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AFAL-TR-75-64

Part 1

AD B008909

ER: YLF LASER DEVELOPMENT

SANDERS ASSOCIATES, INC.
NASHUA, NH 03060

DECEMBER 1975



TECHNICAL REPORT AFAL-TR-75-64, Part I
INTERIM REPORT FOR PERIOD 1 MAY 1974 — 31 OCTOBER 1974

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This technical report has been reviewed and is approved for publication.

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20. ABSTRACT (Cont'd.)

Large, uniform diameter boules of Er^{3+} :YLF with high optical quality have been grown. An inert (argon) atmosphere and automatic diameter control were critical elements of the growth process. Boule 106Yb had a diameter variation of only ± 0.20 mm (diameter 16.69 mm) over the central 85 mm of length.

Improved optical quality was obtained by hydrofluorination of the molten feed at 900°C . Additional feed preparation procedures have been investigated for obtaining LiREF_4 feed of uniformly high purity starting with commercial Li_2CO_3 and RE_2O_3 .

Preliminary Q-switch data is repeated using KD*P and a Brewster plate polarizer. Improved efficiency is predicted with increasing Er^{3+} concentration (up to 10% Er). The index of refraction has been measured from 0.225 to $2.6\ \mu\text{m}$ and found to differ from earlier measurements,

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FOREWORD

This is the first ARPA Technical Report of Contract F33615-74-C-1194 covering the work performed to evaluate the 0.85 micron solid state laser material Erbium:Yttrium Lithium Fluoride. This work is sponsored by the Advanced Research Projects Agency under ARPA Order 2075 00 02, AMD No. 3. The inclusive dates of the research reported herein are 1 May 1974 through 31 October 1974.

Mr. Richard L. Remski (TEO), Air Force Avionics Laboratory, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio is the Project Monitor of this program.

These studies were carried out at Sanders Associates, Defensive Systems Division, Merrimack Facility. Subcontracting services were provided by the Center of Material Sciences and Engineering, Crystal Physics Laboratory, Department of Electrical Engineering, Massachusetts Institute of Technology.

Dr. C. S. Naiman, Manager - Laser Systems Department is the Project Supervisor and E. P. Chicklis and R. C. Folweiler are the principal investigators. Crystals were grown in the Sanders Associates Fluoride Growth facility under the direction of R. C. Folweiler with the assistance of Mr. L. Terrenzio. Mr. J. C. Doherty assisted with laser measurements. Subcontracting efforts conducted at the Center of Material Sciences and Engineering, Department of Electrical Engineering, MIT, were directed by Dr. A. Linz. Support of the crystal growth effort was provided by Dr. D. R. Gabbe; Dr. H.P. Jenssen, and D. Castleberry assisted with some of the spectroscopic measurements.

The views and conclusions contained in this document are those of the authors and should not be interpreted as necessarily representing the official policies, either expressed or implied, of the Advanced Research Projects Agency or the Air Force Avionics Laboratory of the United States Government.

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Er³⁺

1.0 SUMMARY

The results of the first six months of a fourteen month program entitled "Er³⁺:YLF Solid State Laser Development" are reported. This work is a continuation of Contract F33615-C-72-2065. The program goals are:

- Growth of 0.25 inch diameter by 3-inch length laser rods with scattering less than $0.5\% \text{ cm}^{-1}$;
- Q-switched operation at a PRF of 10 Hz with an output power in excess of 1 watt; beam divergence less than 5 milliradians at full angle, 90% energy points; and
- Operation at an overall efficiency of 0.5% in the Q-switched mode.

During the first phase of this two phase program efforts were focused on crystal growth and material improvement. Q-switched measurements were made with emphasis on resonator design. The major accomplishments of this phase of the program are:

- Growth of large, uniform diameter boules of Er³⁺:YLF, of high optical quality using a larger furnace and computer operated process control has been successful. The most recent boule has a diameter of $16.69 \pm 0.20 \text{ mm}$ over the central 85 mm of length.
- Scatter-free, undoped YLF has been grown in an argon atmosphere from commercial fluoride feed material by treatment of the compound under HF at 900°C .
- Development of a purification process for commercial rare earth oxides and their conversion to anhydrous fluorides has been carried out.

- Q-switched operation of $\text{Er}^{3+}:\text{YLF}$ using KD^*P and a Brewster plate indicate a loss of about 40% in output compared to long pulse data in an open resonator.
- Increased laser efficiency with concentration for up to 10% Er is predicted in flash pumped operation when the pulse width is maintained below the upper level lifetime.

2.0 CRYSTAL GROWTH

Crystals were grown for this program using Sanders Fluoride Crystal Growth Furnace at the company's Merrimack, New Hampshire Facility shown in Figures 1 and 2. The crystal growth equipment has performed particularly well, with few problems because of extensive shakedown operations performed as part of the installation process that detected the usual problems associated with new, complex apparatus of this nature. The crystal growth apparatus has operated continuously for days completely unattended, permitting greater attention to other aspects of the growth process. The process and the results are described in the following sections.

2.1 REVIEW OF GROWTH TECHNIQUE

The Sanders' fluoride crystal growth equipment and process evolved from the work done at MIT in the Crystal Physics Laboratory.⁽¹⁻³⁾ We use a nonstoichiometric melt composition, nominally 0.52 LiF and 0.48 YF₃ on a molecular weight basis, to avoid the peritectic at the stoichiometric composition. Feed material is prepared by hydrofluorinating the rare earth trifluoride at 800°C in flowing HF, and cooling in Ar. The rare earth is then combined with the stoichiometric proportion of LiF, then melted and frozen under flowing HF. The hydrofluorination is performed in an Inconel reactor, using a vitreous carbon boat to hold the feed. The feed is subsequently transferred to the growth furnace where it is outgassed at 600°C and 10⁻⁶ Torr, then melted under an atmosphere of high purity argon at 20 PSIA (140 kN/m²) in a platinum crucible.

A new melt invariably has a surface coating of carbon; recent melts have had as little as 30% of their surface area covered. The material probably is pyrolyzed organic solvents used in processing the rare earths since the hydrofluorination process does not affect carbon. The carbonaceous material has the appearance of soot. A "scum" crystal is grown on a platinum wire in a manner that causes

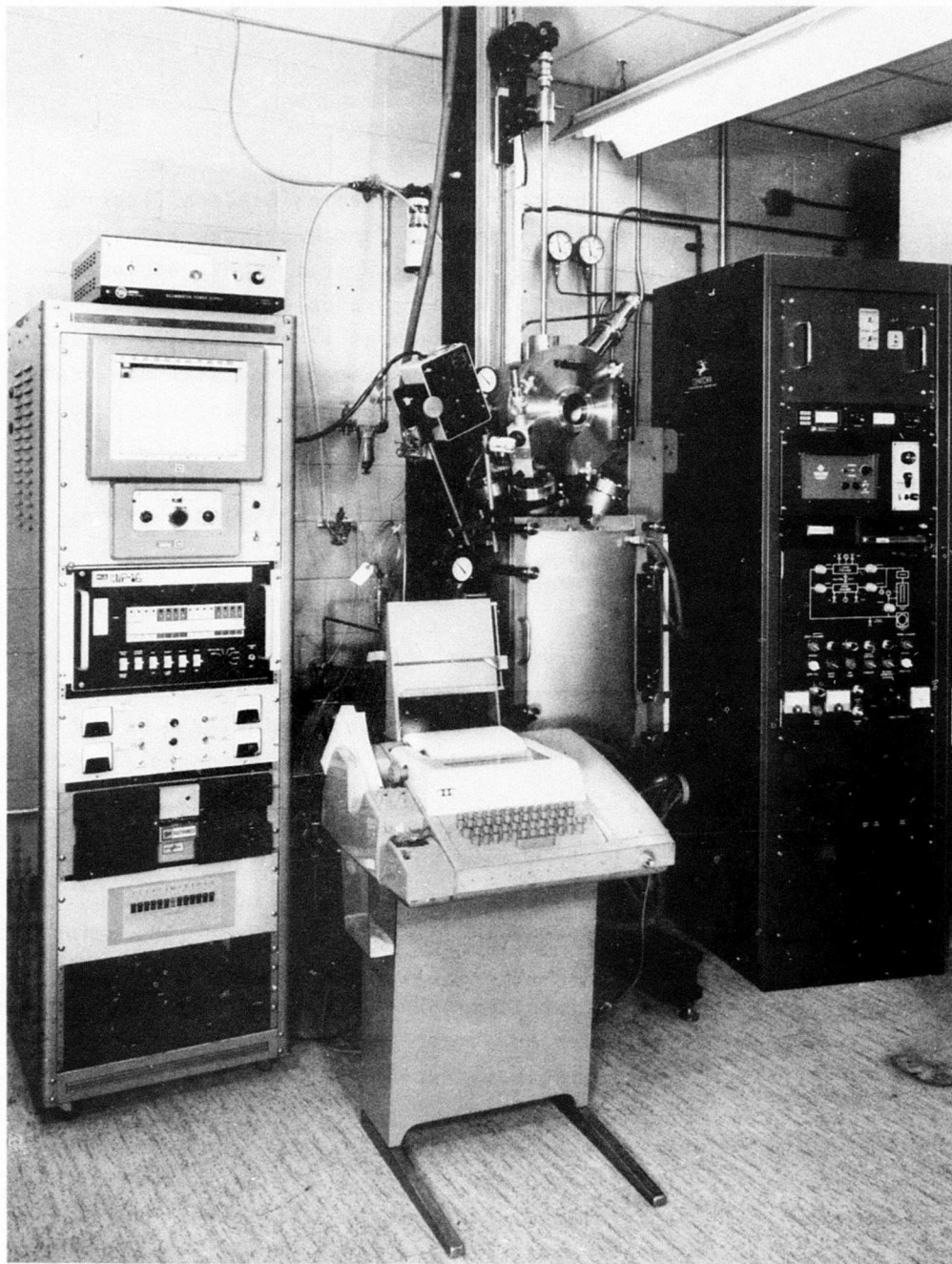


Figure 1. Sanders automated fluoride growth facility.

