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Resource Letter NO-1: Nonlinear Optics

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This Resource Letter provides a guide to the literature on nonlinear optics. Books, journals, and websites are introduced that cover the general subject. Journal articles and websites are cited covering the following topics: second-order nonlinearities in transparent media including second-harmonic generation and optical parametric oscillation, third-order and higher nonlinearities, nonlinear refractive index, absorptive nonlinearities such as saturable absorption and multiphoton absorption, and scattering nonlinearities such as stimulated Raman scattering and stimulated Brillouin scattering. Steady-state and transient phenomena, fiber optics, solitons, nonlinear wave mixing, optical phase conjugation, nonlinear spectroscopy, and multiphoton microscopy are all outlined.
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I. INTRODUCTION

Nonlinear optical phenomena occur when the optical properties of a medium depend on the intensity of coherent light traveling through it. The altered absorption or refractive index affects either the laser beam itself or other beams that probe the local environment. In linear optics, light traveling through media induces an oscillating polarization that is linear with its electromagnetic field. In nonlinear optics, coherent light can distort the oscillating polarization; Fourier components of these distortions can create or enhance new fields that then radiate from the oscillating nonlinear polarization.

Nonlinear optics has reached consumer application with the green laser pointer (Fig. 1), which consists of a nonlinear potassium titanyl phosphate (KTP) crystal that creates second-harmonic from a laser consisting of a neodymium-doped vanadate crystal that is pumped by a laser diode. The second-order optical nonlinearity converts the infrared 1064 nm Nd laser line to 532 nm wavelength, which is visible in the green.

A second consumer item based on nonlinear optics is shown in Fig. 2. The 3D display inside the glass block is produced by laser damage that is created by complex nonlinear optical processes.

Beyond enabling a few consumer products, nonlinear optics has been a fascinating field to explore in its own right. It has produced many interesting phenomena, some of which occur naturally and limit the laser power that can be used in practical applications. This is particularly true in optical-fiber communications where long lengths enable small nonlinear effects to build up over distances.

Geometries can be tailored so that the nonlinearities produce useful effects. Harmonic generation, already mentioned, can be generalized to include sum- and difference-frequency generation (DFG), with multiple harmonics extending all the way through the UV. Third-order nonlinearities produce a nonlinear refractive index that can lead to soliton pulses in time or spatial solitons, or even continuum wavelength generation from today's femtosecond lasers. Indeed, third-order nonlinearities are a key to effectively mode-locking femtosecond lasers. Four-wave mixing (FWM) in laser amplifiers enables frequency conversion used for wavelength division multiplexing in optical communications. Nonlinear absorption can create transparent material from initially absorbing materials or can create absorptive materials out of initially transparent materials (which can protect sensitive detectors by limiting how much optical power is transmitted). Stimulated Raman amplifiers and lasers create coherent light at new wavelengths, as do nonlinear optical parametric amplifiers and oscillators. Four-wave mixing and stimulated Brillouin scattering (SBS) both can produce phase-conjugate signals that enable wave-front correction.

Perhaps the greatest impact of nonlinear optics is in the science of spectroscopy, where new techniques have provided hitherto unheard of resolution, enabling new physics and chemistry. This is attested to by the five Nobel Prizes given for such research.

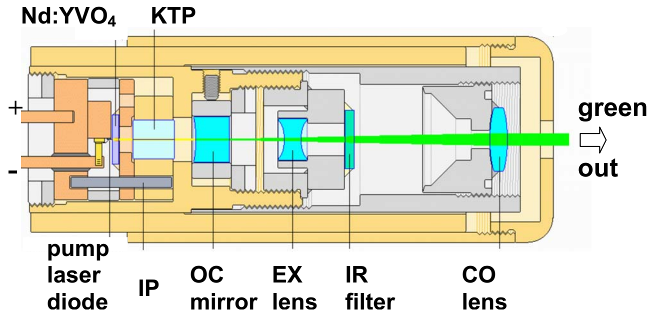

Fig. 1. The inside of a green laser pointer sold by Edmund Scientific as L54-101. Nd:YVO$_4$ is the neodymium vanadate laser crystal that emits infrared 1064 nm light when pumped by a laser diode emitting at 802 nm. The backsurface of the laser crystal is coated with a dielectric mirror that transmits the 802 nm pump light and reflects both 1064 and 532 nm lights to form one laser cavity mirror. KTP is the nonlinear crystal that converts the infrared laser light to green light at 532 nm. The KTP crystal is placed inside the laser cavity to enhance the light's electric field; OC is the output coupling mirror, IP is an indexing pin, EX is the beam-expanding lens, IR is an infrared filter that removes the 1064 nm light and CO is a beam-collimating lens. Drawing adapted from ⟨http://173-23-71-206.client.mchsi.com/repairfaq/sam/l54-101.gif⟩. Permission granted.

