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6. AUTHORS
Robert L. Byer and Robert C. Eckardt

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Ginzton Laboratory, Stanford University
Stanford, CA 94305-4085

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The accomplishments of this program are in the development and application of periodically poled nonlinear optical materials for nonlinear frequency conversion. We have demonstrated the use of periodically poled lithium niobate (PPLN) as a bulk material for external resonant cavity second-harmonic generation with continuous-wave (cw) output power of 1.7 W. Work that is following this investigation is showing that planar waveguides of PPLN may well be the most satisfactory method of generation of 10's of mW of the 659-nm harmonic of the 1.32-μm Nd:YAG laser. We encountered major obstacles obtaining multilayer dielectric coatings necessary to pursue our proposed design of monolithic bulk optical harmonic generators. Additional alternative approaches such as discrete component resonant second harmonic generation employing single domain and periodically poled bulk crystals and monolithic single domain resonators formed by total internal reflection remain under investigation.

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Efficient Nonlinear Optical Conversion of 1.319-micron Laser Radiation

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for the period
1 September 1989 to 30 April 1992

Principal Investigator
Professor Robert L. Byer, Applied Physics Department

Ginzton Laboratory
Stanford University
Stanford, California 94305-4085

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Efficient Nonlinear Optical Conversion of 1.319-micron Laser Radiation

Professor Robert L. Byer, Applied Physics Department
Stanford University, Stanford CA 94305

Abstract

The accomplishments of this program are in the development and application of periodically poled nonlinear optical materials for nonlinear frequency conversion. We have demonstrated the use of periodically poled lithium niobate (PPLN) as a bulk material for external resonant cavity second-harmonic generation with continuous-wave (cw) output power of 1.7 W. Work that is following this investigation is showing that planar waveguides of PPLN may well be the most satisfactory method of generation of 10's of mW of the 659-nm harmonic of the 1.32-μm Nd:YAG laser. We encountered major obstacles obtaining multilayer dielectric coatings necessary to pursue our proposed design of monolithic bulk optical harmonic generators. Additional alternative approaches such as discrete component resonant second harmonic generation employing single domain and periodically poled bulk crystals and monolithic single domain resonators formed by total internal reflection remain under investigation.

Efficient Nonlinear Optical Conversion of
1.319-micron Laser Radiation

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Efficient Nonlinear Optical Conversion of 1.319-micron Laser Radiation

Professor Robert L. Byer, Applied Physics Department

I. Introduction

The goal of this program was to develop techniques for the generation of continuous-wave (cw) single-frequency 659-nm radiation at power levels of approximately 10 mW by optical second harmonic generation of the 1319-nm output of a cw semiconductor-diode-laser-pumped Nd:YAG laser. This goal was motivated both by specific needs for the 659 nm radiation and more general considerations of development and application of sources of coherent radiation at varied wavelengths. Our earlier success with the cw generation of 532-nm radiation by external resonant cavity second-harmonic generation of 1064-nm laser radiation lead us to propose that this method also be used for the generation of 659-nm radiation. We encountered difficulties in this approach and turned instead to an alternate approach of periodically poled nonlinear optical materials. We have made significant progress with the alternative approach, and we believe that it will soon be demonstrated that periodically poled lithium niobate (PPLN) in the configuration of planar waveguides provides a practical solution.

A specific goal that motivated this investigation was the development of a source of coherent cw radiation that could replace helium-neon lasers in surface acoustic wave (saw) devices and in integrated optical devices. The wavelength of the 659-nm harmonic of the 1319-nm Nd:YAG laser is desirable because it results in significantly less damage in the saw devices than would result from shorter wavelengths, and lasers operating at this wavelength could replace 633-nm He-Ne lasers in existing devices. There are additional applications that motivate the continued investigation of second-harmonic generation pumped by the 1319-nm Nd:YAG laser for the generation of coherent cw 659-nm radiation.

