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NONLINEAR OPTICAL FREQUENCY CONVERSION

Mixing laser beams of different frequencies in a crystal with nonlinear polarizability can generate coherent output at sum and difference frequencies for which there are no convenient laser sources.

Martin M. Fejer

The colored rings shown in figure 1 are a spectacular manifestation of one type of nonlinear optical frequency conversion: parametric amplification of quantum noise. The amplification in this case is produced by the propagation of an intense pulse of ultraviolet radiation through a crystal of barium borate. When intense electromagnetic radiation propagates through such a material, whose polarization response at optical frequencies manifests a strongly nonlinear dependence on electric field amplitude, nonlinear mixing of the input radiation generates new spectral components. We can exploit this phenomenon to generate coherent radiation at frequencies for which we have no convenient laser sources.

Since Peter Franken's original demonstration of second-harmonic generation by such means in 1961,[1] nonlinear optics has evolved from a laboratory curiosity into a tool that finds applications in fields as diverse as laser fusion, biomedical instrumentation, femtosecond spectroscopy and precision metrology. Though nonlinear optical technology is now well into its fourth decade, a renaissance is under way, fueled by recent improvements in pump lasers and nonlinear materials and by the demand for efficient sources of coherent radiation for a variety of important applications. In this article I will review the basic ideas of optical frequency conversion in nonlinear crystals and describe some recent advances in the relevant materials and devices.

Nonlinear susceptibility

For the phenomena discussed in this article, the nonlinear response of dielectric media is adequately described by a local, weakly dispersive second-order susceptibility χ_2. (See the introductory article by Elsa Garmire on page 23.) In that approximation $\mathbf{P}$, the dipole moment per unit volume induced by the applied electric field $\mathbf{E}$, is given by[2]

$$\mathbf{P} = \varepsilon_0(\chi_1\mathbf{E} + \chi_2\mathbf{EE})$$

The first term is responsible for ordinary optical phenomena like reflection and absorption, and we have neglected the third-order susceptibility, which is responsible for

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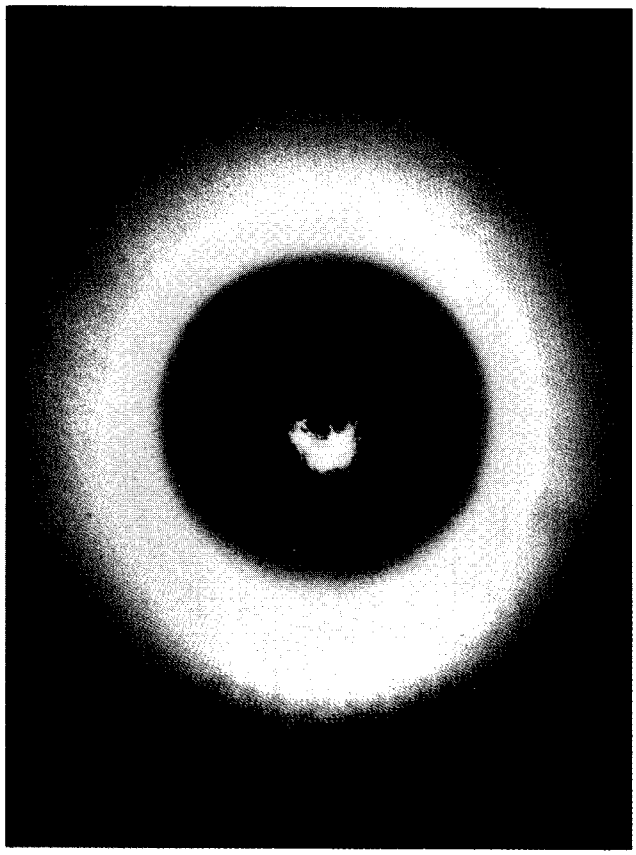


Parametric amplification of quantum noise. This spectrum of concentric rings was generated by the nonlinear interaction of an intense pulse of ultraviolet radiation with a 1-cm-long crystal of barium borate through which it propagated. The angular radii of the rings for the various colors are determined by the requirement that the phase velocities of interacting waves in the anisotropic crystal must be matched. (Courtesy of Walter Bosenberg, Lightwave Electronics.) **Figure 1**

phenomena such as the Kerr effect, the Raman effect and third-harmonic generation. The second-order suceptibility χ_2 is a third-rank tensor and therefore must vanish if the medium is symmetric under inversion through a center of symmetry. Its contribution to polarization component P_i is the sum over coordinate indices j and k of terms $\chi_{2ijk}E_j E_k$.

The origin of the nonlinear susceptibility can be viewed classically as the response of an electron driven by an electromagnetic field in an anharmonic potential well resulting from the interatomic electric field $\mathbf{E}_A$ in the solid. (See figure 2.) The interatomic field is on the order of a volt per angstrom, or 10^8 V/cm. For driving optical fields $\mathbf{E}$ much weaker than that, the polarization response is essentially linear. For optical fields intense enough to drive the electron beyond the quadratic minimum of the interatomic potential, the response becomes increasingly nonlinear: The ratio P_2/P_1 of quadratic to linear polarization response is on the order of E/E_A, so it is of order 10^{-4} even for laser beams with intensities of a megawatt per cm^2, in which E is as high as 10^4 V/cm. Adequately transparent materials of practical importance for nonlinear frequency conversion are found to have χ_2 in the range from about 0.2 to 20 picometers per volt.

