixing furnace to produce the controlled oxygen fugacity needed for the controlled oxidations. Thus a necessary first step will involve identifying a controlled oxidation state furnace using flowing gas mixtures. These furnaces are standard use in metallurgy and geochemistry and utilize mixtures of flowing gases to fix the P_{O2} or partial pressure of oxygen. This is achieved by using gas reactions such as

$$CO_2\ (g) = CO\ (g) + 1/2\ O_2(g)$$

Thus, CO and CO_2 gas reactions can control the oxidation state of the flowing gas, and thus of any solid in the gas stream that equilibrates with that gas. Another way to control the CO and CO_2 ratios is through the use of $H_2 + CO_2$ mixtures, and reactions such as

$$H_2(g) + CO_2(g) = CO\ (g) + H_2O\ (g)$$

Because these gases are poisonous and/or flammable, significant health, safety, and environmental planning needs to be done before commissioning such a furnace. Crystal Systems has identified a manufacturer of such a furnace that permits high temperature and controlled oxidation. This manufacturer also performs toll experiments for customers using their in-house furnace. CSI will perform the scoping experiments with controlled oxidation on a toll basis at this manufacturer's facility, and based on these results and the resulting economic conditions either use this manufacturer as an outside vendor or purchase and bring in house this capability. .

The controlled oxidation will be tested on a variety of concentrations and sizes, and the rates of oxidation, and the nature of the diffusion profiles will be determined. Test coupons will be constructed from varying concentrations of Ti. A run which has produced highly doped sapphire has been done, and the run products are shown below. Although this run fractured, this may not be a problem for fabricating small test coupons of varying high dopant level for the tests. The

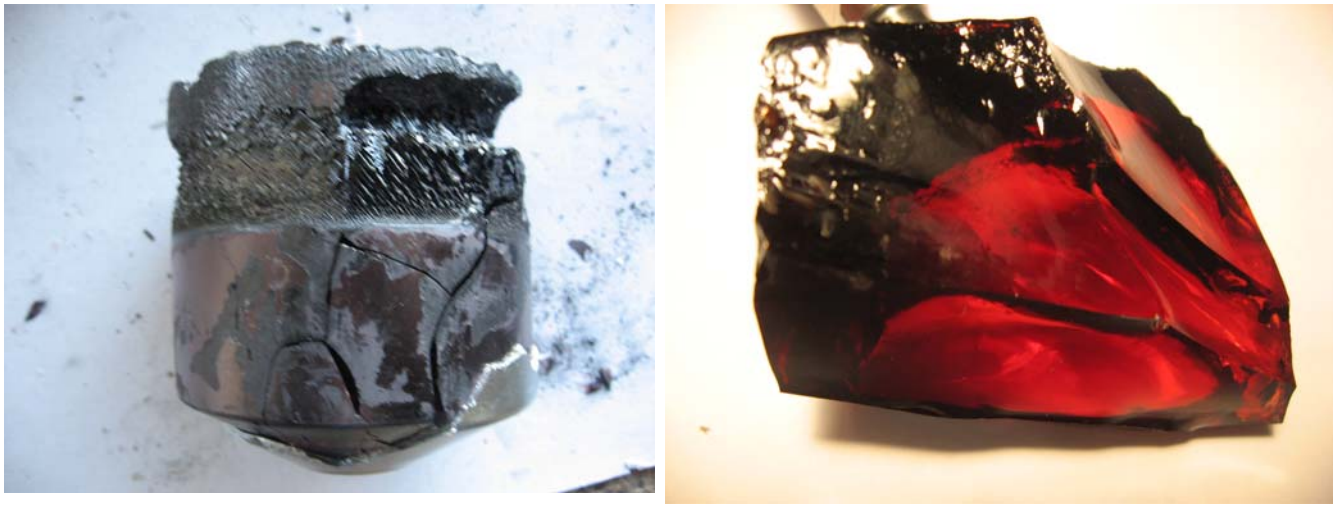

Figure 6a &b. Very high dopant Ti:sapphire fabrication run. This boule fractured (6a) and went polycrystalline at the top due to extreme levels of Ti2O3 due to segregation. However, it is likely that this boule will successfully yield very high doped test pieces for the controlled oxidation studies(6b) as well as the study of the effect of initial α_{514} level on the ultimate α_{800} level, as described in the next section.

Effect of initial α_{514} level on the ultimate α_{800} level

A major part of the work in Phase II will be aimed at developing the proper relationship between thickness, concentration of Ti, and oxidation level to produce the optimal level of absorption at 800 nm in overgrowths. To that end, a better understanding of the interactions between Ti^{3+} Ti^{4+} and the overall concentration needs to be developed at higher concentrations than have been previously studied. Material has been grown for this purpose and is being fabricated for further oxidation studies. The results of this study will allow better confirmation of the curve shown in Figure 7, which demonstrates the non-linear nature of the effect of initial Alpha 514 on the Maximum achievable Alpha800 in Ti:sapphire. This will allow precise prediction of the effects of oxidation on the absorption of the parasitic lasing energy by Ti:Sapphire of various dopant levels.