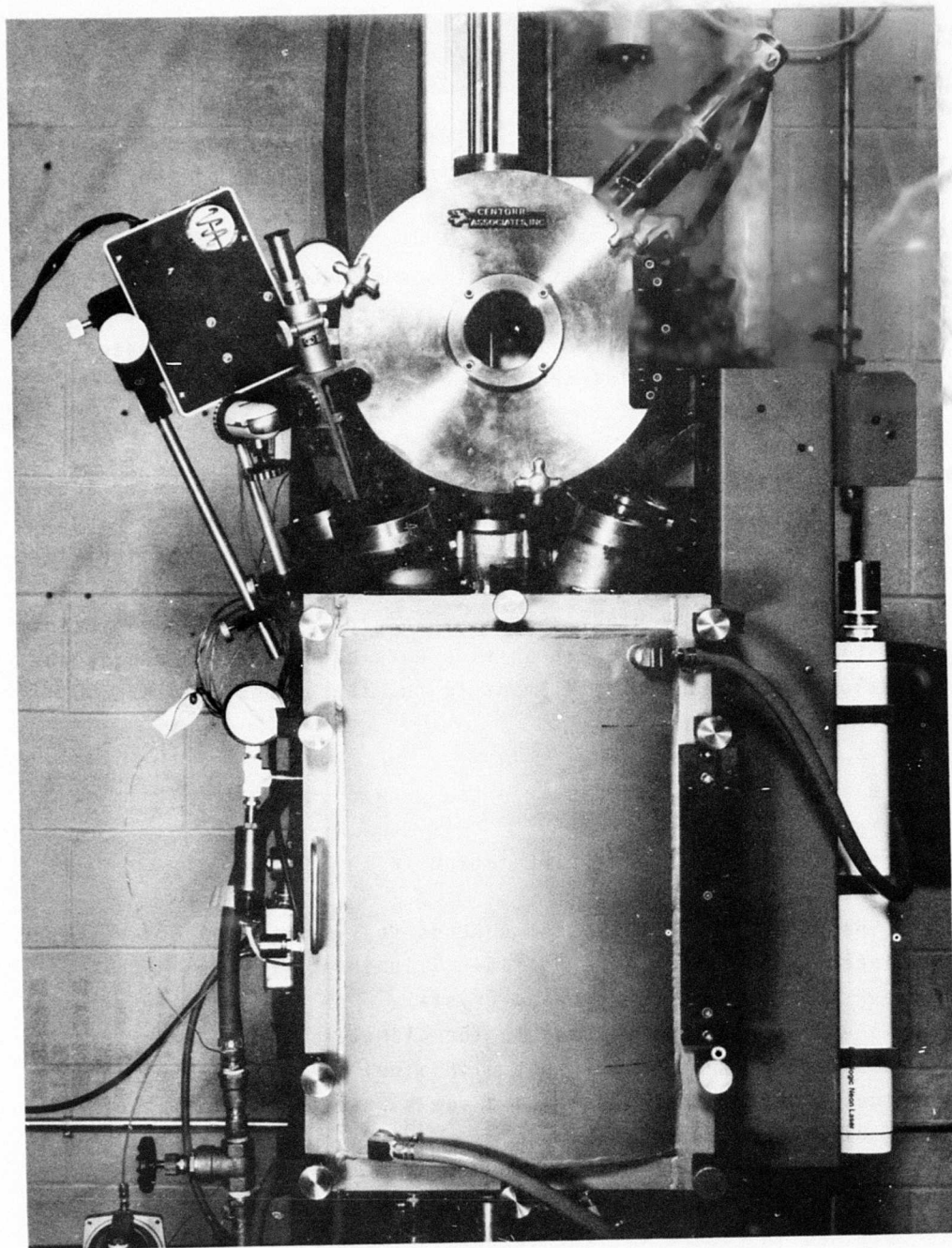


Figure 2. Exterior view of growth furnace.

the carbon to adhere to the surface of the crystal; the scum crystal is subsequently removed through the interlock system of the Sanders furnace.

A seed of pure LiYF_4 , attached to a molybdenum adapter with platinum wire, is then introduced through the interlock of the furnace system. The adapter is mounted on the end of the water cooled seed rod to provide the thermal gradient necessary for controlled growth.

The furnace is constructed of stainless steel with water cooled walls using vacuum fabrication techniques and sealed with O-rings. The main (growth) chamber and the interlock chamber can be individually pumped with a conventional LN_2 trapped diffusion pump. The heating element is machined graphite, insulated with five molybdenum heat shields. Auxiliary heat shields have been added to reduce the radiation from the heating element to the space above the crucible to:

- a. increase the gradient in the growing crystal
- b. develop effective circulation in the melt.

The process control system depends heavily on a diameter sensor developed by Sanders. A He-Ne laser illuminates the meniscus between the melt and the growing crystal; a bright spot is reflected from one point that is related to the diameter. A receiver measures the position of this bright spot with a null sensing system and provides a difference signal and a sum signal to the computer. A block diagram is shown in Figure 3.

The control system centered around the Sanders MIP-16 computer is outlined in Figure 4. The system controls all functions except rotation of the seed rod. The basic (inner) control loop uses the thermocouple input for temperature control, while the

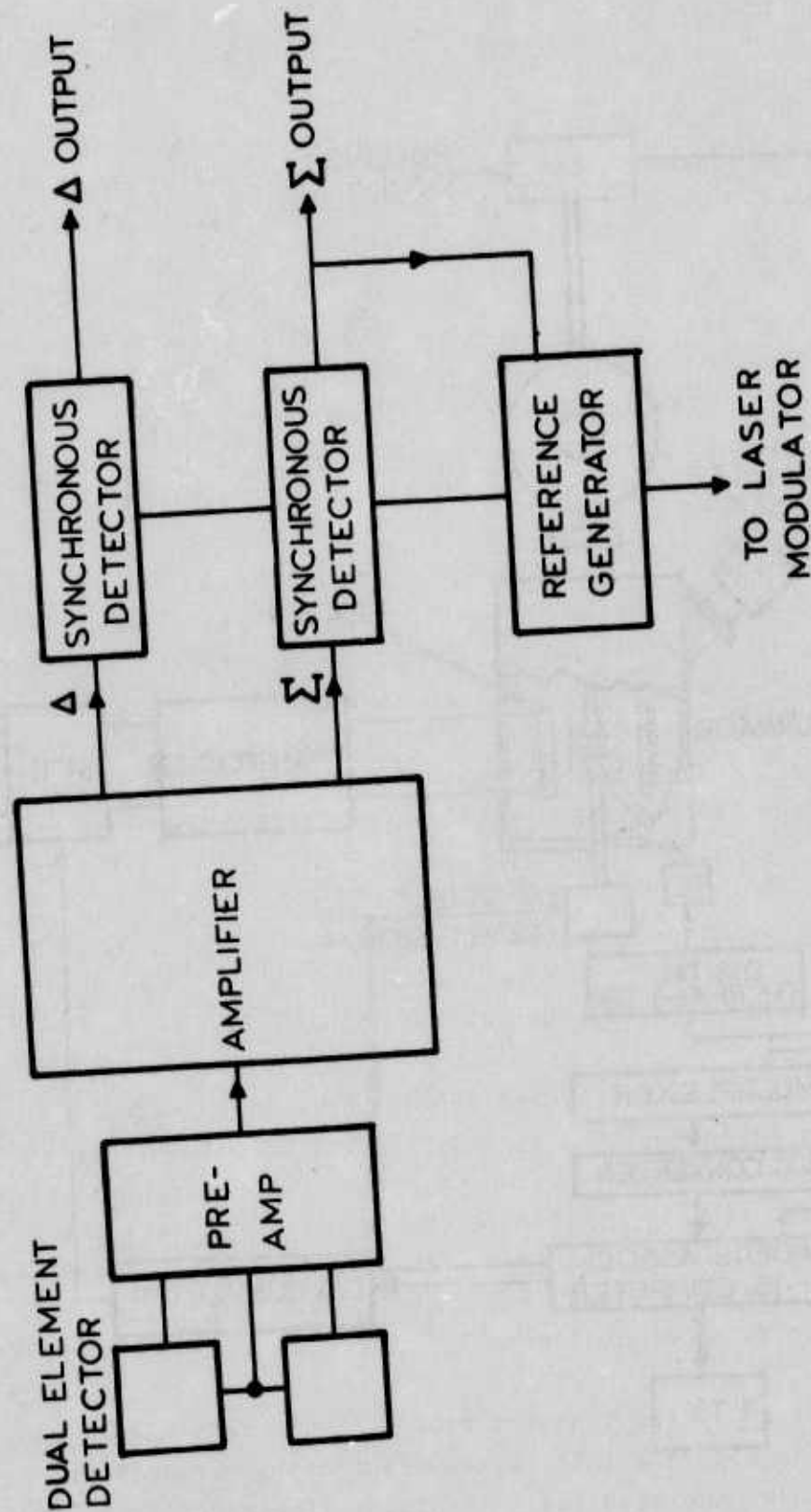


Figure 3. Block diagram of diameter sensor.

