Nonlinear Optics and Solid-State Lasers: 2000

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Invited Paper

***Abstract*—Progress in solid-state laser sources and the nonlinear frequency conversion of lasers has been impressive over the first forty years of their development. This paper reviews the progress with an emphasis on the interactions of the scientists and engineers involved in the work and the motivation for the research. This account [1], is highly personal and necessarily incomplete. The references cited point to the key results and to reviews of progress in nonlinear optics and solid-state laser sources and should assist those seeking to learn about the field as it developed.**

***Index Terms*—Lasers, nonlinear optics, optical parametric oscillators, remote sensing, reviews, solid state lasers.**

I. HISTORICAL PERSPECTIVE

THE FIELD of nonlinear optics began nearly simultaneously in the United States and in Russia in 1962. The first decade of discovery led to an understanding of the requirements and methods to achieve phasematching. In the second half of the 1970s, tunable parametric oscillators driven by high-peak-power Q-switched lasers made the transition to tools useful for spectroscopy and remote sensing. In the late 1980s, engineered nonlinear materials were introduced with the successful implementation of quasi-phasematching by periodic inversion of ferroelectric domains in lithium niobate. Lithographic processing techniques enabled the fabrication of quasi-phasematched nonlinear "chips" using electric field poling of lithium niobate at the wafer scale.

Parallel to the developments in nonlinear optics was the discovery and development of solid-state laser sources. The first solid-state laser was the flashlamp-pumped chromium-doped sapphire crystal now referred to as the Ruby laser. Following the demonstration of the Ruby laser by Maiman [2], the $\mathrm{Nd:CaWO_4}$ laser and then the Nd : YAG laser were developed [3]. Flashlamp-pumped solid-state lasers were the preferred source for nonlinear optical experiments and applications for the next two decades. However, as early as 1962, laser diode (LD) pumped solid-state lasers were proposed [4] and demonstrated [5]. The first LD-pumped solid-state laser operated at cryogenic temperatures, but the promise of the approach was recognized. It was not until 1978, with the demonstration of a 1-W continuous wave (CW) LD bar by Scifres *et al.* [6] that LD-pumped solid-state laser began their rapid evolution that continues today.

I am asked often by students why scientific progress in the past took many paths, some of which were dead ends. The students are interested in the motivation for past work, the investigators, and the challenges that confronted the research. In this brief overview of the intertwined fields of nonlinear optics and solid-state lasers, I will describe the motivations that led to progress. At times, I will trace the path to success and show that it was not always evident and that "the road to good physics is neither paved nor well marked [7]."[1]

I entered the field of quantum electronics with the assistance of a mentor and teacher, Prof. Sumner P. Davis then and still at the University of California at Berkeley. As a youth, I was an amateur astronomer and had built a number of telescopes for astro-photography. Upon arriving on campus as a freshman, I decided to seek an opportunity for research in the physics laboratories, preferably in optics. Prof. Davis provided that opportunity, but only after I had studied and passed an oral examination about gratings, interferometers, and optical spectroscopy. Following graduation four years later, Prof. Davis pointed me to a small company located in Mountain View, CA, and suggested that I might consider taking a position there. I arrived for my interview at a small building located on Terra Bella Street. Since no one was at the door to meet me, I wandered in. I was greeted by a very excited group of individuals. I was told that the green light visible from the discharge device on the table was a mercury ion laser. The inventor of the ion laser, Dr. Earl Bell, became my supervisor and mentor for the following year. During that year at Spectra Physics, we worked on the argon, krypton, and xenon ion lasers.[2] I learned how to be an inventor from Dr. Bell, and the value of clearly presented theoretical explanations from Dr. Arnold Bloom. Spectra Physics grew from a start-up company to the largest laser company in the world under the leadership of Herbert M. Dwight, Jr.

My employment at Spectra Physics came to an end indirectly because of Wednesday noon brown-bag lunches organized by Prof. A. E. Siegman at nearby Stanford University. I was given permission to attend these discussions held in the Microwave Laboratory. I learned about the studies in nonlinear optics, and more importantly, about the possibility of graduate study at Stanford. The following year, I began my studies under the direction of Prof. Stephen E. Harris in the area of nonlinear optics.

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[1] This is a quote from Dr. Lev Kulevski, General Physics Laboratory, Moscow, Russia, made during his visit to Stanford University in 1976.

[2] This work led to a trip to Bell Labs to meet the laser group led by Gene Gordon. There I met Ed Labuda and was shown his progress on argon ion lasers. Later I met William Bridges, inventor of the argon ion laser, then at Hughes Research Laboratory.

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The early progress in nonlinear optics has been the subject of numerous monographs, review papers, and texts. It will suffice for this introduction to direct the reader to the monograph by Bloembergen [8] and to the books by Akhmanov and Khokhlov [9], and Shen [10]. The first nonlinear optics experiment was conducted by Franken *et al.* with the demonstration of second harmonic generation in crystal quartz in 1961[3] [11], using a normal-mode Ruby laser. The conversion efficiency was less than one part per billion because of the lack of phasematching. The UV output was recorded photographically and was supposed to be published as a small white dot in the key figure of the paper. The editor of the journal had the dot removed prior to publication because it was thought to be a smudge.

Efficient nonlinear interactions require, in addition to a nonlinear response in the medium, a means of achieving phase-velocity matching of the interacting waves over an interaction distance of many optical wavelengths. Two methods of achieving phasematching and, thus higher conversion efficiency, are the use of crystal birefringence to offset dispersion [12]–[14] and the use of a periodic modulation of the sign of the nonlinear coefficient to reset the optical phase. The latter approach, suggested by Armstrong *et al.* [15, Figure 10] of their seminal paper, is now referred to as quasi-phasematching (QPM).

The idea of parametric amplification and generation of tunable light was proposed and analyzed by Armstrong *et al.* [15], Kingston [16], Kroll [17], and by Akhmanov and Khokhlov [18] in 1962. Three years later in 1965, the first experimental demonstration parametric gain by three-wave mixing was achieved by Wang and Rachetti [19]. (We shall return later to applications of a three-wave parametric mixer to telecommunications.) In the same year, parametric oscillation was achieved by Giordmaine and Miller [20]. They used a Q-switched $\mathrm{Nd:CaWO_4}$ laser, frequency-doubled to the green in $\mathrm{LiNbO_3}$, to pump a monolithic $\mathrm{LiNbO_3}$ tunable parametric oscillator. Shortly thereafter, the parametric oscillator was demonstrated in Russia at Moscow State University by Akhmanov *et al.* [21]. The Russian parametric oscillator, based on the nonlinear crystal KDP, had a tuning range that extended from 957.5 to 1177.5 nm. It was pumped by a Q-switched frequency-doubled Nd : Glass laser. Earlier work on parametric oscillators was reviewed in an article published in 1968 [22].

In 1966, Boyd and Askin [23] suggested that CW parametric oscillation might be possible in the crystal $\mathrm{LiNbO_3}$ with a threshold as low as 10 mW. This paper led the Harris group at Stanford to pursue CW parametric oscillation based on the recently discovered crystal $\mathrm{LiNbO_3}$. The research program led to the growth of $\mathrm{LiNbO_3}$ by Prof. Feigelson and to significant improvements in $\mathrm{LiNbO_3}$ as a nonlinear crystal. Two years later in 1968, Smith *et al.*achieved CW oscillation in the near infrared in $\mathrm{Ba_2NaNb_5O_{15}}$ [24]. Almost simultaneously, the first CW visible parametric oscillator was demonstrated by Byer *et al.* [25]. The CW $\mathrm{LiNbO_3}$ optical parametric oscillator (OPO) was pumped by the 514-nm output from an argon ion laser. The OPO threshold at greater than 400 mW was just below the maximum power generated by the experimental argon ion laser source built at Stanford that was based on technology from Spectra Physics. The experiment was reported in a memorable post-deadline paper session at the International Quantum Electronics Conference held in Miami, FL, in 1968.

In a joint experiment with Dr. A. (Shasa) Kovrigin, a visiting scientist from Moscow State University, the work was extended to demonstrate an efficient CW ring-cavity parametric oscillator [26]. For the first time, pump depletion greater than 60% was observed in a CW–OPO. Dr. Kovrigin's visit to Stanford University followed in the footsteps of his advisor and the founding director of the Moscow State University research program in nonlinear optics, Dr. Khokhlov. Prof. Khokhlov had visited Stanford ten years earlier in 1959 as a guest of Prof. H. Heffner, later chair of the applied physics department. During his stay at Stanford, Prof. Khokhlov was introduced to professor of history, Richard W. Lyman. Slightly more than one decade following their meeting at Stanford, Prof. Khokhlov and his friend Prof. Lyman would become the Rector and the President of their respective universities.

Parametric amplifiers, like all linear amplifiers, have inherent quantum noise [27]. It was realized by Harris that the parametric noise was intense enough to be visible as fluorescence [28]. Quantum noise from a parametric amplifier was studied nearly simultaneously by Tang [29], by Kleinman [30], and by Klyshko [31]. Further, this visible fluorescence could be tuned by altering the phasematching conditions of the parametric interaction. A set of striking color photographs of visible parametric fluorescence was published by Giordmaine in 1969 [32]. Fig. 1 shows the parametric fluorescence emitted by a crystal of $\mathrm{LiNbO_3}$ pumped by an argon ion laser. The center wavelength is tuned by changing the crystal birefringence with temperature. Today, photon "fission," as it was called by Klyshko [31], is the basis for the generation of correlated paired photons useful for the production of squeezed states of light and for the study of quantum optics.

The research at Stanford University led to the first commercial tunable laser source based on a $\mathrm{LiNbO_3}$ parametric oscillator. The parametric oscillator, introduced by Chromatix in 1971, tuned from 550 nm to greater than 4.8 μm when pumped by the 659, 561, 532, and 473.5 nm outputs of an internal frequency-doubled Q-switched Nd : YAG source. More than 50 of these tunable parametric oscillators were sold worldwide. Fig. 2 shows the tuning range of the $\mathrm{LiNbO_3}$ parametric oscillator. A few these devices are still in operation today, more than 25 years after their introduction.

The progress in nonlinear optical devices was reviewed by Harris [33] in 1969 as the decade drew to a close. Harris also reviewed the status of nonlinear materials. Only four nonlinear crystals had been used successfully to achieve parametric oscillation by 1969. Efforts were initiated in Europe and the U.S. to discover, grow, and improve nonlinear materials. The status of parametric and nonlinear optics was reviewed by Smith [34] a few years later.

Early progress in nonlinear optics was held back by the lack of useful nonlinear crystals. Research to discover new nonlinear materials progressed slowly, limited by the difficulty in finding crystals with adequate birefringence to achieve phasematching. In a study over the summer of 1969, I discovered that of 22 000 crystals surveyed, fewer than 100 had adequate birefringence

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[3] Many years later, Prof. Franken explained to a young assistant professor why my just-graduated Ph.D. students earned more than their professor and that it would be 20 years before a professor's salary would catch up.

for phasematching. Of these, only a handful could be grown and prepared for characterization of their nonlinear optical properties. Nevertheless, work in Europe and the U.S. led to the discovery of new classes of nonlinear materials, including $LiIO_3$, the semiconductors CdSe and proustite Ag_3AsS_3, and the chalcopyrites $AgGaS_2$, $AgGaSe_2$, $ZnGeP_2$, and $CdGeAs_2$. Progress in nonlinear materials and devices was reviewed by Byer in 1973 [35].