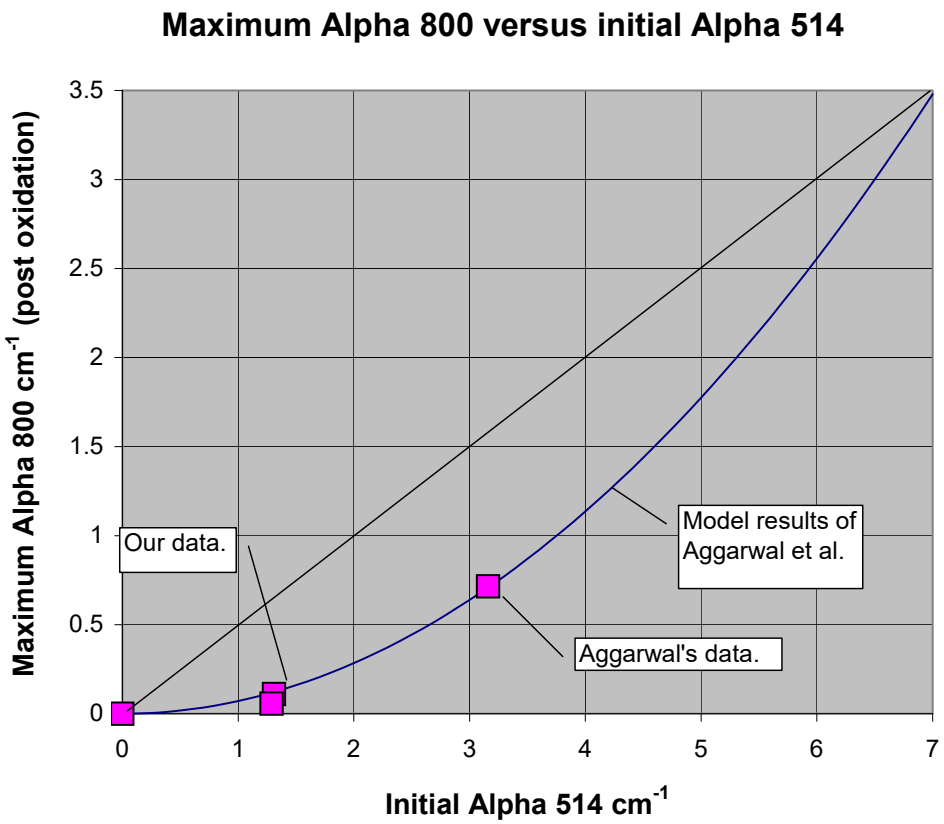

Figure 7. Maximum alpha at 800 nm after oxidation versus initial α_{514} (Ti concentration). Initial alpha is a measure of Ti concentration, as virtually all the Ti is in 3+ before oxidation.

In order to extend this model to concentrations required to produce the needed absorption affects high doped Ti:Sapphire overgrowths were to be performed in Year 2 so that α_{514} and α_{800} measurements can be evaluated. These data will be used to extend the model to much higher values of initial α_{514}. Being able to predict these effects will be a major tool in proceeding to the optimal condition of doping, overgrowth thickness and quality, and final absorption.

Grooved surfaces with index matched thermoplastic with additives

One of the most interesting results from the Phase I was the sensitivity of the parasitic lasing to the roughness of the outside surface, as well as the blackening of the outside of the crystal barrel, using a simple black magic marker. This sparked the approach of developing light trapping features using in house capability at CSI for slicing sapphire, namely the Fixed Abrasive Slicing Technique (FAST). FAST is a proprietary technology developed at CSI under a successful SBIR program, and is currently in commercial operation slicing sapphire. Slicing with FAST is accomplished rotating the workpiece while feeding it down into reciprocating wires that have diamond abrasive fixed to them (Figure 8). The diamonds are plated onto the

wire, and water is used as a coolant. The diamonds cut the work piece and protect the wire from wire wear. (See Figure 9.) By operating the FAST for to slice to different depths, wire pitches and diameters, it is possible to provide a wide range of groove widths and depths.

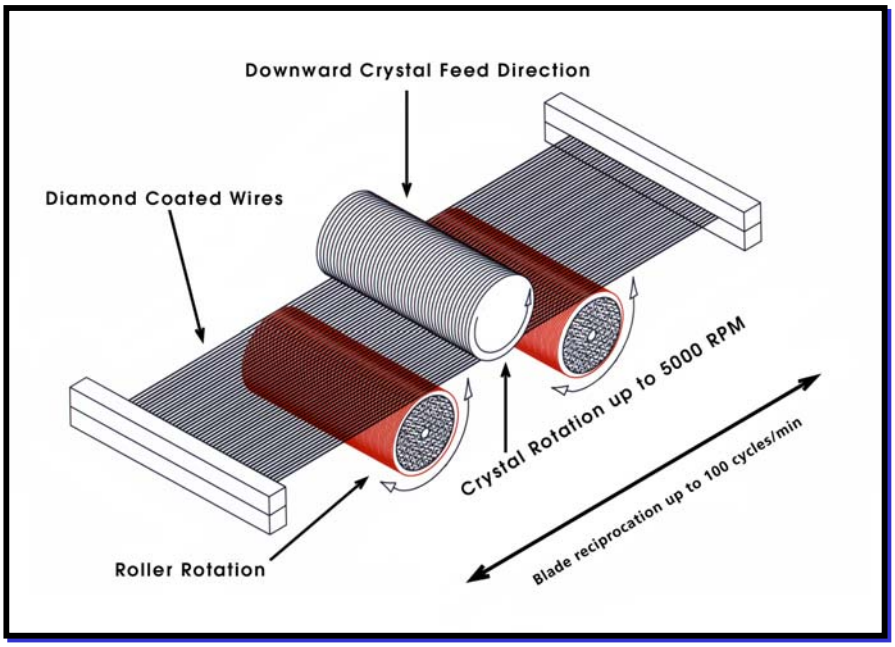

Figure 8. Schematic of the FAST machine showing the technique for grooving the light trapping features in cylindrical crystals of Ti:Sapphire.

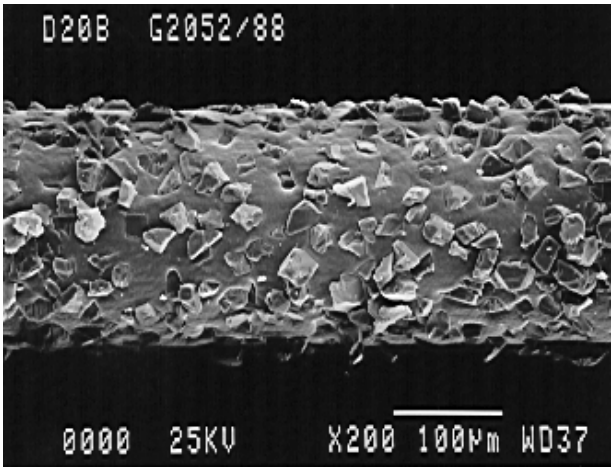

Figure 9. Photomicrograph of FAST wire with plated (fixed) diamond abrasive.

