AD-A278 408

UMENTATION PAGE

Form Approved
OMB No. 0704-0188

ion is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, eting and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this ducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

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2. REPORT DATE
Feb. 28, 1994

3. REPORT TYPE AND DATES COVERED
Final Report 6/16/93-2/15/94

4. TITLE AND SUBTITLE
Development of Materials for Spectral Hole Burning Applications (SBIR-93)

5. FUNDING NUMBERS
F49620-93-C-0035

6. AUTHOR(S)
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Crystal Systems, Inc.
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8. PERFORMING ORGANIZATION REPORT NUMBER
AFOSR-TR- 94 0242

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Air Force Office of Scientific Research
110 Duncan Avenue, Suite B115
Bolling AFB, D.C. 20332-0001

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
3005/SS

11. SUPPLEMENTARY NOTES

DTIC
ELECTE
APR 20 1994
S B D

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release;
distribution unlimited.

unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

It was intended to evaluate growth of europium-doped yttrium aluminum oxide ($Eu:Y_3Al_5O_{12}$, Eu:YAG) and europium-doped yttrium silicate ($Eu:Y_2SiO_5$, Eu:YSO) crystals to characterize these crystals for spectral hole-burning application. After technical discussions at a topical Workshop and agreement of the Technical Monitor, the scope of the program was enlarged to include growth of rare-earth doped mixed-oxide crystals and growth of Eu:YSO was de-emphasized as these crystals were available for characterization. The experimental effort involved producing YAG, YSO and $YAlO_3$ (YALO) host crystals and the dopants Eu, Tm, Tb and Ce. High optical quality crystals of Eu:YAG, Tm;YAG, Tb:YAG and Ce:YAG were fabricated from 6 cm diameter boules grown using the Heat Exchanger Method (HEM). The samples were characterized for absorption as a function of wavelength at SRI International, Menlo Park, CA. Previously-grown $Ce:Gd_3Sc_2Al_3O_{12}$ (Ce:GSAG) crystal was characterized also. The available data is insufficient for choosing a crystal for spectral hole burning application. It is necessary to explore other rare-earth doped mixed oxide crystals and carry out more characterization.

DTIC QUALITY INSPECTED 3

14. SUBJECT TERMS
spectral hole burning, yttrium aluminum garnet, Heat Exchanger Method, crystal growth, dephasing time

15. NUMBER OF PAGES

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	**18. SECURITY CLASSIFICATION OF THIS PAGE**	**19. SECURITY CLASSIFICATION OF ABSTRACT**	**20. LIMITATION OF ABSTRACT**
Unclassified	Unclassified	Unclassified	UL

AFOSR-TR- 94 0242

Approved for public release;
distribution unlimited.

PREFACE

The effort entitled "Development of Materials for Spectral Hole Burning Applications" (Contract Number F49620-93-C-0035) was carried out as a Phase I program for the Air Force Office of Scientific Research, Bolling AFB, D.C. with Dr. Alan E. Craig as the Technical Monitor.

The key personnel at Crystal Systems involved in the effort were:

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John A. Lesiczka, Engineer

Frederick Schmid, Program Manager

The authors would like to thank Dr. Alan Craig of AF OSR and Dr. Ravinder Kachru of SRI International, Menlo Park, CA for their input, discussions and characterization.

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DEVELOPMENT OF MATERIALS FOR SPECTRAL HOLE BURNING APPLICATIONS

1.0 Introduction

High resolution nonlinear spectroscopy can be combined with modern computer and processor architecture to offer an extra degree of freedom to enhance optical memory and processing. This can be achieved by utilizing frequency-selective optical memory both in the frequency domain (hole-burning memory)[1] and the time domain (photon-echo memory)[2-4]. Such spectral domain can be implemented for optical memory and processing by using a material that supports spectral hole burning. These materials show an absorption spectra which is inhomogeneously broadened.

So far, the understanding of spectral hole burning materials is limited and these materials have not had practical application. It was intended to identify the basic characteristics required in materials to achieve persistent spectral hole burning and to evaluate these materials on the basis of practical constraints so that single crystals can be produced. Later, candidate materials would be characterized and evaluated for spectral hole burning to identify potential for devices.

The understanding of persistent spectral hole burning materials for optical memory and processing applications is rather limited, and the requirements are quite extensive. Every known material that demonstrates spectral hole burning has been inadequate in some characteristic important for either memory or processing. The need for spectral hole burning materials is new, and the general criterion is material whose absorption spectrum is inhomogeneously broadened. Therefore, it was intended to identify spectral hole burning materials based upon the data available on these materials. The procedure will be useful for identifying these materials; however, it will be necessary to optimize this procedure as more information is obtained from literature or from the results of the experimental program. The intent was to develop a systematic program to produce materials for persistent hole burning applications.

During the Phase I program, attempts were made to produce several materials in single-crystal form using different hosts and different dopant ions. Some of these materials were envisioned prior to the start of the Phase I program; however, other materials were added to the list based upon discussions within the technical community. The host crystals grown included Y_2SiO_5(YSO),

$YAlO_3$(YAlO), $Y_3Al_5O_{12}$(YAG) and the dopant ions were from the family of rare earths: Eu,Tm, Tb and Ce. In addition, data obtained on these materials included meltpoint, melting behavior, reaction with crucible, vapor pressure, phase formation and single-crystal growth. Good optical quality samples were fabricated from the crystals and after polishing were sent to Stanford Research Institute (SRI) for characterization and potential use for spectral hole burning application.