The difficulty in discovering and developing new nonlinear crystals has led researchers to revisit the idea first suggested by Bloembergen [8], [15] that phasematching might be achieved by periodically altering the sign of the nonlinear coefficient or quasi-phasematching. In 1968, Bloembergen applied for and received a patent on the idea of quasi-phasematching[4] [36]. Like many fundamental patents, Bloembergen's patent ran its 17-year course before the idea could be implemented. The next two decades would achieve nonlinear interactions using birefringent phasematching. The lack of adequate gain in the birefringent phase-matched nonlinear interactions would mean that high-peak power pump laser sources would be required for efficient nonlinear frequency conversion.

II. HIGH PEAK POWER LASERS AND PARAMETRIC OSCILLATORS

Progress in the second decade of nonlinear optics was in the area of high-peak power nonlinear interactions based on birefringent phasematching. Although efforts were applied to finding a practical approach to achieve quasi-phasematching, it would be two decades before significant progress was made. Thus, research was directed toward the combination of improved nonlinear optical materials coupled with improved laser sources. Research took place in many laboratories around the world. I summarize briefly the progress in tunable parametric oscillators at Stanford University and demonstrate their capabilities by describing the applications of tunable sources with remote sensing and coherent anti-Stokes Raman spectroscopy (CARS).

A. CARS

The availability of tunable laser radiation immediately opened the door to tunable laser spectroscopy. The advantages were the high brightness of the laser source compared with traditional discharge lamp sources, and the potential for very high-resolution spectroscopy based on narrow linewidth laser sources compared with the resolution of grating and interferometer based spectrometers. A further advantage of the tunable laser source was the potential for high-peak power levels. This opened the possibility of nonlinear spectroscopy. At Stanford University, in collaboration with visiting scientist Albert B. Harvey and chemistry professor Bruce Hudson, we initiated a program in nonlinear Raman spectroscopy. Our goals were to show that nonlinear Raman spectroscopy could find uses in chemistry and that we could achieve the promised very high spectral resolution.

Nonlinear Raman spectroscopy had been demonstrated earlier by Terhune [37] and had been studied by Bloembergen [38], [39]. However, progress had been slow because of the lack of tunable laser sources. By 1973, many groups had initiated research in nonlinear Raman spectroscopy including Taran in France [40], and Tolles [41], Nibler [42], and Eckbreth [43]–[45] in the United States. In our laboratory, we had available a chromatix frequency-doubled Nd : YAG laser, which pumped a tunable-dye laser source. We set out to detect and to study the anti-Stokes Raman spectrum generated by a four-wave-mixing Raman process in liquid benzene. After hours of futile searching for the blue anti-Stokes output, I acquired the anti-Stokes signal by looking directly into the beam. After minor adjustments, the coherent diffraction-limited anti-Stokes beam was observed and projected across the room. Clearly, nonlinear Raman spectroscopy had significant signal-to-noise advantages compared to spontaneous Raman spectroscopy.

Our next task was to prepare the paper for publication. A discussion took place in the Ginzton Laboratory courtyard at morning coffee break regarding the title of the paper and the name for the nonlinear Raman spectroscopy. After some consideration we titled the paper "Coherent anti-Stokes Raman Spectroscopy" [46], [47]. That name was adopted along with many other forms of nonlinear Raman spectroscopy, including Raman-induced Kerr effect (RIKES), stimulated Raman gain spectroscopy (SRG), and box-CARS. In the words of Marc Levenson, "*At midnight on January 1st of every year, the coherent Raman physicists have conspired to permute the acronyms for these techniques … just to keep the chemist confused*."

The field has been extended by the efforts of many scientists and is now firmly established. The introduction to nonlinear spectroscopy is provided by Levenson [48], a summary of all of the early publications and applications is given by Eesley [49], and the first excellent overview of chemical applications of nonlinear Raman spectroscopy is provided in a book edited by Harvey that appeared in 1981 [50].

B. Remote Sensing of the Atmosphere

The availability of the tunable optical parametric oscillator with a tuning range that extended into the near infrared molecular fingerprint region of the spectrum opened the possibility for lasers to become the critical tool for remote sensing applications. We set about to study the laser transmitter requirements and the possible approaches to laser remote sensing in 1969. My first student agreed to this direction for his thesis work, even though it was very challenging technically and, at that time, unexplored territory. In 1971, Kildal and Byer [51] published a review paper on laser methods for remote sensing in the *Proceedings of the IEEE*. This review paper generated widespread interest as indicated by the hundreds of postcards received requesting copies of the paper. This was before the days of copy machines and well before the days of word processing on a computer.

The requirements for wavelength tuning with linewidth control, especially in the near infrared molecular fingerprint spectral region, led the Stanford group to develop an infrared (IR)

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[4] Prof. Bloomberg informed me that he had followed the advice of a friend and had retained a patent attorney early in his career as an assistant professor at Harvard University.

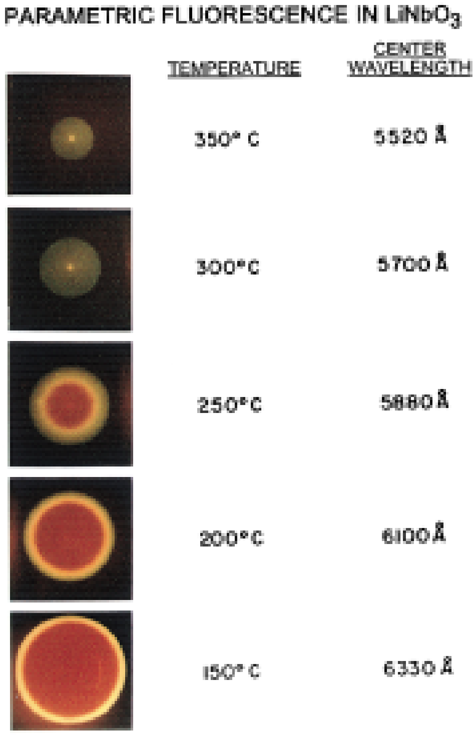


Fig. 1. Visible parametric fluorescence in $LiNbO_3$ pumped by and argon ion laser. Center wavelength is tuned with crystal temperature. Wavelength varies with radius according to the phasematching conditions.

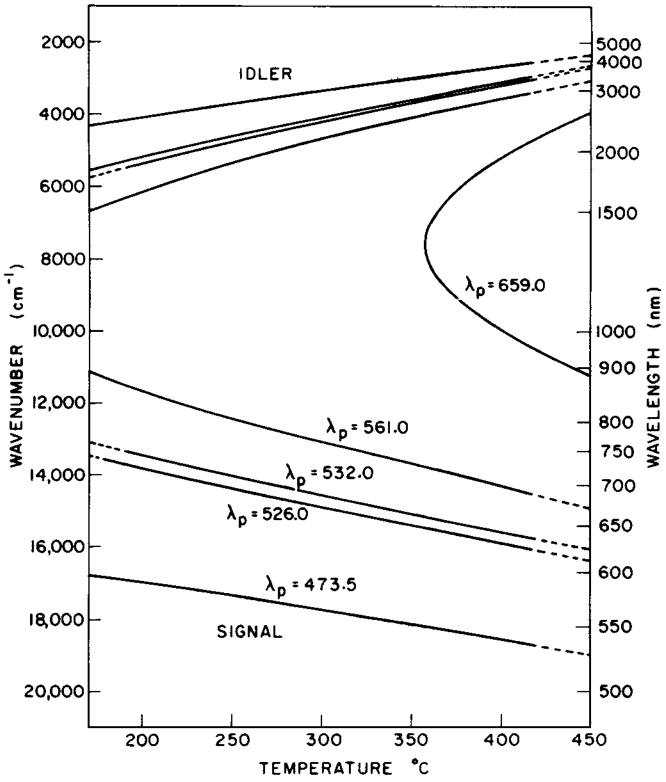
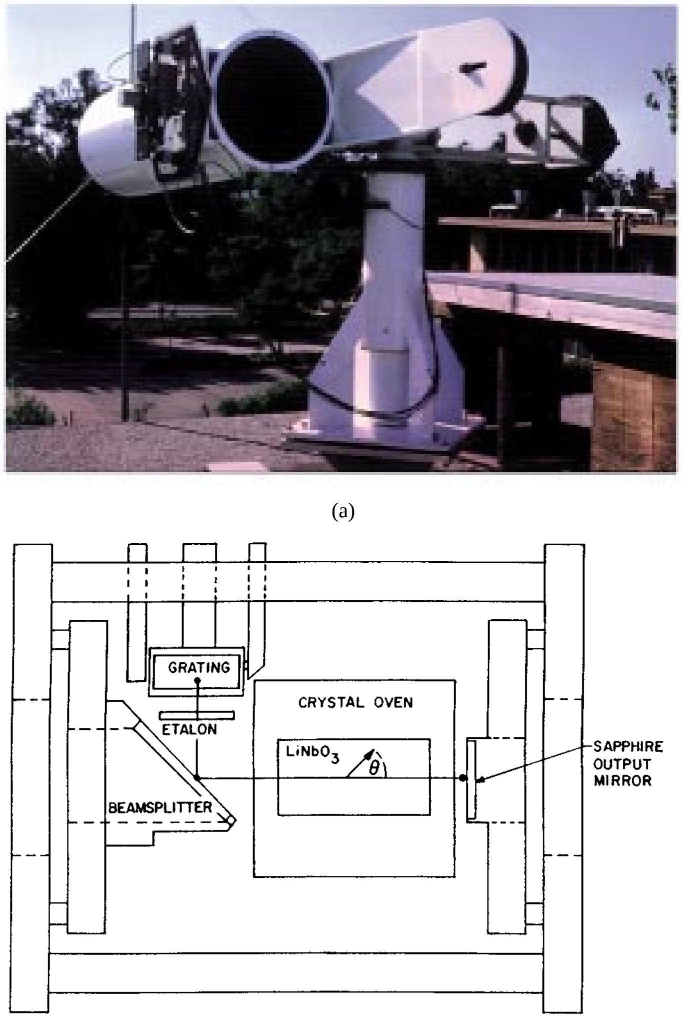


Fig. 2. $LiNbO_3$ parametric oscillator wavelength tuning range versus temperature. Frequency-doubled Q-switched Nd : YAG laser-pump wavelengths are indicated.

tunable parametric oscillator. The first tunable IR–OPO was based in CdSe and pumped by Nd : YAG operating at an unusual 1.83-μm wavelength [52]. A review of parametric oscillators and nonlinear materials by the mid 1970s was published in 1973 [53]. The review covered recently discovered IR nonlinear materials, such as the chalcopyrites, and the devices based on these nonlinear materials. The second tunable IR–OPO was an angle phase-matched $LiNbO_3$ OPO pumped by a Q-switched Nd : YAG laser. Prof. Feigelson provided the [01.4] growth direction lithium niobate crystals[5] [54], [55].