An example of rod that has been grooved is shown in Figure 10

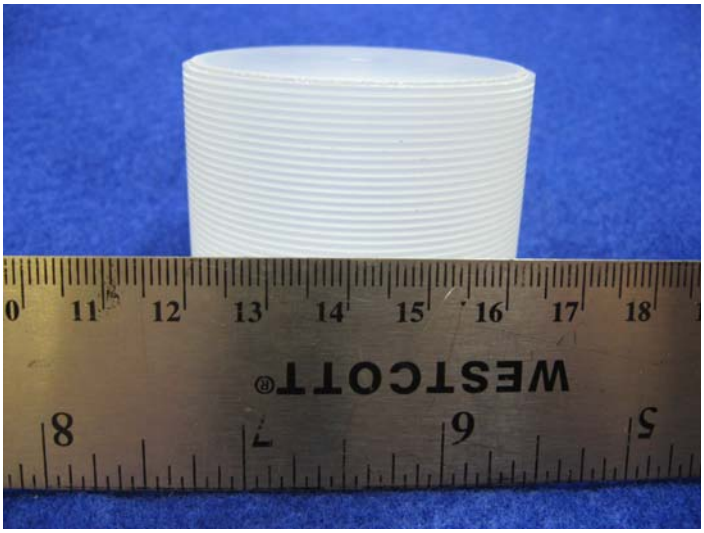

Figure 10. Grooved sapphire piece to produce light trapping structures, using the FAST slicer.

The further application of an near index matched thermoplastic to which various amounts of carbon black has been added yields distinct light trapping features, especially for light reflecting at near 90 degrees to the cylinder axis, which is the most deleterious light from a parasitic lasing point of view. An example of a grooved sapphire rod treated with thermoplastic to which has been added carbon black is shown in Figures, 11, 12, and 13. Figure 14 is a grooved treated rod which has been sliced down the cylinder axis and polished, to allow examination of the light reflection near normal to the cylinder axis.

Figure 11. Grooved sapphire piece # 1.

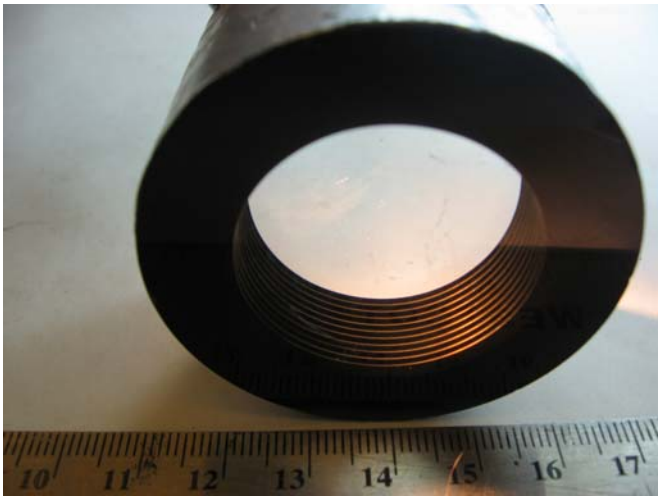

Figure 12. At low enough angle, the insides of the groove show some reflectivity, but this is not evident at higher angles. Grooved sapphire piece # 2.

Figure 13. Grooved sapphire piece # 3. With an improved formulation, there is less reflectivity from the sides.

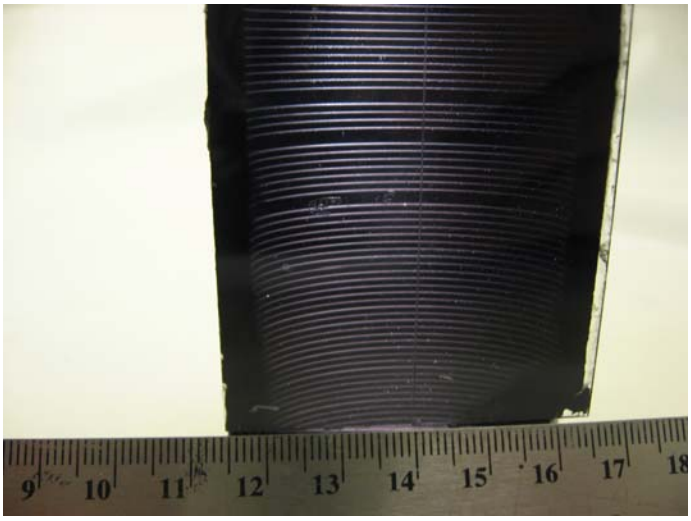

Figure 14. Grooved sapphire piece # 4. This piece was sliced parallel to the cylinder axis., then polished to examine the effects of light direction on reflectivity. Note that at low angles, the grooves bottoms have relatively greater reflectivity than the thermoplastic. This application was not optimized to thoroughly impregnate the thermoplastic into the grooves.

Figure 15. Ungrooved coated sapphire rod. The ungrooved rod appeared to have higher reflectivity than the grooved rod. Reflectance measurements will be made to quantify these effects.

The proper amount of carbon black to use in the thermoplastic was a subject of significant research. The absorbing ability of different mixes of thermoplastic with carbon black was ascertained by mixing known amounts of carbon black with the thermoplastic, then making mounts on glass or sapphire substrates of known thicknesses and analyzing the transmission

using a Cary 50 UV-Visible Spectrophotometer. Some of these samples are shown in Figure 16, and the results are shown in Figure 17, below.

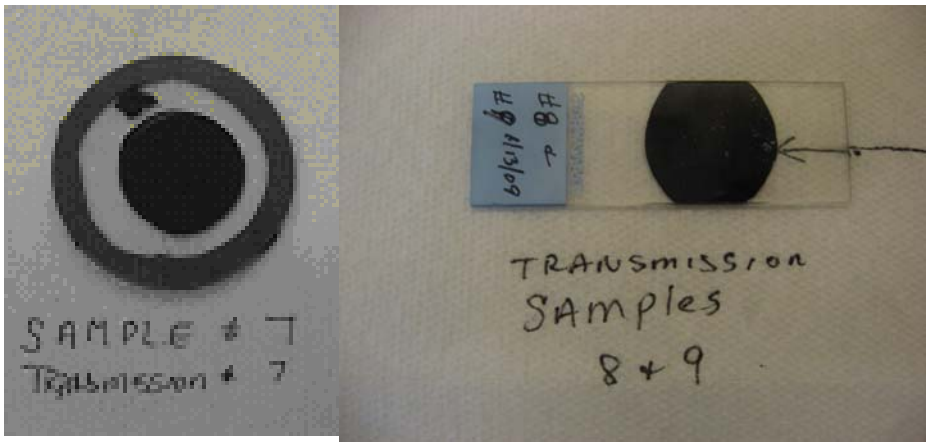

Figure 16 Transmission samples of thermoplastic with different wt. % carbon black with sapphire and glass substrates.