The nonlinear optics topics included here represent my views of what will be most interesting to students and teachers. Undoubtedly I am swayed by my own research interests and I apologize in advance for any important aspects I may have missed.

II. GENERAL INFORMATION ON NONLINEAR OPTICS

A. Journals

Papers on nonlinear optics are spread throughout many journals—no single journal in this field has built up enough clientele to have a significant impact factor. Breakthrough papers tend to appear in *Nature* and *Science*, with *Physical Review Letters* falling slightly behind. Another journal that initially published a large fraction of nonlinear optics papers was *Applied Physics Letters*, although fewer in recent times. Besides these, authors publish either in optics journals or in general-physics journals, the most important of which are listed below in order of impact factor, which roughly means in order of perceived research value.

Optics and lasers journals:

Optics Letters
Optics Express
IEEE Journal of Quantum Electronics
IEEE Journal of Selected Topics in Quantum Electronics
Journal of the Optical Society of America B
IEEE Photonics Technology Letters
Applied Physics B: Lasers and Optics
Journal of Lightwave Technology
Journal of Optics A—Pure and Applied Optics
Applied Optics
Optical Materials
Optics Communications

General-physics journals:

Reviews of Modern Physics
Physical Review B
Journal of Chemical Physics
Journal of Applied Physics

In addition to these optics and physics-based journals, specific topics in nonlinear optics are published in a wide variety of other journals, extending from theoretical physics to electrical engineering, from materials to chemistry (particularly polymer chemistry), and as applications in such fields such as biology, materials processing, and spectroscopy. Finally, a few important nonlinear optics papers will be found in a variety of review journals, too varied to list here, such as those found in the citations in Sec. III.

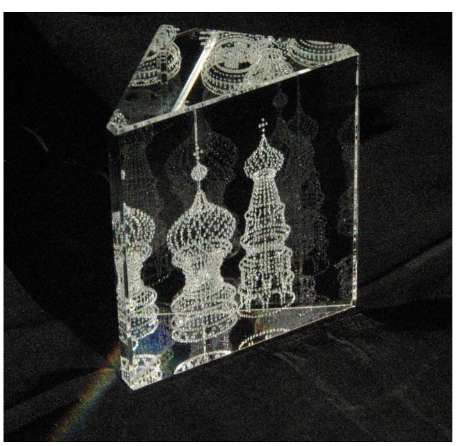

Fig. 2. Three-dimensional image within a glass prism purchased by the author in Russia in 1995. Each microscopic glass bubble was created by a single pulse from a laser beam focused to a point. The nonlinear optical process of multiphoton ionization results in localized plasma formation and internal melting. This image was created on a lathe; today with computer control any 3D image can be inscribed inside a block, with a process that originates from nonlinear optics. See Ref. 143.

B. Books

Readers have the option to peruse an entire textbook or handbook specializing in nonlinear optics or to explore specific chapters on nonlinear optics in more general textbooks and handbooks that cover optics and lasers (sometimes called photonics). Over 40 books have been published on nonlinear optics (not including those related to specific applications), most of which I have evaluated. Here are listed only those I believe will be of greatest value in teaching the principles of this subject. Thus the list does not include some historically important books, because newer texts include more recent developments and use more up-to-date units. The list also omits compendia of papers since these have limited success as learning tools.

1. Textbooks and handbooks specializing in nonlinear optics

1. **Physics of Nonlinear Optics**, Kuang-Sheng Ho, Song H. Liu, and Guang S. He (World Scientific, Singapore, 2000). A textbook written for undergraduate seniors, much of it seems more at graduate level. Originally written in Chinese, the English is not perfect, but the physical explanations are clear, as are the mathematical calculations carried out to support the physics. (I)
2. **Nonlinear Optics: Theory, Numerical Modeling, and**

Applications, Partha P. Banerjee (Marcel Dekker, New York, 2004). This undergraduate textbook is geared to engineering. It does not cover experimental results, but the concepts are easier to follow than the above book (Ref. 1), and classical approaches are emphasized. Some topics are included that might be considered more specialized, such as liquid crystals, self-organization, and photonic band gaps; available as a Kindle book. (E)