(a)

(b)

Fig. 3. (a) Photograph of the 16-in diameter transmitter/reciever telescope on the Stanford campus. (b) Schematic of the parametric oscillator cavity showing the grating and etalon linewidth control elements. The 5-cm-long $LiNbO_3$ crystal is mounted in the crystal oven and the beam exits to the right. Computer-controlled tuning range extends from 1.4 to 4.4 μm.

The Q-switched Nd : YAG pumped tunable 1.4 to 4.4 μm $LiNbO_3$ OPO was reported in the *Proceedings of the 2nd International Conference on Laser Spectroscopy* held in Megeve, France, in 1975 [56]. This meeting has been held every other year since the first meeting held in 1973 in Vail, CO. The series of meetings has led to the published proceedings that captured the excitement and advances in laser spectroscopy as it unfolded over more than a quarter of a century ago.

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[5] The first [01.4] $LiNbO_3$ crystal was grown by accident. I gave the unusually shaped crystal to my wife as a pendant, but later realized its importance as a seed crystal and had to ask for it back.

The $LiNbO_3$ OPO had an extended 1.4 to 4.2 μm IR tuning range required for remote sensing, but was well short of the required energy. We set about to develop a high-peak power Nd : YAG laser master-oscillator power-amplifier pump source. The flash-lamp pumped laser system consisted of a TEM_{00} mode oscillator followed by a Faraday isolator, two 1/4 in diameter Nd : YAG rod amplifiers followed by a second Faraday isolator, and a final 3/8 in diameter Nd : YAG rod amplifier. The output energy at 5 Hz repetition rate was 350 mJ in a 10-ns Q-switched amplified pulse. The system worked as designed, but was complex and costly to operate. Clearly, a simpler, more direct approach to high pulse energy output was needed.

Progress is made often by asking the right question. The unstable resonator concept had been invented and studied by Siegman,[6] but had not been successfully applied to a high average power Nd : YAG laser source. Asking why this was the case led to the realization that low magnification unstable resonators that had been used previously would become *stable* resonators with the onset of thermal focusing in the Nd : YAG rod. On a flight from Pittsburgh to San Francisco, I calculated the high-magnification cavity parameters. Over the weekend, Richard Herbst, then at Stanford University, designed a high-magnification unstable resonator. By the following Friday, graduate student Hiroshe Komine fabricated the 1.8-mm diameter 50-cm radius of curvature output coupler by grinding a high-reflector mirror and mounting it onto a fused silica Brewster plate. Within a short time, we had a 200 mJ diffraction-limited Nd : YAG unstable resonator oscillator with a characteristic hole in the center of the plane wave beam [58]. With the addition of a single stage of amplification, the output energy reached 700 mJ at 10-Hz repetition rate. We were on our way toward meeting the remote sensing transmitter requirements using this high-peak power near-diffraction-limited laser to pump a parametric oscillator [59] followed by a parametric amplifier [60].

The combined breakthroughs of the Nd : YAG unstable resonator oscillator/amplifier and the growth of $LiNbO_3$ crystals along the [01.4] crystalline axis, which was parallel to the phasematching angle, led to 1.5-cm diameter by 5-cm long nonlinear crystals [54], [55] and to the transmitter for remote sensing. With the addition of an intracavity grating in combination with a tilted etalon, the singly resonant parametric oscillator operated in a narrow bandwidth and even in a single axial mode required for remote detection of molecular species. The addition of the PDP-11 minicomputer (with 8 kB of core memory!) allowed computer controlled tuning over the entire 1.4- to 4.4-μm spectral range. Fig. 3(a) shows a photograph of the 16-in telescope transmitter/receiver on the roof of the Ginzton Lab. Fig. 3(b) shows a schematic of the $LiNbO_3$ parametric oscillator resonator with computer-controlled crystal orientation and grating, and etalon tilt for controlled linewidth and tuning.

The Nd : YAG laser-pumped computer-tuned parametric oscillator was applied to remote sensing of atmosphere, including the detection of SO_2 [61] and CH_4 [62]. Improvements in the tunable source led to simultaneous measurements of atmospheric temperature and humidity using on–off tuning between two levels in water vapor with split ground-state levels [63], [64]. Lidar measurement of atmospheric temperature and humidity over a four-hour period demonstrated the reliability of this first computer-tuned parametric oscillator. Progress in optical parametric oscillators and nonlinear materials and the characteristics of the Nd : YAG unstable-resonator-pumped computer-controlled $LiNbO_3$ parametric oscillator were reviewed by Byer in 1975 [65].

The research led to the commercialization of the unstable-resonator Nd : YAG laser by Quanta-Ray in 1976.[7] Although the intent was to introduce a tunable IR OPO as a product, a market survey dictated that the high-peak power harmonically converted Nd : YAG unstable-resonator Quanta-Ray laser should be used to pump a dye laser tunable source.

In 1985, Chuangtian Chen introduced the new nonlinear crystal beta barium borate (BBO). In 1986, Chen visited Stanford, where the crystals were evaluated by second-harmonic generation [66], [67]. The visible BBO–OPO was demonstrated at Stanford and elsewhere in 1988 [68], [69]. Research at Cornell University by Prof. Chung Tang *et al.*led to early advances in BBO parametric devices and to improvements in the material properties. Tang *et al.*reviewed progress in parametric oscillators in 1992 [70]. Improvement in the growth and the optical quality of BBO led to the reintroduction of a commercial tunable parametric oscillator product by Spectra Physics in 1993. Continued progress in the frequency control of the BBO–OPO by injection seeding [71]–[74] led to the application of these all-solid-state tunable sources to spectroscopy, including CARS. Today, BBO parametric oscillators, with their broad 0.41- to 2.5-μm tuning range, have replaced high-peak power dye lasers as the preferred tunable coherent source. More than 5000 Quanta-Ray lasers have been sold, with many applied to the well-established fields of laser remote sensing and CARS spectroscopy.

The availability of the $LiNbO_3$ OPO led to a research program to study the detailed spectra of liquid water by four-wave mixing Raman spectroscopy. Again, the research at Stanford was conducted jointly with a visiting scientist from Moscow, Prof. Nikolai Koroteev. Using polarization CARS, the details of the spectra of liquid water were investigated [75]. Prof. Koroteev had served as the Vice Rector of Moscow State University and the Director of the International Laser Center at Moscow State University until his untimely death last year.

Continued collaborations with scientists from Russia were facilitated by the early Vavilov conferences on nonlinear optics. The conference, held at the Akademgorodok in Novosibirsk, Siberia, Russia, in 1976, was attended by many of the founders in Russia of the field of nonlinear optics, including Rem V. Khokhlov, Sergie A. Akhmanov, A. (Sasha) I. Kovrigin, D. N. Klyshko, and Algis S. Piskarskas. As was tradition, all participated in the Asian-Russian versus European-Russian football (soccer) match, including Rem Khokhlov. The author refereed the game.

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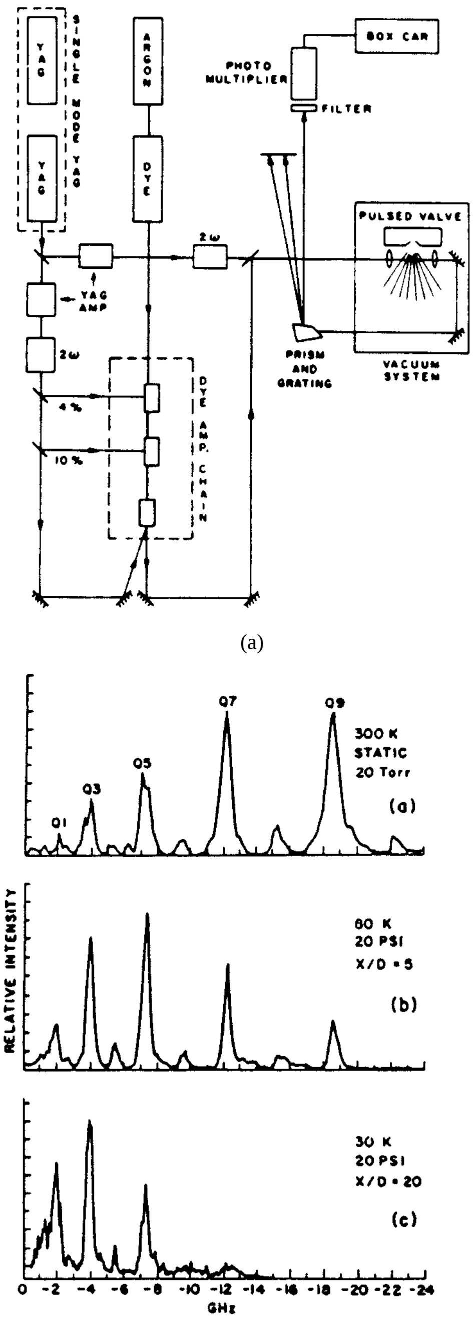
[6] For a description of the unstable resonator and its application to gas and solid-state lasers from the inventor of the unstable resonator concept, see [57].

[7] Quanta-Ray was founded by Earl Bell and Gene Watson in 1975. Bob Mortensen was the CEO. Bell's motivation was to found a successful company to generate funds to pay for the continued operation of the privately owned narrow-gauge railroad at Chalma, NM. The goal was achieved.

High-spectral resolution CARS was the next challenge. We undertook the study of high-resolution Raman spectroscopy of gases cooled by expansion in supersonic gas jets. The spectroscopy demanded that the unstable resonator Nd : YAG Q-switched laser source operate reliably in a single axial mode because the frequency-doubled Nd : YAG output was mixed with the tunable dye laser to achieve the CARS anti-Stokes spectra. The demand for single-mode operation on every pulse from the laser was very difficult to achieve and took more than three years of unsuccessfully exploring various approaches, including electronic-controlled Q-switching, tilted etalon axial mode selection, and output-coupler etalon mode selection. Two theses depended upon finding a solution to this problem such that the pressure was felt both by the students and the professor. Finally, in near desperation, we returned to an idea that had been rejected earlier: injection seeding. We had rejected injection locking, the steady state control of an oscillator frequency by gain saturation as originally described by Adler in 1946 [76]. The calculated injection locking range was less than 1 MHz and our Fourier transform limited laser linewidth for the 10-ns Q-switched pulses was near 100 MHz. However, when we tried frequency injection into the high-gain Q-switched unstable-resonator Nd : YAG oscillator, a single axial mode was selected immediately and on every shot. The single-axial mode operation of Nd : YAG by injection seeding [77], the transient solution of the Adler equations coupled with the development of a pulsed supersonic nozzle [78], led to successful high-resolution Raman spectroscopy of acetylene in a molecular beam [79]. Fig. 4(a) shows a schematic of the injection-seeded, single-axial mode unstable resonator Nd : YAG laser source, and Fig. 4(b) shows the resolved and cooled rotational CARS spectra of acetylene. High-resolution CW CARS was successfully demonstrated in a continuous jet expansion and applied to the spectroscopy of H_2 and D_2 [80] and methane [81]. In an elegant experiment, Gustafson showed that high-pressure gases could be safely studied in 60-μm diameter glass spheres using CW CARS through a microscope [82].