Frequency mixing

If two monochromatic optical fields, with frequencies ω_a and ω_b and amplitudes E_a and E_b, respectively, are applied to a medium with nonvanishing second-order susceptibility χ_2, the induced dipole response will contain, in addition to the ordinary linear response terms at frequencies ω_a and ω_b, a nonlinear response P_2 proportional to $\chi_2 E_a E_b$, with spectral components at $\omega_a \pm \omega_b$. In the important special case where the two input frequencies are the same, the nonlinear polarization contains the second harmonic of the driving frequency and a dc "optical rectification" term.

The essential goal of practical nonlinear optical frequency conversion is the efficient generation of radiation at such sum, difference and second-harmonic frequencies. At microwave frequencies, the availability of diode rectifiers and other strongly nonlinear elements permits the straightforward design of efficient "lumped" mixers, smaller than a wavelength in size. At optical frequencies, by contrast, nonlinear responses are quite weak, so that efficient mixing requires "distributed" devices many wavelengths long.

Many of the practical problems associated with optical frequency mixing stem from the distributed nature of the mixing process. In particular, the difference in the phase velocities of the interacting waves of different frequencies in the nonlinear medium produces a phase difference that accumulates along the length of the device and significantly limits the efficiency of the mixing process. Consider the convenient special case of second-harmonic generation:[2] A plane wave at the fundamental frequency ω (the "pump" wave) propagates with a phase velocity $c/n(\omega)$ through a nonlinear medium of length L, where n is the frequency-dependent refractive index. A polarization wave proportional to the nonlinear susceptibility and the square of the incident pump field is thus generated, with twice the temporal and spatial frequency of the pump. This polarization wave serves as an oscillatory current density that generates an electromagnetic wave at the second-harmonic frequency 2ω, propagating with phase velocity $c/n(2\omega)$.

In an ideal "phase matched" interaction, the velocities of the fundamental and second-harmonic waves are equal, the relative phase of the driving polarization and the generated field remains constant throughout the nonlinear medium, and the magnitude of the second-harmonic field grows linearly along the length of the medium. (See the red line in figure 3.) But if the phase velocities differ, the relative phase of the interacting waves varies along the medium, so that the direction of power flow oscillates, initially generating the second harmonic but then converting the second harmonic back into fundamental radiation after the relative phase reaches π. Thus efficient interaction requires that that the wavenumbers k satisfy the condition $k_{2\omega} - 2k_\omega < \pi/L$.

Quantum mechanically, second-harmonic generation involves the destruction of two photons at the fundamental frequency and generation of one photon at the second harmonic, so that the phase-matching requirement is equivalent to conservation of the photon momentum $\hbar k$ within the Heisenberg uncertainty limit in a medium of

length L. Similar considerations apply to the more general case of generating sum and difference frequencies.

A characteristic length for nonlinear second-harmonic generation, L_{NL}, is given by $\lambda n_\omega / E_\omega \chi_2$, where λ and E_ω are the wavelength and amplitude of the pump radiation. For a phase-matched interaction in which the pump remains undepleted, the conversion efficiency $I_{2\omega}/I_\omega$ from pump to second harmonic (where I_ω and $I_{2\omega}$ are the intensities of the input and output waves) is $(L/\pi L_{NL})^2$. For a centimeter length of nonlinear medium with a second-order susceptibility χ_2 of 1 pm/V, one needs pump intensities on the order of 0.2 GW/cm^2 for efficient harmonic generation.

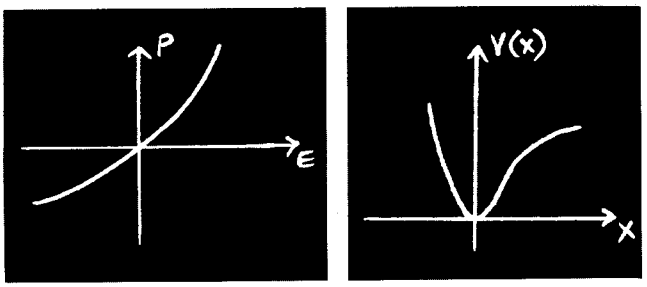
A measure of the phase-velocity mismatch is the coherence length $L_c = \lambda/4[n(2\omega) - n(\omega)]$, the distance over which the relative phase of the interacting waves shifts by π. In an interaction that is not phase matched, the efficiency is reduced by a factor $\sin^2(\pi L/2L_c)/(\pi L/2L_c)$. For a typical coherence length of 5 μm, that's a reduction of 10^{-6} in a 1-cm-long medium. For minimal reduction of the conversion efficiency, one needs $L_c > L$. That requires matching the refractive indexes to a part in 10^5 for L on the order of 1 cm. For typical media the indexes differ by about 10% between the fundamental and second-harmonic frequencies. So phase matching does not occur without special steps.

Franken's first demonstration of nonlinear optical frequency conversion in 1961 employed a quartz crystal to double the frequency of 694-nm light from a ruby laser.[1] Because that interaction was not phase matched, the ultraviolet output power was so small that the editors at *Physical Review Letters* mistook for a blemish the spot on Franken's spectrograph plate that demonstrated the new effect. They airbrushed it out of the published version, rendering the first evidence of nonlinear frequency conversion truly invisible. Subsequent progress in the field was rapid. By the end of 1962 the classic paper[3] had appeared in which Nicolaas Bloembergen and coworkers gave the theoretical underpinnings for both the microscopic origins of the nonlinear susceptibilities and the propagation effects governing macroscopic nonlinear interactions between electromagnetic waves. (See the article by Bloembergen in PHYSICS TODAY, October 1993, page 28.)