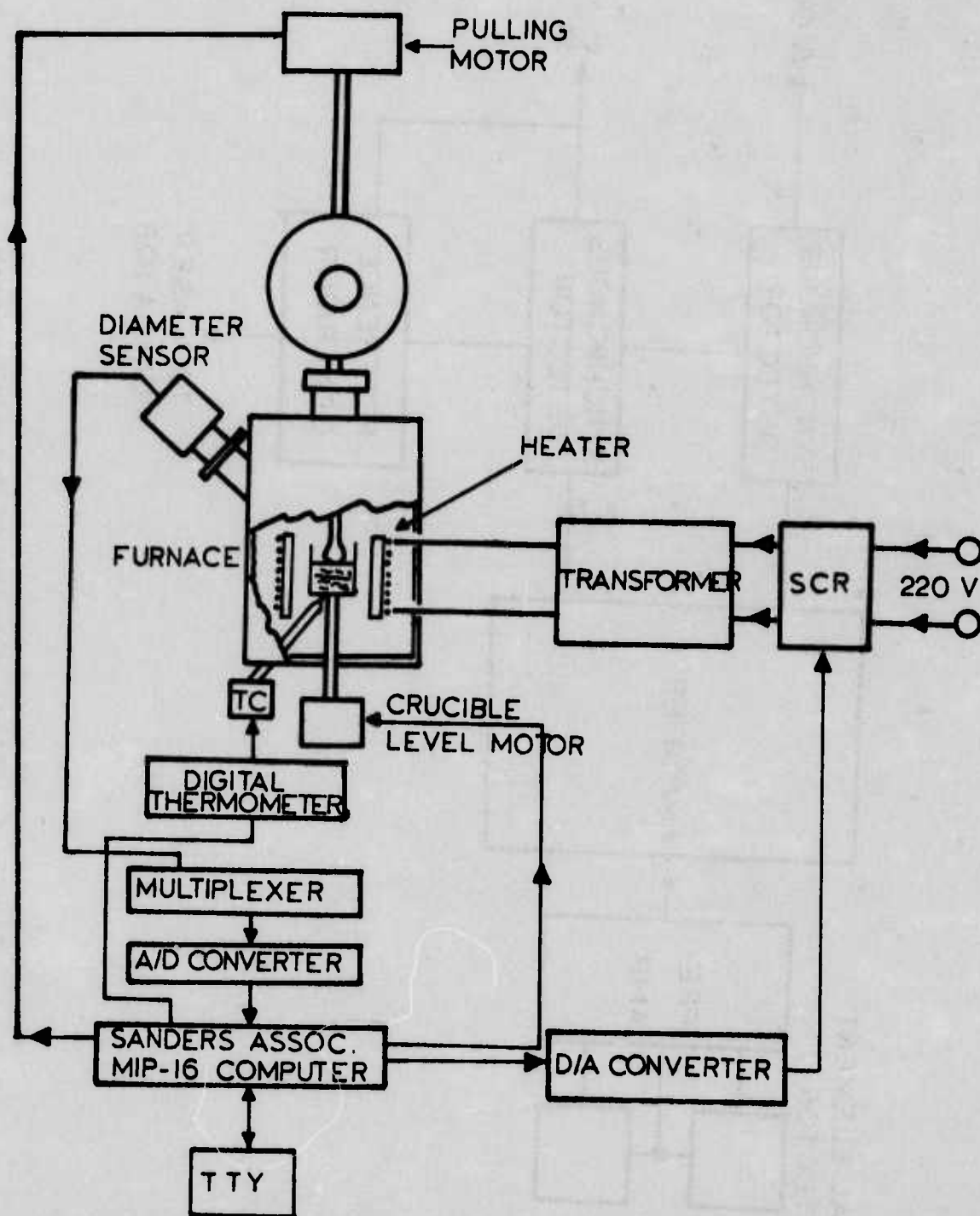


Figure 4. Block diagram of computer control system.

auxiliary (outer) control loop adjusts the temperature depending on the observed deviations from the desired diameter. The computer controls the motion of the pulling motor and the crucible position motor. The latter motor is provided to maintain a constant melt level for the diameter sensor, since a tapered crystal would otherwise be grown.

The computer has provided a number of operational features not easily available in an analog control system, especially when combined with the diameter control. A number of protective routines are built in, which continuously monitor the observed parameters and respond to out of tolerance conditions in a manner to protect the crystal and furnace. These routines have functioned a number of times during actual crystal growth and have prevented any unnecessary loss. Several growth runs have occurred over two to four day intervals with no operator attention.

2.2 CRYSTAL GROWTH

All crystals grown for this program at Sanders Associates used rare earth trifluorides purchased from Research Chemicals, plus LiF from Harshaw Chemical. The materials were treated with HF to remove contaminants. Other sources of rare earths were examined prior to this program but sufficient problems were encountered that they were abandoned. Minimal weight change has been experienced when Research Chemicals material has been utilized, indicating that their process is effective in preparing nearly anhydrous, stoichiometric material.

The crystals grown for this program, their doping level, and the weight with seed are listed in Table I. The steady progression in boule size and appearance is apparent in Figures 5, 6, and 7; boule 104Yb was similar in appearance to 103Yc. Four laser rods were fabricated from boule 106Yb, two 6.4 x 76 mm (0.25 x 3.0 inches) and two 5 x 60 mm.