(a)

(b)

Fig. 4. (a) Schematic of the injection-seeded single-axial-mode unstable-resonator Nd : YAG laser source and tunable dye laser. (b) Resolved expansion-cooled rotational CARS spectra of acetylene.

III. Slab Geometry Solid-State Lasers

The decade of the 1980s began with the recognition that future applications of lasers for remote sensing, specific spectroscopic measurements of short lived isotopes, and laser plasma generation of X-ray microscopy and lithography required high-peak and *average*-power laser sources. It was evident that average-power scaling of rod geometry lasers was limited by thermal-induced focusing and ultimately by thermal-induced fracture. Counter to intuition, the average-power scaling of rod lasers is independent of the rod diameter and scales only linearly with rod length. The zigzag slab laser, invented by Chernoch at General Electric Laboratories in New York [83], [84], and illustrated in Fig. 5, showed promise as an approach to average-power scaling solid-state lasers.

We initiated a small research program in 1979 jointly with a visiting scientist from Switzerland, Dr. Sep Unternahrer, to investigate the slab laser. We were motivated by the need for a tunable IR source for the spectroscopy of short-lived muonium. Within a few years, we demonstrated a flash-lamp pumped Nd : Glass slab laser [85] and later Nd : YAG lamp-pumped slab lasers. This work led to the publication of the analytical model of slab lasers [86] and to a useful computer model of the slab performance [87]. We extended the study to a novel moving slab-laser concept [88] and to a study of power scaling-to-megawatt average-power levels [89].[8] The research led to the successful application of an 11-J, 4-Hz repetition-rate Nd : Glass slab laser [90], [91] by Reed, to laser plasma X-ray generation, and to a demonstration of X-ray lithography using

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[8] The title as submitted included the word “megawatt,” but was rejected because it exceeded the classification limit at that time.

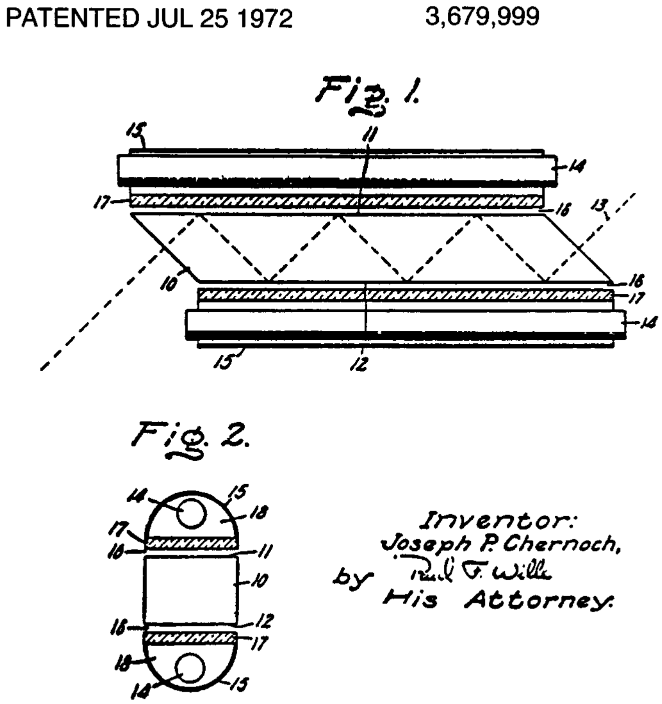

Fig. 5. Schematic diagram of the zigzag slab laser.

a mask loaned to us by IBM. Trail,[9] the other New Zealand "kiwi" in the research group, applied the laser plasma X-ray source to successfully demonstrate a reflective objective X-ray scanning microscope [92].

This early work demonstrated the advantages of the slab geometry with power scaling proportional to the area of the slab. Further, we realized that the slab geometry could enable both high-energy storage and high-average power operation that were required for remote sensing on a global scale. Our research was guided by the question: What is the most important global remote sensing measurement? The answer was provided by R. M. Huffaker two years earlier [93], [94] and published in 1984: remote sensing of the global-wind field.

Global-wind sensing using coherent laser radar to detect the Doppler shift of the returned light scattered from dust in the atmosphere is very challenging. A NASA study called laser atmospheric wind sounder (LAWS) showed that the transmitter must operate at a 10-Hz repetition rate at 10-J per pulse, and at less than 1-MHz linewidth for a 1-m diameter receiver aperture at an 800-km orbit. Further, the laser source must be 10% electrically efficient and operate for the four-year mission duration. In 1980, these specifications were very daunting and met all but the impossible criteria as stated by Edwin Land, "Don't undertake a project unless it is manifestly important and nearly impossible."

The challenge could not be met by flash-lamp pumped solid-state lasers, so we began, in a modest way, to explore laser diode (LD) pumped solid-state lasers.

IV. LD-PUMPED SOLID-STATE LASERS

A. *Coherent Laser Radar*

We set about to evaluate the laser requirements for a more modest ground-based Doppler wind velocity sensing using a Nd : YAG-based coherent laser radar [95]. The study showed that we would need to invent or develop a CW narrow-linewidth Nd : YAG local oscillator, a Nd : YAG amplifier to meet the transmitter power requirements, and a coherent detection system that would allow efficient mixing of the returned beam with the local oscillator for coherent detection of wind velocity.

Our experience showed that lamp-pumped Nd : YAG lasers were limited to linewidths near 1 MHz due to induced vibrations [96]. We asked the question of whether laser diode pumping might open the way to narrow-linewidth local oscillators. Our first experiment used a 10-mW LD borrowed from Toshiba, a borrowed small imaging lens, and a fabricated standing-wave monolithic Nd : YAG oscillator. The laser reached threshold at 2 mW of pump power, operated with a 25% slope efficiency and reached the maximum output power of less than 2 mW. Our research into LD-pumped solid-state lasers had taken a small step, jointly with Prof. Binkun Zhou of Tsinghua University, Beijing, China, to begin the journey [97]. A photograph of this LD-pumped laser, shown in a 1988 review article on LD pumped solid-state lasers [98], has a penny in the background that represents the cost of doing research in the university environment.

We set out to improve on this standing-wave Nd : YAG oscillator and asked: How do we obtain single-axial mode operation and maintain the inherent stability of the monolithic device? That question led Kane to the concept of the monolithic unidirectional ring Nd : YAG oscillator [99], a device that we now refer to as the nonplanar-ring oscillator (NPRO). The very first operation of the NPRO, pumped by an argon-ion laser, showed single-axial mode operation. The first LD-pumped NPRO operated at 3-mW of output power with a 10-kHz linewidth. Over a decade, the power was increased to 40 mW and then to 600 mW. At the same time, the linewidth was reduced from 10 kHz to less than 1 Hz by locking to a reference cavity [100], [101]. Further, Dr. Ady Arie, then a visiting postdoctoral scholar and now a professor at Tel Aviv University, stabilized the frequency-doubled NPRO by Doppler-free spectroscopy to reference lines of the iodine molecule at 532 nm [102]. He demonstrated an absolute stability of better than one part in 10^{-13}. The engineers at Lightwave Electronics have improved the reliability and performance of the NPRO such that more than 4000 oscillators have been working in the field for greater than a four-year duration.

Fig. 6 shows a photograph of the NPRO Nd : YAG crystal next to the 1-cm diameter coin, the American dime. The NPRO is the "quartz crystal" of the optical frequency region. This highly coherent local oscillator has become the key element of almost every laser experiment in our laboratory for the past decade. Nilsson has prepared a beautiful theoretical description of the NPRO based on symmetry arguments [103]. And, in a step of some significance, Day succeeded in phaselocking two Nd : YAG NPRO oscillators using a low bandwidth locking loop [104]. On congratulating Day on his success, I informed

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[9] Trail's thesis defense presentation was attended by the pioneers of X-ray microscopy at Stanford University, Dr. Albert Baez and Prof. (emeritus) Kirkpatrick.

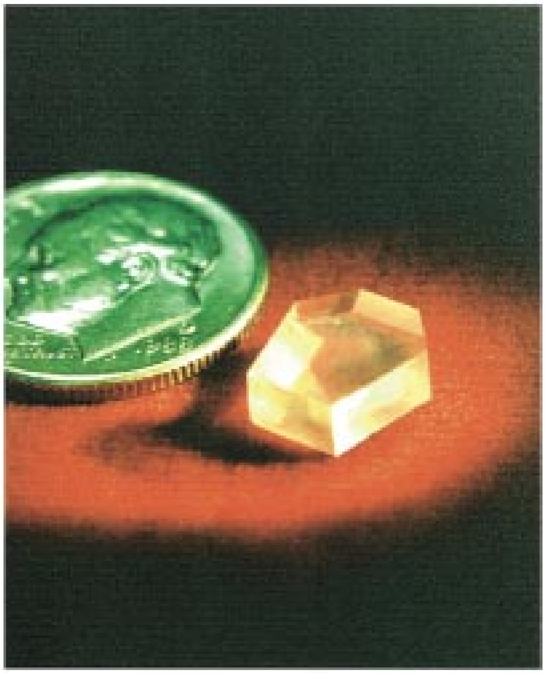


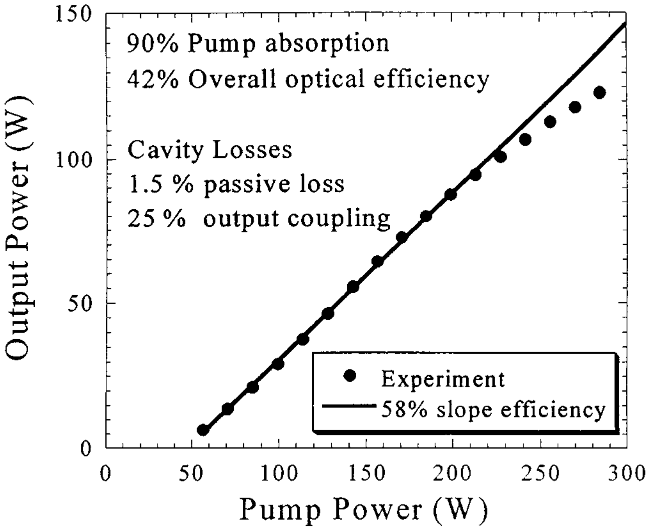
Fig. 6. Photograph of a monolithic Nd : YAG NPRO next to a 1-cm diameter coin. Small size of the monolithic laser isolates it from acoustic noise. Ring configuration assures single-frequency oscillation with freedom from back reflection-induced instabilities.

him that we could now begin the investigation of a laser-driven particle accelerator.

The NPRO provided the local oscillator for the coherent detection of wind velocity. A lamp-pumped Nd : YAG slab-laser amplifier with 62 dB of gain was also demonstrated [105]. The final element of the system was the fiber-based receiver for beam-combining the received signal with the local oscillator. The successful measurement of wind velocity using a Nd : YAG-based coherent laser radar was reported in 1987 in collaboration with Byvik, a visiting scientist from NASA [106].