3. **Nonlinear Optics, Third Edition**, Robert W. Boyd (Academic/Elsevier, Burlington, MA, 2008). This is the nonlinear optics textbook most often used in graduate courses and includes most of the nonlinear phenomena discussed here. The explanations are clear and physical, but rapidly become very high-level, going into the optical Bloch equations and Rabi oscillations that result from the quantum mechanical two-level atom approximation; available as an e-book. (A)
4. **Light-Matter Interaction: Atoms and Molecules in External Fields and Nonlinear Optics**, Wendell T. Hill III and Chi H. Lee (Wiley, Hoboken, 2007). This books is half quantum mechanics and quite theoretical, but the second half is relatively practical; available in paperback. (I)
5. **Handbook of Nonlinear Optics (Optical Science and Engineering)**, Richard L. Sutherland (Marcel Dekker, New York, 2003). This extensive compendium was put together for researchers and graduate students, summarizing a massive amount of information—basically all that is outlined in this Resource Letter as well as a vast array of more advanced topics, such as quantum mechanics, modeling and numerical analyses, optical chaos, photonic crystals, quantum coherence, nonlinear optics in random media, in free atoms and molecules, in guided waves, in cavities and quantum optics. Probably a bit too advanced for beginning students, but a handy reference to know about. (I)

2. General textbooks and handbooks that include a chapter on nonlinear optics

These textbooks, designed for undergraduate seniors and graduate students, cover optics in general, but have valuable coverage of nonlinear optics.

6. **Photonics: Linear and Nonlinear Interactions of Laser Light and Matter**, Ralf Menzel (Springer, New York, 2007). An excellent, rather concise but clear compendium of most nonlinear optical effects. (I)
7. **Photonics and Laser Engineering: Principles, Devices and Applications**, Alphyan Sennaroglu (McGraw Hill, New York, 2010). An engineering approach to nonlinear optics. (E)
8. **Fundamentals of Photonics**, B. E. A. Saleh and M. C. Teich (Wiley, Hoboken, 2007). A commonly used textbook; the material on nonlinear optics is rather condensed and may need a fair amount of in-class explanation. (I)
9. **Photonics: Optical Electronics in Modern Communications**, Amnon Yariv and Pochi Yeh (The Oxford Series in Electrical and Computer Engineering, Oxford, 2006). Covers only the basics of nonlinear optics, but in a clear way. (E)
10. **Handbook of Optics Vol. IV: Optical Properties of Materials, Nonlinear Optics, Quantum Optics**, **Third Edition**, edited by M. Bass, sponsored by the Optical Society of America (McGraw Hill, New York, 2010). Contains separately written chapters that cover most of the specific topics in nonlinear optics discussed in this paper. (I)

3. Monographs on specific nonlinear optics topics

Optical nonlinearities have become particularly important in cases of extremely long lengths, extremely high optical intensities, or in semiconductors, where collective behavior within bands produces unusually large nonlinearities. The first case involves glass fibers, where kilometer lengths compensate for very small nonlinearities at low laser powers. The second is made possible by extremely short pulse lasers providing very high instantaneous intensities. Taken to its limits, nonlinear optics has enabled lasers in the visible regime to generate x rays, among other exciting phenomena. Semiconductors, particularly quantum wells, have optical nonlinearities that are particularly large; absorption of light causes electrons to fill available states or to move within internal electric fields.

Specialized monographs are available on a wide variety of topics, such as quantum theory of optical nonlinearities, optically nonlinear polymers, photorefractivity, optical phase conjugation, nonlinear spectroscopy, and biomedical nonlinear microscopy. The list is too long to include here and will probably interest only those who are researching those specific areas. Search engines on the web can find such books, given appropriate key words.

11. **Nonlinear Fiber Optics**, G. P. Agrawal (Academic, New York, 2007); **Applications of Nonlinear Fiber Optics**, G. P. Agrawal (Academic, New York, 2008). These monographs are comprehensive and tend to be quite mathematical, but with experimental discussion mixed in. They should be understandable to advanced undergraduates as well as graduate students. (I)
12. **Nonlinear Optics in Telecom**, T. Schneider (Springer, New York, 2004). Focused more on the practical effects of nonlinear optics in telecommunications, the engineering approach makes it reasonably accessible. (I)
13. **Extreme Nonlinear Optics: An Introduction**, Martin Wegener (Springer, New York, 2005). Well written, begins with elementary discussions, and should inspire students who are interested in fundamental physics and how lasers can get us into new regimes of physics. (E)
14. **Nonlinear Optics in Semiconductors, I and II**, edited by E. Garmire and A. Kost (Academic, New York, 1999). A two-volume set covering the origin and application of nonlinearities in semiconductors; chapters vary from elementary to very advanced. (E-A)