Diode-pumped solid-state lasers such as the 1319-nm Nd:YAG laser provide unique characteristics, and these characteristics are preserved in nonlinear frequency conversion devices such as harmonic generators, optical parametric oscillators, and sum and difference frequency generators. The semiconductor-diode-laser-pumped solid-state lasers are highly efficient and reliable. The solid-state laser has improved coherence and stability compared to the array of semiconductor-diode lasers used for pumping. Monolithic solid-state lasers in the ring configuration have a high resistance to perturbation by optical feedback, and the output of these lasers can be precisely tuned either slowly by changing temperature or rapidly by piezoelectric control of the laser resonator length. Second-harmonic generation results in the production of light at twice the frequency or half the wavelength as that of the laser. Optical parametric oscillation results in the generation of two output beams with frequencies that sum to the frequency of the pump beam; usually the direct laser output or a harmonic of the laser output is used for pumping an OPO. Extending the frequency of lasers in this way has important applications in spectroscopy, materials processing, chemistry, and the establishment of optical frequency standards.

We originally planned to use congruent $LiNbO_3$ (lithium niobate) to fabricate monolithic resonators for second harmonic generation. This composition of $LiNbO_3$ has noncritical phase matching for 1319- to 659-nm harmonic generation at a temperature of 364°C. For noncritical phase matching, propagation is at an angle of 90° to the crystal optic axis. With propagation in this direction, phase matching is not sensitive to small angular changes in the direction of propagation, and there is no birefringent walkoff of the pump and harmonic beams. These conditions are important for the relatively low power and for the tightly focused beams that we used. The high operating temperature has advantages of increased optical damage threshold in the lithium niobate material, but the high temperature also made the planned monolithic second harmonic generators difficult to fabricate. The required multilayer dielectric coatings on the surface of the monolithic resonator could not withstand the thermal cycle due to differences of thermal expansion.

After the problem of coating failure was encountered, we began to investigate alternate approaches, and we have made substantial progress with the technique of periodic poling of lithium niobate. More common birefringent phase matching is achieved by balancing dispersion with birefringence so that the fundamental and harmonic waves remain phase matched while propagating through the nonlinear material. Without phase matching, the harmonic and fundamental waves dephase by π radians after propagation through one coherence length, and the flow of energy reverses with the harmonic depleted and the fundamental increased. The coherence length for harmonic and fundamental waves of extraordinary polarization propagating along the x axis in $LiNbO_3$ is 6.5 μm for 1319- to 659-nm conversion and 3.4 μm for 1064- to 532-nm conversion. If the poling of the ferroelectric domain is reversed after each coherence length, the sign of the nonlinear optical coefficient is also reversed, and the nonlinear optical conversion will add to that of the preceding domains. Having both harmonic and fundamental extraordinary polarization allows access to the larger d_{33} tensor component of the nonlinear coefficient of $LiNbO_3$.

Periodically-poled lithium niobate can be produced by several techniques. The technique used in this investigation involved modulation of heating during a miniature pedestal growth method. This produced a periodic segregation in MgO dopant in the crystal which in turn produced a periodic reversal of the ferroelectric domains of the crystal. Typical samples were 800 μm in diameter and 1 or 2 mm long. Using these samples we were able to generate 1.7 watts of cw second-harmonic radiation at 532 nm. Harmonic generation of 659-nm radiation was also demonstrated, but only at microwatt power levels.

These demonstrations show that periodically poled lithium niobate is capable of producing tens to hundreds of milliwatts of 659-nm radiation. We were not able to reach this goal during this investigation, but work is continuing in related projects. We believe that planar waveguides of periodically poled lithium niobate will perform well in this application, and a demonstration is expected in the next couple of months. We are also continuing to investigate thin slabs of lithium niobate periodically poled by the application of electric field. Bulk material prepared by electric field poling is expected to be superior to the material grown by the pedestal growth technique. Waveguide devices will be useful at lower power levels, but bulk material will retain advantages for high power and highly coherent second-harmonic generation. The results of our investigation are described more fully in the next section.