Birefringent phase matching

An essential step toward efficient frequency conversion, taken in 1962, was the use of birefringent phase matching in second-harmonic generation.[4] In uniaxially birefringent crystals the two orthogonal polarization eigenmodes, known as the ordinary and the extraordinary waves, in general have different phase velocities, c/n_o and c/n_e. The velocity of the extraordinary wave depends also on the propagation direction. It varies continuously between the two extremal eigenmode velocities. In crystals with an appropriate balance of birefringence and dispersion, one can achieve phase matching by choosing the propagation direction such that the phase velocity of one polarization mode at the second-harmonic frequency equals that of the other polarization mode at the fundamental frequency.

An important consideration in such "angle phase-matching" schemes is that the dependence of refractive index on propagation direction used to tune the phase matching also limits the angular acceptance of the process.



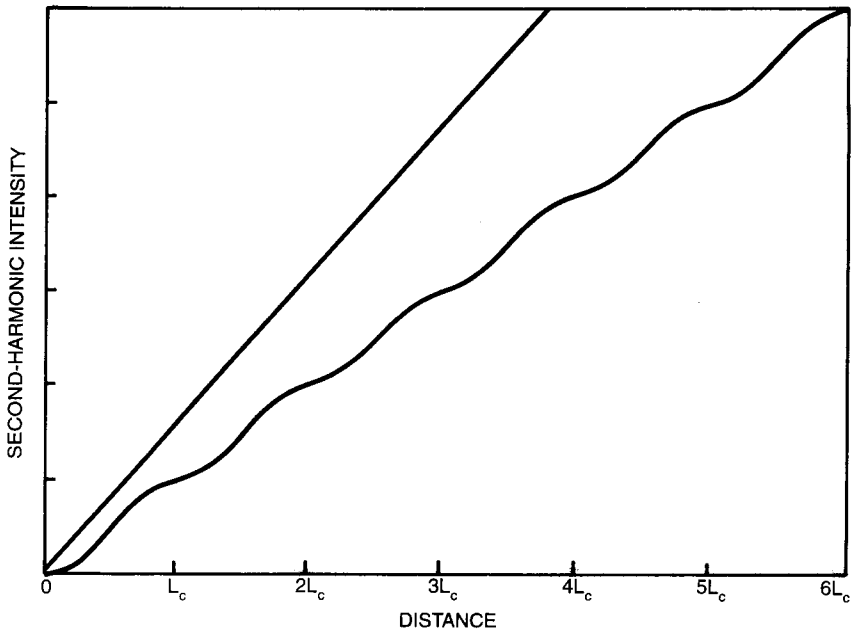
Nonlinear dependence of polarization density P in a crystal on the imposed electric field E (left) can be viewed classically as the forced response of electrons moving in the crystal's anharmonic interatomic potential wells (right). When the imposed field is laser light passing through the crystal, this nonlinear susceptibility can generate harmonics of the laser frequency. **Figure 2**

With a 1-cm-long crystal the acceptance angle is typically only a milliradian. That makes it necessary to use a pump laser with high spatial coherence. In the special case where the phase-matching direction is perpendicular to the optical axis, there is no first-order dependence of the extraordinary refractive index n_e on the propagation direction. Therefore the angular acceptance is much larger, typically 20 milliradians in a 1-cm-long crystal. Such "noncritically phase matched" interactions offer significant practical advantages over the general angle-phase-matched interactions, but appropriate media are even more difficult to find. Despite the severe constraints it imposes on the selection of materials, birefringent phase matching is employed in the vast majority of nonlinear frequency conversion devices with bulk crystals.

Early nonlinear optical experiments

By 1965 all the basic nonlinear optical frequency conversion phenomena—second-harmonic generation, sum and difference frequencies, optical rectification, and parametric amplification and oscillation—had been demonstrated. Practical applications, however, were hampered by the limitations of available pump lasers and nonlinear materials. Over the next ten years the inefficient, low-coherence ruby lasers of the earliest frequency conversion demonstrations were supplanted by newly developed laser systems. Notable among these were the lamp-pumped 1-micron infrared lasers based on Nd^{3+} ions in glass or crystal hosts. By the mid-1970s, 1-μm lasers with peak power in excess of 10 MW and beam quality close to the diffraction limit were readily available.

The development of improved nonlinear materials[5] paralleled the evolution of improved pump lasers. The nonlinear medium must meet several basic requirements. A reasonably large nonlinear susceptibility is, of course, a prime consideration, but adequate birefringence for phase matching is equally important. Particularly attractive are media that can be temperature-tuned to deliver noncritical phase matching at the desired wavelength. Because second-order susceptibility must vanish in inversion-symmetric media, almost all practical frequency conversion is done with single crystals that lack inversion symmetry. Growth of crystals in cubic-centimeter volumes with refractive index inhomogeneities smaller than a part in 10^{-5}, as required for phase matching, is generally difficult. Therefore the early frequency conversion experiments took advantage of available crystals such as am-



Growth of second-harmonic radiation in nonlinear media with perfect phase matching (red), with no phase matching (black) and with quasi-phase-matching (blue). In the third, quasi-phase-matched case, the sign of the medium's linear susceptibility is reversed after each coherence length L_c (blue shading) to compensate for dispersion. **Figure 3**

monium dihydrogen phosphate (ADP) and potassium dihydrogen phosphate (KDP). These crystals, developed for their piezoelectric properties (primarily as acoustic transducers in sonar applications), fortuitously had birefringence adequate for phase matching second-harmonic generation of 1-μm radiation. Extensive materials research over the next decade yielded several other suitable nonlinear crystals, including some still in use today: For example, two ferroelectric niobates, $LiNbO_3$ and $Ba_2NaNb_5O_{15}$, both have nonlinear susceptibilities an order of magnitude larger than that of KDP, and they can be noncritically phase matched to double the frequency of 1-μm radiation. Lithium iodate, another of the new materials of that generation, could be phase matched to wavelengths as short as 380 nm.