The results of the investigations have shown that some of the materials are easy to grow in single-crystal form using the Heat Exchanger Method (HEM) and some crystals show potential for use in spectral hole burning applications. Based upon preliminary data, no one material could be qualified or disqualified. Extensive basic work is necessary to fully explore this new area of importance to produce high intensity memory storage media using spectral hole burning materials.

2.0 Background

2.1 Spectral Hole Burning

There are many known mechanisms for spectral hole burning: two-level saturation, storage of population in multi-stable optical levels, population storage in long lived ground state components (hyperfine, super-hyperfine or Zeeman) photoionization, charge transfer, photo-assisted barrier crossing and modification of the local structural environment[5]. To achieve these characteristics, doped materials are used, and rare earth materials offer a good model system based upon their characteristics and the volume of information available[6,7]. Similar to laser materials, spectral hole burning materials also involve using a dopant ion and a host lattice. Therefore, the information in the search for new laser materials[8-15] could be useful for evaluation of materials for spectral hole burning applications. Similarly data on phosphors could be utilized[16-19].

The general criteria for selecting a material for spectral hole burning application includes:

- (i) choice of dopant ion,
- (ii) choice of host crystal,
- (iii) ability to grow crystals of selected materials for spectral hole burning characteristics,
- (iv) storage density,
- (v) access times, erasability, reliability, error rate, etc.

In the search for the ultimate computer memory, the desired characteristics may be that one bit of information is stored for each atom or molecule in the storage material. This will give a storage capability of the order of 10^{22} bits/cm^3. Other features desired could be totally random, instantaneous access, erasability for reuse, stability under all conditions, etc. Presently, the understanding of spectral hole burning materials is so limited that compromises are made in material selection. It is important to develop a data base for these new materials and applications to arrive at an appropriate selection of materials for use in spectral hole burning applications and within the application for appropriate storage media.

Based on published information on properties, two materials were selected: Eu:Y_2SiO_5 and Eu:YAG. Eu:Y_2SiO_5 has shown the longest dephasing time (822 μs) of one site combined with the ultra-long hole lifetime; hence it has been recommended as one of the best candidates for frequency-selective optical memory of either the hole-burning or the photon-echo type[20]. Similarly, Eu:YAG was selected because among laser oxides, there is maximum experience with the growth of YAG crystals. Among other materials containing similar ions, Eu:$YAlO_3$ has been characterized for dephasing times [21-25]; however, from a crystal growth point of view, it is easier to grow YAG than $YAlO_3$ crystals.

Soon after the start of the Phase I program, a Workshop on spectral hole burning materials was held at the IBM Almaden Research Center, San Jose, California. Discussions at this Workshop and subsequent interactions were used to select additional materials which could be evaluated for spectral hole burning applications. These materials include adding Tm,Tb and Ce to the dopant list and also adding $YAlO_3$ to the crystal host list for the experimental effort.

2.2 Heat Exchanger Method

The Heat Exchanger Method (HEM™) is being used for the commercial production of 10" diameter sapphire crystals. These crystals are of very high quality and it is possible to grow sapphire by HEM free of scattering centers for stringent optical applications. HEM is also in commercial production for multi-crystalline silicon ingots for photovoltaic and optical applications. Titanium doped sapphire (Ti:Sapphire) boules are grown routinely for cw and pulsed laser applications. Mixed oxides, fluorides and compound semiconductors have also been grown by HEM.

The salient features of the process are shown in Figure 1. The crucible with the seed positioned at the bottom is loaded with charge and placed on top of the heat exchanger. After evacuation, heat is supplied by the graphite heater and the charge is melted. The seed is prevented from melting by forcing gaseous helium through the heat exchanger.

After sufficient meltback of the seed is achieved, growth is started by increasing the flow of helium to decrease the heat exchanger temperature. The liquid temperature gradients are controlled by the furnace temperature, while the temperature gradient in the solid is controlled by the heat exchanger temperature. Crystal growth is achieved by controlling the heat input as well as the heat extraction. After solidification is complete, the gas flow through the heat exchanger is decreased to equilibrate the temperature throughout the crystal during the annealing and cooldown stage.

The HEM is the only crystal growth process in which both the heat input and heat extraction are controlled. The independent liquid and solid temperature gradients are achieved without movement of the crucible, heat zone or crystal. After the crystal is grown, it is still in the heat zone and can be cooled at a controlled rate to relieve solidification stresses. This unique capability allows the growth of sapphire up to 32 cm diameter and weighing about 50 kg without cracking due to thermal stresses associated with such large sizes.