II. Research Results

It is necessary to use some technique beyond focusing to confine the beam to achieve efficient optical harmonic generation at power levels less than kilowatts. This requirement is a result of the size of the optical nonlinearity and limitations that arise from diffraction and divergence in a single pass through a nonlinear optical material. There is continuing effort to increase the size of the optical nonlinearity; increased nonlinearity is one of the advantages of periodically poled nonlinear material. The increases in nonlinearity, however, have not eliminated the need for additional beam confinement. Harmonic conversion in an optical resonator is a demonstrated method for achieving efficient conversion at lower power levels. The laser resonator can be used for this purpose, but this results in troublesome interference effects between the laser oscillation and the nonlinear optical conversion process. Harmonic conversion in a resonant cavity external to the laser resonator can eliminate feedback effects on the laser oscillation at the expense of additional complication, and has advantages for higher coherence applications. The investigations performed in this program used external resonant cavities. Another technique for providing the required beam confinement is harmonic conversion in an optical waveguide. Beam coupling, uniformity, and losses become important issues in waveguide devices.

In an externally resonant second harmonic generator, the fundamental wave is resonant which increases its intensity inside the cavity resulting in higher harmonic conversion efficiency on a single pass through the nonlinear crystal. Since the fundamental radiation effectively has many passes through the cavity, a modest single pass conversion efficiency can yield high overall conversion to the harmonic. Monolithic devices with dielectric mirrors deposited directly on the crystal surfaces offer advantages of greater mechanical stability and lower loss compared to discrete component harmonic generators. Low loss is important in resonant cavity devices; losses of only a couple of tenths of a percent measurably lower the harmonic conversion efficiency. The mechanical stability of the monolithic device is also very useful because the resonance of the harmonic generator must be matched to the fundamental laser frequency, and locking the frequencies is more easily achieved with greater cavity stability.

The high phase matching temperature of 364°C for 1320- to 659-nm second-harmonic generation made it difficult to obtain dielectric coatings. We obtained dielectric coating for this application from two vendors. The coatings consisted of typically 18 TiO_2/SiO_2 layers deposited on $LiNbO_3$ substrates with 3-mm square surfaces that were convex with 1-cm radius of curvature. The coatings had greater than 99.5% reflectivity at 1320 nm. Coatings from both vendors failed during thermal cycling tests. Two coatings from one vendor failed at 200°C. One coating from the other vendor failed at 250°C. A second coating, however, survived a cycle to 280°C without damage. The nature of the coating failure was crazing parallel to the $LiNbO_3$.z axis on the y-z surface. The small cracks seemed to propagate from the larger irregularities in the chamfered edge of the polished surface. There are indications that with further effort, coatings could be developed to withstand the thermal cycle from room temperature to 364°C. We did not pursue coating development, however, because of the cost and time considerations.

The problems with obtaining the coatings necessary for the proposed monolithic cavity design lead us to consider other cavity configurations and nonlinear optical material development. We began work on a number of discrete component resonant cavities for harmonic generation. In a discrete component cavity, the highly reflecting mirrors are on substrates separate from the nonlinear crystal, and the nonlinear crystal must be anti-reflection coated. The antireflection coatings for lithium niobate are a single SiO_2 layer that withstands the thermal cycle to 364°C with no problem. Discrete component cavities have disadvantages of higher losses and reduced mechanical stability. Entering and exiting an oven with a crystal at 364°C with the other discrete components of the cavity at room temperature poses some additional problems. We considered standing-wave and ring-geometry cavities for temperature tuned noncritically phase matched congruent $LiNbO_3$. Work on these cavities has not been completed because of the emphasis placed on periodically poled lithium niobate.