By 1975 several commercial nonlinear optical devices had appeared: Q-switched Nd lasers with KDP crystals generating second, third and fourth harmonics at the level of a hundred millijoules per pulse; continuous-mode Nd lasers producing green light by intracavity second-harmonic generation in barium sodium niobate; and even the first commercial optical parametric oscillator, a nonlinear lithium niobate device pumped by the second harmonic of a Q-switched Nd laser.

The essential ideas of nonlinear optical frequency conversion had been developed by the mid-1960s and commercial applications were well established by the mid-1970s. Let us now discuss some of the recent progress in materials and pump lasers that has fueled the resurgence of academic and industrial interest over the last decade, and some of the device applications made possible by these advances.

Pump lasers

Flashlamp-pumped Nd lasers remain the most widely used pumps for nonlinear devices. Improvement in the spatial and temporal coherence of these lasers has been essential to the development of more sophisticated nonlinear sources, especially optical parametric oscillators. A variety of other lasers—argon, dye, excimer, CO_2, copper vapor, color center, helium–cadmium, helium–neon, semiconductor diode and Ti:sapphire lasers, for example—have achieved widespread use in research, industrial and commercial applications. While almost all of these laser types have served as pumps for nonlinear optical devices, the Ti:sapphire lasers, diode lasers and solid-state lasers pumped by diode lasers stand out as particularly important for recent developments.

High-quality, room temperature diode lasers are commercially available with output wavelengths from 0.65 to 2 microns. Single-mode outputs of 100 milliwatts are available at selected wavelengths in this range. Typical packaged devices are several cubic centimeters in volume, and their efficiencies[6] for conversion of electrical to optical power are typically 40%. These lasers can serve as useful inputs to nonlinear frequency convertors, and they also can be used to pump other solid-state lasers. Such diode-pumped solid-state lasers[7] can function as "brightness convertors," absorbing low-coherence radiation from multimode diode lasers and then emitting it in a single transverse mode. The temporal coherence of these lasers can also be extremely high: Commercial devices with kilohertz linewidths are readily available. Rare-earth-activated host crystals such as Nd:YAG, for example, can produce outputs at various discrete infrared wavelengths between 1 and 2 microns. The energy storage possible in such media also makes it possible to convert continuous radiation from diode pump lasers into high-peak-power Q-switched nanosecond pulses.

A third major development in laser technology has been the emergence of broadly tunable solid-state lasers[8] such as $Ti:Al_2O_3$, whose output can be tuned from 0.7 to 1.06 μm. The large gain bandwidth of these lasers also makes it possible to generate mode-locked pulses as short as 10 femtoseconds.

All these lasers operate in the near infrared; similar performance has not been possible farther out in the infrared or at visible and ultraviolet wavelengths. There is therefore considerable interest in using nonlinear optical methods to transfer the outputs of these near-infrared lasers to other wavelengths so that all-solid-state sources of coherent radiation will be available from the ultraviolet to the far infrared.

Materials

As increasingly sophisticated frequency conversion applications require operation at higher efficiencies over a broader range of peak and average powers and over

extended spectral ranges, the demands on nonlinear materials place increasing emphasis on parameters besides birefringence and nonlinear susceptibility.[9] Notable among these are low absorption and scatter losses, high surface-damage threshold, high thermal conductivity, low thermo-optic coefficients and environmental stability. Another, often crucial issue is ease of crystal growth and processing. The continued importance of KDP, a nonlinear material that is rather undistinguished except for the size and quality of the crystals that can readily be grown in low-temperature aqueous solution, bears testimony to this point.

Among the materials to emerge as important for device applications in recent years are potassium niobate, which phase matches for second-harmonic generation into the blue and has twice the nonlinear susceptibility of $LiNbO_3$; potassium titanyl phosphate (KTP), which phase matches nearly noncritically for second-harmonic generation of the 1.064-micron output of Nd:YAG lasers; damage-resistant lithium and barium borates that can do phase-matched conversion into the ultraviolet and are transparent to wavelengths shorter than 200 nm; and several chalcopyrite crystals that are transparent and can be phase matched over much of the mid- and far-infrared spectrum.[9]