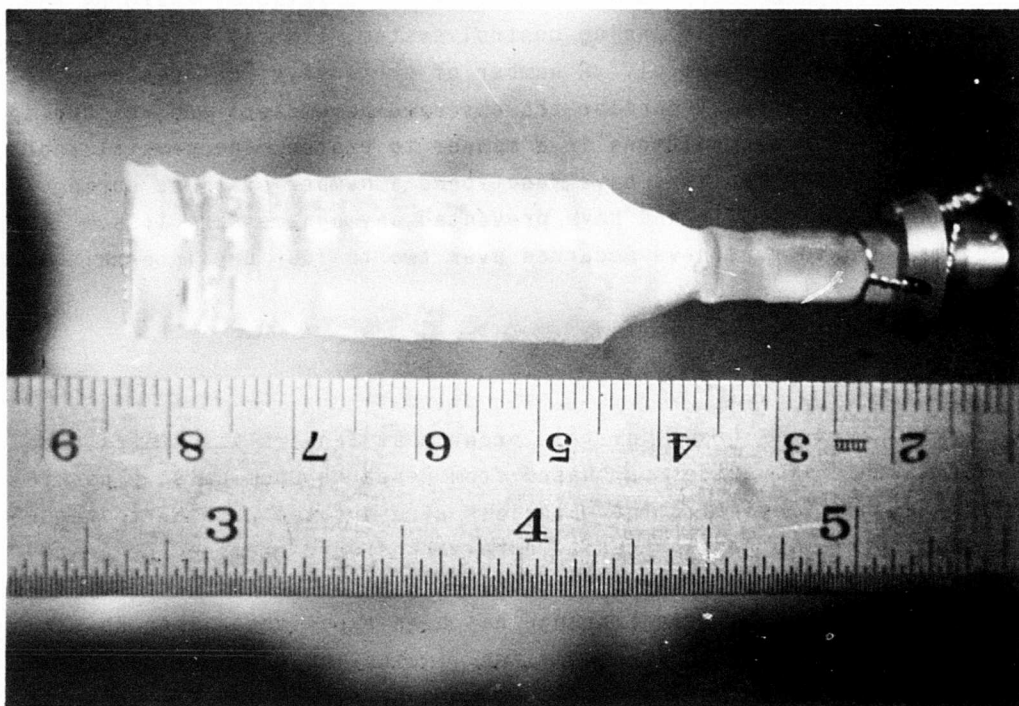


Figure 5. Boule 103 Yc



Figure 6. Boule 105 Yc

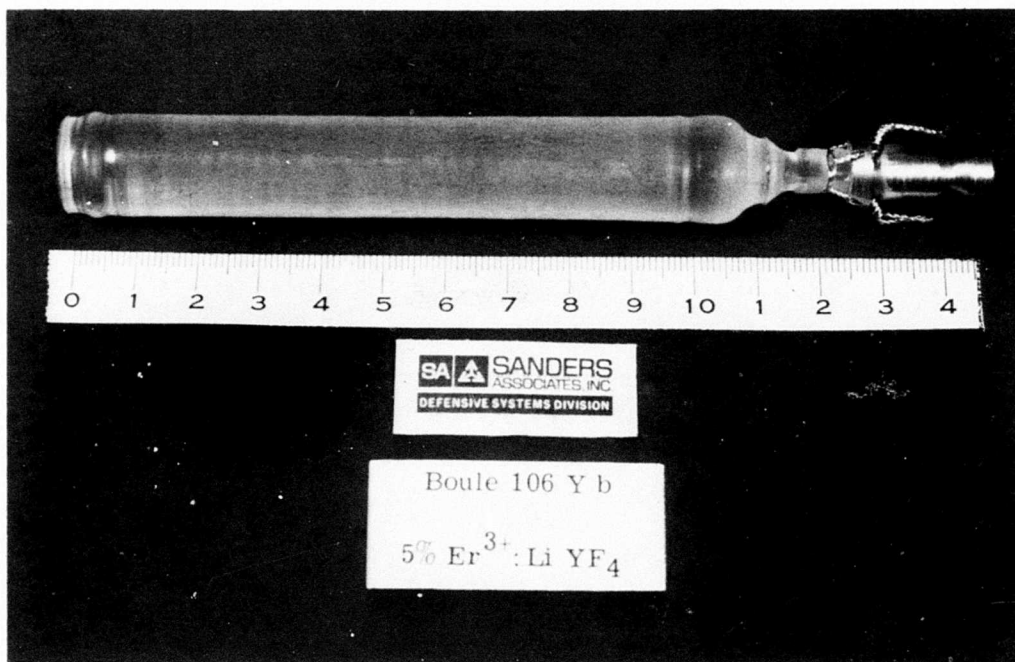


Figure 7. Boule 106 Yb

TABLE I

LIST OF LASER BOULES GROWN

Boule No.	Dopant Concentration (percent)	Weight w/seed (gm)	Diameter (mm)	Length (mm)	Remarks
103Yc	3.5	23	13	38	First crystal grown with diameter control. Unstable control loop. Concave interface.
104Yb	5	40			Air leak. Concave interface.
105Yb	5	91	19	95	Rehydrofluorinated melt from 104Y. Noncircular cross-section. Concave interface.
106Yb	5	99	17	115	Extremely straight walls. Slightly convex interface. Laser rods fabricated. Used remaining melt from 105Y.

Several changes were made in the thermal configuration of the furnace in an effort to increase the thermal gradient along the axis of the growing crystal. All of the earlier crystals showed evidence of inadequate gradient because of their concave melt/crystal interface. This form of interface leads to stagnation, and thus constitutional super cooling, with increased potential for incorporation of defects in the growing crystal. The desirable configuration is slightly convex, providing the necessary balance between the stable circulation of the melt below the crystal, and the grown-in strain. Our resistance heated furnace has the opposite problem from an R-F heated unit. In the latter system, afterheaters are often used to reduce the gradient because the system is heated only at the crucible wall, while our furnace tends to be isothermal, thus requiring an arrangement of heat shield baffles and openings to increase the gradient in the direction desired. The initial attempt involved the enlargement of the openings in the top shield, and subsequently the addition of a single molybdenum heat shield concentric with the graphite crucible support, and extending above it. This shield was removed for run 106Y and replaced by a two layer shield hanging from the top shield. The internal arrangement is shown in Figures 8 and 9.

Crystals have been grown at pulling rates between 0.1 and 2.0 mm/hr with little observable effect other than smoother surfaces on the boules at higher rates. No change has been observed in the internal optical properties. The majority of growth time has been at rates between 1.0 and 1.6 mm/hr. At present, no upper limit has been determined. The rotation rate of all crystals thus far was approximately 60 RPM.

Pure YLF seeds have been used for all growth runs, with a 5 x 5 mm cross-section, and typically 10 mm long. All seeds are carefully checked in a polarizing microscope for grain boundaries and oriented by x-ray with an a-axis in the direction of growth. They are notched with an air abrasive tool, and wired for the

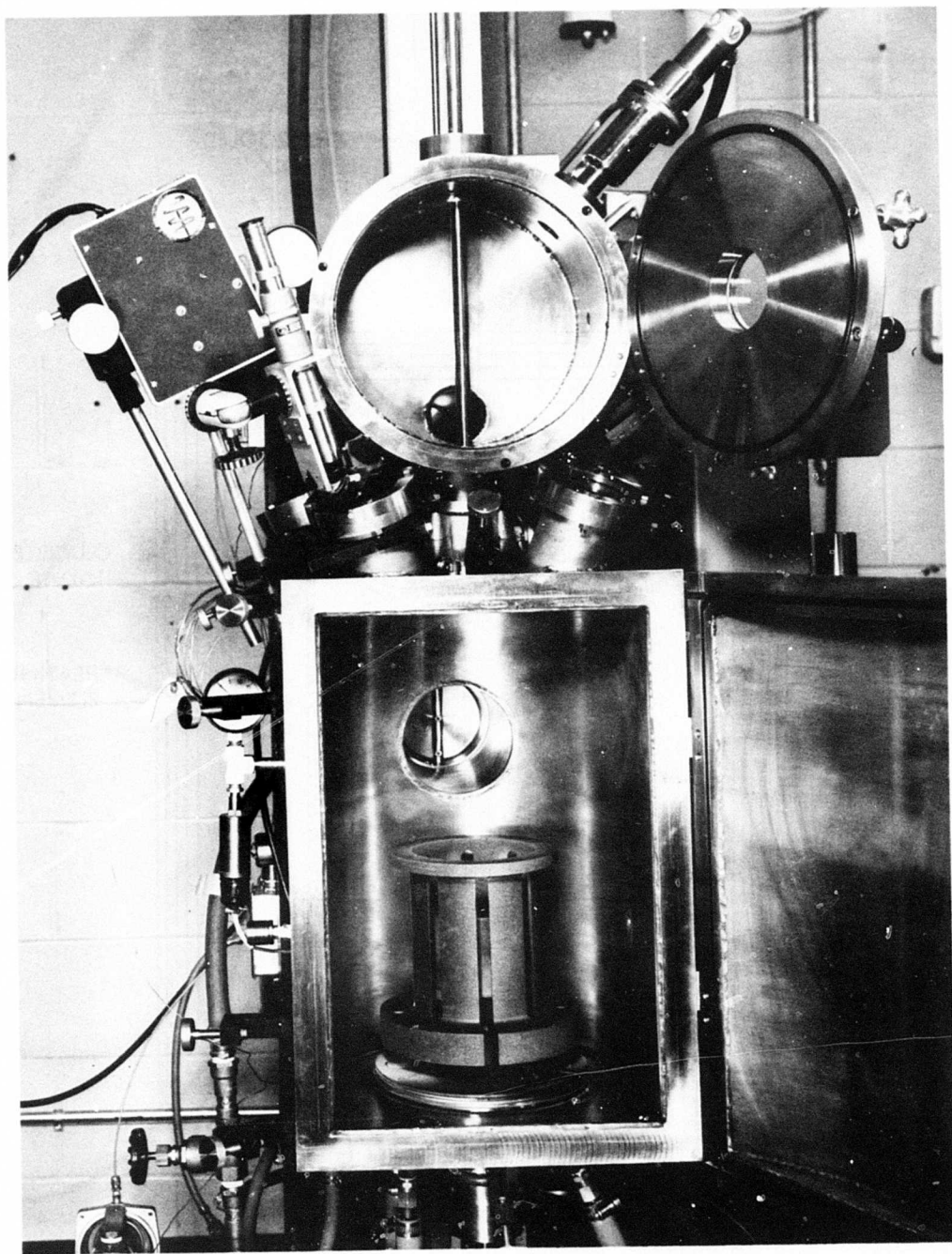
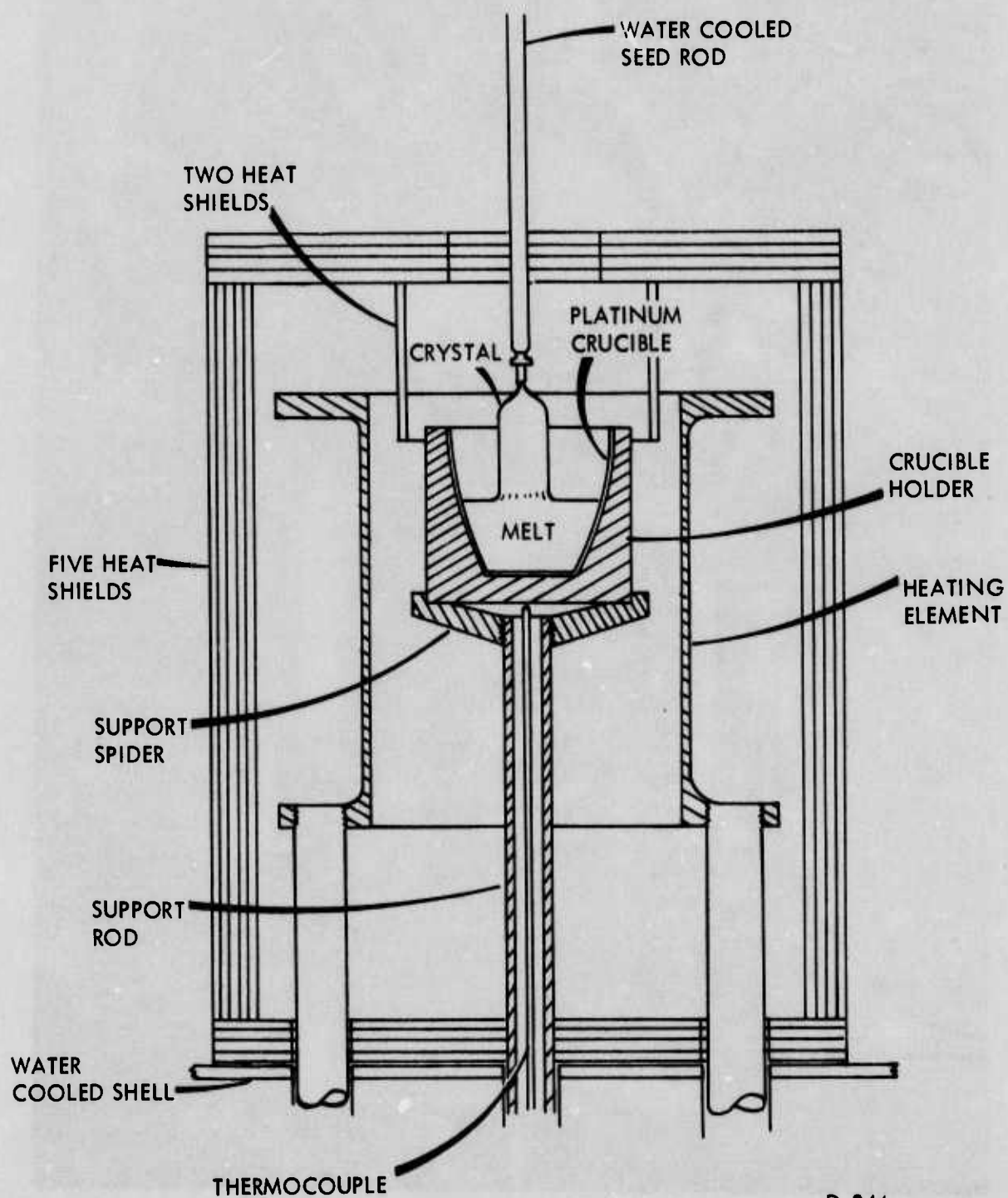