The demonstration of coherent wind-velocity measurements was the first step in meeting the nearly impossible goal of wind-field measurements on a global scale. The challenge had led to LD pumping studies and to the single-frequency monolithic Nd : YAG ring laser oscillator. The highly coherent LD-pumped sources opened the door to research on highly coherent parametric oscillators and laser gravitational wave interferometers. The research in remote sensing at Stanford took a sabbatical leave for more than a decade as other opportunities were pursued. However, three books published between 1976 and 1984 summarized the progress in laser remote sensing [107]–[109]. The last of these books reported the proceedings of a conference held at Stanford University in October 1984. At that meeting, papers on the progress in diode-array development [110] and LD-pumped solid-state lasers [111] pointed the way to a very promising future in LD-pumped solid-state lasers.

In 1988, progress in LD-pumped solid-state lasers was reviewed by Fan and Byer [112]. By that time, an LD-pumped CW Nd : Glass laser [113], the first LD-pumped three-level laser [114], and the LD-pumped 2-μm Ho : YAG laser had been demonstrated [115]. The 2-μm wavelength eye-safe laser was considered a possible source for wind sensing. The experiment was conducted in collaboration with Prof. Gunter Huber, who was visiting Stanford from Hamburg in 1987. The early work also included LD pumping of a Nd : YAG and Nd : Glass miniature slab lasers using a borrowed LD array from Spectra Diode Laboratory [116].



(a)

(b)

Fig. 7. (a) The measured output power versus LD-pump power for an edge-pumped conduction-cooled Nd : YAG slab laser. (b) Photograph of the edge-pumped conduction-cooled Nd : YAG slab laser.

B. Highly Coherent CW Parametric Oscillators

The progress in nonlinear optical devices was equally rapid, driven by the availability of a stable single-axial-mode laser source and improved MgO : LiNbO$_3$ nonlinear crystals. Taking advantage of the single-frequency laser source, Kozlovsky and

Nabors reinvestigated external-resonant second-harmonic generation and obtained 56% second harmonic generation (SHG) conversion efficiency by harmonic generation of 52.6 mW and 1064 nm to generate 29.7 mW of 532 nm. The low loss of the monolithic external-resonant ring-doubler led to the high efficiency [117]. This was followed by a CW monolithic OPO that operated in a single frequency at both the signal and the idler [118]. The coherence properties of the CW–OPO were studied in detail. The conclusion was that the OPO reproduced the 10-kHz linewidth of the highly coherent Nd : YAG–NPRO pump laser [119]. The frequency and tuning control properties of CW parametric oscillators were studied extensively [120]. Further, the nonlinear optical coefficients of many standard nonlinear crystals were carefully remeasured by Eckardt. The surprising result, after two years of careful experimental work, was significantly lowered values of nonlinear coefficients of many crystals [121].

The potential for very wide wavelength tuning and the difficulty and delay in obtaining dielectric coatings for the monolithic nonlinear crystal resonators led to the study of total internal reflection (TIR) parametric oscillators. The work was preceded by the study of whispering gallery modes of a fused silica sphere by Schiller [122]. The NPRO provided the narrow linewidth probe, and the Gravity Probe-B program provided a highly spherical fused silica optical resonator. A quadruply resonant submilliwatt-threshold CW–OPO followed [123]. Nearly simultaneously, Darwin K. Serkland demonstrated a CW angle phase-matched $LiNbO_3$ OPO pumped at 1064 nm and oscillating near 2 μm. The measured finesse of the TIR $LiNbO_3$ cavity at 2 μm was 6000 and the tuning range extended from 2040 to 2225 nm. This monolithic OPO operated in a single axial mode at the signal and idler for tens of minutes [124].

Harris had predicted the benefits of high conversion efficiency and single-frequency operation of the singly resonant OPO more than 25 years earlier [33]. The operation of a CW singly resonant OPO had yet to be achieved because of the greater than 2-W threshold pump power. The high threshold of the SRO led A. Nilsson to write in his thesis that “a nightmarish dissertation might involve using a low power (Nd : YAG) nonplanar ring oscillator to injection lock a high-power oscillator that is then resonantly doubled to drive a CW singly resonant OPO.” The successful demonstration of injection locking[10] [125] of a 13-W lamp-pumped CW Nd : YAG laser oscillator by the Nd : YAG NPRO opened the door to the successful operation of a CW singly resonant optical parametric oscillator (SRO) by Yang. The “nightmarish” experiment was successfully completed using a lamp-pumped 20-W CW Nd : YAG laser injection locked to produce 19 W of single-axial-mode output. The laser was then frequency doubled in an external resonant cavity using Lithium Borate (LBO) to produce 11 W of CW 532-nm output. The 532 nm was used to pump a potassium titanyl phosphate (KTP) singly resonant OPO that operated with a threshold of less than 4 W and generated 1.9 W of CW output with 70% slope efficiency [126], [127]. Further, the SRO operated in a single-axial mode at the resonated wave as had been predicted. The CW SRO in KTP was demonstrated a short time before periodically poled $LiNbO_3$ crystals became available with their order of magnitude higher parametric gain.

C. Solid-State Lasers for Laser Interferometer Gravitational Wave Detection

During a visit to the Laboratory for Astrophysics at the University of Colorado in 1988, Prof. Peter Bender introduced me to the challenge of laser interferometer gravitational wave detection. Specifically, he showed me the concept for a three-spacecraft interferometer with 1.5 million-km arm lengths in orbit around the sun, trailing earth orbit by 20 degrees. This interferometer, now called laser interferometer space antenna, or LISA, is designed to detect gravitational waves at frequencies in the range of 0.1 mHz to 0.1 Hz [128]. Prof. Bender had learned about the NPRO Nd : YAG laser oscillator and explained that the LISA mission required a 1-W power single-frequency laser source. I assured him that by the date of the proposed LISA launch, 2010, the LD-pumped single-frequency oscillator would meet the LISA requirements. The LISA project has obtained a high priority by both NASA and by ESA as a medium-scale joint mission for the coming decade. The science goal is to observe gravitational waves emitted by binary star systems in our galaxy and by massive black holes that are now known to lie at the core of most galaxies.

Shortly after the visit with Prof. Bender, I was invited in 1988 to attend a laser interferometer gravitational-wave observatory (LIGO) proposal review by the National Science Foundation (NSF). There I learned about the proposed LIGO project to search for gravitational waves using 4-km arm-length Michelson–Fabry–Perot interferometers, located on the ground at two sites in the United States. The LIGO project, funded by the NSF, is now nearing completion of the first generation interferometer receiver. Articles by Barish and Weiss [129], [130] describe the LIGO project and progress toward operation. Stanford University faculty members have become part of the LIGO collaboration and are working to support the project in laser source development, interferometer architecture studies, material studies, and suspension and control system development. The measurement task is very challenging. The optical phase must be resolved to one-part per billion in the frequency range between 10 and 1000 Hz. Furthermore, the laser source must provide single-frequency output at 10 W of average power at 1064 nm with diffraction-limited spatial mode and within one-half percent of the quantum noise limit at the 10 MHz intermodulation measurement frequency. Taken together, LISA and LIGO present a nearly impossible challenge to the laser source and the optical system.

We began our research on the laser source for LIGO by investigating LD-pumped slab-geometry Nd : YAG laser oscillators. The first LD-pumped Nd : YAG slab was pumped by 52 fiber-coupled[11] 1 W laser diodes. The Nd : YAG miniature slab laser generated 5.5 W of CW injection-locked single-axial-mode output [131]. Farinas studied the frequency

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[10] The injection-locked laser is shown with a Faraday isolator in the cavity. A nickel bet was placed that the Faraday isolator was not necessary. The author collected the nickel.

[11] The 52 fiber-coupled 1-W laser diodes were loaned to us by the Sony Corporation.

and intensity noise of the injection locked laser oscillator and showed that it met the LIGO specifications [132]. The injection-locked oscillator approach has been selected by the gravitational-wave interferometer projects in France and Italy (VIRGO), in Germany (GEO), and in Japan (TAMA). The LIGO project took an alternative approach and selected an NPRO master oscillator followed by a power amplifier to generate the CW 10-W output power required for the LIGO-I receiver.

It is clear that future LIGO interferometer receivers will require greater power to reduce the shot noise. It is also clear that new interferometer designs may require other than a fixed-frequency single-frequency output laser source. We set about to increase the power of the LD-pumped slab-geometry laser. Two years following the first laser demonstration, we purchased more than 250 W of fiber-coupled LDs for pumping a next-generation slab laser. The resulting laser design was a direct water-cooled face-pumped Nd : YAG slab laser [133]. This laser generated more than 75 W multimode output and 40 W TEM_{00} mode output. The laser represented state-of-the-art performance in 1995, but suffered from coolant-induced noise and the lack of long-term reliability because of the direct water cooling of the zigzag laser slab surface. However, the laser developed by Shine showed that the LD-pumped Nd : YAG had adequate gain to operate as a CW amplifier as well as a power oscillator. This led to the possibility of constructing a master-oscillator power-amplifier (MOPA) laser source for LIGO. The advantages of the MOPA approach were freedom to control the laser coherence, the ability to add amplifier stages for future power scaling, and the ability to engineer the laser for long-term operational reliability.

The master oscillator power amplifier design approach was adopted by the LIGO project and a commercial laser company engineered the laser system.[12] The single-frequency 10-W Nd : YAG lasers were installed into the LIGO receiver in September 1998. The installed lasers have proven to be very reliable and have now operated for two years during the installation, alignment, and commissioning of the LIGO receiver. The LIGO receiver is planned to begin operation in early 2001.

The next generation LIGO laser interferometer receiver, LIGO-II, will require a TEM_{00} mode laser source with 200 W of output power. An advanced LIGO receiver will require in excess of 1 kW of laser power to reach the projected shot noise sensitivity level. Thus a laser design is required that is not only scalable in power, but offers simplicity and reliability as well. The challenge of the LIGO project forced us to reexamine the assumptions of solid-state laser design. Our goal was to design a reliable conduction-cooled low-noise power-scalable laser. This led us to evaluate the idea of pumping the zigzag slab laser not on the zigzag face but on the edge. At first glance, this approach is counter to the slab design goal of uniform pumping and cooling from the slab face. However, the edge-pumping approach had the potential advantages of high brightness pumping and the capability to conduction cool the slab faces.

We undertook a study of edge pumping and learned that the advantages far outweighed the potential disadvantages [134]. The study showed that edge pumping not only worked for Nd : YAG, but also for the quasi-three-level Yb : YAG laser system introduced by Fan in 1991 [135]. The Yb : YAG laser offers higher quantum efficiency, lower heat load, and the potential for power scaling to hundreds of kilowatts of average power. The Yb : YAG has been operated successfully at greater than the 1-kW power level by three groups including the Giesen group [136], [137]. Recently, the Yb : YAG laser has been operated as a single-axial-mode tunable oscillator with a 10 THz tuning range [138] and has been modelocked with subpicosecond pulse duration. The Yb : YAG laser is nearly ideal but requires high brightness pumping to reach its potential. The fiber-coupled laser-diode edge-pumped design allows the high brightness pumping required for efficient operation of Yb : YAG.

