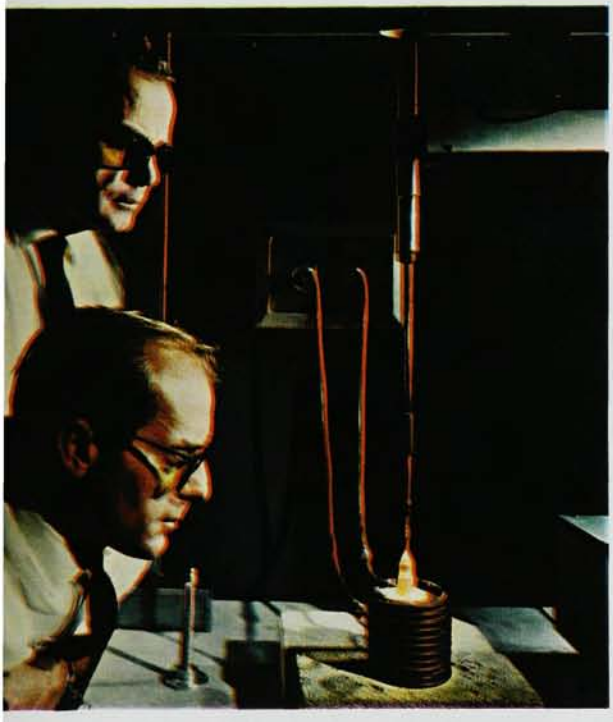
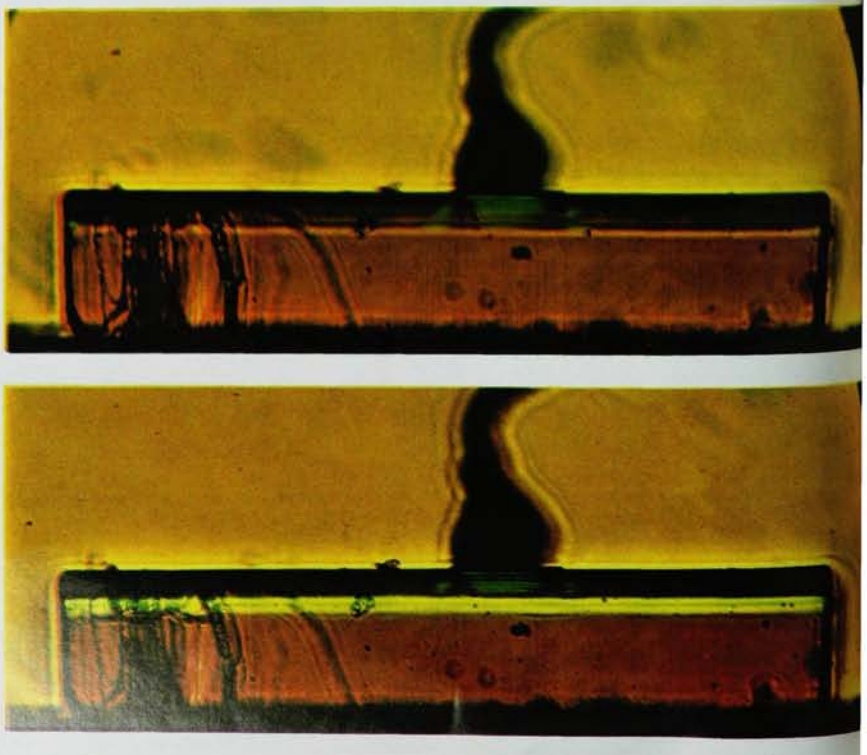
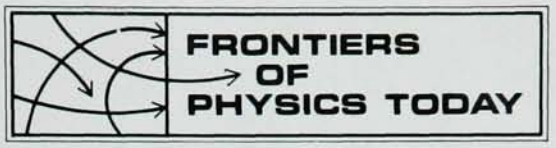
RED-TO-GREEN CONVERSION (above). Infrared laser light (1.06 microns) changes to green second-harmonic light (0.53 micron) in a crystal of barium sodium niobate. This photograph was made with the aid of a card treated with infrared-sensitive phosphor. In work by J. E. Geusic, H. J. Levinstein, S. Singh, R. G. Smith and L. G. Van Uitert at Bell Telephone Laboratories a similar crystal mounted in a 1-watt 1.06-micron continuous-wave laser has produced 1 watt of continuous 0.53-micron light.



CZOCHRALSKI GROWTH (left) occurs as nonlinear optical crystal emerges from 1500°C melt. L. G. Van Uitert (standing) and A. A. Ballman watch. Nonlinear materials are commanding much attention.