Figure 8. Interior of furnace.



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Figure 9. Geometry of furnace hot zone.

molybdenum seed adapter with 0.5 mm platinum wire. Prior to the attachment of the seed, it is cleaned in a solution of HCl and boric acid, a mild etchant. The seed is rinsed in distilled water and blown dry with a clean gas. Care is taken to snug the crystal tightly against the adapter for good thermal contact.

Prior to growth, the furnace is checked with a He mass spectrometer leak detector to 10^{-9} std cm³/sec air leak rate and all leaks repaired. A number of problems have arisen because of seals in the furnace, primarily the seed rod seal. This is obviously the most difficult area to seal because of the combined reciprocating and rotating motion, with no allowance for leakage. Other leaks have been more easily repaired. The seed rod is plated with hard chromium and ground to a fine finish. The original seal design utilized two opposing graphite loaded Teflon lip seals with a mechanical pump evacuating the space between the seals. The design was changed to permit a more heavily loaded seal after leaks were observed during operation. The new seal assembly had inadequate clearance, bound on the seed rod and damaged it. The lip seals were replaced with lubricated O-rings that appear to provide the required integrity on the inner seal at least, although the outer seal leaks occasionally without apparent detriment to the atmosphere inside the furnace. A new seed rod is anticipated in the near future that can be used with the unlubricated lip seals.

YLF crystals have been found to be amenable to having growth stopped deliberately, or even melting back and restarting, without detriment. In several instances, it has been necessary to melt back a portion of a partially grown crystal because of a problem such as a power failure. No optical defects have been observed in crystals where this has taken place, as long as the melt/crystal interface is allowed to adequately stabilize before recommencing growth.

2.3 GROWTH OF SCATTER-FREE YLF

A scatter free boule of pure YLF, plus a nearly scatter-free one, has been grown. Figure 10 shows scattering of a He-Ne laser beam in a specimen showing some scattering compared with no scattering in a sample where the feed was prepared by a slightly modified process. The essential features of the process are outlined below:

- a. Crystals are grown in an inert atmosphere of high purity argon.
- b. Commercially available YF_3 powder and LiF single crystal chips.
- c. The LiYF_3 feed is pretreated with undiluted flowing HF for 5 - 10 hours in an Inconel reactor before transfer to the growth furnace.
- d. The growth technique is the same as used in earlier programs.

Undoped YLF grown in this fashion shows no scattering from the He-Ne in a darkened room, while doped YLF shows some scattering, probably due to the presence of more impurities in the ErF_3 .

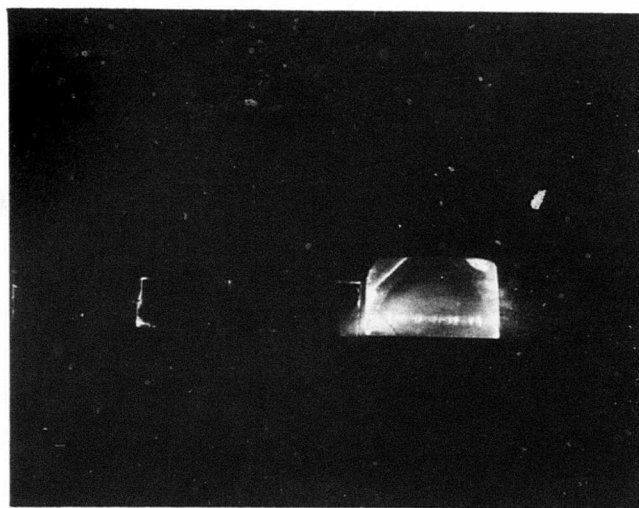


Figure 10. Photograph of scattering by YLF
of a He-Ne laser beam.

The crystal on the left is undoped YLF grown from a melt pretreated with HF. The crystal on the right is YLF 2.5% Er grown without melt pretreatment. Both crystals were grown at MIT/CPL.

3.0 CHEMISTRY AND CHEMICAL PRODUCTION TECHNOLOGY

Growth of laser material for this program was carried out at the Sanders Associates Automated Fluoride Growth facility. Simultaneous efforts were carried out by members of the technical staff of the Crystal Physics Laboratory (CPL) of the Massachusetts Institute of Technology to accomplish the following specific tasks:

a. Analysis

Experimental studies to identify impurities associated with inclusions to include scanning electron microscopy, and electron microprobe analysis.

b. Feed Purification

Investigation of new feed purification techniques which will lead to growth of higher quality material.

c. Technology Transfer

On site consultation to assist in growth of YLF boules at the Sanders' Furnace.

Efforts directed at material improvement have emphasized feed purification during this phase of the program. Evidence of widely varying feed purity from commercial sources and of the influence of feed purity on crystalline quality, reported previously^(1,2), strongly suggest the need for improved purification procedures. This is not to minimize the importance of growth conditions. However, growth conditions are operator controllable and optimization of growth technique, while not fully realized, is believed to be a far less serious problem.

The most significant result of this investigation (and the motivation for continued investigation of feed purification techniques) has been the growth of "scatter-free" YLF samples using a relatively simple purification procedure. "Scatter-free" in this context means that side scatter of a He-Ne (1 mw) probe laser illuminating a YLF sample was not visible in a dark room to the unaided eye.

In previous work, zone-refining and recrystallization in the crystal furnace have been used at MIT/CPL as YLF purification techniques. The difference between the two procedures is due to kinetic factors which inhibit segregation of certain impurities at the relatively high crystallization rate encountered in zone-refining and to the greater purification efficiency of normal freezing (crystal growth configuration) compared to a few zone-refining passes. Recrystallization in the furnace has resulted in the growth of scatter-free YLF⁽¹⁾; zone-refining has been shown to purify the feed with respect to other rare earths⁽²⁾ and has resulted in improved crystalline quality (although the results have not been as dramatic as with recrystallization).

In section 2 of this report growth of "scatter-free" YLF is described using yet another "purification" procedure. In this case molten LiREF₄ is hydrofluorinated in an Inconel reactor prior to growth. While comparisons of the effectiveness of this procedure compared to recrystallization are premature, hydrofluorination of the molten feed is much less time consuming and does not tie up crystal growing apparatus.

The object of this part of the investigation is to develop low cost feed purification procedures which, when used with hydrofluorination of the molten LiREF₄ (LiYF₄), will result in reproducible growth of scatter-free YLF. Hydrofluorination of the molten feed very effectively removes moisture and oxygen (oxides). In addition purification with respect to certain metallic compounds may be obtained.

However, to obtain uniformly high quality crystals, additional procedures are required to accommodate the widely varying feed purity of commercially available rare earth fluorides (and LiF). In the following section the results of chemical purification techniques for production of high purity feed are described. The goal of this investigation is the development of a procedure for obtaining LiREF_4 (LiYF_4) feed of uniformly high purity starting with commercial Li_2CO_3 and RE_2O_3 (Y_2O_3).