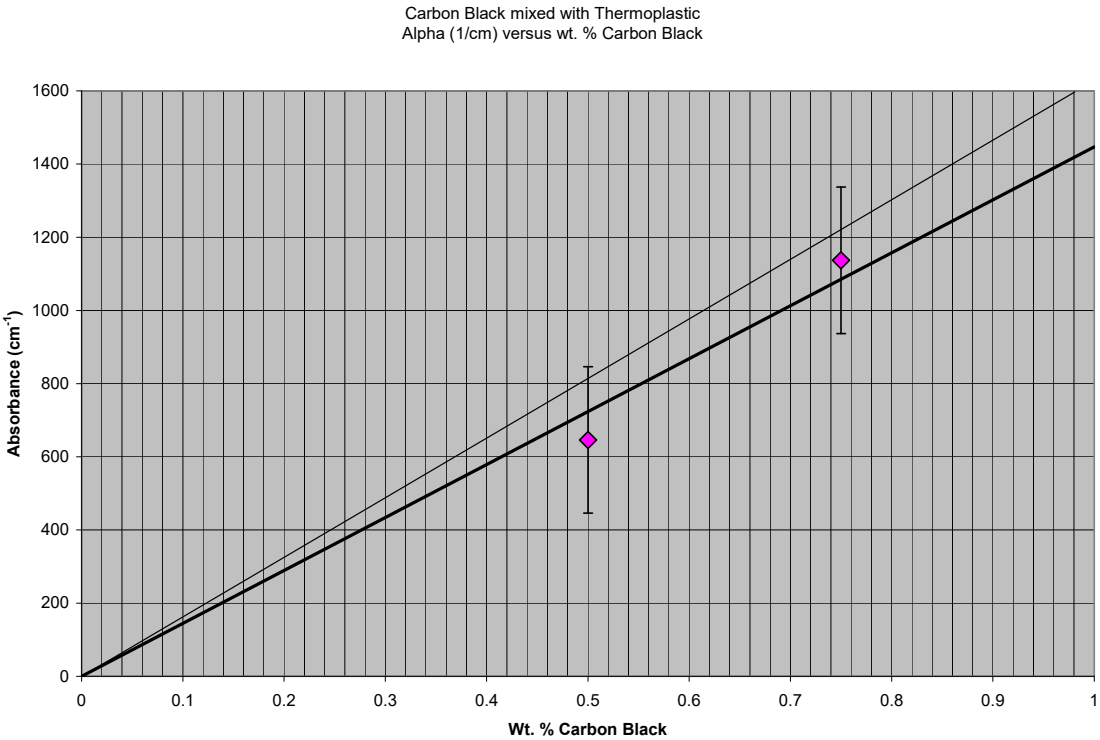

Figure 17. Absorbance of thermoplastic at 800 nm versus wt. % carbon black.

YEAR 2 PHASE II RESULTS

Due to the modeling and experimental results showing the limitation of the oxidation approach, the initial promising results from the overgrowth and the limited grooving experiments, this approach was focused on in Year 2.

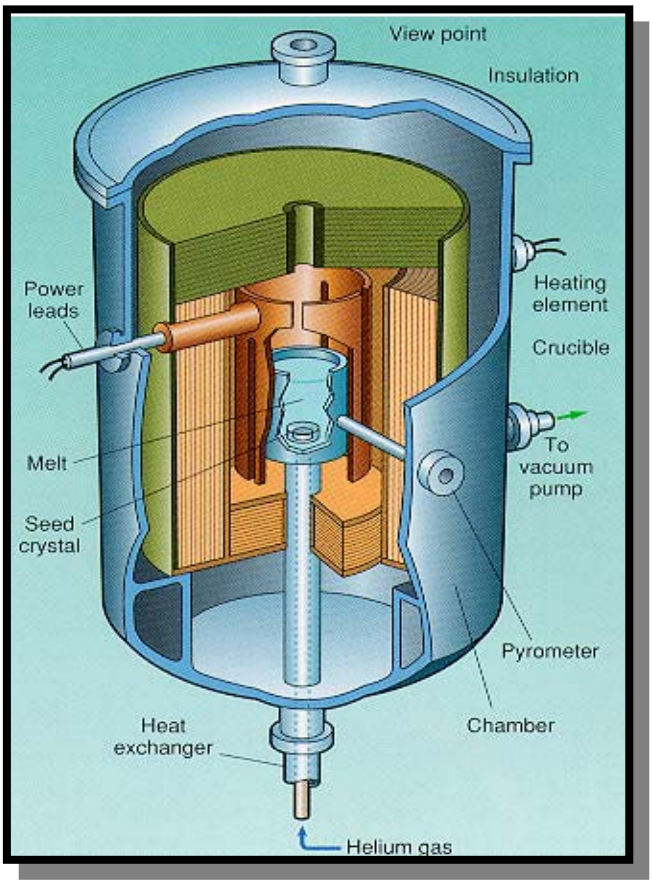

Figure 2. Schematic of an HEM furnace.

Perform Overgrowths

Overgrowth tests have been conducted both in the R&D furnace used in Phase I, as well as in a retrofitted larger production furnace (Figure 1). Six runs were performed in the experimental furnace, and five runs were made in the crystal growth furnace during this reporting period.

Much of the work of the overgrowths involves proprietary parameter optimization. The overgrowths are essentially specialized crystal growth runs with highly doped systems.

The overgrowth approach consists of optimizing growth parameters of time, temperature, pressure, crucible size, dopant concentration and annealing conditions to produce highly doped overgrowths of high optical quality. Several issues with the quality of the overgrowths were hypothesized to be due to the crucible interacting with the highly doped sapphire liquid. In order to improve these results, W crucibles were tried on these small scale runs. The results were encouraging, and the shift to W crucibles was made.