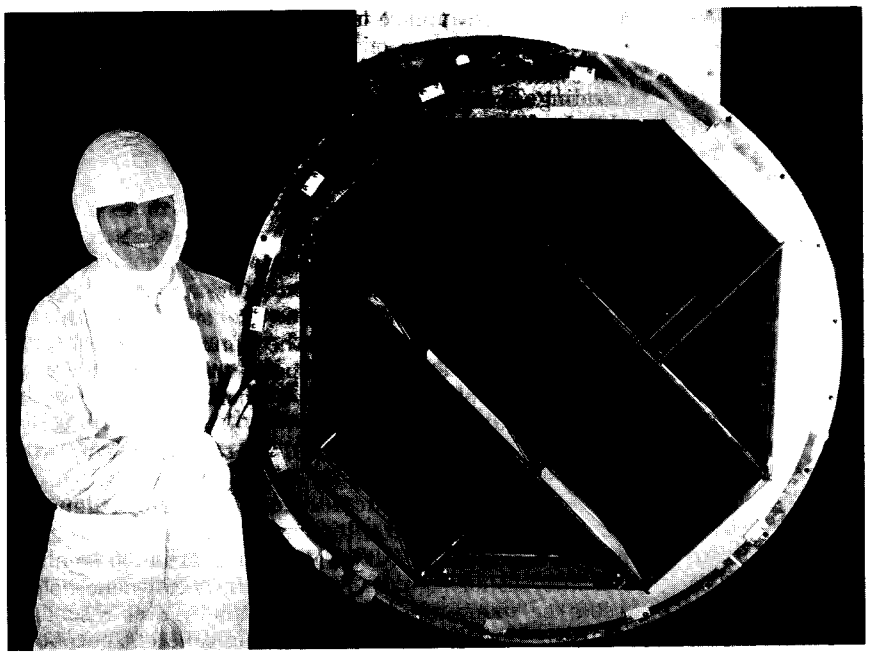
Organic nonlinear materials with outstanding promise for extremely large nonlinear susceptibilities, designed with molecular-engineering techniques, have long been plagued by difficulties with purification, crystallization, fabrication and environmental stability. (See the article by Anthony Garito, Rui Fang Shi and Marvin Wu on page 51.) A large research effort is addressing these problems, and recent developments in both single-crystal and polymeric media suggest that practical applications will grow rapidly in the future.[10]

Because important properties of nonlinear crystals, especially loss coefficients and surface-damage thresholds, tend not to be intrinsic to the materials, development of "new" materials often amounts to no more than improvements in the growth and processing of older materials. It is not uncommon for 10 or 15 years to pass between the time a crystal is shown to be potentially useful and the appearance of a viable material for device applications.

It is not possible to present here a comprehensive review of the many types of frequency conversion devices now in use. Reference 11 offers such a review. I will touch on a few results illustrating the range of performance parameters and experimental techniques being explored in several rapidly developing areas.

High-power pulsed harmonic generation

Harmonic generation of visible light from the infrared output of 1-μm lamp-pumped pulsed Nd lasers has been a common laboratory tool for 20 years, and it remains the most widespread application of nonlinear frequency conversion. KDP crystals are used to generate the second, third and fourth harmonics of 1-micron input radiation from various combinations of fundamental and second-harmonic beams. One routinely gets 10-nanosecond pulses with 100 megawatts of peak power and 10 W of average power. Assuming that adequate pump power is available, the major practical issues in such harmonic generation are surface damage and parasitic processes



Array of KDP crystals at Lawrence Livermore's Nova inertial-confinement fusion facility converts terawatt pulses of 1050-nm laser light to 350 nm to drive nuclear fusion in small targets. The array of nine single crystals provides the 75-cm aperture required to avoid damage in the terawatt beams. The KDP crystals have damage-resistant surfaces prepared by single-point diamond turning. (Courtesy of Lawrence Livermore National Laboratory.) **Figure 4**

such as self-focusing and two-photon absorption due to higher-order nonlinear effects.

An extreme example of these problems is found in the drivers for the large-scale inertial-confinement fusion experiments. These systems use intense pulses of ultraviolet radiation to compress small targets of hydrogen isotopes to densities high enough to initiate thermonuclear fusion. (See the article by John Lindl, Robert McCrory and Michael Campbell in PHYSICS TODAY, September 1992, page 32.)

The Nova system at the Lawrence Livermore National Laboratory, for example, generates 120-kilojoule, 2.5-nsec pulses of 1.05-μm radiation in an array of ten arc-lamp-pumped Nd:glass oscillator–amplifier systems. The system converts pulses to the third harmonic (350 nm) with an efficiency of about 70% by first generating the second harmonic and then summing its frequency with that of the primary beam.[12] To accommodate the 10-kJ pulse energies and terawatt peak powers in each of Nova's ten arms, the KDP crystals used for both frequency-mixing steps have apertures 75 cm wide and damage-resistant surfaces prepared by single-point diamond turning. Because single KDP crystals of that size were not available, techniques were developed for tiling an array of 27-cm crystals (as shown in figure 4) to present the requisite aperture. The National Ignition Facility, the proposed next step in inertial-confinement fusion, calls for the production of 1.8-*mega*joule pulses of 355-nm radiation, to be generated in 200 arms, each with a 34-cm-aperture KDP crystal. Improved growth technology nowadays makes it possible to grow significantly larger single crystals of KDP.

Continuous visible and ultraviolet sources

At the other extreme in frequency conversion technology are continuous-wave sources at visible and ultraviolet wavelengths. This regime is important for applications such as biomedical instrumentation, spectroscopy, graphics and optical data storage. At present, coherent sources in this part of the spectrum are based on gas-discharge lasers that are bulky, extremely inefficient and require high-voltage power supplies. There is considerable interest in replacing them with compact, solid-state sources.

