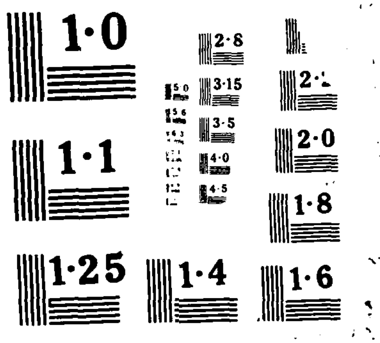
AD-A190 874 TUNABLE OPTICAL SOURCES/GROWTH AND CHARACTERIZATION OF NONLINEAR OPTICAL..(U) STANFORD UNIV CA EDWARD L GINZTON LAB OF PHYSICS R L BYER ET AL. JAN 88 GL-4313 1/1

UNCLASSIFIED ARO-21327.17-PH DAAG29-84-K-0071 F/G 20/6 NL


1·0
2·8
3·15
3·5
4·0
4·5
5·0
2·0
1·8
1·1
1·25
1·4
1·6

UNCLASSIFIED

SECURITY CLASS

(2)

REPORT DOCUMENTATION PAGE

AD-A190 874

DTIC FILE COPY

1a. REPORT SECURITY CLASSIFICATION: Uncl	1b. RESTRICTIVE MARKINGS
2a. SECURITY CLASSIFICATION AUTHORITY	3. DISTRIBUTION/AVAILABILITY OF REPORT: Approved for public release; distribution unlimited.
2b. DECLASSIFICATION / DOWNGRADING SCHEDULE	
4. PERFORMING ORGANIZATION REPORT NUMBER(S): Ginzton Laboratory 4313	5. MONITORING ORGANIZATION REPORT NUMBER(S): ARO 21327.17-PH
6a. NAME OF PERFORMING ORGANIZATION: Stanford University Ginzton Laboratory, — 6b. OFFICE SYMBOL *(If applicable)*	7a. NAME OF MONITORING ORGANIZATION: U. S. Army Research Office
6c. ADDRESS *(City, State, and ZIP Code)*: Stanford, California 94305	7b. ADDRESS *(City, State, and ZIP Code)*: P. O. Box 12211 Research Triangle Park, NC 27709-2211
8a. NAME OF FUNDING/SPONSORING ORGANIZATION: U. S. Army Research Office — 8b. OFFICE SYMBOL *(If applicable)*	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER: DAAG29-84-K-0071
8c. ADDRESS *(City, State, and ZIP Code)*: P. O. Box 12211 Research Triangle Park, NC 27709-2211	10. SOURCE OF FUNDING NUMBERS: PROGRAM ELEMENT NO. / PROJECT NO. / TASK NO. / WORK UNIT ACCESSION NO.

11. TITLE *(Include Security Classification)*
Tunable Optical Sources/
Growth and Characterization of Nonlinear Optical Materials

12. PERSONAL AUTHOR(S) Robert L. Byer and Robert S. Feigelson

13a. TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT *(Year, Month, Day)*	15. PAGE COUNT
Final	FROM 8/1/84 TO 10/31/87	January, 1988	17

16. SUPPLEMENTARY NOTATION The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

17. COSATI CODES		
FIELD	GROUP	SUB-GROUP

18. SUBJECT TERMS *(Continue on reverse if necessary and identify by block number)*
Nonlinear optics, nonlinear optical materials, barium borate, silver gallium selenide, lithium niobate, harmonic generation, optical parametric oscillation, slab laser (cont on reverse)

19. ABSTRACT *(Continue on reverse if necessary and identify by block number)*

Progress in solid-state laser research, nonlinear optical material growth science and nonlinear optical frequency conversion techniques is reported. High quality crystals of silver gallium selenide in lengths up to 3.5 cm are now being grwon for nonlinear infrared applications. The growth of barium borate for ultraviolet and high-intensity visible applications has pursued. With these new materials we have made first demonstrations of optical parametric oscillation and demonstrations of high-average-power frequency conversion capabilities. The stability of the single-axial-mode diode-pumped solid-state lasers is proving vital for the reliable operation of nonlinear optical devices such as cw second harmonic generators and single-mode optical parametric oscillators. Zig-zag slab laser development has continued to provide sources of high-average-power pump radiation.

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT: ☐ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☐ DTIC USERS	21. ABSTRACT SECURITY CLASSIFICATION: Unclassified
22a. NAME OF RESPONSIBLE INDIVIDUAL	22b. TELEPHONE *(Include Area Code)* / 22c. OFFICE SYMBOL

DD FORM 1473, 84 MAR — 83 APR edition may be used until exhausted. All other editions are obsolete.