Another parameter that has been difficult to control is the amount of meltback. After melt and growth parameters were optimized, it was realized that variations in dopant were causing variations in meltback. This has led to process development that will be aimed at better control of the dopant concentration.

Some results from these runs are shown below.

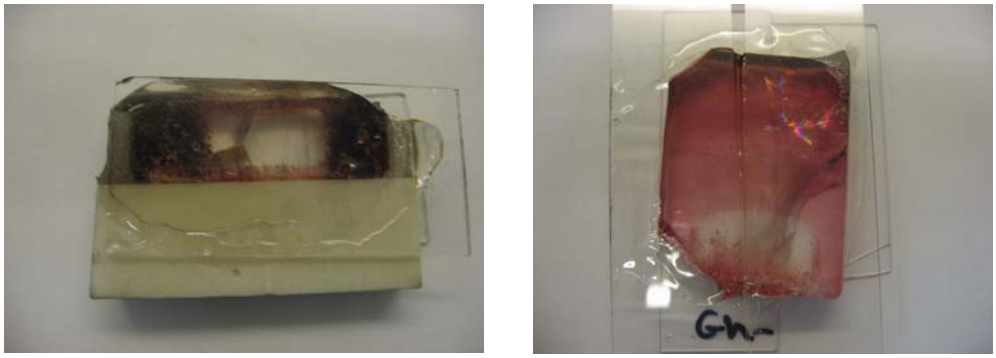

Fig. 3a and b. Gh-29, and Gh-30 W crucibles. Parameter optimization and the crucible allowed improvement in the quality of the overgrowth. The degree of meltback of the crystal was the target of several runs.

Fig. 4a and b. Gh-28 and Gh-32. However, inconsistent results indicated the importance of controlling the process better through better dopant control.

Following the small scale experiments, the results were taken to a full scale crystal growth furnace. Some examples of the results of the 5 runs performed are shown below. The fracturing is due to shortened anneals, and will be one of the last parameters to be optimized. Overall there

has been good progress in improving the quality of the overgrowth. However, reproducibility of the dopant will be the focus of the next series of runs.

Fig 5 a, b and c. Runs E-81, 84 and 85 respectively showing cross sections taken from these 100 mm runs. This figure shows the trend in improving the nature of the overgrowths. Fracturing is still a function of the short cooldowns, and will be addressed last.

The thermodynamic modeling and of the experimental overgrowth attempts clearly showed that for the very high absorption, very high Alpha 800 values would require very high initial Alpha 514 values. However the overgrowths were more difficult the higher the doping value, and this approach was considered to not be as efficient as the index matched thermoplastic approach that is described in Year 2 Phase II results.

Grooved surfaces with index matched thermoplastic with additives

One of the most interesting results from the Phase I was the sensitivity of the parasitic lasing to the roughness of the outside surface, as well as the blackening of the outside of the crystal barrel, using a simple black magic marker. This sparked the approach of developing light trapping features using in house capability at CSI for slicing sapphire, namely the Fixed Abrasive Slicing Technique (FAST). FAST is a proprietary technology developed at CSI under a successful SBIR program, and is currently in commercial operation slicing sapphire. Slicing with FAST is accomplished rotating the workpiece while feeding it down into reciprocating wires that have diamond abrasive fixed to them (Figure 6). The diamonds are plated onto the wire, and water is used as a coolant. The diamonds cut the work piece and protect the wire from wire wear. (See Figure 7.) By operating the FAST for to slice to different depths, wire pitches and diameters, it is possible to provide a wide range of groove widths and depths.

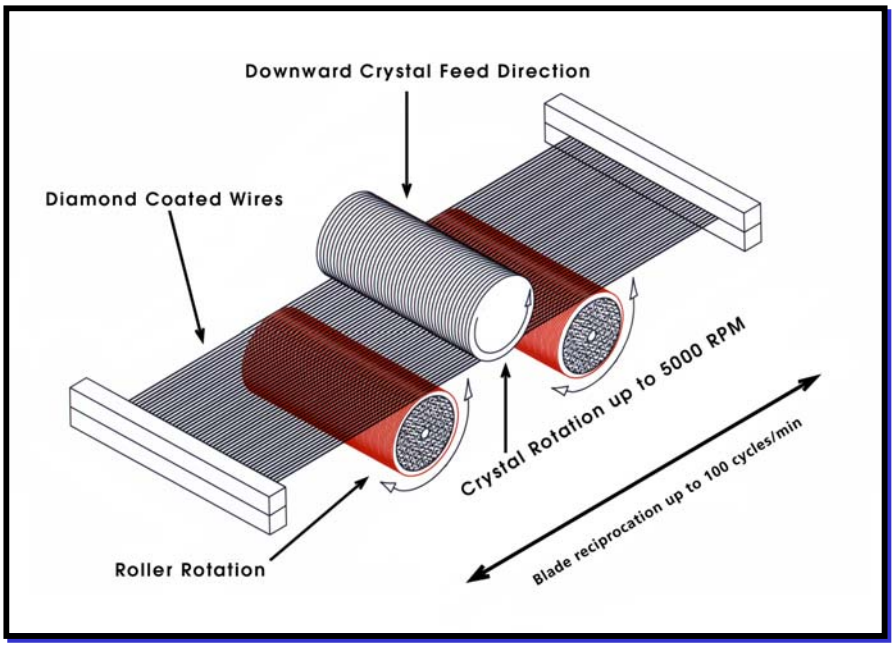

Figure 6. Schematic of the FAST machine showing the technique for grooving the light trapping features in cylindrical crystals of Ti:Sapphire.

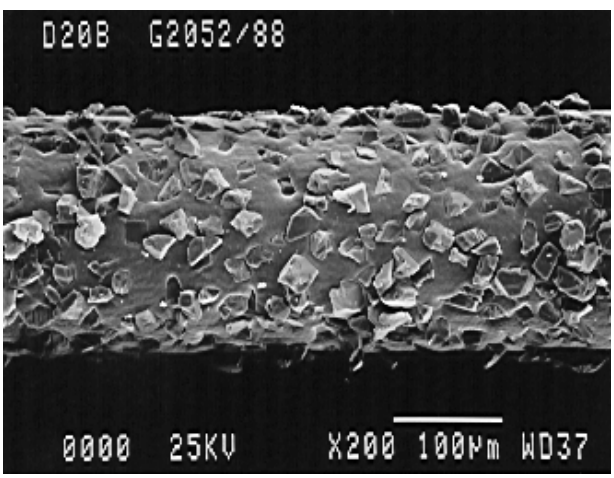

Figure 7. Photomicrograph of FAST wire with plated (fixed) diamond abrasive.