The essential difficulty is that the nonlinear susceptibilities of available media are too small to achieve adequate conversion efficiency in a single pass. Resonant devices, where the nonlinear crystal is placed either in a Fabry–Perot cavity external to the pump laser[13] or inside the pump-laser cavity itself,[14] yield much larger conversion efficiencies, because of the enhancement of the radiation circulating inside the cavity. A well-designed Fabry–Perot resonator can increase conversion efficiency by a factor of 10^4, making possible efficient devices with pump powers as small as 50 milliwatts.[15]

The strong dependence of this resonant enhancement on the cavity losses makes low-loss crystals and careful cavity design essential. Although these resonant techniques were already demonstrated in the 1960s,[13] the low quality of first-generation nonlinear crystals and the lack of appropriate pump lasers prevented widespread application at that time. Devices are now commercially available that can generate more than 100 mW of 532-nm radiation by intra- or extracavity resonant second-harmonic generation with diode-pumped Nd:YAG lasers and either KTP or $MgO:LiNbO_3$ mixing crystals.[16]

An interesting variant of these techniques makes use of a monolithic ring Fabry–Perot resonator and total internal reflection to produce an extremely low-loss cavity that yields efficient second-harmonic generation of 1.06-μm beams with input power of less than a milliwatt.[17]

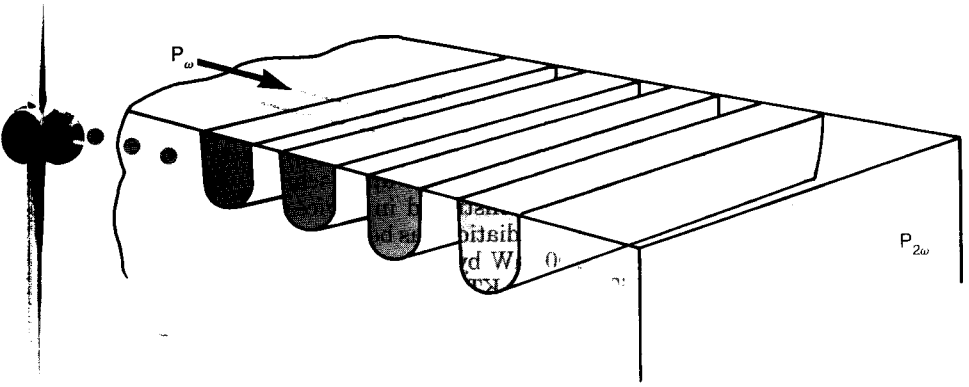
Optical parametric amplifiers and oscillators

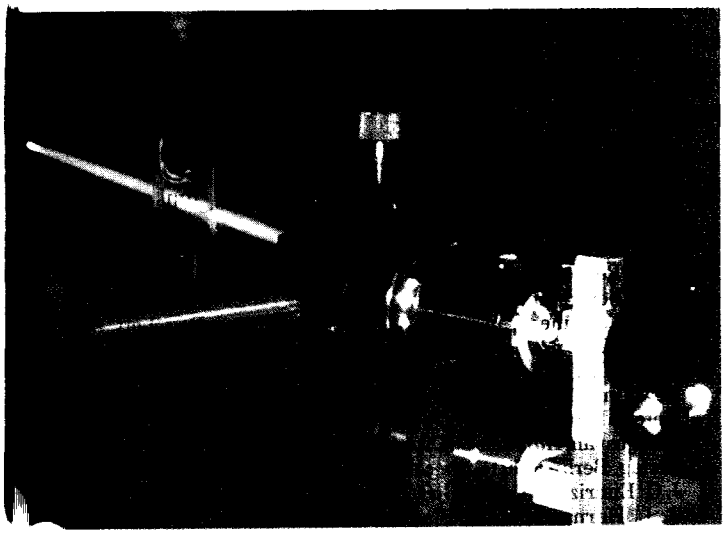
Tunable coherent radiation has long been used for high-resolution spectroscopy. Recently it has attracted renewed interest in connection with a variety of applications in combustion diagnostics, process control, remote sensing and environmental monitoring. Picosecond and femtosecond pulses of tunable radiation can be used for time-resolved studies of chemical reactions or carrier dynamics in semiconductors. Replacement of the widely used dye and color-center lasers with more convenient sources based on Ti:sapphire and diode-pumped solid-state lasers is therefore of considerable practical utility; it has become one of the most active areas of research in nonlinear frequency conversion.

While second-harmonic generation and sum-frequency mixing with Ti:sapphire lasers have become useful methods for the generation of visible and ultrafast ultraviolet pulses, and difference-frequency generation techniques are extending Ti:sapphire tunability further into the infrared,[11] I will focus here on a particularly active area of research: optical parametric oscillators.

In a difference frequency interaction, where a pump at frequency ω_3 mixes with a "signal" at ω_2 to generate an "idler" at $\omega_1 = \omega_3 - \omega_2$, conservation of energy dictates that for every photon created at ω_1, a pump photon is destroyed at ω_3 and another signal photon is created at frequency ω_2. Because mixing efficiency is proportional to the intensities of both the pump and the signal, the signal wave grows exponentially at the expense of the pump wave (until pump depletion sets in), a phenomenon known as parametric gain.

For a degenerate optical parametric amplifier ($\omega_1 = \omega_2 = \omega_3/2$), the small signal gain is numerically equal to the conversion efficiency for second-harmonic generation. So the gain can be as high as 1% per watt of pumping power in the visible. Although such gain is too small to be of interest for signal amplification, it can nonetheless exceed the round-trip losses in a Fabry–Perot resonator containing the gain medium, allowing coherent output to





Quasi-phase-matched waveguide frequency converter. Top: In this schematic diagram the orange periodic structures are ferroelectric domains of inverted nonlinear susceptibility, of which a typical device will have several hundred. The blue strip traversing the domains is a region of raised refractive index that forms the core of the dielectric waveguide along which the second-harmonic radiation is generated. Bottom: Quasi-phase-matched second-harmonic generation of 532-nm radiation in a periodically pole-reversed $LiNbO_3$ waveguide. This early device uses a planar waveguide and prism coupling. (Courtesy of Eric Lim, Uniphase.) **Figure 5**

build up from a noise input.