LIGHT MODULATION in gallium-phosphide p-n junction modulator diode (right) developed by F. K. Reinhart and D. F. Nelson. Green light is focused on the junction region, which is viewed between 45-deg crossed polarizers. In upper picture no voltage is applied to junction and diode is biased "off." In lower picture about 30 volts applied to the junction produces a half-wave phase shift through the Pockels effect, and green light is transmitted. Thickness of this diode is about 0.2 mm.



This review starts a new series that experts will write for PHYSICS TODAY. The series is inspired by the success of our 20th-anniversary issue last May, which reviewed the past 20 years in 13 brief, broad, qualitative articles. The new series will continue to survey the present and near future in the same manner. —*The Editors*

NONLINEAR OPTICS

Intense, coherent laser beams permit study of media polarized as the square or third power of field strength. Phenomena include harmonic generation, modulation, stimulated scattering and self-focusing. Newly available tools utilize picosecond optical pulses.

J. A. GIORDMAINE

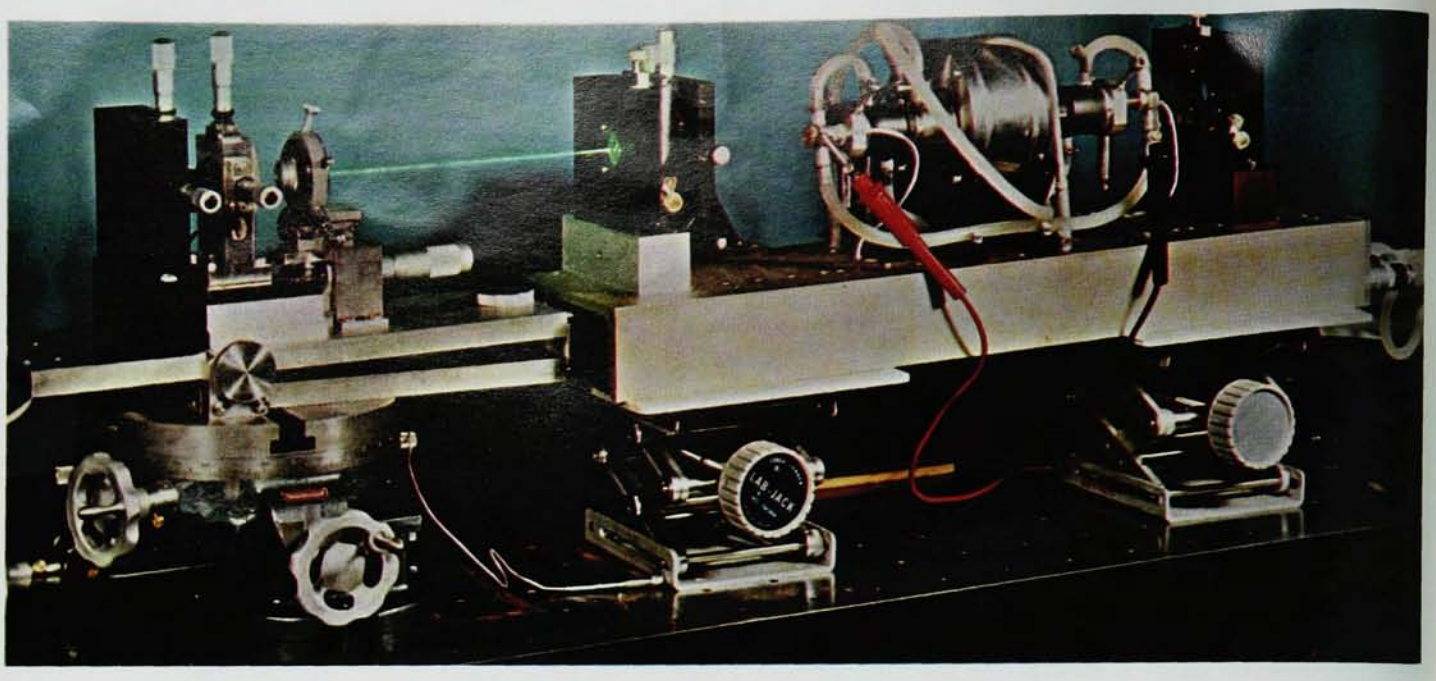
IN THE TEN YEARS since the invention of the laser, there has occurred a vigorous expansion of research and development in optics comparable with the growth of nuclear physics following the invention of the cyclotron. The impact of the laser on optical physics can be seen not only in the flowering of such previously established fields as inelastic light scattering from excitations in solids, holography, and optical coherence and photon statistics, but also in the birth of entirely new fields such as nonlinear optics, the present subject.