The edge-pumped slab laser design was demonstrated experimentally for Nd : YAG by Bill Tulloch and Todd Rutherford. Fig. 7(a) shows the measured 125-W output power for an edge-pumped Nd : YAG slab laser that operates at 55% slope efficiency with 40% optical efficiency. Fig. 7(b) shows a photograph of the edge-pumped Nd : YAG slab laser with Todd Rutherford overseeing his invention.

The rapid progress in both LD-pumped solid-state lasers and nonlinear devices occurred in the 1987 to 1993 time, during which I had been asked to serve in an administrative position at Stanford University. Because of these commitments, I suggested to the students in my group that the group size might have to be reduced. The students were motivated to undertake a study to determine productivity versus group size. They concluded that the group size was near optimum and that efficient progress was made because the senior students had time to introduce new students to the ongoing research and to the underlying technology. I observed that the productivity of the group increased with my absence, a disturbing finding to say the least.

V. ENGINEERED NONLINEAR MATERIALS

A. Progress in Nonlinear Materials and Devices

The progress in optical parametric oscillation and amplification was the focus of special issues of the *Journal of the Optical Society of America B* [139]. The articles featured the considerable progress and achievements in parametric devices made during the previous two decades. Progress in CW parametric devices, pulsed high-peak-power infrared tunable sources, visible parametric sources, and spectroscopic applications was covered. A section was devoted to synchronously pumped and travelling wave picosecond and femtosecond parametric devices, an area pioneered by Piskarskas, Laubereau, and Tang and actively pursued by Hanna, Ferguson, Wallenstein, and others.

A second special issue on optical parametric devices was published in the *Journal of the Optical Society of America B* in November 1995 [140]. The progress in parametric devices highlighted in that issue included highly efficient devices, frequency control and spectroscopic applications, picosecond and femtosecond synchronously pumped parametric devices,

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[12] Lightwave Electronics, Mountain View, CA.

parametric amplifiers, quantum optical effects, and squeezing in parametric processes. Also included was a contribution on quasi-phasematched (QPM) optical parametric oscillators in bulk periodically poled $LiNbO_3$ [141]. Progress in QPM nonlinear optical materials during the preceding five years had allowed QPM parametric oscillator devices to be demonstrated based on ferroelectric domain inversion in $LiNbO_3$.

B. Quasi-Phasematching in Periodically Poled Nonlinear Crystals

In 1988, 20 years after the idea of quasi-phasematching had been patented by Bloembergen [36], the possibility of fabricating nonlinear crystals to obtain periodic modulation of the nonlinear coefficient was considered seriously by at least three groups around the world. The work had been preceded by earlier studies [142]–[147] but crystal quality and technology had not allowed progress to be made in the control of ferroelectric domains to the precision required.

The work at Stanford University on laser-assisted single-crystal fiber growth had been initiated in 1974 with the goal of preparing and studying nonlinear interactions in guided-wave devices. The growth of single-crystal fibers is described by Fejer [148], [149]. In 1987, Fejer noticed that c-axis $LiNbO_3$ single-crystal fibers grew with preferential domain orientation. Shortly after, we discovered that domain orientation could be controlled during growth by modulation of the growth interface temperature or modulation of the crystal-fiber displacement during growth [150], [151]. This led to 0.5-mm diameter periodic-poled $LiNbO_3$ single-crystal-fiber devices with micrometer-scale domains for the generation of blue [152], [153] and green [154], [155] by second-harmonic generation.

Simultaneously, Fejer and his group investigated the possibility of chemical diffusion at the surface of $LiNbO_3$ to control domain inversion. The effort was successful and led to the first demonstration of a QPM waveguide device to generate green [156] and blue output [157]. There was simultaneous progress in Japan [158], [159] and Sweden [160] and in periodically poled KTP by Bierlein *et al.* [161], [162]. In a particularly striking experiment, Prof. Hiromasa Ito of Tohoku University in Sendai, Japan, demonstrated electron-beam poling of 1-μm domains with propagation distances of nearly 1 mm, an aspect ratio of 1000 [163]–[165]. The work was followed by electric-field poling by Yamada *et al.* of Sony [166], [167] which overcame the slow writing speed of electron-beam poling.

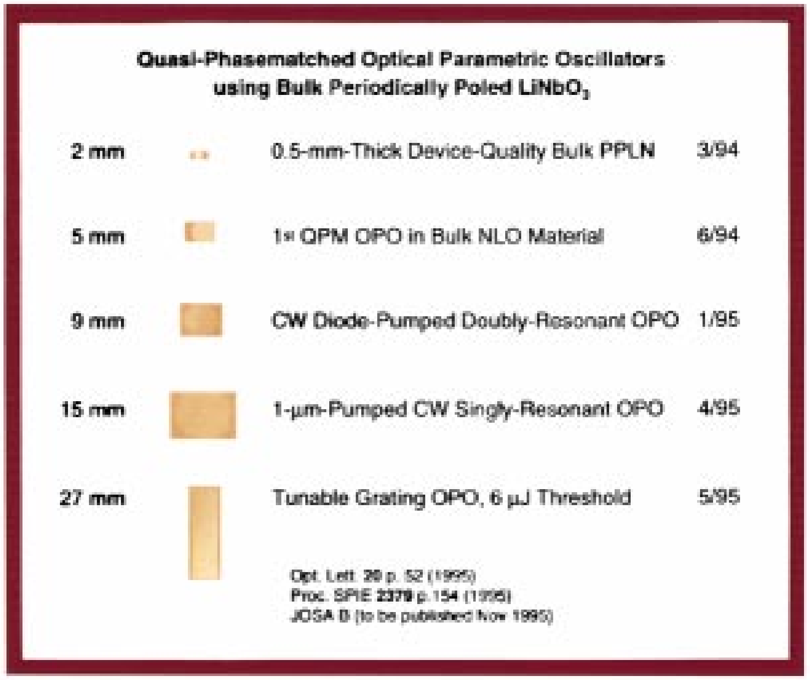
The early work in quasi-phasematching was reviewed by Fejer [168] and by Byer [169]. A broad overview of nonlinear optics was prepared by Fejer [170]. The recent overview of nonlinear optics with an emphasis on quasi-phasematching was prepared by Byer [171] for the Rem V. Khokhlov 70th Jubilee Celebration held in Moscow, Russia, from October 14–19, 1996. These reviews provide a complete reference to past work and an introduction to the advantages and progress in quasi-phasematching. Thus, in this paper, only highlights are presented to illustrate progress in nonlinear materials by engineering the ferroelectric domain structure of $LiNbO_3$ to obtain optimum interactions.

Quasi-phasematching provides many advantages for nonlinear interactions. Crystals that cannot be phasematched because of the lack of adequate birefringence to offset dispersion can be phasematched by modulating the sign of the nonlinear coefficient. Periodically poled $LiNbO_3$, for example, can be quasi-phasematched over the entire transparency range from less than 400 nm to greater than 4000 nm. Further, the crystal orientation can be selected to optimize the nonlinear interaction. In $LiNbO_3$, the d_{33} nonlinear coefficient is 7 times larger than the d_{31} nonlinear coefficient used in birefringent phasematching. This yields a factor to 20 improvement in nonlinear gain. Quasi-phasematching allows the tailoring of the phasematching interaction to broaden the wavelength acceptance, generate multiple output frequencies, or lengthen or shorten pulses. These advantages of quasi-phasematching were analyzed by Fejer *et al.* in 1992 [172].

Although domain modulation during crystal growth produced samples adequate for device demonstration, the technique was not suitable for volume production of QPM crystals. Therefore, one goal of the research programs was to develop a lithographic-patterned electric-field-based poling process for creating inverted domains with controlled period and duty cycle at a wafer scale. The long-range goal was to produce, at low cost, QPM "nonlinear chips" engineered to optimize device performance from wafers of $LiNbO_3$. Control of domain inversion and duty cycle proved to be much more difficult than expected. However, persistence coupled with understanding and modeling of the electric-field-induced domain-inversion process led to success in 1994 [173]–[175].

The steps taken to define an appropriate processing recipe for lithographic-patterned electrodes and room-temperature electric-field poling of $LiNbO_3$ are described by Myers [141], [176]. Briefly, the wafer of $LiNbO_3$ is thoroughly cleaned, patterned, and coated with metal electrodes on the $+z$ surface to provide electric-field contact for poling. The metal electrodes are overcoated with a thin dielectric layer to inhibit growth of the domains between the metal electrodes. The field is applied to the sample using a liquid electrolyte contact to achieve field uniformity. Domain inversion is initiated when the applied electric field exceeds the coercive field of 21 kV/mm for $LiNbO_3$ at room temperature. The field is applied for up to 1-s duration by a high-voltage supply through a current-limiting resistor of 100 MΩ. For a properly configured circuit, the poling current terminates on completion of poling at a domain duty cycle set by the choice of the electrode width to spacing ratio. The domain duty cycle can be controlled by the correct choice of electrode duty cycle and the applied field in excess of the coercive field. It has been experimentally observed that domains can be inverted without dielectric breakdown for properly prepared wafers. To date, poling has been accomplished in wafers up to 1-mm thickness [177]. Domains revert to their original orientation or can be reversed repeatedly if the field is applied for less than 50 ms. However, the inverted domain regions are permanent for fields applied longer than approximately 50 ms after completion of poling. The domains are made visible by etching with hydrofluoric acid or by observation through crossed polarizers.

Domain periods of 10 to 30 μm that are suitable for infrared devices were first generated in 2-mm lengths of 0.5-mm-thick



Quasi-Phasematched Optical Parametric Oscillators using Bulk Periodically Poled $LiNbO_3$

2 mm	0.5-mm-Thick Device-Quality Bulk PPLN	3/94
5 mm	1st QPM OPO in Bulk NLO Material	6/94
9 mm	CW Diode-Pumped Doubly-Resonant OPO	1/95
15 mm	1-µm-Pumped CW Singly-Resonant OPO	4/95
27 mm	Tunable Grating OPO, 6 µJ Threshold	5/95

Opt. Lett. 20 p. 52 (1995)
Proc. SPIE 2379 p. 154 (1995)
JOSA B (to be published Nov 1995)

Fig. 8. Progress in periodically poling of $LiNbO_3$ "chips" as illustrated by photographs of the devices over an 18-month period. Not shown, but illustrated on the cover photo of *Laser Focus World*, May 1997, is the periodic poled 3-in diameter wafer of $LiNbO_3$. The 27-mm-long chip contains 25 OPOs, each with a different grating period. When Larry Myers was asked how many of the 25 OPOs operated, he replied "all but one: it was designed to be below cut-off near degeneracy."

$LiNbO_3$ in March 1994. By May 1995, the QPM interaction length had been extended to 27 mm. By September of 1995, wafer-scale processing of lithium niobate was achieved and nonlinear device "chips" were being fabricated with high yield and reproducible characteristics in 0.5-mm-thick by 50-mm-long samples based on processing of 3-in-diameter $LiNbO_3$ wafers [176]. Fig. 8 illustrates the progress in room-temperature electric-field poling of $LiNbO_3$. The 27-mm-long sample produced in May 1995 is special in that it contains 25 parametric-oscillator QPM gain regions, each with a different grating period.