3.1 LITHIUM FLUORIDE

Although the LiF currently used is obtained from Harshaw as highly purified single crystal chips, we undertook the development of an inhouse LiF preparation capability to not only preclude difficulty in the event of a disruption in supply, but also to investigate the possibility of making purer material than is now available. The latter goal, upon close inspection, is not unreasonable. Reports in the literature on Harshaw-grown NaF indicate that this material is not as pure as can be made⁽⁴⁾, suggesting that commercial LiF may be subject to the same deficiencies. Harshaw claims that the only impurities in LiF are Al, Ca, Mg, and Fe in the 1-3 ppm range, but subjective evidence derived from zone-refining experiments on LiYF_4 , using Harshaw LiF and MIT/CPL zone-refined commercial LiF, indicates much better crystallization; i.e., larger grains, using our material.

As an alternative to zone-refining, a chemical purification method is under investigation which involves dissolving reagent grade Li_2CO_3 in water under CO_2 pressure, solvent extraction to remove transition metals and possibly aluminum, followed by repeated recrystallization to remove Ca, Mg and Na. Lithium carbonate is slightly soluble in water and yields a solution of pH 11-12, but under CO_2 pressure the CO_3 ion concentration is greatly reduced, the pH decreases to about 6, and a considerable amount of Li_2CO_3 dissolves. The solution is stable at ambient CO_2 pressures and may be decomposed by heating with

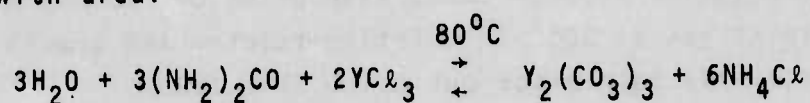
reprecipitation of Li_2CO_3 and evolution of CO_2 gas. Solvent extraction of transition metals from LiHCO_3 solution (lithium bicarbonate) with diethylammonium diethyldithiocarbamate has been demonstrated and extraction of aluminum, if any, is soluble in a bicarbonate medium with 8-hydroxyquinoline and should pose no problem. Slow recrystallization of Li_2CO_3 driven by gentle heating to liberate CO_2 , when allowed to stop at pH 9, will allow separation of Ca, Mg and Na, which will remain in the solution phase. Conversion of Li_2CO_3 to LiF will be carried out in HF gas at 300° in a Teflon reactor and growth runs at Sanders and MIT will be carried out using this feed.

3.2 RARE EARTH FLUORIDES

Direct use of commercial rare earth fluorides for YLF feed has been problematic; suppliers have withdrawn from the market; there is variation in the purity and quality and utility of the laser crystals. Calcium, for example, is a well-known and difficultly controlled impurity in rare earth compounds⁽⁵⁾. Aluminum, iron, and sodium are also prominent impurities in rare earth fluorides. It is felt that these impurities, possibly in conjunction with oxygen, contribute to formation of residual light scattering centers in YLF crystals. Yet by hydrofluorinating molten LiREF_4 , prepared from commercial fluorides, growth of scatter-free YLF has been achieved - but the success of this process, too, is dependent upon the quality of the starting materials. Large amounts of moisture or oxide in the commercial REF_3 are cause for hydrofluorination over an extended period, or repetition of the process, and the presence of other impurities when not precipitated as scattering centers may cause index variations.

Development of chemical processes has been undertaken to enable complete inhouse synthesis of uniform high purity REF_3 , starting from commercial oxides of varying quality to provide material free from contaminants likely to contribute to formation of optical defects. The chemical purification methods being developed start with commercial

rare earth oxide which is dissolved in hydrochloric acid to form a solution of RECl_3 . This solution is purified by filtration, followed by solvent extraction to remove transition metal impurities. Analysis of extracts from an ErCl_3 solution by X-ray fluorescence showed Fe, Cu, Zn and Pb to be the major impurities removed by solvent extraction. The rare earth carbonate is then precipitated homogeneously under CO_2 pressure from the purified chloride solution, using the following reaction with urea:



It is significant, in terms of throughput, that the initial YCl_3 concentration can be 0.5 molar without causing deterioration of the crystallinity, particle size, and bulk density of the precipitate.

The chemical advantages of the urea- CO_2 pressure process follow from the slowness of the reaction, which favors good crystallization, and from the slightly acidic pH which suppresses hydroxycarbonate formation and enables separation of alkaline earth impurities as soluble bicarbonates. Examination of the product yttrium carbonate under the microscope showed 0.02 mm clear crystals with no sign of a second phase. The X-ray powder pattern agreed with published data showing the compound to be $\text{Y}_2(\text{CO}_3)_3 \cdot 2.3 \text{H}_2\text{O}$. Sharp reflections with resolution of Cu $K\alpha_1$ and $K\alpha_2$ indicated excellent crystallization. High bulk density and free flowing behavior characterized the precipitate. These two properties, lacking in yttrium carbonate formed in alternate chemical reactions, allow the material to be easily separated from and washed free of the supernatant liquid from which it was precipitated.

Conversion of rare earth carbonates to fluorides was investigated before development of the urea- CO_2 pressure process, using $\text{Y}_2(\text{CO}_3)_3$ and $\text{Er}_2(\text{CO}_3)_3$ prepared by homogeneous thermal decomposition of

erbium trichloroacetate solution*. Reaction of the carbonates with HF gas at 200-300°C in a Teflon tube produced a fluoride shown by X-ray powder pattern analysis to be single phase. In contrast to the carbonate hydrofluorination of commercial Y_2O_3 under the same conditions yielded an incompletely reacted material showing strong reflections for Y_2O_3 and YF_3 in the X-ray powder pattern.

Subsequent growth of $LiErF_4$ without further high temperature hydrofluorination of the ErF_3 , and using Harshaw LiF , yielded a crystal with a clear, almost scatter-free, top section, bounded by a sharp transition in a plane perpendicular to the growth axis to a two-phase section containing (as shown by X-ray analysis) an oxyfluoride phase. Thus, at low temperatures, in an inexpensive Teflon apparatus, REF_3 has been produced which is nearly dry enough for use as crystal growth feed, further processing by melting $LiREF_4$ in HF at 900°C completes preparation of the feed.

Success of the low temperature reaction follows from use of $RE_2(CO_3)_3$ rather than the oxide as the fluoride precursor. A further advantage starting with $RE_2(CO_3)_3$ is that the reaction leading to its formation also allows separation from alkaline earths which are frequently troublesome impurities in rare earth chemicals.

A large batch of YF_3 is now being produced which will be evaluated in growth runs at Sanders Associates and at MIT.

*This process is not suitable for large scale carbonate synthesis.

4.0 LASER MEASUREMENTS

Although the major effort of this phase of the program was material improvement and growth, some laser, and laser related spectroscopic measurements were made. In addition, the index of refraction of YLF was measured and found to be in serious disagreement with data previously reported by Shand. Index measurements are reported in the Appendix.

4.1 Q-SWITCHED MEASUREMENTS

Experiments were carried out to determine the Q-switched performance of Er^{3+} :YLF. In previous attempts⁽¹⁾ very high resonator losses were observed when a calcite polarizer was inserted in the resonator. To avoid the losses associated with calcite prisms a Brewster plate fabricated from SF-6 was used as the polarizer. The experimental conditions were as follows:

Rod:	470.4; 5 x 45 mm 3.5% Er
Flash Lamp:	ILC 4F2 (450 Torr Xe, 4 mm bore, 2 inch arc)
Pump Cavity:	Silvered elliptical cylinder, $a = 11.5$ mm $b = 5$ mm $\ell = 40$ mm, separate coolant jackets over lamp and rod.
Water Flow Rate:	0.5 gal/min.
Energy Storage:	Single Mesh PFN, $C = 25\mu\text{f}$, $L = 4\mu\text{H}$
Polarizer:	SF-6 plate cut at $\theta = 60.7$ degrees
Q-Switch:	Lasermetrics EOM-812, KD*P. Index matched with Dow Corning 200 fluid, 5CS; window material, Dynasil; A/R coated @ 850 nm

Figure 11 shows a comparison of the long pulse and Q-switched output vs. input; the long pulse data was taken with the polarizer and KD*P in the resonator. Q-switch hold off problems were not encountered at the highest input energies (60J) - that is with 1/4 wave voltage on the KD*P, but not switched, long pulse oscillations were not observed. The Q-switched output consisted of spikes up to 120 ns FWHM - in some cases multiple spikes were observed. Using a KDP switch, Q-switched performance was similar but the output consisted of a single spike over the range shown in Figure 11.

Long pulse data were taken under the same conditions with the substitution of a 3000 Torr Xe lamp (2 mm bore x 45 mm length). The efficiency is considerably improved (see Figure 12) but flash lamp life was seriously degraded.

Figure 12 shows that the insertion loss of the KD*P is quite high - about 35% at 35J input. The source of the high loss is not fully understood - similar results were reported by Steinbruegge with Nd:SOAP⁽⁶⁾. Approximately the same loss was observed with a KDP crystal with broad-band AR coatings. The insertion loss of the SF-6 Brewster plate, however, was extremely low and this polarizing technique (possibly with more plates at higher energies) appears most attractive for Q-switch operation.

Because Er:YLF is naturally polarized, output losses to a (low loss) polarizing element would be expected to be small as is observed. This is also anticipated to be the case in repetitively pulsed operation as the ratio of natural to thermally induced birefringence is expected to be quite high.