With nonlinear optical techniques we can generate harmonics of light frequencies and study new phenomena such as stimulated Raman scattering and self-induced transparency. Inevitably such studies have initiated a search for materials in which such effects occur strongly. We are learning to find such materials and also to calculate and predict their properties. Applications are arising, for example the study of molecules and solids by nonlinear scattering: in some instances information we can not obtain with infrared absorption and Raman scattering becomes accessible through stimulated scattering. Of course some new and unexpected phenomena are turning up also, among them self-focusing of intense beams and the observation of picosecond pulses. In the future some of these effects in nonlinear optics may lead us to new knowledge of astronomy and quantum electrodynamics.

Harmonic generation of light

Phenomenologically, nonlinear optics describes optical effects connected with the small changes of dielectric polarizability or refractive index with electric and magnetic fields. It therefore includes the familiar Pockels and Kerr effects, which arise respectively from refractive-index contributions linear and quadratic in a constant or radio-frequency electric field. The extension of these effects to higher frequencies began in 1961 with the discovery by Peter Franken and his colleagues at the University of Michigan. They found that a minute amount of the second harmonic of 694.3-nanometer ruby-laser light could be generated by focusing the beam in crystalline quartz. The second-harmonic light is radiated from the dielectric polarization component quadratic in the optical electric field.

The descriptions of almost all the classical optical phenomena such as reflection, total internal reflection, refraction, diffraction and birefringence have been generalized to include their newly observed nonlinear optical counterparts. These nonlinear phenomena display novel features not previously familiar in optics, such as the dominating influence of the dispersion of the refractive index. In second-harmonic generation for example, the harmonic intensity is determined by the coherence length over which the fundamental and harmonic light can travel together in phase, a length usually limited by dispersion to about 10 microns. For this reason, efficient optical harmonic generation usually requires birefringent crystals in which the fundamental and harmonic can be polarized in different directions, allowing the birefringence to compensate exactly for dispersion and providing coherence lengths as long as several centimeters. Early second-harmonic experiments made use of kilowatt and megawatt pulsed lasers and achieved conversion efficiencies of a few per cent. Recently, as the result of the successful development of highly nonlinear materials and a better understanding of the



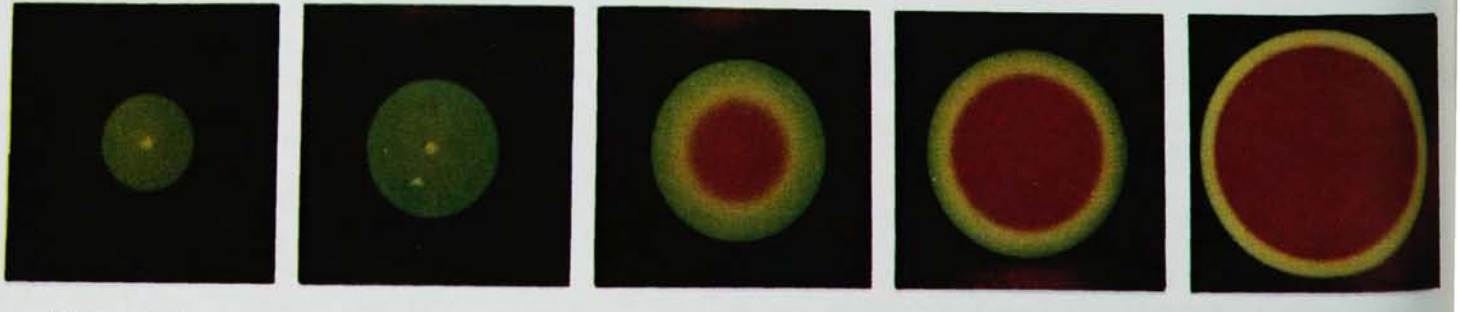
CONTINUOUS OSCILLATION in an optical parametric oscillator developed at Bell Labs by Smith, Geusic, Levinstein, J. J. Rubin, Singh and Van Uitert. Pump light at 0.53 micron is generated in a barium-sodium-niobate crystal mounted in the 1.06-micron Nd:YAG laser at right. Green pump light (visible in photograph) enters parametric oscillator resonator at left. Continuously tunable near-infrared light, not visible in photograph, emerges at left. Nonlinear polarizability mixes pump and signal light.

interaction of focused beams, continuous generation of 1 watt of 532.3-nm harmonic light has been achieved from a crystal of barium sodium niobate mounted within a 1-watt 1.06-micron laser. The usefulness of optical harmonics as ultraviolet sources is shown by the quasi-continuous generation of 0.5-watt harmonic light at 257.3 nm from the fundamental frequency of an argon laser.