C. Conference proceedings

Conference proceedings do not generally provide information in nonlinear optics that is richer than in published journal papers. The list of topics presented at conferences gives a snapshot of what is currently of interest, but archival journals are likely to be of greater research use. Four organizations provide conferences that cover topics in nonlinear optics. The Optical Society of America (OSA) has a primary focus on optics and lasers and is a member of the American Institute of Physics. It has an annual meeting called *Frontiers in Optics*. The electrical engineering side of nonlinear optics is supported by the IEEE Photonics Society (previously called LEOS—Lasers and Electro-Optics Society), which has its own annual conference. Both societies regularly have topical meetings that sometimes explore topics in nonlinear optics. Information on such meetings can be found by exploring their websites at the addresses given below.

The premier international forum for scientific and technical optics that includes sections on nonlinear optics is the Conference on Lasers and Electro-optics: Laser Science to Photonic Applications (CLEO). This conference is held in conjunction with the Quantum Electronics and Laser Science (QELS) Meeting, which is the annual meeting of the Quantum Electronics and Laser Division of the American Physical Society.

The fourth society in the field of lasers and optics is SPIE, which arranges conferences in engineering and applications of optics. They have occasionally technical sessions in nonlinear optics, but this is not a major focus of this society.

15. The Optical Society of America, ⟨www.OSA.org⟩, has an annual meeting, ⟨http://www.frontiersinoptics.com/⟩, which contains a section called "quantum electronics" that includes topics in nonlinear optics.

16. The IEEE Photonics Society, ⟨http://photonicssociety.org/⟩, has an annual meeting, ⟨http://www.photonicsconferences.org/ANNUAL2010/⟩, that includes topics on nonlinear optics.

17. Topical Meeting on Nonlinear Optics: Materials, Fundamentals and Applications (2009, 2007, 2004, 2002, 2000), ⟨http://www.opticsinfobase.org/browseconferences.cfm?meetingid=28⟩. A specialized conference held approximately every two years; conference proceedings are available through this website.

18. The Conference on Lasers and Electro-Optics website: ⟨http://www.cleoconference.org⟩

D. Online resources

Two important websites listed below are reliable and describe each of the major nonlinear optics phenomena in terms that should be understandable to undergraduates. The rest of the web sources are online nonlinear optics course material accessed in July 2010. Several websites present slides on nonlinear optics that are introductory and might serve as outlines that could be supplemented by other written material.

19. ⟨http://www.rp-photonics.com/encyclopedia.html⟩. *An Open Access Encyclopedia for Photonics and Laser Technology*, written by Dr. Rüdiger Paschotta, a consultant in photonics, covers the basics of nonlinear optics rather well, with good search ability and provides the basic equations. Nothing else online compares to it.

20. ⟨http://en.wikipedia.org/wiki/Nonlinear optics⟩. Wikipedia can usually be trusted on scientific topics, and their sections on nonlinear optics are no exception.

21. ⟨ http:// www. pma. caltech. edu/ Courses/ph136/yr2004/books03/chap09/0209.1.pdf ⟩. A chapter written by Kip Thorne at Caltech as part of a course called "Applications of Classical Physics."

22. ⟨ http:// www. physics.gla.ac.uk/Optics/Lectures/Modern Optics/ModOpt2001.pdf ⟩. Extensive notes for a course called "Modern and Nonlinear Optics," taught by Miles Padgett at the University of Glasgow.

23. ⟨http://phys.strath.ac.uk/12-370/⟩. Slides by Allister Ferguson from the University of Strathclyde, Glasgow.

24. ⟨ http://www-vlsi.stanford.edu/papers/aen_nlog_02.ppt ⟩. Slides by Azita Emami of Stanford University.

25. ⟨ www. iasbs. ac. ir/ faculty/khalesi/nonlinear%20optics/nlo1.ppt ⟩. Slides by H. R. Khalesifard from the Institute for Advanced Studies in Basic Sciences, Iran.

26. ⟨http://unjobs.org/tags/nonlinear-optics⟩. Links to other sites on nonlinear optics.