4.2 SPECTROSCOPY

In a previous report⁽¹⁾ laser efficiency versus concentration was analyzed by solving the rate equations of a three level system (four level laser) under the assumption that:

OUTPUT ENERGY (mJ)

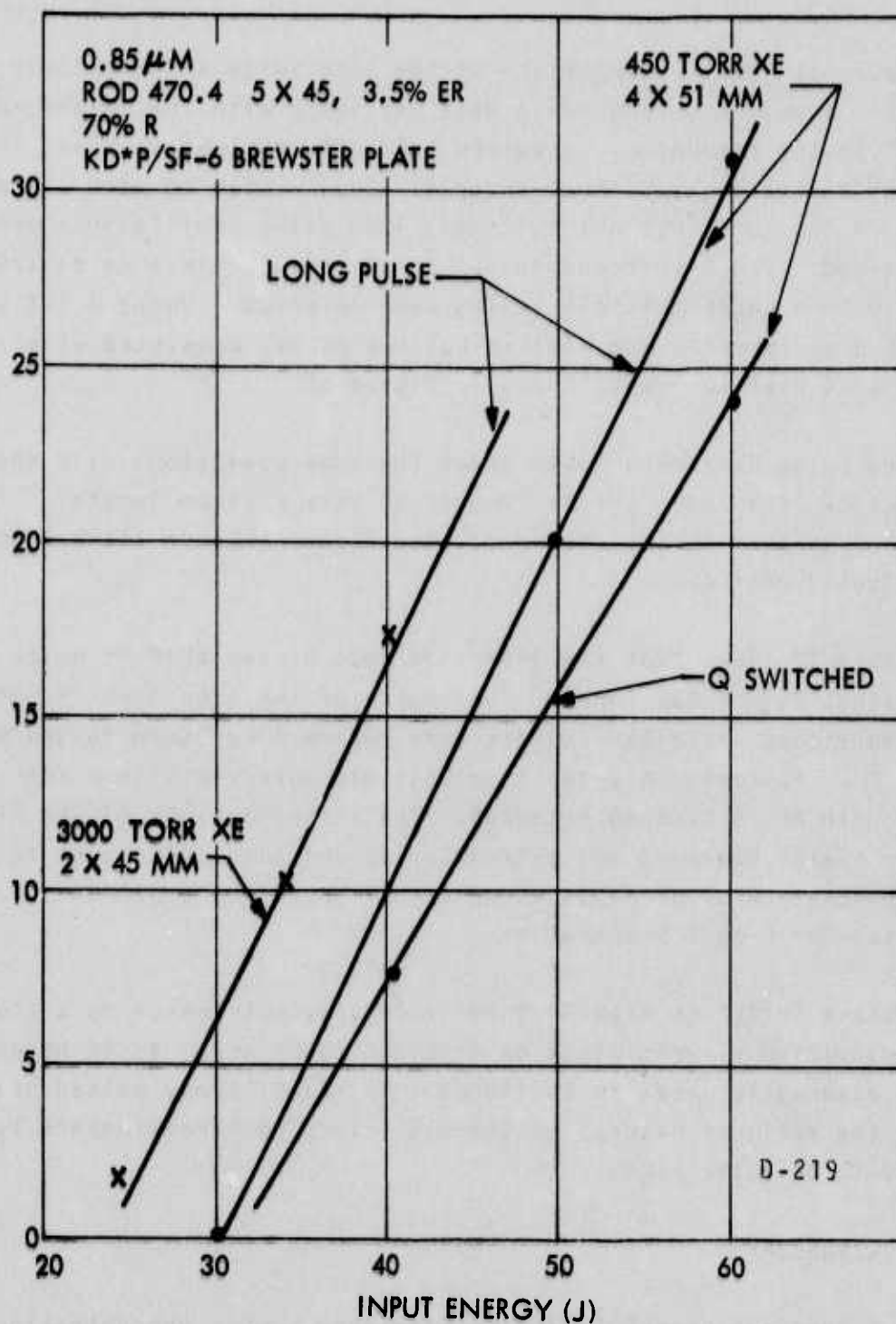


Figure 11. Long pulse and Q-switched performance.

OUTPUT ENERGY (mJ)

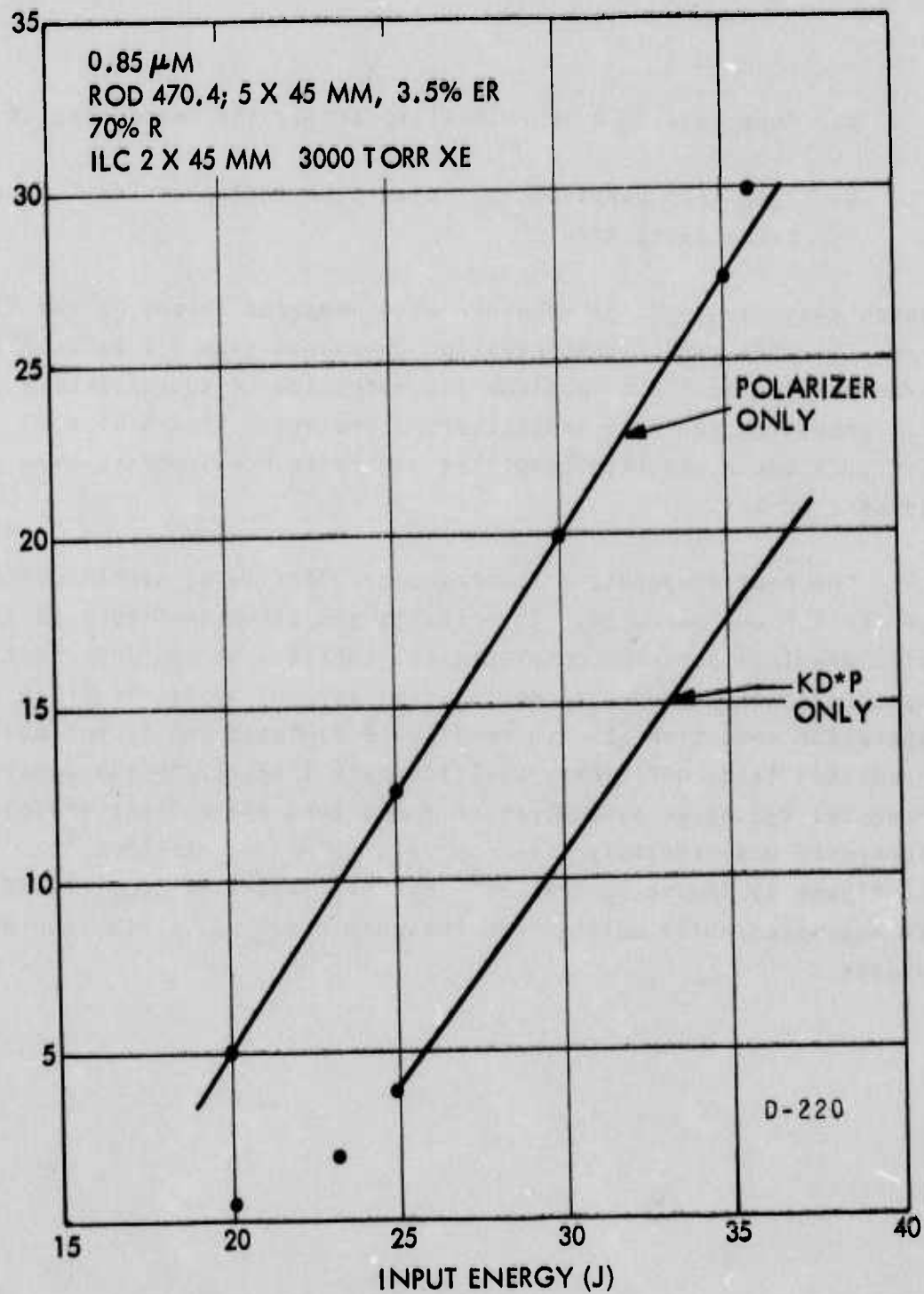


Figure 12. Effects of KD*P on laser performance.

- a. Pump rate is proportional to active ion concentration.
- b. Pump lamp duration is longer than the upper level fluorescence decay time.

Using these assumptions together with measured values of the fluorescence lifetime versus concentration (measured from 0.5 to 3.5% and extrapolated to 5%) an optimum concentration of approximately 5% Er was predicted for room temperature operation. Growth of a 5% Er^{3+} :YLF boule has been completed but laser measurements have yet to be carried out.

The room temperature fluorescence lifetime of samples of 5% and 10% Er:YLF was measured. The results are shown in figure 13 together with previous data for completeness. While a 5% Er concentration appears optimal under assumptions (a) and (b) above; in flash pumped operation condition (b) can readily be violated and it follows that predicted laser efficiency will increase linearly in the concentration. Over the 2-3.5% Er concentration range long pulse slope efficiency increased approximately linearly with Er^{3+} concentration⁽¹⁾. The data of figure 13 indicates that Er^{3+} concentrations up to at least 10% can be attempted while maintaining reasonable (20 μs) flash lamp pulse widths.