Nonlinear materials

The successful search for improved nonlinear optical materials and a better understanding of the physics of the nonlinearities have been the key to the efficient continuous harmonic light sources now available. An important step was the discovery in 1964 that nonlinear polarizability is to a good approximation simply proportional to the cube of linear polarizability. This rule, which is now understood for the case in which the optical properties can be accounted for by a single effective oscillator, illustrates the strong dependence of nonlinear optical properties on refractive index and provides a guide in the search for new materials. For example, tellurium metal, with a refractive index at 10.6 microns about four times higher than that of quartz, has a nonlinear polarizability 10^4 times higher. Useful nonlinear optical crystals need to display the desirable and somewhat improbable properties of asymmetry (because the third-rank tensor nonlinear susceptibility is symmetry-forbidden in centrosymmetric crystals), high refractive index (associated with high nonlinear polarizability), large birefringence, low susceptibility for optical damage (which appears as a refractive-index inhomogeneity and is thought to arise from internal electric fields of charge distributions accumulated through photoconductivity), wide spectral transmission and availability in large single crystals.

Certain groups of materials, such as the oxygen-octahedra ferroelectrics (including lithium niobate and barium sodium niobate) and the iodates (notably iodic acid) tend to favor these properties, and the challenging problems of growth of these crystals are



PARAMETRIC FLUORESCENCE emitted by a lithium-niobate crystal, observed at Stanford by S. E. Harris, M. K. Oshman and R. L. Byer. Crystal is irradiated by 0.4880-micron pump source and fluoresces in red, yellow and green with angular distributions shown in photograph. "Signal" light of a given wavelength is emitted on a conical surface, at angles allowing phase matching among the k-vectors of signal, idler and pump light. Phase-matching angle changes with temperature and wavelength.

receiving considerable attention. We can now measure the nonlinear polarizability of crystals in powder form without the necessity of single crystals, and already cataloged are the nonlinear properties of hundreds of naturally occurring and synthetic crystals.

Methods of calculation

Calculations of nonlinear optical polarizability have been made for a few materials, including III-V semi-conductors and oxygen-octahedra ferroelectrics. In the shell-model approach nonlinear polarizability is calculated from the third-order moment of the molecular bonding orbitals at each atomic site. The band-theory approach, on the other hand, relates the nonlinearity to Stark-like shifts in the interband transitions. Both methods at present rely on the dominant role played by particular orbitals or bands and allow calculations with 20 to 50% accuracy. Higher order (for example, third-harmonic) nonlinearities contain important contributions from conduction as well as bound electrons, and a nonparabolic conduction-band theory has successfully described conduction-electron optical mixing effects in III-V semiconductors at 10.6 microns. A wide variety of nonlinear effects has been studied at 10.6 microns in narrow-bandgap semiconductors; these effects include resonant enhancement of the third harmonic when its frequency coincides with the energy separation between Landau levels of the conduction and valence bands.

Although formal perturbation-theory expressions for nonlinear polarizability are not readily amenable to quantitative calculation, they do reveal a number of important symmetry relations among the various tensor components. For example, they show that the component describing the beating of two nearby optical frequencies to generate microwave difference frequencies or dc optical rectification is identical with the Pockels-effect coefficient. Many such symmetry predictions, which also follow from more general thermodynamic arguments, have been verified. It is interesting that optical-difference-frequency generation appears to provide a useful tunable source of far infrared and millimeter radiation.

Physical applications

Second-harmonic generation is being applied at present as a selective tool for the study of a variety of physical problems. For example, light scattering from noncentrosymmetric molecules in gas or liquid phase includes weak but detectable second-harmonic light and, in addition, components shifted from the second-harmonic frequency by the molecular vibrational frequencies. The selection rules for this "hyper-Raman" scattering differ from those of infrared absorption and Raman scattering and in particular allow the direct observation of "silent" modes that are forbidden in both infrared and Raman scattering.

A second application has been to the study of particular phase transitions in solids for which conventional (linear) light-scattering techniques are ineffective. In the order–disorder transition in ammonium chloride, for example, loss of the ordered orientation of ammonium ions NH_4^+ at 245 K is not marked by the usual refractive-index fluctuations or critical point opalescence since the linear polarizability of NH_4^+ has complete rotational symmetry. But the disordering does cause the disappearance of nonlinear polarizability, which is present below the critical temperature solely because of the ordering of the noncentrosymmetric NH_4^+ ions. Measurements of the temperature dependence of harmonic generation near the critical point have given a measure of the correlation length for the NH_4^+ ordering and, in addition, have revealed the existence of domains of differently oriented ions.

In liquids the lowest-order nonlinear polarizability is inherently connected with optical rotatory power and is a pseudoscalar property of molecules not superimposable on their mirror image. The nonlinearity, which disappears in a racemic (optically inactive) solution of left- and right-handed isomers, arises solely from asymmetry in the orbitals of asymmetric carbon atoms. The relative magnitudes of the nonlinear optical polarizability and the optical rotatory power give information on the mechanism (for example, single-electron or coupled-oscillator model) of optical rotatory power.