SECURITY CLASSIFICATION OF THIS PAGE

UNCLASSIFIED

18. Subject Terms - cont.

diode-laser-pumped solid-state laser, resonant-cavity harmonic generation, infrared, ultraviolet.

ARO 21327.17-PH

Tunable Optical Sources /
Growth & Characterization of Nonlinear Optical Materials

Final Report
for the period
1 August 1984 to 31 October 1987

Principal Investigators
Professor Robert L. Byer, Applied Physics Department
Professor Robert S. Feigelson, Center for Materials Research

Stanford University
Stanford, California 94305.

Ginzton Laboratory
Report Number 4313

Report date
January 1988

Prepared for
U. S. Army Research Office

Contract Number DAAG29-84-K-0071

APPROVED FOR PUBLIC RELEASE;
DISTRIBUTION UNLIMITED.

Tunable Optical Sources / Growth & Characterization of Nonlinear Optical Materials

Professor Robert L. Byer, Applied Physics Department
Professor Robert S. Feigelson, Center for Materials Research
Stanford University, Stanford CA 94305-4085

Abstract

Progress in solid-state laser research, nonlinear optical material growth science, and nonlinear optical frequency conversion techniques is reported. High quality crystals of silver gallium selenide in lengths up to 3.5 cm are now being grown for nonlinear infrared applications. The growth of barium borate for ultraviolet and high-intensity visible applications has been pursued. With these new materials, we have made first demonstrations of optical parametric oscillation and demonstrations of high-average-power frequency conversion capabilities. The stability of the single-axial-mode diode-pumped solid-state lasers is proving vital for the reliable operation of nonlinear optical devices such as cw second harmonic generators and single-mode optical parametric oscillators. Zig-zag slab laser development has continued to provide sources of high-average-power pump radiation.

Tunable Optical Sources /
Growth & Characterization of Nonlinear Optical Materials

Table of Contents

Tunable Optical Sources / Growth and Characterization of Nonlinear Optical Materials

Professor Robert L. Byer, Applied Physics Department
Professor Robert S. Feigelson, Center for Materials Research

I. Introduction

Historically the development of lasers has focussed on the optimization of a few narrow linewidth, single frequency sources. Gas carbon dioxide lasers and solid state neodymium lasers are now efficient, high average power infrared radiation sources at the wavelengths 10.6 and 1.06 μm. Tunable coherent radiation can be generated by the nonlinear optical frequency conversion of these monochromatic sources in appropriate materials. Harmonic generation produces the frequency harmonics of the applied radiation and is now an established technology for Nd:YAG lasers. Optical parametric oscillation and amplification provide a method of continuously tuning the output frequency of the nonlinear conversion across a wide bandwidth. Nd:YAG pumped parametric oscillators using lithium niobate have been well studied, but difficulties with operating these devices has slowed their development.

We proposed a cooperative research program to study tunable coherent sources and nonlinear materials and devices. It is our program aim to develop efficient, high average power tunable coherent radiation by nonlinear optical generation from fixed frequency sources. This requires the development of stable pump laser sources, improved nonlinear materials, and practical nonlinear techniques.

The laser studies have been conducted by the Quantum Electronics group under the direction of Robert L. Byer. Laser research has centered on the slab geometry concept for improved performance of solid state sources. We have developed two unique, high coherence, high average power Nd:glass slab lasers. The fixed slab design has high energy and peak power per pulse whereas the moving slab laser features a high average power potential. Diode pumping of stable miniature rod and slab lasers of both glass and crystal has also been investigated. These small lasers have been used as injection seeding sources to control the output of the larger lasers. In addition a Nd:YAG slab amplifier with over 60 dB of gain has been built for the amplification of diode pumped Nd:YAG laser sources. The laser devices have demonstrated output properties that will allow efficient nonlinear conversion.

Research in the growth technology and characterization of existing and new nonlinear optical materials has been carried out by the Crystal Science group headed by Professor Robert S. Feigelson. Progress in the growth of the chalcopyrites, silver gallium sulfide and silver gallium selenide ($AgGaS_2$ and $AgGaSe_2$), has resulted in high quality angle phasematched crystals of lengths up to 35 mm. The recent development of beta-barium borate (BBO) has spurred an intensive effort to grow this new material here, and a 12-mm-long crystal has been produced. We have demonstrated that both magnesium oxide doping and lithium in-diffusion of lithium niobate ($LiNbO_3$) substantially reduces the photorefractive damage that has limited the use of this material. Each of these materials has unique features which extends the range of performance of available nonlinear materials. As these and new materials are developed they are evaluated experimentally with various nonlinear conversion techniques