The further application of an near index matched thermoplastic to which various amounts of carbon black has been added yields distinct light trapping features, especially for light reflecting at near 90 degrees to the cylinder axis, which is the most deleterious light from a parasitic lasing point of view. .

This work is continuing, focusing on operational issues of applying the coating in a way that excludes bubbles, that are found to increase the reflectivity of the inner surface. This process has evolved to include special jigs and coatings to protect the faces of the superpolished and AR

coated faces of the crystal (Figure 8 and 9). Other issues which have been addressed are methods of encapsulating the rod to protect the coating from damage during handling and installation.

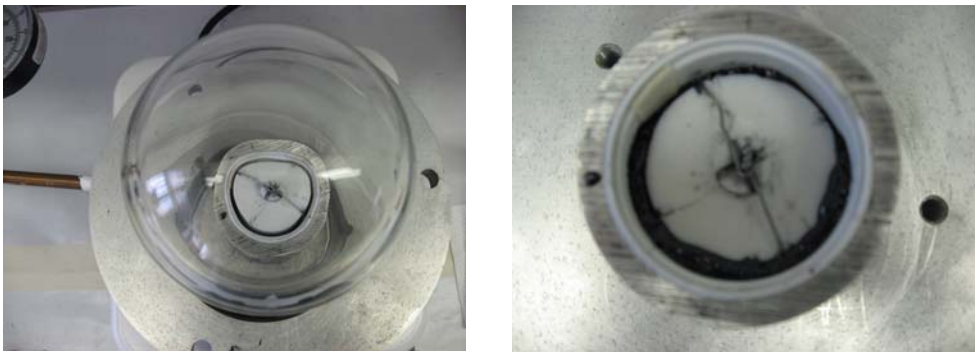

Fig 8. Processing of sapphire rods to apply thermoplastic coating in grooves. a. View of vacuum bell jar showing jig and container for the sapphire piece. b. Closeup of jig in cup showing thermoplastic flowing over the top.

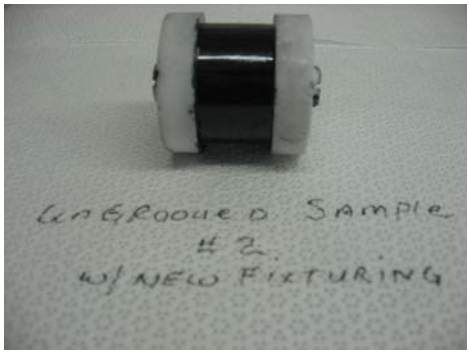

Fig. 9 After processing in the sample holder, the jig and the sapphire test piece are removed, and cleaned before the jig is removed from the test piece. The jig secures the rod, and protects the faces of the sapphire during processing.

During the period since the last progress report eight grooved, and five ungrooved samples have been fabricated, polished and treated, some of them several times over. (Another area investigated is the ability to clean off the coating and reapply without damaging the crystal to allow recovery from process upsets without risking the valuable Ti:sapphire rod.) Several examples of these are shown in the figures below.

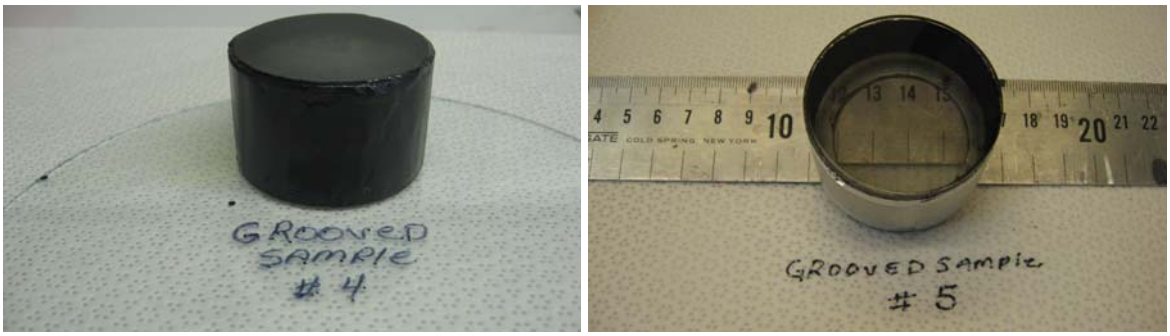

Figure 10 a and b. Grooved Sample #4 and # 5. The goal of these runs was to fill all of the slotted areas with a light-absorbing layer without having bubbles trapped inside. Results: The heated mix and the spacers seemed to allow a faster encapsulation of the work piece and after cleanup there are very few bubbles and good light absorption.

Figure 11 a and b. Part of the process is encapsulating/packaging the rods for shipment and installation without damaging the coating.

Figure 12. Ungrooved samples have been used to allow testing of the effects of surface preparation on the reflectivity, because they allow larger surfaces to be seen and tested.

Reflectivity testing at LBNL.

During this reporting period, progress was made in designing and producing a test chamber for performing reflectivity and operability tests on coated sapphire and Ti:sapphire. This progress is discussed further below.