Optical modulation

A broad area of application of nonlinear optical effects is development of efficient broad-band light modulators for communications. The most useful modulator materials are crystals such as lithium tantalate, which are ferroelectric or "almost ferroelectric." Raman spectroscopy shows that the large electroöptic effect in these crystals arises primarily from the electric-field-induced lattice displacement, which in turn modifies the electronic polarizability and the refractive index. The lattice response is dominated by the same low-frequency "soft" modes that give rise to ferroelectricity and determine the linear low-frequency dielectric properties. Present lithium-tantalate optical modulators can produce 80% modulation of 632.8-nm light with a bandwidth of 220 MHz and a power input of only 0.2 watts.

A different and novel approach to the modulator problem exploits the large electric fields (about 10^6 V/cm) in reverse-biased semiconductor p–n junctions to produce a large Pockels effect with small applied voltage. Gallium-phosphide modulator diodes have achieved 80% modulation with 100-MHz bandwidth at 0.1-watt input.

Fast optical modulators now available make possible the manipulation and modification of light in novel ways. A striking example is their use in frequency shifting and compression of light pulses. In the frequency shifter, repetitive light pulses from a mode-locked helium–neon laser traverse a modulator crystal whose refractive index is swept back and forth in phase with the pulse repetition. When the pulses are passed through the crystal at times of maximum rate of change of refractive index, one obtains Doppler-like frequency shifts as large as 1.5 cm^{-1}. The optical pulse compressor

---

Joseph A. Giordmaine studied at the University of Toronto and at Columbia, where he received a PhD in 1960. After teaching at Columbia for two years, he joined the Bell Telephone Laboratories where he is now head of the Solid State Spectroscopy Research Department. He has worked on paramagnetic resonance, molecular beams, radio astronomy, maser and laser physics, and nonlinear optics. In 1966 he was a visiting professor at the Technische Hochschule in Munich.

makes use of the same crystal, but the pulses cross the crystal at times such that the leading portion of the pulse is upshifted and the trailing portion downshifted. These frequency-swept or "chirped" pulses are then passed through an interferometer having a frequency dispersion appropriate to delay the higher-frequency leading edge slightly longer than the low-frequency tail. The result is to compress the pulse to a width limited only by the spectral width imposed by the modulator. Helium–neon 0.5-nanosecond laser pulses have been compressed by a factor of 2 by this technique; even frequency-swept 5 picosecond (10^{-12} sec) pulses have been compressed to 0.4 psec!

Parametric oscillators

Optical parametric oscillators are broadly tunable sources of coherent light in the visible and near infrared. They use nonlinear polarizability in a way qualitatively different from that of harmonic generators and are being widely studied. As in traveling-wave microwave amplifiers, oscillator pump light is converted to lower-frequency signal and idler light whose frequencies sum to the pump frequency. A traveling packet of signal light beats with pump light through the nonlinear polarizability to produce idler difference frequency light; this light in turn beats with the pump light to produce a polarization wave at the signal frequency with just the proper phase to amplify the original packet.

It follows that the signal (and idler) see exponential gain; a suitable nonlinear crystal within an optical resonator will oscillate when the pump power provides enough gain to offset the surface transmission, scattering and other losses. This type of oscillator makes no use of discrete energy levels in the amplifying crystal and therefore is inherently broadly tunable. Actual frequency of maximal gain is that for which the correct relative phase relation for amplification among the three waves can be maintained over the length of the crystal—the same phase-matching requirement as for harmonic generation. This condition defines a spectral range for gain typically about 10 cm^{-1} wide, with center frequency continuously tunable by crystal rotation, temperature, electric field and other means of changing the refractive index of the crystal.

The first oscillators of this kind (1965) used lithium niobate, were pumped by 10-kW light pulses at 530 nm, were tunable over the range 680 nm to 2.3 microns and produced an output power of about 100 watts. Pulsed oscillators pumped at much higher powers have since attained conversion efficiencies as high as 50%. Recently, carefully designed barium-sodium-niobate and lithium-niobate oscillators pumped by green light have operated continuously with threshold power as low as 5 mW. Special precautions are necessary to avoid optical damage in these oscillators.

Although such oscillators have most of the characteristics of broadly tunable lasers, they differ, for example, in the requirement of modes simultaneously resonant at two frequencies whose sum is the pump frequency. Without special precautions oscillation tends to occur at one or more favorable frequency "clusters" close to the frequency of maximum gain, the spacing between clusters typically about 30 cm^{-1}. Current work in this field, in addition to materials research, is directed toward an understanding of the mode structure and the oscillation dynamics with pulsed pumping and toward developing a practical tunable source for spectroscopy and other applications. Recently megawatt traveling-wave optical oscillators have been developed in the Soviet Union; other oscillators have been reported at a variety of pump frequencies and with optical-resonator feedback at the signal frequency only—a mode of oscillation that eliminates the frequency-clustering effect and achieves a smoothly tunable output at the expense of somewhat higher threshold pump power.