One type of monolithic resonant cavity constructed from congruent $LiNbO_3$ that might be of use for 1319- to 659-nm second-harmonic generation employs total internal reflection instead of dielectric mirrors. Coupling into and out of these cavities is done with frustrated total internal reflection accomplished by bringing a coupling prism near to the surface, and by normal refraction of the non-resonated beam, which can be the result of orthogonal polarization and birefringence of the nonlinear crystal. The total internal reflection cavities have very low loss; we have measured finesse as high as 3000. With losses this low it is possible to use temperature-tuned critically phase-matched crystals. For this particular application in lithium niobate, departing from propagation perpendicular to the crystal optical axis results in phase matching at lower temperatures; for example, the phase-matching temperature for 1319- to 659-nm second-harmonic generation with propagation at 60° in congruent lithium niobate is 101°C . We are investigating total internal reflection cavities for 2.04- to 1.02-μm second-harmonic generation and 1064-nm-pumped optical parametric oscillation in a separate program.

We have investigated two modifications of lithium niobate for nonlinear frequency conversion. In addition to the periodically poled material, we have also investigated variation of the Li-Nb composition. Congruent $LiNbO_3$ has a lithium to niobium ratio of 48.6 : 51.4. When materials are combined in the congruent ratio, the composition of the crystal being grown and the melt from which the crystal is grown are the same and remain constant. The composition of the crystal can be changed by high-temperature solid-state diffusion after crystal growth. Lithium enriched $LiNbO_3$ prepared by this method, called vapor transport equilibration, was used for the generation of 1.6 watts of 532-nm radiation in a discrete-component external-resonant-cavity second-harmonic generator. The material used for 532-nm generation is near the stoichiometric composition whereas a lithium poor composition, 47% to 53% Li:Nb ratio, is required for 659-nm generation. The 47%-Li material will noncritically phase match at 150°C for the generation of 659-nm radiation. A number of difficulties are encountered in processing the material as it becomes further removed from the stoichiometric composition, and the adaptation of periodically poled lithium niobate was chosen as a better approach.

Periodically poled lithium niobate was grown by the laser heated miniature pedestal growth technique. Growth rates for the 800-μm diameter rods were approximately 2 mm/min. The carbon dioxide laser used for laser heating was shuttered off for approximately 40 ms of the 400-ms growth period. The lithium niobate from which the small diameter rod was grown was doped with 5% MgO, which is not a congruent composition. The shuttering of the heating laser beam results in a cycle of fast growth, melt back and slow growth that produce periodic MgO concentration gradients. The dopant gradients in turn produce the periodic poling as the crystal is cooled through the Curie temperature. Due to convection in the molten zone during crystal growth, the liquid-crystalline interface is curved. The surfaces of constant dopant concentration and the domain boundaries have the same curvature as the freezing interface.

When both the fundamental and harmonic waves have extraordinary polarization, the nonlinear optical interaction is described by the d_{33} component of the nonlinear tensor. The magnitude of the $LiNbO_3$ d_{33} component is six times larger than that of the d_{31} component. The d_{33} component can be accessed by quasi phase matching whereas the d_{31} component is accessed in noncritical birefringent phase matching. The effective nonlinear coefficient for first-order quasi phase matching in which the domain thickness is one coherence length is given by

$$d_{\mathrm{eff}} = 2\, d_{33} / \pi.$$

Therefore the effective coefficient for quasi phase matching of an extraordinary fundamental and an extraordinary harmonic is about 4 times larger than the effective nonlinear coefficient for noncritically birefringent phase-matched lithium niobate. This results in a significant decrease in the optimum length of nonlinear material for quasi phase matching.