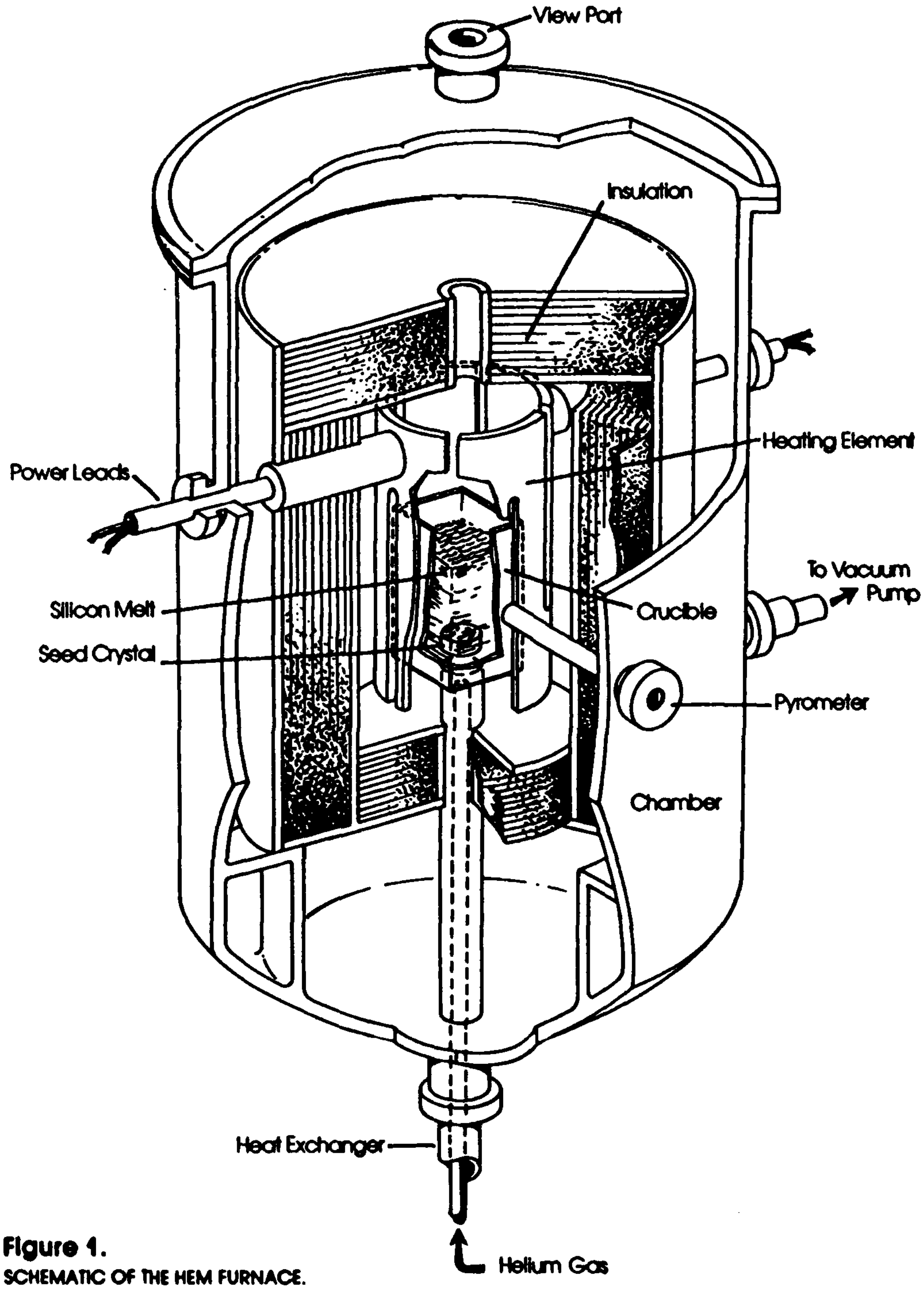

Figure 1.
SCHEMATIC OF THE HEM FURNACE.

A distinguishing feature of the HEM, as compared with Czochralski (Cz) or top-seeded processes, is that the solid-liquid interface is submerged beneath the surface and is surrounded by the melt. Under these conditions the thermal and mechanical perturbations are damped out by the surrounding molten mass before they reach the interface. This results in uniform temperature gradients at the interface. In the Cz process and top-seeded techniques, growth occurs at the melt surface where the local gradients vary sufficiently to cause solidification and remelting of the crystal. Precise control of the furnace and heat exchanger temperatures, combined with minimized thermal perturbations resulting from the submerged interface, gives HEM a big advantage over other growth techniques for growing high-quality crystals.

In the HEM growth, after the crystal is grown, the temperature of the furnace is reduced to just below the solidification temperature and the helium flow is reduced at a desired rate. The whole crystal can therefore be brought to high temperatures to anneal the solidification stresses, followed by uniform cooling to room temperature at a controlled rate. Because *in situ* annealing is part of the solidification cycle, HEM can reduce the defect density. Further, the last and most impure material to solidify is along the crucible walls, where it can be removed. These features of HEM produce uniform growth and the only sapphire free of light scatter.

In the case of sapphire and silicon, it has been demonstrated that once crystal growth parameters are established, large crystals can be grown.

The HEM has been used for the growth of Cr,Nd:$Gd_3Sc_2Al_3O_{12}$ (Cr,Nd:GSAG), Nd:YAG, GaAs, CdTe, Co:MgF_2, aluminum oxynitride (ALON) and BaF_2 crystals.

From an economic point of view, HEM is cost competitive. The furnace is uncomplicated, automated, and well insulated, which results in low equipment, labor and energy costs.

2.3 Proposed Approach for Phase I

It was intended to concentrate on materials doped with rare earth elements which hold promise for use in spectral hole burning applications. Growth of these materials in single-crystal form was to be evaluated and the crystals characterized for identifying potential candidate materials. It was intended to evaluate europium-doped yttrium silicate (Eu:Y_2SiO_5) and europium-doped yttrium aluminum oxide (Eu:$Y_3Al_5O_{12}$, Eu:YAG) for growth of single crystals. Based upon the characteristics of these materials, it was intended to identify other materials for spectral hole burning applications.

Initially attempts were made to produce Eu:Y_2SiO_5 using the Heat Exchanger Method (HEM). Experiments with unseeded, directional solidification of this material from the melt by HEM did not produce a transparent bulk crystal. Based upon technical discussions at a Workshop on Persistent Spectral Hole Burning at IBM, Almaden Research Laboratory in San Jose, California, during July 28 through 30, 1993, it was decided that Eu:Y_2SiO_5 samples were available to the community and, therefore, the emphasis on producing Eu:Y_2SiO_5 during the current program was reduced. Investigations to produce Eu:$YAlO_3$ were pursued instead. The effort to produce Eu:YAG was also pursued with more emphasis. Based upon informal discussions within the technical community and with the Program Manager, it was decided that the feasibility of producing YAG crystals doped with Tm, Tb and Ce should also be investigated.