Such a device, producing signal and idler outputs from a single pump wave, is known as an optical parametric oscillator. Because the wavelength for maximum gain depends on the phase-matching condition in the crystal, one can tune the output wavelengths by varying the angle or the temperature of the nonlinear medium.

If only the signal wave is resonated in the cavity, the device is known as a singly resonant oscillator. In that case the threshold condition is that the parametric gain exceed twice the signal losses. For losses of 1% in the cavity, typical threshold power is on the order of 5 W. If both the signal and the idler are resonated, the device is known as a doubly resonant oscillator and the threshold condition requires only that the gain exceed the product of the signal and idler losses. The threshold power is thus lowered to tens of milliwatts, at the cost of significantly complicating the tuning behavior of the oscillator.

Thresholds for nanosecond operation are higher than for continuous operation. That can result in crystal surface damage before the threshold intensity is reached. For picosecond and femtosecond pulses, synchronous pumping (matching the round-trip time of the pulse in the resonator to the repetition rate of the pump laser) is necessary to allow buildup of the oscillation. At the high pump intensities attainable with short pulses (typically greater than 10 GW/cm^2), the parametric gain can approach e^{10} or even e^{20}, so that amplification of quantum noise without a resonator, as shown in figure 1, can generate outputs comparable to the pump intensity.

The broad gain bandwidths and efficient power conversion characteristic of optical parametric oscillators make them attractive alternatives to lasers in many applications requiring broad tunability or short pulses. The first pulsed and continuous-mode optical parametric oscillators[18] were demonstrated, respectively, in 1965 and 1968. By 1971 a commercial $LiNbO_3$ oscillator pumped with a Nd:YAG laser had become available. The early work is reviewed in reference 19.

Reference 20 covers many of the important recent results. The cover of this issue is a superposition photo showing outputs at various frequencies from a nanosecond-pulsed barium borate optical parametric oscillator pumped by the 355-nm third-harmonic output of a Nd:YAG laser. With a single input frequency such devices are continuously tunable from 400 nm to 2.2 microns in the infrared. One tunes the output frequency by varying the propagation direction of the input beam to alter the phase-matching condition in the barium borate crystal. $AgGaSe_2$ optical parametric oscillators pumped at 2.2 microns can be tuned from 2.5 to 12 microns, and a commercial lithium borate oscillator pumped by a Ti:sapphire laser produces near-infrared output pulses shorter than 130 femtoseconds.

Quasi-phase-matching and microstructured media

The essential difficulty in a non-phase-matched interaction is the difference in the phase velocities between the nonlinear polarization and the output wave it generates, which produces a phase shift of π over every coherence length L_c, with a concomitant reversal of the energy flow between the waves. The power can be made to flow continuously into the output wave by introducing a periodic change in the sign of the medium's nonlinear susceptibility that periodically resets the phase of the polarization wave by π, with half-period equal to the coherence length.[3] (See figure 3.)

This "quasi-phase-matching" scheme can significantly

extend the utility of a single material, because it does not rely on birefringence and can therefore operate noncritically over the material's entire transparency range. In addition one can use quasi-phase-matching with pump and output waves polarized parallel to one another so that they couple via the diagonal components of the nonlinear susceptibility tensor. These diagonal components are often quite large, but they are inaccessible to birefringently phase-matched interactions. Reference 21 is a review of quasi-phase-matched harmonic generation.

The difficulty in implementing this scheme lies in creating a medium with the requisite sign reversals every coherence length—typically 1–100 microns. Early work for infrared interactions involved stacks of thin crystal plates, each rotated 180° with respect to the one before, which proved to be difficult to handle and excessively lossy. Periodically perturbing the growth of ferroelectric crystals can controllably reverse domain orientations, thereby patterning the nonlinear susceptibility. (See figure 5.) Crystals of adequate quality to generate nearly 2 watts of continuous 532-nm radiation have been produced by this method. (See the photo on page 23 in Elsa Garmire's introduction to this special issue.)

Periodic in-diffusion of dopants by means of a patterned mask or application of periodic fields with a periodic electrode provides precise, lithographically controlled methods for creating quasi-phase-matched structures of alternating polarity at the surfaces of ferroelectric substrates. Such surface domain gratings are used primarily in waveguide devices, which will be discussed in the next section.

Quasi-phase-matching opens the search for better nonlinear media to new classes of materials, such as poled-polymer and fused-silica films, diffusion-bonded stacks of plates, laterally patterned semiconductors and asymmetric quantum wells. It is likely that these techniques will lead to significant progress in the performance of existing device types, as well as new devices that wouldn't be possible with conventional media.

Waveguide frequency conversion

The efficiency of single-pass bulk interactions is limited by the the trade-off between tight focusing for the sake of high intensities and loose focusing for the sake of large effective interaction lengths. Dielectric waveguides do away with the need for such tradeoffs: Total internal reflection tightly confines the radiation to the vicinity of the waveguide's axis, and waveguides only 1 cm long can yield single-pass efficiencies two to three orders of magnitude larger than one gets from bulk interactions in devices of that size. Thus one can achieve efficient nonresonant frequency conversion with milliwatts of pump power. Waveguide frequency conversion was first demonstrated in 1969, by P. K. Tien.[22]

Waveguide fabrication requires the creation of a core region of high refractive index surrounded by a low-index cladding. In the oxide crystals widely used for frequency conversion this is typically accomplished by in-diffusion of a dopant through a lithographic mask. Depending on the size and refractive index change of the core region, one or more modes are supported at a given wavelength.[23]