Preparation of experiments for meltmount cooldown and reflectivity change tests:

A Design drawings:

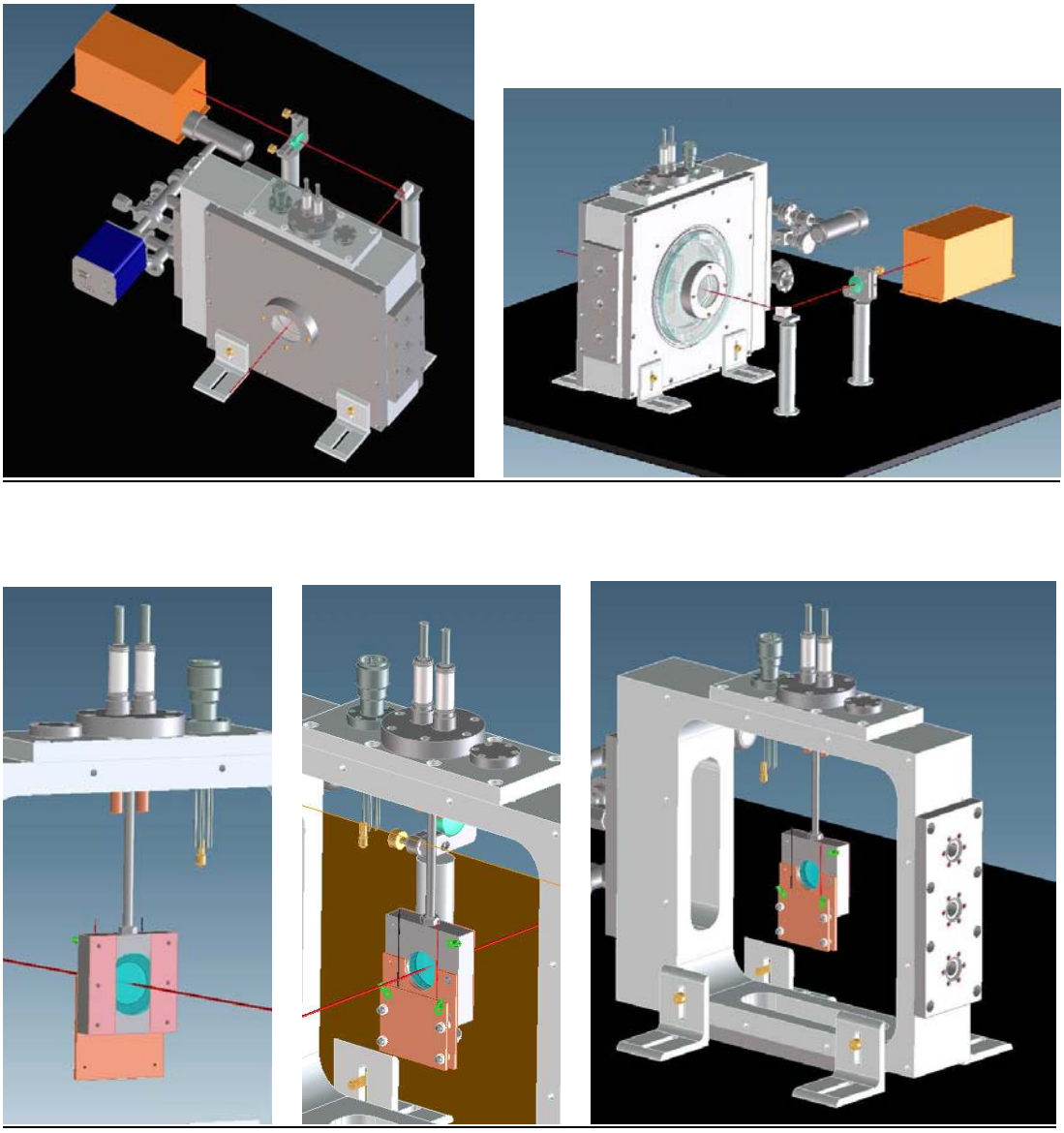

Figure 1. CAD models of cool-down test chamber assembly for laser reflectivity testing at LOASIS, LBNL.

B: Experimental chamber implementation:

To perform reflectivity measurements and to study the temperature dependence of the index matching properties and practical usability of various compounds with sapphire, we have designed and built an experimental chamber seen on Figure 2.
This index-matching compound is to surround the sapphire plates so that light is able to pass through this element just as it does through sapphire, even at cryogenic temperatures. In an investigation we are planning, two sapphire plates containing a sample of this index-matching fluid are to be surrounded by a vacuum chamber. A laser beam is able to pass through the sample at room temperature, but at much lower temperatures it is not guaranteed that the fluid will maintain the same index of refraction and homogeneity/translucency.

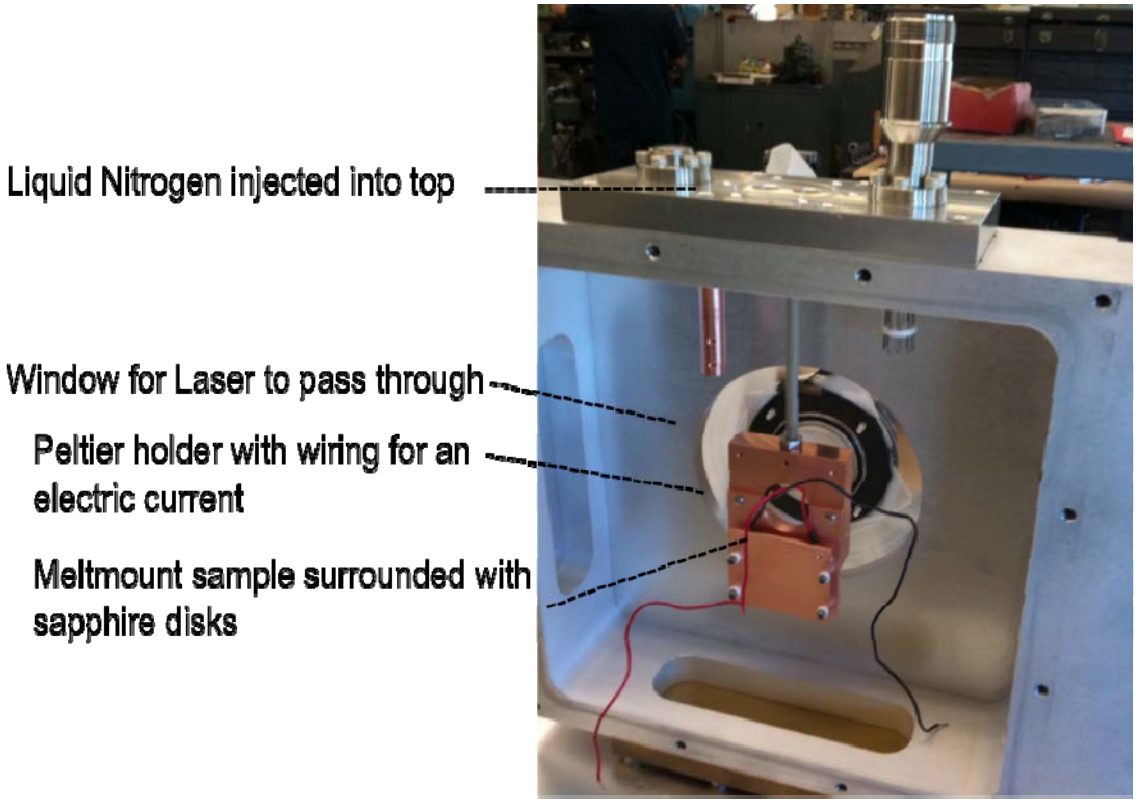

Figure 2. Cooldown test chamber assembly being prepared for laser reflectivity testing at LOASIS, LBNL.