3.0 Experimental Results

3.1 Crystal Growth

The objective of the program was to show feasibility of producing several doped crystals using the Heat Exchanger Method so that large enough samples of good optical quality could be produced for studies on spectral hole burning applications. Since the objective was to produce high temperature rare earth doped mixed oxides, it was decided to use molybdenum crucibles under an inert gas atmosphere for melting and directional solidification since past experience showed good results using this approach with HEM. For the Czochralski (Cz) growth process, more expensive iridium crucibles are generally used. The details of the experiments carried out are shown in Table I.

The first experiment involved preparation of the Eu:Y_2SiO_5 charge using component oxides and evaluation of the reactivity of the charge with the crucible and atmosphere in the HEM furnace. Data was also taken to establish melting point and directional solidification parameters. The charge was initially heated under reduced pressure; however, the pressure in the furnace chamber was increased after melting of the charge. A polycrystalline pellet was produced after directional solidification. In the bulk form, this material was opaque. However, in thin sections it appeared to be translucent in the as-grown state.

In the second experiment the material solidified in experiment #1 was used as charge in a synthesized YSO form. It was heated to slightly higher temperature before directional solidification was started. The pellet appeared to grow as layers, but the material was not clear two phases were formed.

A portion of the product from experiment #2 was melted in a tungsten crucible and directionally solidified thereafter. This material also did not show clarity in the as-grown bulk state. The material appeared to be dark and this discoloration was attributed to oxygen defects because of growth under non-oxidizing conditions. A sample was heated in air at 900°C for 24 hours. This sample turned white, but it was still not clear.

Experiments #4 and 6 were attempts to produce Eu:$YAlO_3$ by directional solidification from the melt. A grayish-white pellet (Figure 2) of this material was produced which was transparent in thin sections. However, it was not transparent in bulk form. The appearance of the pellets produced showed that more than one phase was formed. Based upon the phase diagram of the Y_2O_3-Al_2O_3 system, it is possible to form several phases in addition to the $YAlO_3$ phase.

Experiment #5 involved single-crystal growth of Eu:YAG. An undoped YAG crystal was used as seed and component oxides were used as

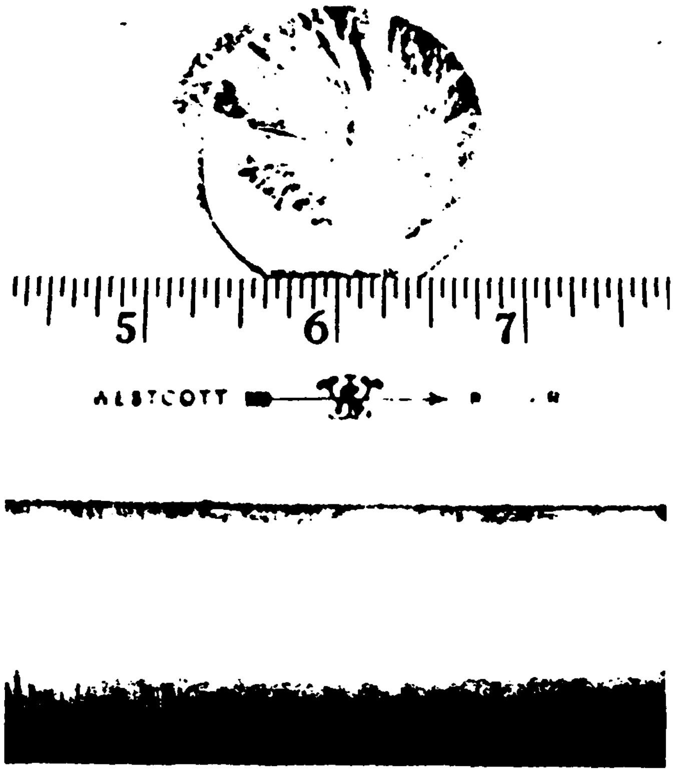

Figure 2 Bottom surface of an Eu:$YAlO_3$ pellet produced by unseeded, directional solidification from the melt. The material showed transparency in thin sections.

Table I

Details of Experiments Carried Out During the Program

Experiment #	Charge	Crucible	Melting Point (°C)	Solidification Time/Hrs	Remarks
1	Eu:YSO	4.5 cm dia Mo	-	-	Evaluate reaction of charge with crucible and atmosphere
2	Eu:YSO	4.5 cm dia Mo	2020	19	Material grown in layers
3	Eu:YSO	3 cm dia W	2020	-	Material from Experiment #2 used as charge
4	Eu:YALO	4.5 cm dia Mo	1910	26.5	Material transparent in thin sections
5	Eu:YAG	6 cm dia Mo	1930	86	Core-free single crystal formed
6	Eu:YALO	4.5 cm dia Mo	1910	74.5	Material not transparent in bulk form
7	Eu:YAG	6 cm dia Mo	1930	117	Single crystal formed
8	Tm:YAG	6 cm dia Mo	1930	52	Single crystal samples fabricated
9	Eu:YSO	4.5 cm dia Mo [tapered bottom]	2020	84.5	Material not transparent
10	Tb:YAG	6 cm dia Mo	1930	111	Single crystal samples fabricated
11	Ce:YAG	6 cm dia Mo	1930	110.5	Yellow crystals formed

charge. The charge was melted and directionally solidified by extracting heat through the seed crystal by the heat exchanger in the HEM furnace. After directional solidification, central section was single crystal (Figure 3). Approximately 1 cm cube crystals could easily be fabricated from this crystal.