Early phase-matching work in waveguides focused on birefringent interactions. That work demonstrated second-harmonic generation, difference-frequency generation and parametric oscillation. But the material requirements for birefringent phase matching, compounded by the requirements of waveguide technology and lithography, severely limited the practicality of these early devices.[23] Promising results are now being obtained in the so-called Čerenkov configuration, where the second-harmonic radiation propagates at an angle to the waveguide axis such that the axial velocity component matches that of the guided fundamental wave.[24]

The advent of lithographic quasi-phase-matching techniques and compact diode-laser pumps has stimulated much of the recent work on waveguide frequency conversion.[25] Conversion efficiencies approaching 1% per milliwatt have been demonstrated in devices 1 cm long, and 10–20 mW of blue radiation has been generated with pump powers below 100 mW by second-harmonic generation in $LiNbO_3$, $LiTaO_3$ and KTP waveguides. One can readily adapt this technology to a variety of interactions simply by changing the lithographic masks. It has already yielded second-harmonic generation of wavelengths as short as 365 nm and difference-frequency generation of infrared wavelengths from 1.3 to 3 μm. For the near future it holds out the promise of high-gain parametric amplification and continuous-mode parametric oscillation.

References

1. P. A. Franken, A. E. Hill, C. W. Peters, G. Weinreich, Phys. Rev. Lett. **7**, 118 (1961).
2. Y. R. Shen, *The Principles of Nonlinear Optics*, Wiley, New York (1984).
3. J. A. Armstrong, N. Bloembergen, J. Ducuing, P. S. Pershan, Phys. Rev. **127**, 1918 (1962).
4. J. A. Giordmaine, Phys. Rev. Lett. **8**, 19 (1962). P. D. Maker, R. W. Terhune, M. Nisenoff, C. M. Savage, Phys. Rev. Lett. **8**, 21 (1962).
5. F. Zernike, J. E. Midwinter, *Applied Nonlinear Optics*, Wiley, New York (1973), contains interesting details of early nonlinear materials research. V. G. Dmitriev, G. G. Gurzadyan, D. N. Nikogosyan, *Handbook of Nonlinear Optical Crystals*, Springer-Verlag, Berlin (1991), is a comprehensive current reference.
6. J. G. Endriz, M. Vakili, G. S. Browder, M. DeVito, J. M. Haden, G. L. Harnagel, W. E. Plano, M. Sakamoto, D. F. Welch, S. Willing, D. P. Worland, H. C. Yao, IEEE J. Quantum Electron. **28**, 952 (1992).
7. D. W. Hughes, J. R. M. Barr, J. Physics D **25**, 563 (1992).
8. IEEE J. Quantum Electron.**28**(10) (1992)ia a special issue on ultrafast optics.
9. P. F. Bordui, M. M. Fejer, Annu. Rev. Mater. Sci. **23**, 321 (1993).
10. J. Zyss, *Molecular Nonlinear Optics: Materials, Devices, and Physics*, Academic, Boston (1994).
11. U. Simon, F.K. Tittel, *Methods of Experimental Physics*, vol. III, R. G. Hulet, F. B. Dunning, eds., Academic, Boston (1994).
12. P. J. Wegener, M. A. Henesian, D. R. Speck, C. Bibeau, R. B. Ehrlich, C. W. Laumann, J. K. Lawson, T. L. Weiland, Appl. Opt. **31**, 6414 (1992).
13. A. Ashkin, G. D. Boyd, J. M. Dziedzic, IEEE J. Quantum Electron. **2**, 109 (1966).
14. R. Smith, IEEE J. Quantum Electron. **6**, 215 (1970).
15. W. J. Kozlovsky, C. D. Nabors, R. L. Byer, IEEE J. Quantum Electron. **24**, 913 (1988).
16. M. A. Persaud, J. M. Tolchard, A. I. Ferguson, IEEE J. Quantum Electron. **6**, 1253 (1990).
17. S. Schiller, R. L. Byer, J. Opt. Soc. Am. B **10**, 1696 (1993).
18. J. A. Giordmaine, R. C. Miller, Phys. Rev. Lett. **14**, 973 (1965). R. G. Smith, J. E. Geusic, H. J. Levinstein, J. J. Rubin, S. Singh, L. G. van Uitert, Appl. Phys. Lett. **12**, 308 (1968).
19. R. L. Byer, in *Nonlinear Optics*, P. G. Harper, B. S. Wherrett, eds., Academic, San Francisco (1977), ch. 2.
20. C. L. Tang, W. R. Bosenberger, T. Ukachi, R. J. Lane, L. K. Cheng, Proc. IEEE **80**, 365 (1992). Two recent special issues on optical parametric oscillators are J. Opt. Sci. Am. **10**(9) and **10**(11) (1993).
21. M. M. Fejer, G. A. Magel, D. H. Jundt, R. L. Byer, IEEE J. Quantum. Electron. **28**, 2631 (1992).
22. P. K. Tien, R. Uhlrich, R. J. Martin, Appl. Phys. Lett. **17**, 447 (1970).
23. G. I. Stegeman, C. T. Seaton, J. Appl. Phys. **58**, R57 (1985).
24. H. Tamada, IEEE J. Quantum Electron. **27**, 502 (1991).
25. M. M. Fejer, in *Guided Wave Nonlinear Optics*, D. B. Ostrowsky, R. Reinisch, eds., Kluwer, Dordrecht, The Netherlands (1992), p. 133. ■