We have grown periodically poled lithium niobate and demonstrated harmonic conversion for 934- to 467-nm, 1064- to 532-nm, and 1319- to 659-nm second harmonic generation. The coherence length for second harmonic generation is given by the relationship

$$l_{\mathrm{c}} = \lambda_{\mathrm{f}} / \{4 \mid n_{\mathrm{h}} - n_{\mathrm{f}} \mid \},$$

where λ_{f} is the free-space wavelength of the fundamental, n_{h} is the refractive index of the nonlinear material at the harmonic wavelength, and n_{f} is the refractive index at the fundamental. The coherence lengths for extraordinary waves in lithium niobate are 2.3 μm, 3.4 μm and 6.4 μm for the generation of 467 nm, 532 nm, and 659 nm respectively. In practice it was necessary to grow a series of samples with domain thickness near the calculated value to bracket the exact domain thickness required for quasi phase matching. Precise quasi phase matching was then achieved by temperature tuning the sample with period thickness closest to the required value. Only 532-nm generation has been demonstrated at watt levels in these materials; the 474-nm and the 659-nm demonstrations have been at microwatt levels. Our high-power cw demonstration of 532-nm radiation was performed in a bow-tie resonant cavity external to the laser. A 1.24-mm-long crystal was used. The surfaces of the crystal were antireflection coated for 1064 nm. The crystal was placed at the small beam waist of the resonator where the fundamental had a 15-μm waist radius. The second-harmonic generator was pumped with the output of a cw 1064-nm Nd:YAG laser. The laser was operated in a single transverse and longitudinal mode by injection locking. The resonance of the external resonant cavity used for second-harmonic generation was locked to the frequency of the single-mode laser output. The green output power reached 1.77 W when 4.25 W of fundamental power was incident on the second harmonic cavity. The overall conversion efficiency of 42% was reduced by 75% mode coupling efficiency of the fundamental radiation to the resonant cavity, 85.6% transmission at 532 nm of the 1064-nm antireflection coating on the crystal, and the residual reflection at 532 nm of the cavity mirror that was highly reflecting at 1064 nm. We observed no degradation of the beam quality due to thermal effects, and there was no evidence of photorefractive damage to the lithium niobate. Circulating fundamental powers up to 65 W were used, corresponding to 10 MW/cm^2 peak cw intensity transmitted through the crystal. This demonstration clearly shows the capability of periodically poled lithium niobate to handle moderate cw powers and high cw intensity. We expect better power handling capability for 1319- to 659-nm second-harmonic generation.

The crystals grown by the miniature pedestal growth technique had a discontinuity down the center of the crystal perpendicular to the optic axis that sometimes evolved into a crack. This discontinuity makes it necessary to propagate at an angle through the curved domains in one half of the crystal. We have performed an analysis that shows this should not significantly reduce conversion efficiency, but it does make alignment more difficult. Observations showed that domain periodicity errors did not limit performance until the crystal length exceeded approximately 680 μm corresponding to 200 domain layers.

We have assembled the components to perform external resonant cavity second-harmonic generation of 659-nm radiation in periodically poled lithium niobate. This demonstration is being delayed, however, to allow the investigation of other techniques for periodically poling. We have started to investigate electric field poling of thin slabs of material as well as continued our investigation of planar waveguides of periodically poled $LiNbO_3$. The efficiency and ease of alignment of quasi-phase-matched interactions can be improved with the flat and parallel domain boundaries. E-beam and electric field poling have been shown to achieve both flat and parallel domain boundaries in lithium niobate.

While E-beam poling has been achieved with samples as thick as 1 mm, yielding material suitable for both bulk and waveguide interactions, the process is slow and may be unsuitable for high volume processing. Electric field poling is quicker and more suitable for volume applications, but it has been demonstrated on samples no thicker than 100 μm, largely restricting it to waveguide interactions. In addition, periodicity variations and difficulties with substrate breakdown and non uniformity of poling have been observed with electric field poling.

Our efforts in electric field poling are being directed toward the uniform poling of samples up to 1-mm thick, with lithography-limited periodicity variations, good uniformity for interaction lengths of 1 cm, and domain grating periods suitable for first-order quasi phase matching for second-harmonic generation of near infrared and visible light. We are investigating domain growth under both pulsed and constant field conditions. Considerations include the influence of surface conditions, the effect of fringe electric fields at the sample surface, and the role of temperature. We have achieved electric field periodic poling near the +z face of 1-mm-thick samples of lithium niobate at room temperature. While depth and periodicity variations appear suitable for waveguide interactions, uniformity is still unsatisfactory. We anticipate that surface preparation is important to achieving satisfactory uniformity. We plan to return to the 1319- to 659-nm second-harmonic generation demonstration for use both as a test bed for remaining periodically poled $LiNbO_3$ grown by the miniature pedestal method and new material obtained by electric field poling.