A unique characteristic of this YAG crystal grown by HEM is that it did not show the core formation routine for such crystals grown using the Czochralski process. The result of this experiment showed that even though several phases can be formed in the Y_2O_3/Al_2O_3 system, a single crystal Eu:YAG crystal was produced because seeded growth was carried out. Under these conditions, the YAG composition is grown off the YAG seed and the secondary phases, if any, are rejected to the melt. Material towards the end of solidification was discolored and was opaque. This may be due to rejection of impurities and secondary phases during crystal growth. There may have been contamination from the heat zone of the HEM furnace which could have also caused this discoloration of the charge during experiment #5. The HEM furnace was therefore baked out at high temperatures under vacuum to remove this contamination from the heat zone before the next experiment.

The molybdenum crucible used in experiment #5 was cleaned and reused for the directional solidification experiment #7. The charge consisted of component oxides pertaining to the Eu:YAG composition. Towards the end of solidification, the crucible cracked. However, a transparent crystal with good optical quality (Figure 4) was still produced.

The heat zone of the HEM furnace was again baked out under vacuum to remove the contamination due to the heat from the cracked crucible.

Based upon technical discussions with SRI, it was felt that Tm:YAG and Tb:YAG are potential candidates for hole burning applications. To substantiate this, a charge consisting of Y_2O_3, Al_2O_3, Tm_2O_3, and an undoped YAG single crystal seed were loaded in approximately 6 cm diameter molybdenum crucible for experiment #8. After directional solidification, the charge was cooled to room temperature. During the cooldown stage, a safety tripped off power to the furnace and the charge was cooled rapidly to room temperature. The fast cooldown cracked the crystal into several large pieces. However, even in the as-grown state, the crystals showed transparency and good optical quality.

In experiment #10, an attempt was made to produce Tb:YAG crystals using procedures similar to experiment #8. The charge as-removed from the crucible after directional solidification is shown in Figure 5. A large enough transparent single crystal sample (Figure 6) was fabricated from this ingot for characterization and evaluation for potential hole burning application.

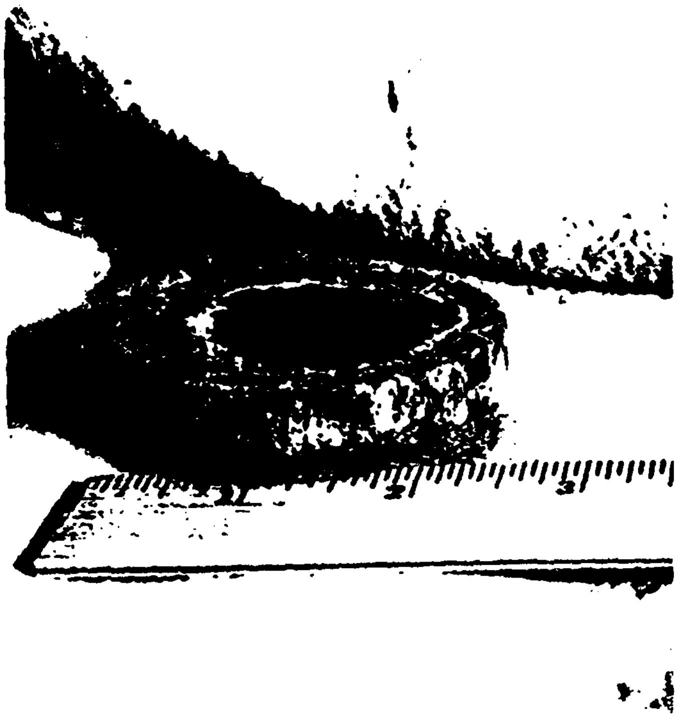

Figure 3 A 6-cm diameter Eu:YAG ingot grown by HEM. The top surface, after lapping, showed large, single-crystal central area. No core was observed in the crystal.

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Figure 4 Approximately 1 cm cube of Eu:YAG fabricated from crystal grown in Experiment #7. The material showed good optical quality.

Figure 5 As-grown ingot of Tb:YAG produced by HEM in a 6 cm diameter crucible in Experiment #8.

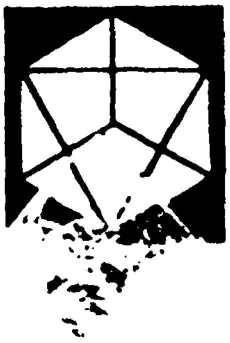

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Figure 6 Approximately 1 cm cube of Tb:YAG fabricated from ingot shown in Figure 5. This sample was sent for characterization for spectral hole-burning application.

Earlier attempts to produce Eu:Y_2SiO_5 and Eu:$YAlO_3$ showed that more than one phase was formed when directional solidification was carried out without a seed. It is likely that more than one phase formed due to deviations in stoichiometry or to volatization of one of the components in the charge. Under these conditions and with unseeded growth, more than one phase may be solidified. To minimize this problem, a conical bottom crucible was used in experiment #9 for directional solidification of Eu:Y_2SiO_5. The intent was to melt the entire charge and during initial stages of directional solidification form a small crystal which could be used as a seed for preferential solidification of the composition of the seed crystal, thereby rejecting the secondary phases during later stages of solidification. This experiment did not yield a transparent single crystal.

In experiment #11 an attempt was made to produce Ce:YAG crystals from an undoped YAG seeded directional solidification. This experiment yielded yellow, clear crystals which could be utilized for evaluation of hole burning application.