III. SPECIFIC TOPICS IN NONLINEAR OPTICS

This section lists mostly reviews rather than original research papers, because the field has been so fast-moving, with so many parallel discoveries, that in most cases the initial papers will not be the most enlightening. For quick overviews of each of these specific topics readers are encouraged to look at the online encyclopedia in Ref. 19.

27. "Recent advances in nonlinear optics," Y. R. Shen, Rev. Mod. Phys. **48**, 1–32 (1976). A fairly complete review of nonlinear optics as of 1976, by which time much of the basic research had been completed; includes the basics of harmonic generation, nonlinear scattering (stimulated Raman and Brillouin), and nonlinear spectroscopy. (I)

A. Second-order nonlinearities in transparent media

Second-order nonlinearities occur in media that have no center of inversion symmetry and are usually assumed nonabsorbing (or weakly absorbing). Nonlinearity in the susceptibility owing to high incident light fields drives new frequencies. The most common effect is the generation of second-harmonic, which doubles the frequency (halves the wavelength), but higher-order harmonics can occur as well. In analogous fashion, two incident beams with different frequencies can interfere and the second-order nonlinearity can provide light at the sum frequency. Successful energy conversion requires matching the phases of the incident waves and the generated wave.

The second-order nonlinearity can also generate a beam at the difference frequency. This is the origin of optical parametric amplification (OPA) and also optical parametric oscillation (OPO) when a second-order nonlinear medium is placed inside a resonant cavity.

1. Second-harmonic generation (SHG) or frequency doubling

Tutorial reviews cited here are followed by early research reviews. References 35–40 introduce some applications; in so doing, they give useful introductions to the field from varying points of view.

28. "Visual methods for interpreting optical nonlinearity at the molecular level," R. D. Wampler, A. J. Moad, C. W. Moad, R. Heiland, and G. J. Simpson, Acc. Chem. Res. **40**, 953–960 (2007). Provides simple pictures of how optical nonlinearities arise, from a molecular point of view. (E)

29. "Second-order nonlinear susceptibilities of various dielectric and semiconductor materials," I. Shoji, T.

Kondo, and R. Ito, Opt. Quantum Electron. **34**, 797–833 (2002). Tutorial review. (E)

30. "2nd-order nonlinearity in poled-polymer systems," D. M. Burland, R. D. Miller, and C. A. Walsh, Chem. Rev. (Washington, D.C.) **94**, 31–75 (1994). Tutorial review. (I)

31. "Role of structural factors in the nonlinear optical properties of phthalocyanines and related compounds," G. de la Torre, P. Vaquez, F. Agullo-Lopez, and T. Torres, Chem. Rev. (Washington, D.C.) **104**, 3723–3750 (2004). Includes general review of second-harmonic generation. (E)

32. "Interactions between light waves in a nonlinear dielectric," J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918–1939 (1962). The classic paper; derives nonlinear optics by quantum mechanics. (A)

33. "Optical harmonics and nonlinear phenomena," P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23–39 (1963). Uses a simple Taylor expansion of the nonlinear polarizability. (I)

34. "Nonlinear optics," R. W. Minck, R. W. Terhune, and C. C. Wang, Appl. Opt. **5**, 1595–1612 (1966). Reviews the status of nonlinear optics only four years after it was discovered, including the relation between second-harmonic and the electro-optic effect; introduces optical parametric amplification (discussed below). (E-I)

35. "Wave mixing spectroscopy for surface studies," Y. R. Shen, Solid State Commun. **102**, 221–229 (1997). Discusses only second-harmonic. (E)

36. "Analytical and device-related applications of nonlinear optics," Joseph A. Miragliotta, Johns Hopkins APL Tech. Dig. **16**, 348–357 (1995). Accessed at ⟨http://techdigest.jhuapl.edu/td1604/Miraglio.pdf⟩. (E)

37. "Bulk characterization methods for non-centrosymmetric materials: Second-harmonic generation, piezoelectricity, pyroelectricity, and ferroelectricity," K. M. Ok, E. O. Chi, and P. S. Halasyamani, Chem. Soc. Rev. **35**, 710–717 (2006). A tutorial. (E)

38. "Second-harmonic generation as a tool for studying electronic and magnetic structures of crystals: Review," M. Fiebig, V. V. Pavlov, and R. V. Pisarev, J. Opt. Soc. Am. B **22**, 96–118 (2005). (E)

39. "Optical 2nd-harmonic generation as a probe of surface-chemistry," R. M. Corn and D. A. Higgins, Chem. Rev. (Washington, D.C.) **94**, 107–125 (1994). (E)