Cooling methods planned to be investigated:

- Peltier elements:

Thermoelectric cooling uses the Peltier effect to create a heat flux between the junction of two different types of materials. A Peltier cooler is a solid-state active head pump which transfers heat from one side of the device to the other side against the temperature gradient (from cold to hot), with consumption of electrical energy. This Peltier effect will be used to cool the meltmount to see its behavior at lower temperatures.

- Liquid Nitrogen:

Liquid Nitrogen is a cryogenic fluid that causes rapid freezing. Liquid Nitrogen boils at a temperature of -196 C. Cryogenic fluids such as Liquid Nitrogen are able to get the contents of the chamber much colder, much faster than the Peltier elements. If the index of refraction does not change too much, even at cryogenic temperatures, there should be no problems with using this index matching fluid.

C: Preliminary experiments: Index Matching Fluid Cold Test

The goal was to determine the change in the refractive index differential between sapphire and Meltmount, a fluid designed to have the same refractive index as sapphire, as their temperatures were lowered. We have cooled these substances inside a pumped-down, and subsequently dry-N2 filled container using a Peltier and water-cooling system. The relative amplitudes of the 800 nm diode laser light reflected off the surfaces of contacts between the sapphire and Meltmount were measured with a photodiode and an oscilloscope setup. The diagram of the setup showing the various surfaces investigated can be seen on Figure 3, initial results on the function of measured target temperature are shown in Table 1. The results indicate, that the reflectivity from the meltmount/sapphire interface remains below 10^{-4}, while the temperature of the samples decreases from room temperature to below -7 ^{0}C.

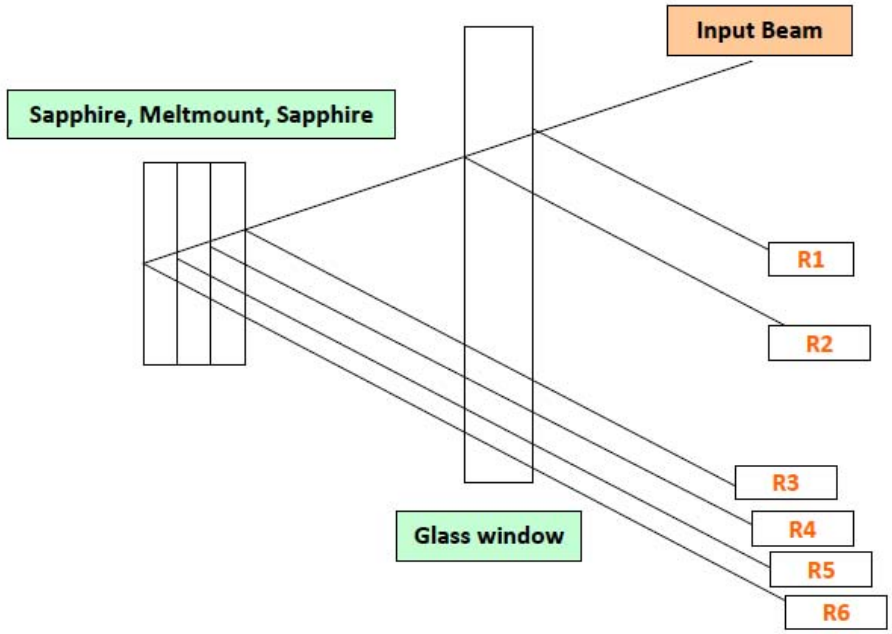

Figure 3. Schematics of investigated surfaces in a vacuum-enclosed sapphire/meltmount/sapphire system. R1 and R2 are reflections from the window of the vacuum chamber, they served as calibration reference.

Table 1. First raw data of initial reflectivity measurements of sapphire/meltmount interfaces at various temperatures. Further measurements and calibration needed for precise reflectivity data. Full size laser beam diameter corresponds to ~ 3 mm.

Reflection	**Voltage (mV)**	**Temperature (^{0}C)**	**Beam Diameter (mm)**
R6	63	22	Full Size
R3	22	22	Full Size
R3	2	22	1 mm
R3	40	22	Full Size
Incident Ray	1000	22	Full Size
R3	3	7	1 mm
R6	4	7	1 mm
R4+R5	0.5	7	1 mm
R3	3	-6	1 mm
R4+R5	0.1	-5	1 mm
R4+R5	0.1	-7	1 mm
R4+R5	0.1	22	1 mm

Conclusion

Significant progress has been made during this period in producing a commercially viable process for producing light trapping features, and filling these features with an optimized mix of thermoplastic and absorber to reduce internal reflections leading to parasitic lasing. The test chamber for analyzing these features at LBNL has been fabricated and tested. Rods have been supplied for testing in this system. Also, the experimental furnace has been successfully used to optimize parameters and test the crucible at a small scale, and these results have been transitioned and scaled up to the larger crystal growth furnace. Significant dopant control issues have been discovered, and are being addressed. These issues have implications for control of normal Ti:sapphire growth, and could represent a major side benefit from this project. A Ti:sapphire rod has been fabricated and a commercial coating has been applied. This rod has been supplied to LBNL for baseline testing of parasitic lasing . Further work will focus on alternative absorbents, improving the final packaging of the Ti:sapphire product to make it acceptable for use in high power systems, to improve the dopant level control, and to begin to perform overgrowths on large diameter Ti:sapphire rods.