C. Engineered QPM Nonlinear Devices

1) Guided-Wave QPM Nonlinear Devices: The single-pass second-harmonic conversion efficiency in bulk nonlinear devices is limited by diffraction spreading of the focused laser beam. For nonlinear interactions in channel waveguides, the parametric gain is expected to be high because the guided wave extends the interaction length. The progress in QPM-waveguide nonlinear interactions is highlighted here.

Bortz *et al.* [178] conducted the first demonstration of a QPM parametric oscillator and amplifier. The guided-wave OPO/OPA operated at single-pass gains of 4.1 dB corresponding to 18% W^{-1} gain at a signal wavelength of 1.55 μm when pumped by a 782.2-nm source. Parametric oscillation was observed at wavelengths between 1.4 and 1.7 μm with a peak output power of 700 mW for an incident peak pump power of 7.7 W coupled into the waveguide.

The QPM waveguide OPO performance has been extended recently by Arbore [179], who reported a singly resonant optical parametric oscillator in a QPM guided-wave structure. In this case, electric-field poling of $LiNbO_3$ was used for the QPM structure. Parametric gains as high as 250%/W and an oscillation threshold of 1.6 W was measured. Pump depletion of 40% was observed. With further optimization of the waveguide, the singly resonant oscillator is predicted to have a threshold of $\sim$100 mW, which is accessible to LD-pumping.

Progress in LD frequency doubling in guided-wave QPM–$LiTaO_3$ was made by Yamamoto *et al.* [180], who demonstrated 4.5 mW of blue light with a 13% conversion efficiency. In Rubidium Titanyl Arsenate (RTA), an isomorph to KTP, Risk and Loiacono [181] demonstrated 225%/Wcm^2 normalized doubling efficiency in a waveguide structure. 8.7 μW of blue at 437.5 nm was generated for 21.1 mW of input. The advantage of RTA compared to KTP is that it can be electric-field poled.

The high conversion efficiency of guided-wave QPM devices allows the interaction to be engineered for other factors that may be important in device performance. For example, the narrow wavelength and temperature acceptance of the nonlinear interaction may limit practical applications of the QPM devices. Fejer *et al.* [172] suggested in their theoretical description of QPM interactions that the phase synchronism bandwidths could be synthesized using Fourier synthesis. The phasematching acceptance bandwidth is the Fourier transform of the nonlinear coefficient distribution or, equivalently, the crystal length for a uniform nonlinear interaction length. A QPM tuning curve, up to a scale factor dependent on the dispersion, is shifted by the periodic modulation of the nonlinear coefficient and has the same bandwidth as the nonshifted structure. This approach artificially broadens the synchronism phasematching curve through a-periodic modification of the QPM structure.

This idea was experimentally demonstrated by Bortz *et al.* [182], [183]. An analysis was presented by Fujimura *et al.* [184] and extended by Mizuuchi *et al.* [185]. Details of synthesizing novel tuning curves using nonuniform QPM gratings is presented by Bortz [182], [183].

Modulation of the QPM gratings can produce sidebands and multiple phasematching peaks in addition to the principle phasematcing peak. Thus, multiple output frequencies in mixers or parametric amplifiers can be generated. It is also possible, and has been demonstrated recently, that chirped QPM gratings can be used to compress frequency-chirped pulses while generating the second harmonic. This is a fundamentally different approach to pulse compression using a nonlinear interaction in a media with an a-periodic QPM grating [186]. Pulse shaping and compression by second-harmonic generation has been treated for the case of an arbitrary dispersion by Imeshev *et al.* [187].

2) Bulk-Wave QPM Nonlinear Devices: Nearly simultaneously with the demonstration of the first QPM–OPO by Bortz *et al.* [178] in a waveguide, Myers *et al.* [188] operated the first bulk crystal QPM–OPO. The QPM–OPO was pumped by a Q-switched Nd : YAG laser and tuned over the 1.66–2.95 μm spectral region. A Chromatix Q-switched Nd : YAG laser was the pump source. The threshold was approximately 0.1 mJ, more than an order of magnitude below the damage level of the QPM–LiNbO$_3$ crystal and more than a factor of 20 below the threshold energy of a birefringently-phasematched LiNbO$_3$ OPO [59]. Larry Myers stated “I see red” on seeing OPO oscillation for the first time. Still today, the generation of red by QPM sum generation of the OPO signal and pump waves is the signature of QPM–OPO oscillation. The QPM–LiNbO$_3$ crystal was an electric-field poled, 0.5-mm-thick sample with a 31-μm period over the 5.2-mm interaction length. The low threshold of the QPM–LiNbO$_3$ OPO has allowed OPO and OPA’s to be pumped by microchip lasers with threshold pump energy near 1 μJ, operation at 1-kHz repetition rates, and with a 25% conversion efficiency over the tuning range from 1.4 to 4.3 μm by Zahowsky [189].

The efficiency of nonlinear frequency conversion is improved by the use of high peak powers available from mode-locked laser sources. Further, extending the wavelength range of mode-locked lasers is important for many applications. In 1996, Pruneri *et al.* [190] used a mode-locked Nd : YLF laser to study second-harmonic generation in QPM–LiNbO$_3$. In a CW mode-locked SHG experiment, 330 mW of green was generated at an average conversion efficiency of 52%. A QPM–LiNbO$_3$ sample of 3.2-mm length was used for frequency doubling the 2.5-ps pulses. The QPM grating with 6.35-μm period was fabricated using electric-field poling with liquid electrolyte electrodes.

The picosecond-pumped interaction was extended by Pruneri *et al.* [191] to synchronously pump at a wavelength of 523.5 nm an OPO with a tuning range from 883 to 1285 nm. The OPO generated 200 mW of average output power within a 10-μs pulse envelope. The QPM–LiNbO$_3$ sample was 3.2-mm long, a suitable length for the 2 ps pump pulses. Pump depletion of 50% was observed during the 10 μs duration of the macropulse. The synchronously pumped QPM–LiNbO$_3$ OPO was extended to CW operation by Butterworth *et al.* [192]. Here, the OPO was pumped at 1.047 μm by a mode-locked Nd : YLF laser. Picosecond pulses were tunable from 1.67 to 2.806 μm. The average output power level was 120 mW with a slope efficiency of 61% at 75% pump depletion when the oscillator was operated at three times threshold. The output power stability of this OPO was excellent, with less than 5% peak-to-peak variation. As noted by the authors, synchronously pumped QPM–OPOs are a promising, compact, and low-cost source of broadly tunable radiation.

Galvanauskas *et al.* [193] demonstrated the first femtosecond QPM–LiNbO$_3$ parametric generator with output tunable from 1 to 3 μm when pumped at 777 nm. The OPO reached 38% internal conversion efficiency at 220 nJ of pump energy at 200-kHz repetition rate. The output pulse length was measured by autocorrelation and found to be 300 fs full-width at half-maximum (FWHM).

The operation of a CW SRO offers many advantages in spectral control, conversion efficiency, tuning range, and stability. However, as is well known, the threshold of the SRO is approximately 200 times higher than for a Doubly Resonant Oscillator (DRO) with the same 1% round-trip losses. Successful operation of a CW–SRO requires QPM–LiNbO$_3$ samples that are near theoretical performance in parametric gain over a gain length of 50 mm. It is a considerable challenge to achieve uniform domain inversion with near 50% duty cycle for the required interaction lengths.

Miller *et al.* [194] studied electric-field poling and developed a model that described the optimized electrode pattern and applied electric field to reach self-terminated electric-field poling with near 50% duty cycle. The model was applied to successfully produce 50-mm-long QPM–LiNbO$_3$ samples with 30 μm periods suitable for infrared OPO operation.

Myers *et al.* [195] demonstrated CW–SRO operation with a 3-W threshold power for a 1.064-μm Nd : YAG pump. The CW–SRO used a 50-mm-long QPM–LiNbO$_3$ gain element. The output power levels generated were greater than 2.5 W at 3.3 μm. The device was tunable over the 1.4–1.6- and 3.1–4.0-μm range. The SRO spectral characteristics were studied by Bosenberg *et al.* [196]. As predicted by Harris [33] 25 years earlier, the SRO operated in a single-axial mode on the resonated signal wave even when the pump was multiaxial mode. The CW–SRO operated with less than 1% amplitude fluctuation when the signal wave was single-axial mode. The maximum output power was 1.25 W at 3.25 μm and 0.36 W at 1.57 μm for 13 W of pump power.

Bosenberg *et al.* [197] extended the CW–SRO performance to achieve a remarkable 93% pump depletion for the Nd : YAG-pumped QPM–LiNbO$_3$ singly resonant OPO. The SRO operated with 86% of the converted pump photons as useful idler photons at 3.25 μm. This SRO was operated in both a standing wave and in a ring cavity. The performance of the SRO was improved, with the ring cavity configuration reaching 93% pump depletion at 2.4 times threshold. The idler output power reached 3.7 W for an input pump power of 14 W. An etalon within the SRO resonator allowed for tuning over a 5 cm^{-1} range with discrete mode hops at each axial mode. A multiple grating QPM–$LiNbO_3$ element with grating periods stepped in quarter micrometer increments from 28.0 to 29.75 μm was also tested in the CW–SRO. Tuning was achieved over a range from 1.45–1.60 and 3.95–3.25 μm by translating the QPM crystal transverse to the SRO resonator axis.

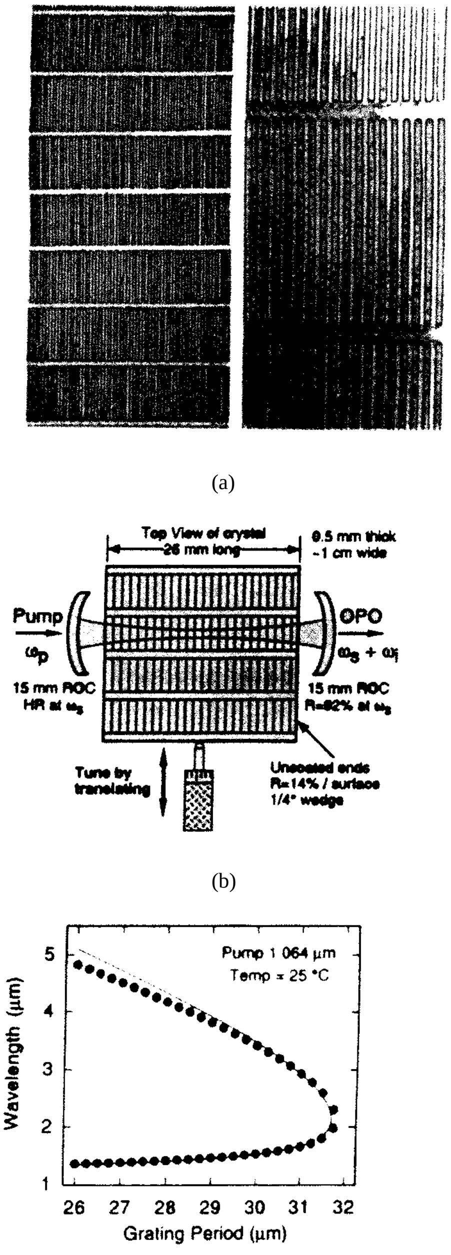
The power of lithographic patterning of QPM–$LiNbO_3$ gain elements is illustrated by the multigrating QPM–OPO demonstrated by Myers *et al.* [198]. Beginning with a 0.5-mm-thick wafer of $LiNbO_3$, Myers designed and then prepared an electric-field-poled 27-mm-long interaction-length QPM–$LiNbO_3$ chip with 25 OPO gratings each of 500-μm width. The grating periods ranged from 26 to 32 μm, thus allowing for tuning across the 1.4- to 4.8-μm spectral range when pumped by an acousto-optic Q-switched Nd : YAG laser source.