3.2 Characterization

From crystal growth experiments large clear crystals of good optical quality were produced of Eu:YAG, Tm:YAG, Tb:YAG and Ce:YAG. All these crystals were directionally solidification attempts from the melt using a seed crystal. In all cases core-free crystals were produced and the secondary phases and/or contamination was rejected to the last material to solidify. Attempts to produce Eu:Y_2SiO_5 and Eu:$YAlO_3$ in single-crystal form were not successful in producing large transparent optical quality crystals. This was attributed to formation of more than one phase and directional solidification of multiple phases resulted in lack of transparency in bulk form.

Transparent samples of Eu:YAG, Tm:YAG, Tb:YAG and Ce:YAG were sent to SRI for characterization. The transmission spectra for the crystals was measured in the wave length range of 400 to 900 nm. In addition to the samples grown during the program, a single crystal sample of Ce:$Gd_3Sc_2Al_3O_{12}$ (Ce:GSAG) was also included for characterization. The data for Tm:YAG and Ce:GSAG is shown in Figures 7 and 8.

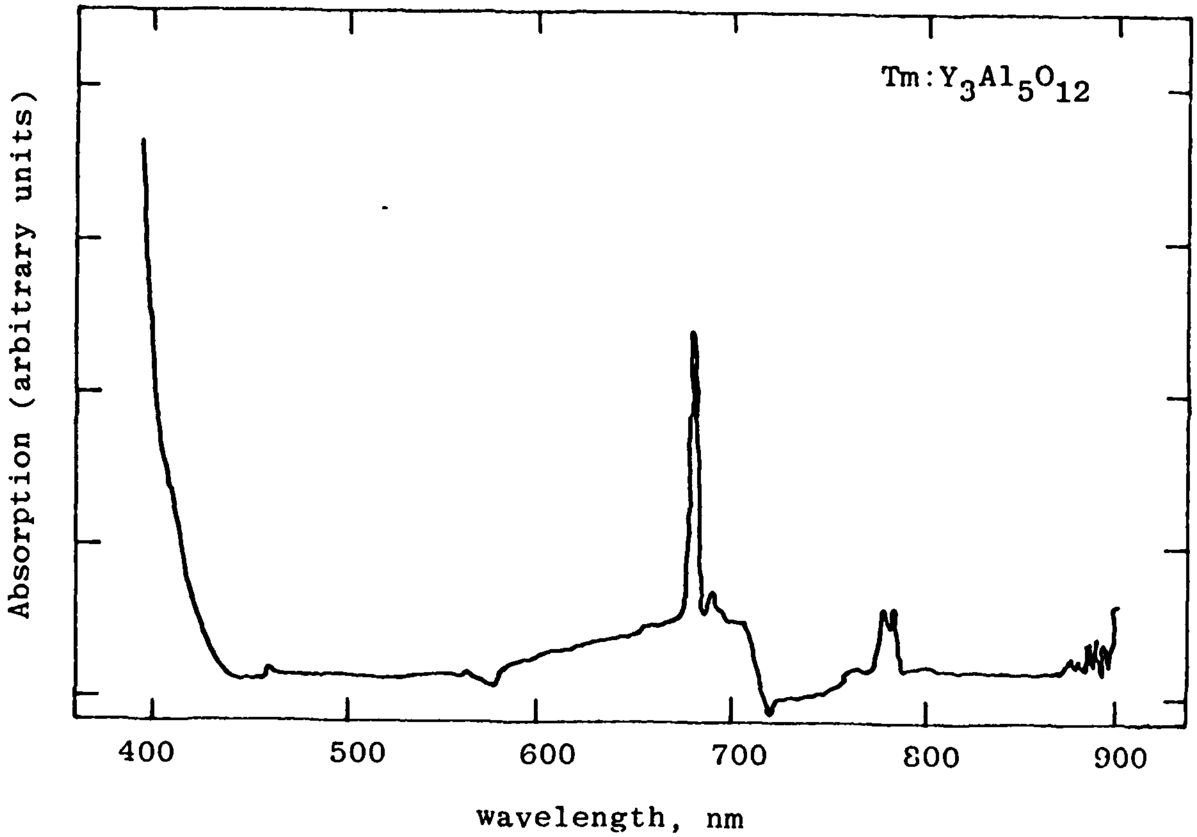

Figure 7 Absorption spectra for a Tm:YAG sample for 500-900 nm wavelength range.