Professor Byer has overseen the investigation of the application of these crystals in nonlinear devices. Efficient second harmonic generation of a CO_2 laser and optical parametric oscillation have been demonstrated with $AgGaSe_2$. The second through fifth harmonics of 1.06-μm Nd laser radiation have been generated with high efficiency in BBO. An external resonant cavity second harmonic generator using $MgO:LiNbO_3$ has shown significantly enhanced harmonic generation of a low intensity diode pumped Nd:YAG laser. A monolithic optical parametric oscillator of $MgO:LiNbO_3$ has demonstrated stable single axial mode operation. Our cooperative arrangement of growth and evaluation has led to much progress in the improvement of nonlinear materials and we anticipate continued progress in the practical application of nonlinear crystals to tunable coherent devices.

II. Research Results

A. Laser Development

The zigzag optical path slab geometry is an established method of reducing thermal focusing and birefringence in solid state lasers. By using the slab geometry, solid state laser systems can be scaled to higher repetition rates without sacrificing beam quality. The goal of our fixed slab laser design is a high peak and average power Nd:glass laser with a polarized, low divergence output. This laser will act to generate a source of incoherent x-rays when focussed to produce high temperature plasmas. It will also be a pump laser for nonlinear frequency conversion studies at high average power levels.

A small test bed Nd:glass slab laser was first built at Stanford in 1981. That laser confirmed computer model performance predictions but indicated the need for improvement in the control of the thermal distribution in the slab and for preventing degradation of the total internal reflection surfaces[1]. The concept of protecting the slab faces from the coolant water by cooling the slab by conduction through a thin protective layer was implemented in the following two generations of lasers. The present laser[2], designed in 1985, also includes the optimization of the slab temperature distribution by uniformly pumping and cooling and controlling the boundary conditions on the slab.

The 350- x 64- x 6-mm Nd phosphate glass slab in our laser is conduction cooled through a thin helium layer and sapphire windows. The electrical pulse-forming system for this laser is capable of delivering energies up to 5 kJ in 200-μs pulses at 20 kW of average power to the four flashlamps used to pump the slab. Normal conservative operation is presently at 2 kJ per pulse at 2 Hz. The laser operates as a flat-flat oscillator in the bottom 2 centimeters with a relay imaged multipass amplifier in the remaining slab aperture. With a 2-kJ electrical input, this extracts 5 J of laser energy in a single spatial mode with divergence about three times the diffraction limit and a Q-switched pulse length of 50 ns. For multi-transverse-mode stable oscillation, the output is 20 J. The uniformity of pumping and cooling results in only small thermal optical distortions except near the slab edges and we predict that operation at 10 Hz will be relatively unaffected by these effects. So far average power tests have been conservative to reduce the chance of fracturing the slab.

We have recently implemented the first experimental version of the moving slab Nd:glass laser. In this design a large Nd:glass slab is repetitively moved across a small pumping area to spread the thermal load while localizing the gain. This offers the potential for high average power and shall be more completely tested with the arrival of appropriate power supplies in the near future.

Recent progress in diode lasers and laser arrays has allowed their use as effective pumps for small solid state lasers. Diode pumping can result in exceptionally stable laser output. We developed cw diode-pumped monolithic Nd:YAG ring lasers for the injection seeding of our higher power lasers. These are small crystals that are polished and coated to create an internal laser cavity of a ring geometry. Careful design allows high power single-axial-mode output. Single-axial-mode seeding of the larger Q-switched lasers smooths the

---

1. J. M. Eggelson, T. J. Kane, K. Kuhn, J. Unternahrer and R. L. Byer, "The Slab Geometry Laser - Part I: Theory," IEEE J. Quantum Electron. QE-20, 289 (1984); T. J. Kane, J. M. Eggleston and R. L. Byer, "The Slab Geometry Laser - Part II: Thermal Effects in a Finite Slab," IEEE J. Quantum Electron. QE-21, 1195 (1985).
2. M. K. Reed and R. L. Byer, "Performance of a Conduction Cooled Nd:Glass Slab Laser," Proc. SPIE 736, New Slab and Solid-State Laser Technologies and Applications, 38 (1987).

Q-switched temporal pulse, removing random high intensity spikes and stabilizing the output. We have also developed a diode-pumped mode-locked Nd:glass laser with 10-ps output pulses. This has been used to injection mode-lock the Q-switched moving slab laser. The technique of injection seeding allows the desirable characteristics of the diode pumped systems to be scaled up to high power for effective nonlinear conversion.