Fig. 9(a) shows a photograph of a section of the QPM–$LiNbO_3$ multigrating domain array. On the left are seven gratings of 500-μm width separated by 50 μm. On the right is an enlargement of the 29-μm-period grating showing in detail the exquisite uniformity of the electric-field poling process. Fig. 9(b) shows a schematic of the experimental setup for the multigrating QPM–OPO. The OPO was tuned by translating the crystal perpendicular to the OPO resonator optical axis. No realignment was needed, and all grating sections oscillated with good efficiency. Fig. 9(c) shows the OPO tuning range versus the grating period. The tuning gaps can be covered by a small shift in the crystal temperature. In the future, the gratings can be fabricated with closer pitch spacing or with a wedged pitch for continuous tuning. This multigrating OPO had a threshold of 6 μJ for the 26-mm interaction length. The OPO reached 70% pump depletion at 8 times threshold and operated well below the damage level of the QPM–$LiNbO_3$ crystal.

Continued progress in electric-field poling by Miller *et al.* [199], [200] led to the preparation of a 5.3-cm-long 6.5-μm-domain period first-order QPM–$LiNbO_3$ sample for bulk CW–SHG of an Nd : YAG laser. Miller *et al.* [199], [200] reported single pass CW–SHG experiment generated 2.7 W of 532 nm output for 7.5 W of 1064 nm input. This CW 42% single-pass conversion efficiency is the highest reported to date and represents a significant breakthrough in nonlinear optical frequency conversion.

In an experiment that used the same 5.3-cm-long crystal chip, Batchko *et al.* [201] demonstrated a 532-nm CW-pumped singly resonant OPO based on the same 5.2-cm-long first-order QPM–$LiNbO_3$ samples. The CW–SRO had a threshold of less than 1 W and operated with 64% quantum efficiency. At three times threshold, the pump depletion reached 60% and the idler output power was 0.9 W. Work is continuing on QPM–OPOs with the goal of intracavity frequency conversion of the signal and idler wave to generate blue and red for possible application to RGB displays.

The generation of red, green, and blue laser output was a goal of research from the beginning [154], [155] and was of special interest to both Greg Miller and Rob Batchko. The interest in RGB displays led to collaboration with the Sony Research Laboratory directed by Dr. Shigeo Kubota. During a visit to Sony, I stated that our laser design approach followed the KISS principal. Asked to explain what that meant, I quickly translated the slang term into the phrase “Keep it simple, straightforward, and elegant.”



(a)

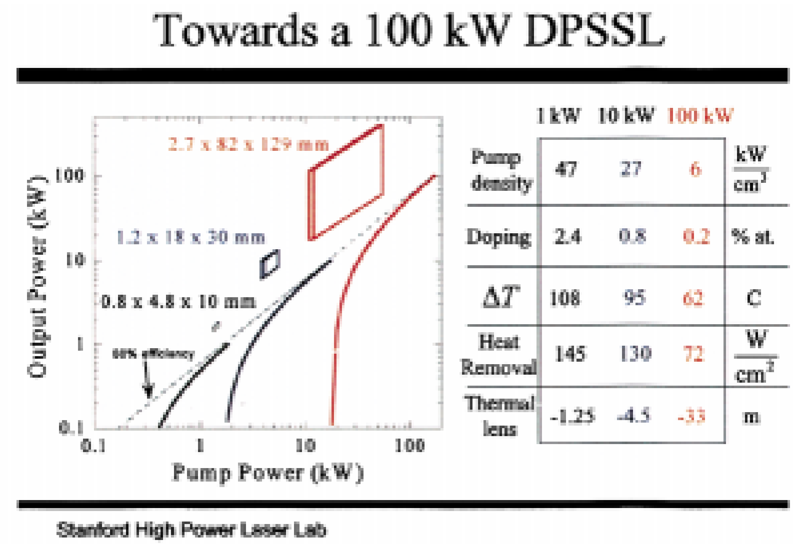
(b)

(c)

Fig. 9. (a) Section of the 26-mm-long multigrating QPM–OPO chip. On the left is a series of gratings with 26–32 μm periods; on the right is an enlarged section showing the fidelity of the periodic poled regions. (b) Tuning was accomplished by translating the gratings through the pump beam. No realignment was necessary. (c) Multigrating OPO tuning curve that extended from 1.36 to 4.83 μm over 24 gratings. The OPO threshold was 6 μJ for the 7-ns-long Nd : YAG pump pulses.

D. Engineered Nonlinear Materials and Devices

The progress in engineered nonlinear materials and nonlinear devices continues at an expanded pace. The possibility of electro-optic control of linear optical devices, such as prisms for beam steering [202] and lenses [203], has been explored. However, future progress, such as an electro-optic controlled Bragg mirror, depends upon the capability to write submicrometer domains. Progress in nanometer-scale domains is being made, with the introduction of backswitch poling by Batchko


Towards a 100 kW DPSSL

	1 kW	10 kW	100 kW	
Pump density	47	27	6	$\frac{kW}{cm^2}$
Doping	2.4	0.8	0.2	% at.
ΔT	108	95	62	C
Heat Removal	145	130	72	$\frac{W}{cm^2}$
Thermal lens	-1.25	-4.5	-33	m

Stanford High Power Laser Lab

Fig. 10. Design for a 100-kW LD-pumped Yb : YAG amplifer based on the edge-pumped slab design approach. Design parameters become easier to meet as the size of the slab and power increases. However, engineering for power is always a challenge.

et al. [204]. The poling method allows precise control of ferroelectric domains and, for the first time, “domain engineering” [205]. Progress had been rapid and it now appears possible to create and control domains at the nanoscale [206]. Ferroelectric domain engineering for QPM nonlinear interactions has been reviewed by Rosenman *et al.* [207].

Engineered nonlinear interactions include the cascaded nonlinear interactions first proposed by Stegeman [208] and demonstrated by others [209], [210]. The application of quasi-phasematching to semiconductor crystals has been explored both by stacking plates of GaAs [211], [212] and by patterned epitaxial growth on a modulated GaAs substrate. After many years of work, Eyres has succeeded in growing orientation patterned GaAs [213]. It is still early in the program, but the promise of patterned GaAs as a QPM nonlinear material from 1 to 16 μm is exciting.

Perhaps the most promising application of engineered nonlinear materials and interactions is the use of QPM–$LiNbO_3$ as a mixer to meet optical telecommunication applications. The image my colleague Marty Fejer projects is that of a simple radio-frequency mixer, which is the key element in electronics processing of radio-frequency signals. The QPM–$LiNbO_3$ mixer is equally promising and more versatile as described in the “best seller” Ph.D. dissertation of Ming-Hsien Chou [214], [215] and extensions of the work by current students in the Fejer group [216]. Thirty-five years after the first demonstration of parametric mixing by Wang and Racetti, the “nonlinear chips” fabricated on a wafer scale by lithographic methods are poised to make significant contributions to optical signal processing in the optical telecommunications world.

Progress in device performance often depends on progress and understanding of fundamental material properties. The development of nonlinear optics is not an exception. At each stage, progress was dependent upon the understanding and resolution of materials issues. In the case of periodic poling, the Center for Nonlinear Optical Materials, initiated in 1992, assisted the process by providing a neutral meeting ground for research scientists and engineers in the private sector to meet with and learn about students and the research progress in university laboratories. The progress continues with new materials challenges and new devices to be designed and tested.

VI. FUTURE CHALLENGES

The beauty of science is that we build on and learn from those who made past contributions, and there are always new challenges to ride on the expanding wave of knowledge. For students entering the field of quantum electronics, there is much to learn in reading about past successes, conceptual breakthroughs [217], and lucky breaks. However, there are future paths to be followed and new challenges to be met. Here I mention a few of the “unpaved roads” that are of interest to me and come close to meeting the Edwin Land criteria of being nearly impossible but manifestly important.

Precision measurement has been a hallmark of quantum phenomena and, in particular, of lasers for the past half century. The goal to detect and to observe the universe with gravitational waves is a precision measurement challenge of first order. Perhaps by the end of this decade, with LIGO and later with LISA interferometers operational, we will succeed in observing with gravitational wave signals. Gravitational waves, in our galaxy, are likely to be generated by the 3000 or so white-dwarf and neutron binary star systems. Gravitational waves from the more than 50 billion galaxies in the universe are likely to be generated by black holes that lie in their core. A new window of the universe may open because of the contributions of many scientists and engineers on the art of precision interferometry using advanced laser sources.

Highly coherent laser sources, operating at high peak power in picosecond pulses and phaselocked to better than one degree of optical phase, are required to drive future laser accelerators [218]. If successful, the SLAC 30-GeV accelerator, driven by klystrons invented more than 70 years ago, may give way to a teraelectronvolt machine driven by lasers invented only 40 years ago. The challenge is nearly impossible, but the rewards are worth the pursuit. Imagine attosecond bunches of electrons surfing an optical wave for a kilometer distance. Each electron microbunch is less than 100 nm in diameter and less than 10 nm in length, and must negotiate 4-μm-wide slits of one-million 1-mm-long accelerator cells. The 1-MW average power mode-locked laser must remain phase-locked over the entire accelerator length.

After more than 20 years, the nearly impossible task of global remote sensing of atmospheric molecular constituents and the global wind field may become a reality. The combined invention of the Yb : YAG laser and the edge-pumped slab laser has opened the door to high-average power LD-pumped solid-state lasers that also store energy. Fig. 10 shows the power scaling of an edge-pumped Yb : YAG slab-laser amplifier. A Yb : YAG slab laser that fits in a breadbox has the potential to emit 100 kW of average power. A much smaller slab, only 3 cm in length, can store more than 10 J of energy, which can be extracted in 1-μs pulses with a Fourier transform-limited linewidth. This source meets the transmitter requirements for coherent wind detection on a global scale if the wavelength is shifted from Yb : YAG 1.03-μm wavelength to the eyesafe 1.5-μm wavelength region [219]. For this, a periodically poled $LiNbO_3$ parametric amplifier is required. After nearly a decade, the sabbatical leave from research in global remote sensing is about to come to an end. Keep it simple, straightforward, and elegant.

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