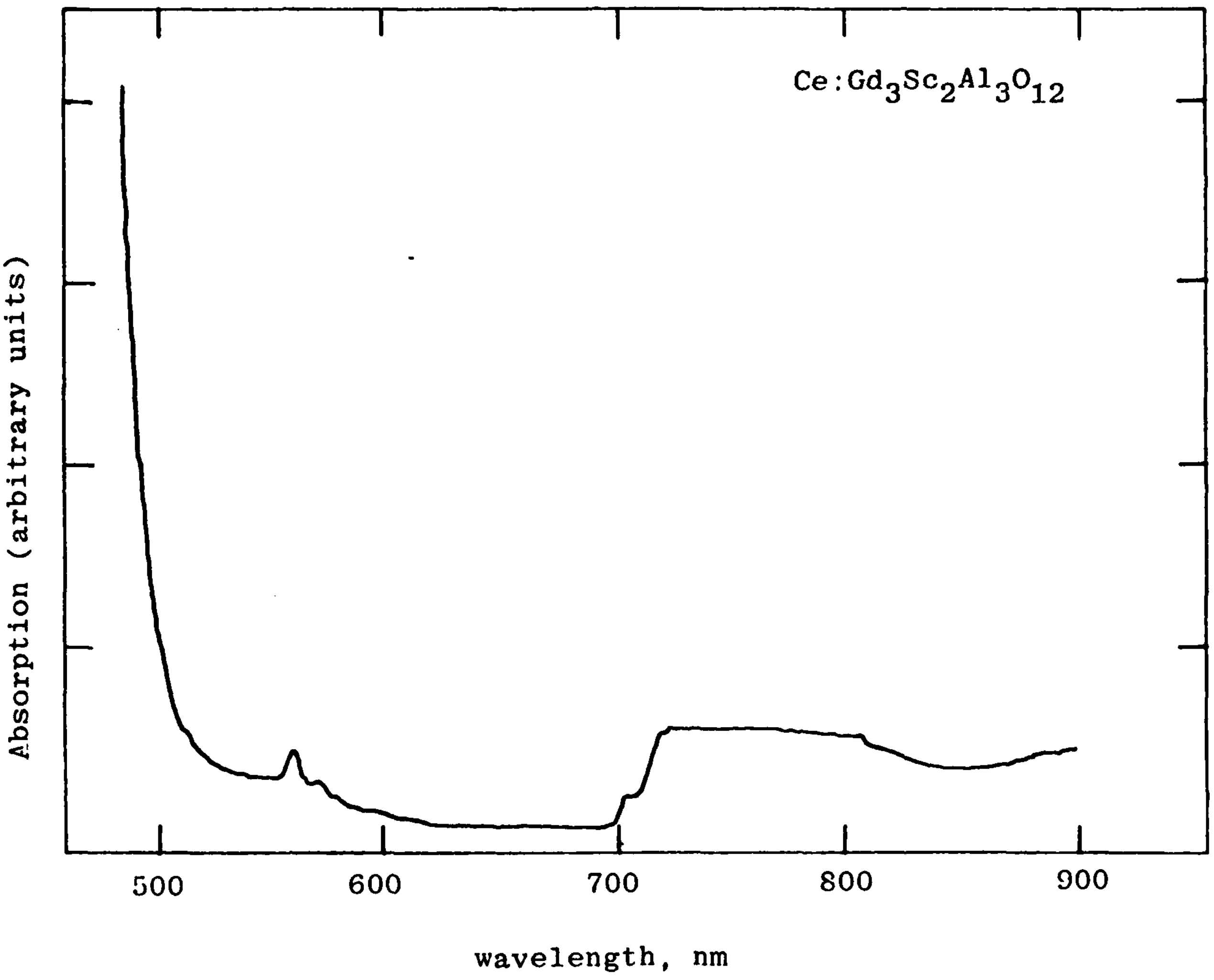

Figure 8 Absorption spectra for a Ce:GSAG sample grown by HEM.

4.0 Conclusions and Discussion

The objective of the experimental program was to grow Eu:$Y_3Al_5O_{12}$ (Eu:YAG) and Eu:Y_2SiO_5 (Eu:YSO) crystals. During the program Eu:YAG, Tm:YAG, Tb:YAG, Ce:YAG single crystals were grown and attempts were made to produce Eu:YSO and Eu:YLO crystals. Large single crystals were produced for YAG hosts using HEM. These crystals were grown in approximately 6 cm diameter crucibles using directional solidification, initiating off a melted-back single crystal seed of undoped YAG. The crystals were transparent in the as-grown state and large samples could be fabricated. These samples appeared to be of high optical quality after polishing.

For the YSO and YAlO host, transparent single crystals could not be produced because more than one phase was formed. This was attributed to directional solidification without a seed crystal to ensure that the right composition corresponding to the seed crystal nucleated off it. Due to time constraints, unseeded single crystal growth for these hosts could not be carried out.

YAG crystals were grown with dopants Eu, Tm, Tb and Ce. In all cases, optical quality samples were produced and sent for characterization for spectral hole burning application. A sample of previously-grown Ce:GSAG crystal was also included for characterization.

In addition to the crystals described above, large crystals of Eu:YSO are available to the scientific community for detailed characterization of hole burning application.

Based upon the data generated on crystals grown during the program as well as other single crystals available to the scientific community, neither material can be ruled out for use in spectral hole burning applications, nor has it been firmly established that a particular crystal is ideally suited for this application.

Cooperative discussions with SRI has resulted in preference for producing Ce:YAG and Tb:YAG crystals for potential use in spectral hole burning applications. Ce^{2+} as a dopant is an interesting dopant ion as it is expected to behave in a manner similar to Pr^{3+}. Based upon spectroscopic data, it was observed that in Ce:YAG, the dopant is mostly Ce^{3+} state and no sharp lines were observed in the visible part of the spectrum. This is consistent with earlier efforts to produce a tunable UV laser. Because of the importance of Ce as a dopant, a sample of Ce:GSAG produced earlier at Crystal Systems was also sent to SRI for characterization. The data on this sample has not been received.

Tb:YAG is of interest because the Tb^{3+} ion shows an isolated absorption line at approximately 800 nm. This absorption is important for hole burning memory applications because it allows use of this material with currently available semiconductor diode lasers. These lasers are compact, efficient, and expected to be available at very low cost in the near future. Systems combining a diode laser with a hole burning material could be very compact, low-cost, efficient and quickly commercialized. The sample of Tb:YAG showed a large absorption near 800 nm spectral region; however, detailed characterization is necessary to evaluate this material for hole burning memory devices. The Tb^{3+} ion is an odd electron system with a large electron magnetic moment and a two-fold degeneracy in the ground state. Therefore, it is expected that this material will lack permanent memory. Further experiments to characterize and deal with this problem are necessary.