Other aspects of tunable optical oscillators that are currently of interest are, first, the coherence properties of the emission, which are expected to differ from those of the laser, and, second, parametric fluorescence, the spontaneous emission of light at signal and idler frequencies when the pump intensity is below the threshold for oscillation. This fluorescence shows the same tuning characteristics as the parametric oscillator emission and is analogous to ordinary fluorescence from an atomic excited state. In a typical nonlinear crystal, about 10^{-12} of an incident light beam is converted to fluorescence at the phase-matched frequencies.

Stimulated scattering

A fruitful area of nonlinear optics has been a wide variety of stimulated scattering processes. Experimental work in this area began in 1962 with the discovery that liquids with relatively strong lines in their vibrational Raman spectra show gain, stimulated emission and oscillation at the frequency of the most intense Raman–Stokes scattering. This radiation was quickly recognized as the stimulated emission predicted 30 years earlier to accompany scattering processes in general. The probability of transfer of a photon from mode a of the field to mode b at the lower Stokes frequency, promoting a molecule from its ground state to an excited state of energy $h(\nu_a - \nu_b)$, is proportional to $\eta_a(\eta_b + 1)$ where η is the photon-mode occupation number. For spontaneous optical Raman scattering, we have for almost all b, $\eta_b = 0$, and the scattered intensity is proportional to the incident intensity. At kilowatt and megawatt pulse laser pump powers, η_b is greater than 1, and the rate of scattering becomes proportional to the scattered intensity itself. This is the condition for exponential gain and efficient conversion; indeed in the presence of an optical resonator or simply the small feedback due to scattering, oscillation can occur. The relation between spontaneous and stimulated scattering for the Raman case, as for the others mentioned below, is, of course, the same as given by the Einstein A and B coefficients for resonant emission.

In practice, it is much more useful to look at stimulated scattering processes classically as the parametric interaction of three coupled waves: the laser pump, the stimulated Stokes scattering and the molecular vibrational displacement wave serving as the idler. The quantum and classical views approach each other properly in the limit of intense light, provided one is careful to describe the fields in the "coherent-state" representation rather than by photon numbers as done above. In the coupled-wave view one predicts secondary scattering processes as the result of modulation of the refractive index by the vibrational displacement wave; these processes include both anti-Stokes and Stokes scattering shifted from the laser frequency by multiples of the molecular resonant frequency. The secondary scattering is usually constrained by phase matching to discrete angles, and the focusing of megawatt 694.3-nm ruby-laser radiation into liquids often results in spectacular yellow, orange and green anti-Stokes emission cones.

Two anomalies

Stimulated Raman gain is isotropic; highly collimated forward and backward stimulated emission is observed simply because the gain path is longest for these directions. In strongly Raman-active liquids the gain, which is proportional to the Raman scattering cross section, typically is about 1 cm^{-1} for laser light of 10^8 W/cm^2, and the conversion efficiency is usually between 1 and 10%. Since a gain of about e^{20} is required to amplify spontaneous Raman emission to the observed 10^5-watt levels, one might expect to need a 20-cm sample length to obtain substantial conversion. Surprisingly a much shorter length proves adequate, in part because of residual feedback but primarily because of the collapse of the incident light beam into self-trapped filaments of extreme power densities, with diameter of 5 to 50 microns. This self-focusing phenomenon, which was overlooked in the early work on stimulated scattering, is the result of molecular alignment in the intense optical electric fields, that is, the molecular Kerr effect. Above a certain threshold power the beam finds itself unstably coupled to its own dielectric waveguide and collapses. The apparent stimulated Raman gain anomaly now appears to be resolved, and careful observations of Raman gain in gases without self-focusing agree fairly well with theory.

A second apparent anomaly occurs in backward stimulated Raman scattering. Although the backward and forward gains are the same, the backward-traveling Raman light is continuously supplied with fresh laser pump light, while forward Raman light has access only to the pump light traveling in the same element of volume. It follows that forward Raman emission saturates while the backward emission can reach a peak power in excess of the incident laser power. Such backward pulses have been observed in carbon disulfide with a duration of 3×10^{-11} sec and peak power ten times the incident laser power.

A practical application of stimulated Raman scattering has been as a source of intense, pulsed monochromatic radiation at a wide variety of wavelengths with the use of a single laser. Other applications, as yet hardly explored, are suggested by the high level of excitation of the optical branch of lattice vibrations during stimulated Raman scattering. Infrared emission at 5 microns radiated from stimulated infrared-active vibrations in benzene has been observed; the study in solids of phonon interactions and lifetimes seems an attractive possibility. Lifetimes of excited vibrational states have already been measured in hydrogen gas excited by the stimulated Raman effect and observed by ordinary Raman scattering.