Slab laser research with crystalline materials has continued. We have designed a multiple-pass slab geometry Nd:YAG laser for the amplification of the diode-pumped monolithic Nd:YAG oscillators. Angular multiplexing was achieved by utilizing internal paths through the slab of varying numbers of internal reflections. This flashlamp pumped laser displayed a 4-pass gain of 62 dB which remained linear up to 12 mJ of extracted energy.

B. Growth & Characterization of Nonlinear Optical Materials

The objective of this portion of the program has been the growth of high optical quality crystals of the important nonlinear optical materials silver gallium selenide ($AgGaSe_2$) and beta barium borate (BBO) which are useful for IR and UV applications, respectively. $AgGaSe_2$ received the major emphasis and was studied continuously throughout the program. Research on BBO began late in the first semi-annual reporting period.

Silver gallium selenide ($AgGaSe_2$)

Two major objectives were undertaken concerning $AgGaSe_2$. The first involved scaling up the boule size to 40-mm diameter, so that longer, more efficient crystals could be prepared. The second objective was to develop a better understanding of the types of optical defects which form in this material, and the mechanisms by which they form, so that crystals with lower residual absorption can be produced. The scaling of boule size was successfully accomplished midway through the program. Boules of 37-mm diameter can now be produced with good yield rates. Minor cracking in the top regions of the boules is still a problem and is due to the formation of a second Ag_2Se rich phase caused by segregation effects. This problem reduces the yield of 3.5-cm-long fabricated bars. Decanting, or pouring off the last liquid to freeze would solve the problem, but the mechanical complications in doing this with a 900-C furnace loaded with a delicate quartz ampoule did not seem to be an attractive solution. Increasing the length of the charge should alleviate the problem without major perturbations to the growth system and this approach will be pursued at a later date. Gaining an increased understanding of the nature of the optical scattering defects that form in $AgGaSe_2$ and how they affect its optical properties and device performance, was considered a more important aspect of the program.

The optical defects found in as-grown $AgGaSe_2$ crystals, and which are discussed in detail in the publications resulting from this research program[3], are known to be Ga_2Se_3-rich precipitates. The density and distribution of these precipitates vary with growth conditions. Fortunately these can be removed by heat treatment procedures with excellent results. Residual scattering defects, which can be observed only after heat treatment, appear to be internally facetted voids, or negative crystals and usually remain in varying concentrations. Experiments with reduced growth rates and Ag_2Se-rich melts have suggested that these residual defects are caused by interface instability (a condition leading to cellular breakdown), and are most likely silver-rich in the as-grown condition. They appear similar in morphology to macroscopic silver–rich inclusions that had previously been identified .

The conventional approaches to dealing with inclusions at the growth interface, which are usually due to impurities or non-stoichiometry effects, involve modified melt composition, slower growth rates, steeper temperature gradients at the growth interface and melt stirring to remove the Ag_2Se-rich material or impurities that build up there. In the last four month reporting interval of this program , (7-1-87 through 10-31-87) slower growth rates have been shown to yield qualitatively better material with fewer and smaller residual scattering defects. A series of experiments has recently been launched to gain quantitative measurements of this effect. We have also completed one melt-stirred growth using a new method developed in our laboratory, known as coupled perpendicular vibrational stirring[4]. This boule, upon preliminary inspection, also looked to be of good optical quality. Preliminary fabrication and heat treatment must first be carried out before a reliable qualitative or quantitative assessment can be made.

Beta barium borate (BBO)

At the beginning of the program, there was literally no "hands-on" experience with the growth of beta-barium borate (BBO) in this country. Our early objective was, therefore, to rapidly catch up with the workers in the Peoples.Republic of China who were growing relatively large crystals by the top-seeded solution growth method. After some

3. R.S.Feigelson and R.K.Route,"Recent Development in the Growth of Chalcopyrite Crystals for Nonlinear Infrared Applications,"Optical Engineering 26, 113, (1987).
4. W.-S. Liu, M. F. Wolf, D. F. Elwell and R. S. Feigelson, "Vibrational Stirring: A New Method for Rapidly Mixing Solutions and Melts During Growth," J. Crystal Growth 82, 589 (1987).

experimentation and false starts, due to purposefully vague scientific reporting by the Fujian Institute of Research on the Structure and Chemistry of Matter, we found that sodium oxide was a suitable solvent for the growth of BBO. From 300 gram solutions we were then able to grow 5-cm-diameter by 12-mm-thick single crystals much like those grown in the P.R.C. The real problem with BBO faced here and abroad is controlling solvent inclusions. We have since been experimenting with flux modifiers and new solvents in order to reduce solvent viscosities and in theory, reduce the density of inclusions. We have also studied growth furnaces having a wide range of thermal parameters, such as axial temperature gradients above the melts and radial gradients within the melts. While we have not found a generic growth method superior to the top-seeded solution growth technique, we have reduced growth time for a 40-gram crystal to under one week, a three-fold increase over the rates reported in the literature. Selected crystal specimens from low defect density sectors of our best crystals are now being tested by Professor Byer's research group.