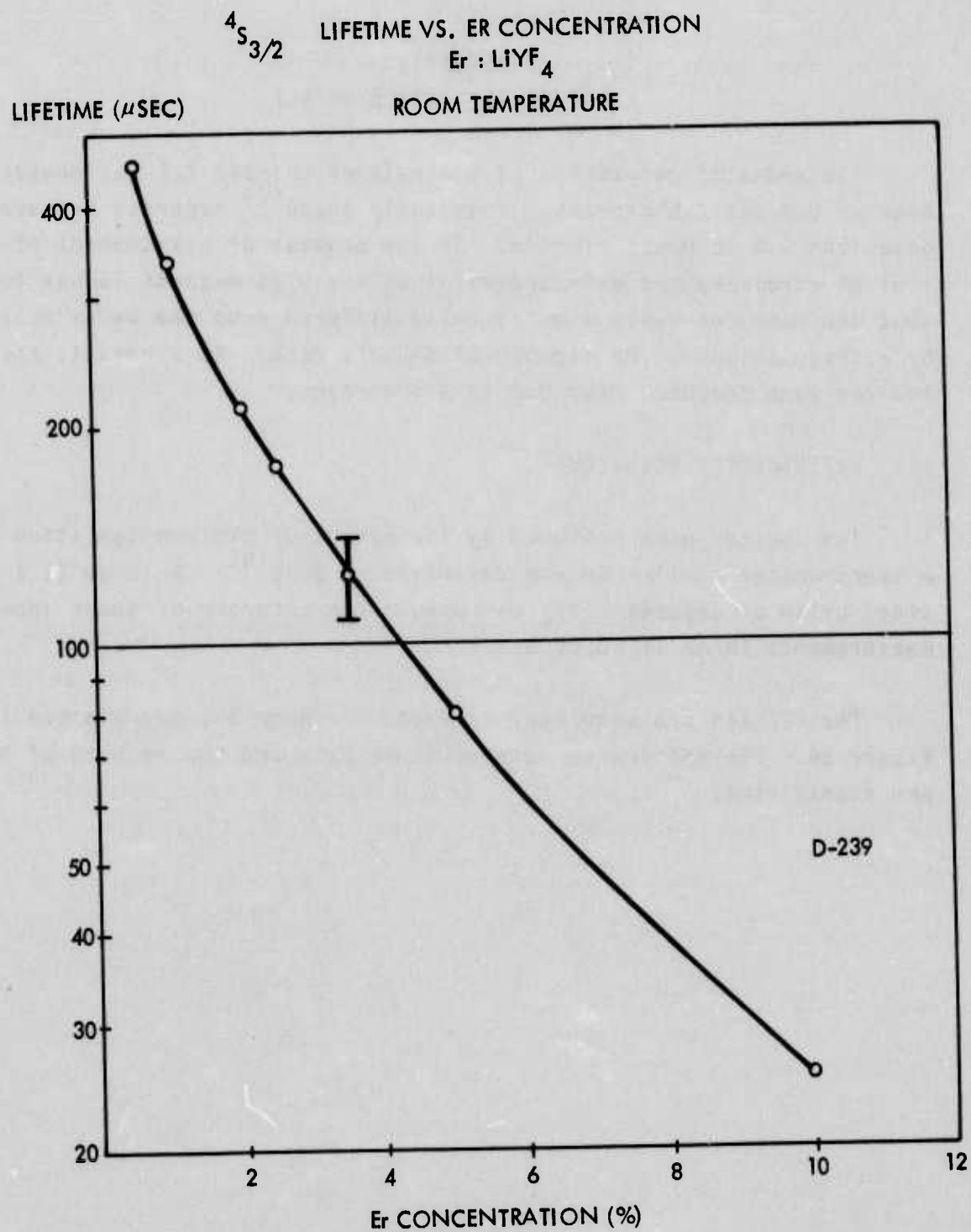


Figure 13. $^4S_{3/2}$ Lifetime versus Er concentration.

APPENDIX

INDEX OF REFRACTION OF YLF

The index of refraction of a sample of undoped YLF was measured between 0.2 and 2.6 microns. Previously Shand⁽⁷⁾ reported measured data from 0.4 to 0.625 microns. In the process of measurement of the indices (ordinary and extraordinary) at $\lambda = 0.85$ microns it was found that the measured value significantly differed from the value obtained by extrapolation to .85 microns of Shand's data. As a result, the indices were measured from 0.2 to 2.6 microns.

A.1 EXPERIMENTAL TECHNIQUE

The indices were measured by the method of minimum deviation on a spectrometer similar to one described by Bond⁽⁸⁾. A large (1.4 cm side) prism of undoped LiYF_4 was used. The accuracy of these index measurements in Δn is ± 0.0002 .

The results are tabulated in Table II, page 33, and plotted in figure 14. The difference between these data and the results of Shand are significant.

TABLE II
Refractive Index of YLF
 Index Tolerance ± 0.0002

Wavelength (μm)	n_e	n_o
2.600	1.4602	1.4381
2.400	1.4617	1.4397
2.200	1.4632	1.4411
2.000	1.4645	1.4424
1.800	1.4658	1.4437
1.600	1.4670	1.4449
1.400	1.4681	1.4460
1.200	1.4693	1.4471
1.000	1.4708	1.4485
.900	1.4716	1.4492
.800	1.4726	1.4502
.700	1.4741	1.4516
.600	1.4762	1.4535
.500	1.4796	1.4567
.450	1.4822	1.4590
.400	1.4858	1.4624
.350	1.4913	1.4674
.300	1.5001	1.4754
.250	1.5156	1.4895
.225	1.5287	1.5014

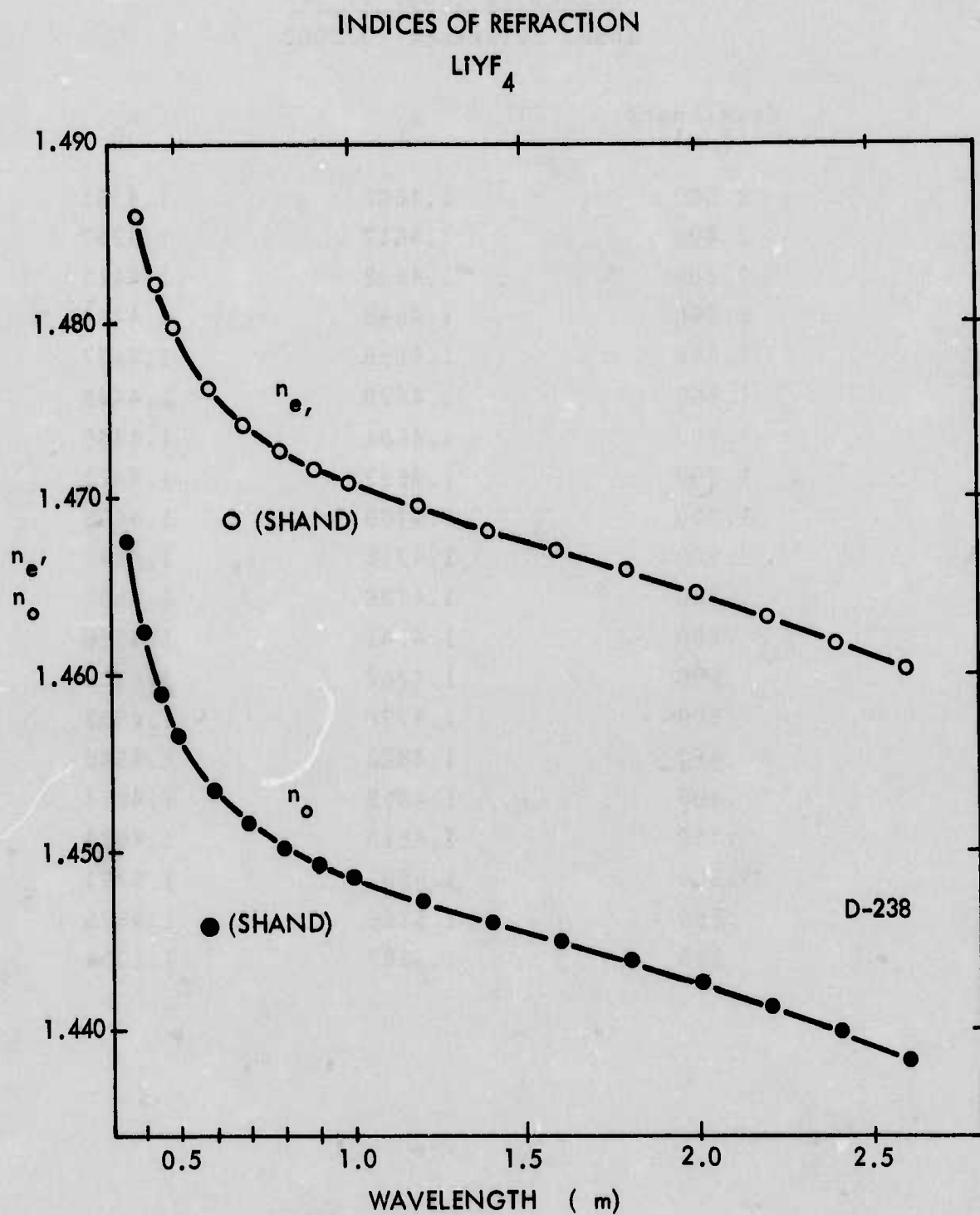


Figure 14. Index of refraction versus λ of YLF.

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