The crystals grown during the program offer unique characteristics for use in hole burning applications. However, detailed characterization is necessary to explore the full potential of these materials. In addition, it is necessary to explore other materials which could be utilized for this application. The state of development of hole burning materials is still in its infancy, and the requirements are many so that it is necessary to carry out further basic studies and combine the devices physics with the materials effort so that the next generation memory devices can be fabricated on a commercial basis.

Combining investigation of several crystals during the program with background experience with HEM, it can be concluded that large single crystals of multicomponent oxide systems can be produced by HEM provided seeded crystal growth is carried out. From these crystals, samples can be fabricated large enough to be characterized for hole burning application.

Other advantages to using HEM also exist; for example, for crystals like YAG, core-free crystals are produced more easily by HEM than by the Czochralski process. Once initial parameters are established, large crystals can be grown by HEM.

In the absence of available single crystal seed material, considerably more effort is required to produce single crystals of complex oxide materials.

5.0 Suggestions for Further Work

Results of the program suggest further development:

- (i) investigate approaches to produce long-term (a few seconds) storage in the Tb^{3+} doped crystals;
- (ii) evaluate growth of Tb^{3+} doped crystals using other hosts and characterize the material for hole burning applications;
- (iii) extend the effort to produce Ce doped crystals in Ce^{3+} ionic state. In hosts such as YAG, the Ce^{3+} should be the predominant state to maintain charge balance. To form Ce^{2+}, either a compensating 4+ ion could be added or a host crystal can be selected so that Ce^{2+} is the stable state.
- (iv) investigate two photon step-wise writing from ground state leading to ionization of the ion. This phenomenon is also known as photon-gating and can lead to "fixing" written data and avoid the inherent risk of erasure during reading prevalent in single photon writing. If achieved at the device level, some requirements on the material could be relaxed to permit hole burning applications.

In addition to the above, there is the possibility of developing new avenues based upon discussions between the device physicist and the material scientist.

6.0 References

1. W. E. Moerner, ed., *Persistant Spectral Hole-Burning: Science and Applications*, Vol. 44 of Topics in Current Physics (Springer-Verlag, Berlin, 1988).
2. T. W. Mossberg, *Opt. Lett.* 7, 77 (1982).
3. M. Mitsunega, M. K. Kim, E. Xu, D. J. Gauthier, J. Huang and T. W. Mossberg, *Opt. Lett.* 16, 103 (1991).
4. M. Mitsunaga, R. Yano and N. Uesugi, *Opt. Lett.* 16, 264 (1991).
5. R. M. Macfarlane and R. M. Shelby, in *Spectroscopy of Solids Containing Rare Earth Ions*, A. A Kaplyanskii and R. M. Macfarlane, eds. (North Holland, Amsterdam, 1987).
6. K. A. Gschneidner, Jr. and L. Eyring, eds., *Handbook on the Physics and Chemistry of Rare-Earths Series* (North Holland, Amsterdam).
7. N. A. Toropov, I. A. Bondar, A. N. Lazarev and Yu. I. Smolin, *Rare Earth Silicates and Their Analogues* (Nauka, Leningrad, 1971).
8. M. J. Weber, ed., *Handbook of Laser Science and Technology*, (CRC Press, Boca Raton, 1981).
9. A. A. Kaminskii, *Laser Crystals--Their Physics and Properties*, Springer Series in Optical Sciences, Vol. 14 (1981).
10. P. Hammerling, A. B. Budgor and A. Pinto, eds., *Tunable Solid State Lasers*, Springer Series in Optical Sciences, Vol. 47 (1985).
11. A. B. Budgor, L. Esterowitz and L. G. DeShazer, eds., *Tunable Solid-State Lasers II*, *Springer Series in Optical Sciences*, Vol. 52 (1986).
12. IEEE J. Quantum Electron. 24 (1988).
13. M. L. Shand and H. P. Jenssen, eds., *OSA Proc. on Tunable Solid State Lasers*, Vol. 5 (1989).
14. H. P. Jenssen and G. Dubé, eds., *OSA Proc. on Advanced Solid State Lasers*, Vol. 6 (1990).
15. G. Dubé and L. Chase, eds., *OSA Proc. on Advanced Solid State Lasers*, Vol. 10 (1991).
16. A. H. Gomes di Mesquita and A. Bril, *Mater. Res. Bull.* 3, 643 (1969).
17. T. E. Peters, *J. Electrochem. Soc. Solid State Sci.*, 116, 985 (1969).
18. I. A. Bondar, A. A. Kolpakova, L. Ya. Markovskii, A. N. Sokolov, L. E. Tarasova and N. A. Toropov, *Bull. Acad. Sci. USSR Phys. Ser.* 33, 977 (1969).
19. A. M. Morotov, M. V. Petrov, V. R. Startsev, A. M. Tkachuk and P. F. Feofilov, *Opt. Spectrosc.* 41, 641 (1976).
20. R. Yano, M. Mitsunaga and N. Uesugi, *Opt. Lett.* 16, 1884 (1991).
21. R. M. Macfarlane, R. M. Shelby and R. L. Shoemaker, *Phys. Rev. Lett.* 43, 1726 (1979).

22. R. M. Shelby and R. M. Macfarlane, *Phys. Rev. Lett.* 45, 1098

(1980); R. M. Macfarlane and R. M. Shelby, *Opt. Commun.* 59, 169 (1981).
23. J. Huang, J. M. Zhang, A. Lezama and T. W. Mossberg, *Phys. Rev. Lett.* 63, 78 (1989).
24. J. Huang, J. M. Zhang and T. W. Mossberg, *Opt. Commun.* 75, 29 (1990).
25. M. Mitsunaga and N. Uesugi, *Opt. Lett.* 15, 195 (1990).