In cooperation with a visiting scholar from the P.R.C. we have also grown BBO single crystal fibers by the traveling solvent zone method using the laser-heated pedestal grower to generate the molten solvent zone[5]. Interestingly, B_2O_3, a relatively viscous flux, was found to be the most suitable for fiber growth. Off-axis growth was possible and the optical quality of the fibers appeared to be good. Further development of this method in a more mechanically stable grower than the one presently available should also be pursued.

C. Tunable Nonlinear Optical Conversion

Recent nonlinear optical materials developments have been paralleled by the investigation of the application of these crystals in nonlinear devices. The silver gallium selenide, lithium niobate and barium borate grown at the Center for Materials Research have been studied with the laser facilities of the Applied Physics Department. This cooperative arrangement of growth and evaluation has led to a practical understanding of the areas in which material development is most important. By working in this fashion we anticipate continued progress in the practical application of nonlinear crystals to tunable coherent devices.

---

5. R. K. Route and R. S. Feigelson, "Growth of Barium Metaborate (BaB_2O_4) Single Crystal Fibers by the Laser-Heated Pedestal Method," submitted to J. Crystal Growth.

$AgGaSe_2$ has been developed to fill the need for nonlinear materials with applications between 4 and 12 μm. The production of high optical quality crystals at CMR has allowed demonstration of its uses. We have produced the second harmonic of a CO_2 laser pulse with 14% energy conversion in a 20-mm long crystal. Using a 2.05-μm Q-switched Ho:YLF laser as a pump, optical parametric oscillation continuously tunable from 2.65 to 9.0 μm was obtained[6]. With this 20-mm-long crystal the peak efficiency involved 16% energy conversion of the pump.

The advantages of beta-barium borate as a nonlinear material have only recently been demonstrated. Optical quality material was first grown under the direction of C.-T. Chen at the Fujian Institute of Research on the Structure of Matter in the Peoples Republic of China. Professor Chen supplied several crystals of excellent quality for temporary use at Stanford. With this material we investigated generation of the second through fifth harmonics of Nd:YAG laser radiation[7] and demonstrated the first BBO optical parametric oscillator[8]. In the generation of second, third and fourth harmonics of a Nd:YAG laser, BBO performance compared favorably with that of KDP and ADP crystals. Transparency in the ultraviolet also allows effective fifth harmonic generation. This material has a high damage threshold and very low absorption giving excellent potential for high average power applications.

Even though lithium niobate has been used as a nonlinear material since the mid 1960's, development is continuing to improve and extend its properties. Recently it has been shown that doping the material with 5% MgO substantially reduces the photorefractive damage that has plagued $LiNbO_3$ applications with high intensity light. An external resonant cavity second harmonic generator using $MgO:LiNbO_3$ has provided stable and efficient harmonic conversion of a low intensity diode-pumped Nd:YAG laser[9]. The external resonator in this system consists of a small $LiNbO_3$ rod with end faces that are curved and coated to create a monolithic ring geometry cavity. A SHG conversion efficiency of 55% was obtained with only 50 mW of 1.06-μm laser power. The very low transmission loss and good optical quality of the $MgO:LiNbO_3$ and the exceptional stability of the diode pumped laser were important properties in this application.

---

6. R. C. Eckardt, Y. X. Fan, R. L. Byer, C. L. Marquardt, M. E. Storm, and L. Esterowitz, "Broadly tunable infrared parametric oscillation in $AgGaSe_2$", Appl. Phys. Lett. **49**, 608 (Sept. 15, 1986).
7. Chuangtian Chen, Y. X. Fan, R. C. Eckardt and R. L. Byer, "Recent developments on barium borate," SPIE Proc. **681**, 12 (1986).
8. Y. X. Fan, R. C. Eckardt and R. L. Byer, Barium Borate Optical Parametric Oscillator," submitted to Applied Physics Letters.
9. W. J. Kozlovsky, C. D. Nabors and R. L. Byer, "Second-harmonic generation of a continuous-wave diode-pumped Nd:YAG laser using an external resonant cavity," Opt. Lett. **12**, 1014 (1987).