Some other processes

A number of other stimulated scattering processes have recently been studied. Stimulated Brillouin scattering, which is the acoustic-phonon analog of the stimulated Raman effect, occurs at a pump threshold below that of the latter in many materials, and the two processes often compete. In certain experiments 90% of incident laser light is converted to stimulated Brillouin light. Wave-vector conservation and beam geometry cause stimulated Brillouin light to be emitted primarily in the backward direction with frequency shifts typically between 10^9 and 10^{11} Hz. Precise determination of the frequency shifts gives a measurement of hypersonic acoustic velocity, and indirectly of acoustic attenuation, that is in some cases more accurate than from spontaneous Brillouin scattering. Acoustic phonons have been stimulated in liquid helium by this technique and their lifetime measured by light scattering of subsequent laser pulses of variable delay.

Stimulated Rayleigh wing scattering differs from the stimulated Raman and Brillouin processes in being associated with no discrete elementary excitation. To visualize this process consider the effect of two light waves of slightly different frequency on a liquid. Through the Kerr effect, the molecules experience a torque oscillating at the difference frequency of the light waves. The resulting molecular rotation leads to a periodic refractive-index variation at the difference frequency that couples the two waves and provides gain at the lower frequency. Maximal gain occurs when the rotation component out of phase with the torque is highest, that is, when the difference frequency corresponds to the Debye time τ for molecular orientation. Stimulated Rayleigh wing scattering is in fact observed with a frequency shift about $1/\tau$, typically 10^{10} to 10^{11} Hz, showing the expected temperature dependence and providing a direct measure of τ.

A related Rayleigh process, stimulated thermal Rayleigh scattering, occurs in liquids that absorb a part of the incident laser light. It arises from coupling between two light waves induced by thermally driven refractive-index oscillations due to *entropy* variation and is the stimulated analog of Rayleigh scattering. The frequency shifts are characteristically of the order of 10^8 to 10^9 Hz. Thermally driven *density* variations in absorbing liquids give rise to stimulated thermal Brillouin scattering at frequencies 10^8–10^9 Hz away from the usual Brillouin lines.

Other stimulated processes, including stimulated scattering from surface vibration modes (or "ripplons"), have been proposed but not observed.

Self-focusing of light

As I have suggested already, a dominant feature of high-power optical experiments is the self-focusing of the laser beam to form short-lived (up to about 10^{-9} sec) filaments. The primary mechanism of self-focusing is undoubtedly the Kerr effect; there is evidence, however, that electrostriction as well as molecular clustering from induced dipole–dipole interactions also play a role. The dynamics of self-focusing is being intensively studied, but a number of experimental results remain unexplained. Still not understood is the mechanism that limits the filament diameter to about 5 microns. It has been pointed out that filament formation has some of the features of a field-induced phase transition and vortex formation in type-II superconductors. The light power in small-scale filaments may exceed 10^{12} W/cm^2 with optical fields in excess of 10^8 V/cm.

As light propagates through a liquid having an intensity-dependent refractive index, not only is its transverse structure modified by self-focusing, but its pulse shape changes, and its spectrum broadens. Frequency broadening arises from several related effects, including stimulated Rayleigh and Brillouin scattering, development of a sharp trailing edge or shock wave as the more slowly traveling peak tends to lag behind the main body of the pulse and, perhaps most important, a pseudo-Doppler broadening due to the changing refractive index accompanying changes in the intensity of the pulse itself. The spectral broadening of a powerful pulse may exceed 2000 cm^{-1}. The intense broad-band light generated in this way has been mixed

with monochromatic light in a narrow-band mixer crystal to produce a useful tunable coherent light source.

Resonance phenomena

A third area of nonlinear-optics research is the resonant interaction of short light pulses with matter through the photon echo, self-induced transparency, transient nutation and related effects that are the optical analogs of their magnetic-resonance namesakes. These effects become important in resonant absorption of light pulses intense enough to excite all the absorbing atoms in a time short compared to the (homogeneous) relaxation time T_2, which is often the excited-state lifetime.

When the transition electric dipole moment excited by a resonant pump pulse decays as a result of dephasing of the individual atoms, a suitable interrogating pulse can initiate rephasing of the individual dipoles and subsequent radiation of an echo pulse. As in nuclear magnetic resonance, the measurement of echo intensity as a function of pulse spacing gives both T_2 and information on the line-broadening mechanism. Photon echo observations have been reported in ruby at 694.3 nm and in a sulfur-hexafluoride vibrational line at 10.6 microns.