In a separate program, we have been investigating the development of waveguide nonlinear optical devices constructed from periodically poled $LiNbO_3$. These devices are prepared by titanium diffusion on wafer surfaces. The Ti is deposited by lithographic techniques. The quality of these devices is improving as we develop the fabrication techniques. Recent improvements have been encouraging for the application of these devices to 659-nm generation. A conversion efficiency of 190% per watt per cm^2 was achieved in a 1-cm-long waveguide for 488 nm generation. The amount of blue light generated in this process was only 0.1 mW, but this was a limitation due to available pump power. At low powers before fundamental power depletion becomes significant, harmonic conversion efficiency scales with the pump power and with the square of the interaction length. There is good indication that power can be increased considerably in these waveguides; transmission of 200 mW of fundamental power through a waveguide without damage has been demonstrated in the near infrared. Theoretically the efficiency of the harmonic conversion process scales inversely with the square of the wavelength. The quality of the waveguide, however, improves at longer wavelengths due to relaxed tolerances. We are beginning to fabricate waveguide devices for the 1319- to 659-nm second-harmonic-generation process and will be evaluating them over the next few months It is reasonable to expect conversion efficiencies of 100% per watt per cm^2 on the basis of previous experience. We expect to see 40 mW of 659-nm harmonic generated from 175 mW of 1319-nm power available in the output of our Nd:YAG laser.

We were not successful in demonstrating the monolithic external resonator technique we originally proposed for second harmonic generation of 659-nm radiation. We have, however, identified an alternative technique of waveguide second-harmonic generation that we believe will provide the required performance, and we expect to demonstrate this performance in the next couple of months. We have made progress in periodically poling lithium niobate nonlinear optical material. This investigation is continuing with the use of electric field poling of periodic material. Additional investigations that in part grew out of this program are continuing. These include the development of different types of external resonant cavity second-harmonic generators including discrete component cavities employing both periodically poled lithium niobate and single-domain bulk crystals and monlithic caviries formed by total internal refelction in congruent lithium niobate.

III. Scientific Personnel Supported by this Contract

Principal Investigator

Professor Robert L. Byer		Applied Physics Department

Other Scientific Faculty and Staff

Martin M. Fejer	Assistant Professor	Applied Physics
Robert C. Eckardt	Sr. Research Assoc.	Ginzton Laboratory
Eric K. Gustafson	Research Associate	Ginzton Laboratory

Students and Degrees Awarded

Deiter H. Jundt, Applied Physics, Ph.D., 1991

Gregory Miller, Electrical Engineering, M.S., 1992

Lawrence Myers, Electrical Engineering

Christopher Polhalski, Electrical Engineering

IV. List of all Publications Supported by ARO Contract DAAL03-89-K-0155

1. D. H. Jundt, M. M. Fejer and R. L. Byer, "Periodically poled $LiNbO_3$ for high efficiency room-temperature frequency conversion," post deadline paper, Conference on Lasers and Electro-optics, Baltimore, MD (13-17 May, 1991).

2. D. H. Jundt, G. A. Magel, M. M. Fejer and R. L. Byer, "Periodically-poled $LiNbO_3$ for high efficiency room temperature second harmonic generation," Appl. Phys. Lett. **59**, pp. 2657-2659 (1991).

3. D. H. Jundt, M. M. Fejer, R. L. Byer, R. G. Worwood and P. F. Bordui, "69% efficient continuous-wave second-harmonic generation in lithium-rich lithium niobate," Opt. Lett. **16**, pp. 1856-1858 (1991).

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