A monolithic optical parametric oscillator (OPO) of $MgO:LiNbO_3$ has demonstrated stable single-axial-mode operation. The diode-pumped Nd:YAG laser was amplified in a 500-ns-long pulse by the multipass Nd:YAG slab, doubled in a 2.5-cm-long $MgO:LiNbO_3$ crystal and used to pump the OPO. Control of the $LiNbO_3$ temperature allowed tuning of the signal output from 834 to 958 nm.

We have also demonstrated simultaneous single-axial-mode laser oscillation and second harmonic generation in a single crystal of $Nd:MgO:LiNbO_3$ and accurate optical index measurements have resulted in a full Sellmeier fit for KTP.

D Future Directions

The development and application of the fixed slab Nd:glass laser is continuing. The fabrication of new slabs will permit maximum average power testing to be completed in the near future. Using this laser as an amplifier for a Nd:YAG laser with a Q-switched pulse length of only 8 ns was studied but the reduced gain at the Nd:YAG wavelength of 1.064 μm compared to the peak gain in phosphate glass at 1.054 μm made this unfeasible. Studies of optical damage in Nd:glass slabs indicate the pulse length can be safely reduced to about 10 ns for a 10-J output in a 2-cm^2 area without risk of damage. We are investigating the use of a Nd:YLF oscillator or a Nd:glass seed laser to shorten the pulse length. The replacement of the helium layer and sapphire window conduction cooling with a coating of magnesium fluoride is under development and will improve the laser efficiency by over 50%. Improvements in glass fracture resistance are possible with new high average power glasses and with surface treatments now available from manufacturers. Using the new glasses will allow us to increase the average power of this laser even further. A collaboration with Sumitomo mining company of Japan should allow us to test a large Nd:GGG slab with very high average power capability in the fixed slab laser.

Arrival of 24-kW, high repetition rate power supplies for the moving slab laser will allow the testing of high power output. With the development of a single spatial mode extraction scheme we plan to use this laser as a laser plasma source for soft X-ray lithography. Diode-pumped injection mode-locking produces very high intensity output with high repetition rate, low energy pulses and will allow efficient nonlinear conversion of the output of this laser. We plan to continue the improvement of the diode-pumped mode-locked seeder. Replacement of the low power pump diode with recently available high power diode arrays will result in an significant increase in output power. Fiber expansion and grating compression of the laser output will result in subpicosecond pulse lengths. Researchers at the University of Rochester have recently demonstrated an exciting technique for increasing the peak power available from a laser amplifier. Temporally expanding a short pulse allows safe amplification to high energy without nonlinear optical damage. The long, high energy pulse can then be contracted in a grating compressor back to its original length with good efficiency and the resulting intensity is far above the damage limit in the amplifier. Applying this scheme to the moving slab laser will allow us to generate extremely hot and efficient laser plasmas at high repetition rate.

Avenues of research for future work on the growth of $AgGaSe_2$ are also clear. Subtle changes in melt compositions coupled with growth parameter modifications (of temperature gradient, growth rate, melt stirring, etc.) must be continued until the optimum conditions are determined. A detailed understanding of the chemistry and crystallization mechanism is very important. Due to the extended duration of each experiment, of up to twelve weeks, it is a relatively slow process to accumulate the requisite data. The new stirring technique may allow us to decrease the preparation time by permitting faster growth rates without degradation of properties. The influence of carbon in the Ag_2Se-Ga_2Se_3 system must also be re-evaluated. We have definite evidence of finite carbon solubility in $AgGaSe_2$ melts at temperatures used in the growth process. Since carbon is used to coat the synthesis and growth ampoules, a better understanding is required of its role, if any, in the formation of optical scattering defects .

The growth and characterization of BBO is being continued under Army Research Contract DAAL03-86-K-0129, which is focused totally on solving some of the difficult growth problems in this very important material. Extensive experiments are planned in the area of quantitative measurements of viscosities in a number of potential solvent systems such as BaF_2 and mixed oxide/fluoride systems. Preliminary microchemical studies of the inclusions in Na_2O-grown BBO have confirmed the presence of sodium. We have not yet identified the chemical compounds involved and their relationship to the phase equilibrium in the system. Additional studies are under way.

During the course of this program we have continuously made available our growth technology to interested parties, including Cleveland Crystals Inc., which is now in pilot production of $AgGaSe_2$. With a rapid transfer of technology to the private sector we hope to improve the speed with which BBO will become a readily available commercial product in this country.

With the positive findings for $AgGaSe_2$ growth we now anticipate being able to produce over-3-cm-long crystals with residual absorptions low enough for efficient operation in resonant (OPO) devices. Improved performance of both harmonic generation and parametric oscillation will soon be demonstrated with these crystals.