A novel and related technique uses the self-induced transparency effect. It has been demonstrated in ruby and sulfur hexafluoride that short optical pulses can propagate without loss through a resonant absorber, provided that they have the proper shape and integrated intensity to excite transitions from the ground state through the excited state and back *precisely* to the ground state. A striking feature of self-induced propagation is the low velocity of the light pulse, about 10^8 cm/sec in ruby; this behavior is expected since such pulses are in fact only to a small degree electromagnetic, with most of the energy stored in the transition. Like the photon echo, self-induced transparency is a useful probe of line-broadening mechanisms and relaxation times.

Picosecond pulse measurements

Finally I would like to describe a recent and somewhat unanticipated application of nonlinear optical effects: measurement of fast light pulses. Conventional photodetectors of single-pulse events have a resolution of several hundred picoseconds and provide no information on the time dependence of picosecond-duration mode-locked laser light pulses. (However, image-converter cameras newly developed in the Soviet Union have achieved a resolution of 2–3 psec.) Early experiments were successful in obtaining indirect information on laser pulse widths by observing harmonic generation efficiency. Recently, however, optical pulse widths have been directly measured by mixing the pulses with delayed replicas of themselves in a harmonic-generator crystal. The intensity autocorrelation function is extracted simply by recording the harmonic-generator output, as observed with a conventional detector, as a function of delay. An even simpler technique, which also measures the intensity autocorrelation function, is based on two-photon fluorescence. The pulse to be measured divides at a beam splitter into two pulses, which pass each other in opposite directions in a suitable indicator medium. The indicator medium, usually a liquid, absorbs a small portion of the pulse energy solely by two-photon absorption and subsequently fluoresces. A photograph of the two-photon fluorescence display shows a background track together with a region of enhanced brightness that delineates where the two pulses cross one another. The length of the bright region is a measure of the pulse length and duration. This method has been applied to pulses as short as 0.4 psec, or about 100 optical cycles. It should be understood that purely "linear" spectroscopic methods such as the spectrometer provide only the spectral intensity distribution of a pulse, and with frequency modulation cannot be used to infer the pulse duration.

Fast nonlinear optical pulse techniques have already made possible direct measurement of 10^{-11}-sec lifetimes for nonradiative decay of excited states of organic molecules and the observation of molecular-orientation time effects in liquids. Picosecond pulses turn out to be too short to produce the usual Kerr intensity-dependent refractive index variation in many liquids; as a result they show little tendency to self-focus, and in fact tend to propagate through liquids with relatively little interaction. There appear to be applications of picosecond pulses in measurement of nonradiative electronic transition rates in solids, in the generation of ultrashort electrical and acoustic pulses, and for information storage.

Space and the vacuum

It will be apparent from these examples that the theme of much nonlinear optics research has been the extension into the optical region of coherent wave techniques developed at radio and microwave frequencies. There have been a number of surprises, however, such as self-focusing and picosecond-pulse effects, which are, of course, peculiar to optical frequencies. It can be predicted that many of these optical techniques can be extended to the ultraviolet and perhaps even to shorter wavelengths. Nonlinear optical effects may play an astrophysical role; it has been suggested that self-focusing and Stokes shifting may account for part of the astronomical red shift. The basic nonlinear properties of the vacuum predicted by quantum electrodynamics are not significant in optical experiments at even the highest presently available fields; they may become observable if present techniques can be extended one or two orders of magnitude in frequency and would appear, for example, as light-by-light scattering and perhaps self-focusing in the vacuum.

* * *

References

The following are recent reviews of nonlinear optics:

S. A. Akhmanov, R. V. Khokhlov, Usp. Fiz. Nauk **88,** 439 (1966), translated as Sov. Phys.–Usp. **9,** 210 (1966); Usp. Fiz. Nauk **95,** 231 (1968), translated as Sov. Phys.–Usp. **11.**

N. Bloembergen, *Nonlinear Optics,* W. A. Benjamin, Inc., New York (1965); Am. J. Phys. **11,** 989 (1967).

J. Ducuing, in Italian Physical Society proceedings of the International School of Physics Enrico Fermi, Course 42, Aug. 1967 (R. J. Glauber ed.), Academic Press, New York, 1968.

J. A. Giordmaine, in Italian Physical Society proceedings of the International School of Physics Enrico Fermi, Course 42, Aug. 1967 (R. J. Glauber ed.), Academic Press, New York, 1968.

P. Pershan, *Progress in Optics,* **5,** 85 (1966), (E. Wolf ed.), North Holland Pub. Co., Amsterdam (1966).

B. Stoicheff, *Italian Physical Society. International School of Physics Enrico Fermi,* Course 31, p. 306, Aug. 1963 (C. H. Townes, P. A. Miles eds.), Academic Press, New York (1964).

A. Yariv, *Quantum Electronics,* John P. Wiley and Sons, New York (1967) p. 340. □