Following the success of the resonant cavity second harmonic generator in $MgO:LiNbO_3$, a similar, doubly resonant, device in stoichiometric $LiNbO_3$ is now being developed. The homogeneity of this material results in very low loss and will allow the use of larger crystals. This should produce improved doubling stability and efficiency and we hcpe to be able to demonstrate the generation of a squeezed optical state.

The stability of the single-axial-mode diode-pumped solid-state lasers is vital for the reliable operation of nonlinear optical devices. Nonlinear optical parametric frequency conversion techniques have the potential of preserving the coherence of the pump source. Preliminary measurements indicate that this is accomplished in our long-pulse singly resonant $LiNbO_3$ OPO. Single-mode OPO operation in this case is achieved by the use of a single-mode pump source and with the frequency selection of a long build up time. The demonstration of stable continuous-wave second harmonic generation is also an important step to reliable and stable tunable nonlinear frequency conversion. We now have available cw second harmonic power in excess of the calculated pump threshold for doubly resonant parametric oscillation in $LiNbO_3$. The same techniques as we have used in externally resonant second harmonic generation are applicable to doubly resonant parametric oscillators. The output of the tunable cw parametric oscillator could be used to injection seed high-peak-power pulsed parametric oscillators. The stability and narrow bandwidth of the miniature solid-state lasers used to injection seed both the nonlinear frequency conversion process and the high-power pump lasers would be preserved in the high-power widely tunable radiation which will be generated.

The use of fixed frequency laser sources to generate tunable radiation by nonlinear optical conversion is rapidly approaching a threshold. For the first time the careful engineering of source lasers to match this application is allowing practical devices to be realized. The recent developments in high power laser diodes and high optical quality slab lasers and the steady improvement in the quality of appropriate nonlinear materials promise to provide pulsed and cw tunable coherent sources covering the spectrum from 0.2 to 20 μm. Our cooperative program has coordinated research into all the components of these systems and allowed the development of the stable pump laser sources, improved nonlinear materials, and practical nonlinear techniques that will make up this technology.

III. Scientific Personnel Supported by this Contract

Principal Investigators

Professor Robert L. Byer	Applied Physics Department
Professor Robert S. Feigelson	Center for Materials Research

Senior Research Associates

Robert C. Eckardt	Ginzton Laboratory of Physics
Roger K. Route	Center for Materials Research

Students

Murray Reed

Yuan Xuan Fan, MS degree granted January 1987.

William Kozlovsky

Alan Nilsson

Gregory Magel

Tso Yee Fan, Ph.D. degree granted September 1987.

Technicians

R. J. Raymakers

IV. List of all Publications Supported by ARO Contract DAAG29-84-K-0071

1. Y.X. Fan, R.C. Eckardt, R.L. Byer, R.K. Route and R.S. Feigelson, "$AgGaS_2$ Infrared Parametric Oscillator," Appl. Phys. Lett. **45**, pp. 313-315 (Aug. 15, 1984).

2. M. Reed, K. Kuhn, J. Unternahrer and Robert L. Byer, "Static Gas Conduction Cooled Slab Geometry Nd:Glass Laser," IEEE J. Quantum Electron. **QE-21**, pp.412-414 (May, 1985).

3. T. J. Kane, J. M. Eggleston and R. L. Byer, "The Slab Geometry Laser - Part II: Thermal Effects in a Finite Slab," IEEE J. Quantum Electron. **QE-21**, pp. 1195-1210 (1985).

4. R.C. Eckardt, Y.X. Fan, R.L. Byer, R.K. Route, R.S. Feigelson, and Jan van der Laan, "Efficient Second Harmonic Generation of 10-Micron Radiation in $AgGaSe_2$," Appl. Phys. Lett. **47**, pp. 786-788 (Oct. 15, 1985).

5. R.S.Feigelson, R.K.Route and T.M.Kao, "Growth of Urea Crystals by Physical Vapor Transport," Journal of Crystal Growth **72**, pp 585-594, (1985).

6. R.S.Feigelson and R.K.Route,"Recent Development in the Growth of Chalcopyrite Crystals for Nonlinear Infrared Applications," Proc. SPIE **567**, *Advances in Materials for Active Optics* , pp. 2-10, (August 1985).

7. R. C. Eckardt, Y. X. Fan, R. L. Byer, C. L. Marquardt, M. E. Storm, and L. Esterowitz, "Broadly Tunable Infrared Parametric Oscillation in AgGaSe2", Appl. Phys. Lett. **49**, pp. 608-610 (Sept. 15, 1986).

8 M.K Reed, T. Yamada and R.L. Byer, "Conduction Cooling of Nd:Glass Slab Lasers," Proc. SPIE **622**, *High Power Solid State Lasers*, p. 622 (Sept. 1986).

9. Chuang-tian Chen, Y. X. Fan, R. C. Eckardt and R. L. Byer, "Recent Developments in Barium Borate," Proc. SPIE **681**, pp. 12-19 (1986).

10. J. L. Nightingale, W. J. Silva, G. E. Reade, A. Rybicki, W. J. Kozlovsky and R. L. Byer, "Fifty Percent Conversion Efficiency Second Harmonic Generation in Magnesium Oxide Doped Lithium Niobate," Proc. SPIE **681**, pp. 20-24 (1986).

11. T. Y. Fan and Robert L. Byer, "Modeling and CW Operation of a Quasi-Three-Level 946 nm Nd:YAG Laser," IEEE J. Quantum Electron. **QE-23**, pp. 605-612 (May 1987).

12. Y. X. Fan, R. C. Eckardt, R. L. Byer, R. K. Route and R. S. Feigelson, "Nonlinear Infrared Frequency Conversion in $AgGaS_2$ and $AgGaSe_2$," in *Tunable Solid-State Lasers II,* A. B. Budgor, L. Esterowitz, and L. G. DeShazer, eds., (Springer-Verlag, Berlin, 1986) pp. 360-363

13. R.S.Feigelson and R.K.Route,"Recent Development in the Growth of Chalcopyrite Crystals for Nonlinear Infrared Applications," Optical Engineering **26**, pp. 113-119, (February 1987).

14. T. Y. Fan, C. E. Huang, B. Q. Hu, R. C. Eckardt, Y. X. Fan, R. L. Byer, and R. S. Feigelson, "Second Harmonic Generation and Accurate Index of Refraction in Flux-Grown $KTiOPO_4$," Appl. Opt. **26**, pp. 2390-2394 (June 15, 1987).

15. M. K. Reed and R. L. Byer, "Performance of a Conduction Cooled Nd:Glass Slab Laser," Proc. SPIE **736**, *New Slab and Solid-State Laser Technologies and Applications*, pp. 38-44 (July 1987).

16. R. L. Byer, S. Basu T.Y. Fan, W. J. Kozlovsky, C. D. Nabors, A. Nilsson and G.Huber "Diode Pumped Solid-State Laser Oscillators for Spectroscopic Applications," in *Laser Spectroscopy VIII*, W. Persson and S. Svanberg, eds., (Springer-Verlag, Berlin, 1987), p. 416.

17. R. C. Eckardt, Y. X. Fan, M. M. Fejer, W. J. Kozlovsky, C. D. Nabors, R. K. Route and R. S. Feigelson, "Recent Developments in Nonlinear Optical Materials," in *Laser Spectroscopy VIII*, W. Persson and S. Svanberg, eds., (Springer-Verlag, Berlin, 1987), p. 426.

18. W.S. Liu., M.F. Wolf., D.F. Elwell and R.S. Feigelson,"Vibrational Stirring : A New Method for Rapidly Mixing Solutions and Melts During Growth," Journal of Crystal Growth **82**, p. 589 (1987).

19. W. J. Kozlovsky, C. D. Nabors and R. L. Byer, "Second-Harmonic Generation of a Continuous-Wave Diode-Pumped Nd:YAG Laser Using an External Resonant Cavity," Opt. Lett. **12**, p. 1014 (Dec. 1987).

20. R. K. Route and R. S. Feigelson, "Growth of Barium Metaborate (BaB_2O_4) Single Crystal Fibers by the Laser-Heated Pedestal Method," submitted to J. Crystal Growth.

21. M. K. Reed, W. J. Kozlovsky and R. L. Byer, "Diode-Laser-Array Pumped Neodymium Slab Oscillators," to be published in Optics Letters.

22. C. D. Nabors, W. J. Kozlovsky, and R. L. Byer, "Efficient Second Harmonic Generation of a Diode-Pumped CW Nd:YAG Laser Using an Externally Resonant Cavity," to be published in Proc. SPIE.

23. Y. X. Fan, R. C. Eckardt and R. L. Byer, "Barium Borate Optical Parametric Oscillator," submitted to Applied Physics Letters.

24. A. Lago, R. Wallenstein, Chuang-tian Chen, Y. X. Fan and R. L. Byer, "Coherent 70.9-nm Radiation Generated in Neon by Frequency Tripling the Fifth Harmonic of a Nd:YAG Laser," submitted to Optics Letters.

END
DATE
FILMED
5-88
DTIC