40. "Imaging techniques for harmonic and multiphoton absorption fluorescence microscopy," R. Carriles, D. N. Schafer, K. E. Sheetz, J. J. Field, R. Cisek, V. Barzda, A. W. Sylvester, and J. A. Squier, Rev. Sci. Instrum. **80**, 081101-081123 (2009). Review article. (E)

2. Quasi-phase-matching

Achieving sizable frequency mixing requires phase-matching. The original technique exploited natural birefringence in nonlinear anisotropic crystals, by propagating the light in a particular direction through the crystal. This approach was briefly explained in the references in the last section, but Ref. 41 provides the details necessary to fully understand phase-matching of anisotropic crystals. Fortunately, commercial vendors have worked out these geometries and sell the proper orientation of crystals to achieve phase-matching, so that only a few materials researchers will need to understand these details.

An important alternative is quasi-phase-matching, which does not require anisotropic crystals. In quasi-phase-matching, the direction of the nonlinear tensor is reversed every time the phase mismatch begins to approach π. This technique has turned out to be used in a great many of the second-harmonic generation applications.

41. ⟨http://www.rp-photonics.com/phase_matching.html⟩. The best source for detailed information on anisotropic crystal phase-matching. (E-I)

42. ⟨http://www.rp-photonics.com/quasi_phase_matching.html⟩. Excellent online description of quasi-phase-matching. (E)

43. "Quasi-phase matching," Karl Tillman, seminar presentation, Kansas State University, ⟨http://jrm.phys.ksu.edu/research/presentations/seminars/fall06/Tillman-Quasi-phasematching_files/v3_document.htm⟩. Provides particularly useful visual representations; also discusses optical parametric oscillation. (E)

44. "Quasi-phasematching," D. S. Hum and M. M. Fejer, C. R. Phys. **8**, 180–198 (2007). Excellent recent review. (E)

45. "Quasi-phase-matched 2nd harmonic-generation—Tuning and tolerances," M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer, IEEE J. Quantum Electron. **28**, 2631–2654 (1992). Research paper that covers the same material as Ref. 44 at a higher level. (I)

46. "An introduction to methods of periodic poling for 2nd-harmonic generation," M. Houe and P. D. Townsend, J. Phys. D: Appl. Phys. **28**, 1747–1763 (1995). Reviews both bulk and periodic harmonic generation in lithium niobate. (E)

47. "Nonlinear frequency conversion in semiconductor optical waveguides using birefringent, modal and quasi-phase-matching techniques," S. V. Rao, K. Moutzouris, and M. Ebrahimzadeh, J. Opt. A, Pure Appl. Opt. **6**, 569–584 (2004). Compares various methods of phase-matching. (E)

3. Difference Frequency Generation (DFG)

The second-order nonlinearity can generate light at a frequency that is the difference between two other frequencies. This has become useful to generating waves at terahertz frequencies, an important new field of study.

48. "Optical THz-wave generation with periodically-inverted GaAs," K. L. Vodopyanov, Laser Photonics Rev. **2**, 11–25 (2008). An easy-to-follow description of how terahertz is produced by DFG. (E)

49. "Terahertz fields and applications," D. Dragoman and M. Dragoman, Prog. Quantum Electron. **28**, 1–66 (2004). A general review of terahertz radiation with only a small, but excellent, section on DFG. (E)

4. Optical parametric amplification (OPA) and oscillation

OPA is a form of DFG in which a higher-frequency pump wave ω_p can amplify a signal at lower frequency ω_s, at the same time generating an "idler" wave at frequency ω_i such that $\omega_p = \omega_s + \omega_i$. Optical parametric *oscillation* occurs when an OPA is placed inside a cavity resonant at the signal or idler frequencies. The usefulness of these processes is their tunability. In wavelength regions where no tunable lasers exist, the OPO can provide coherent light. As with any oscillation, OPOs emit only above a certain threshold of input laser light. Because their threshold tends to be high, most practical OPOs use pulsed light, and they become particularly useful when pumped by ultrafast lasers.

50. ⟨http://www.rp-photonics.com/optical_parametric_oscillators.html⟩. This page of the online encyclopedia contains excellent information on OPOs and OPAs. (E)

51. "Parametric generation of tunable light from continuous-wave to femtosecond pulses," M. H. Dunn and M. Ebrahimzadeh, Science **286**, 1513–1517 (1999). A review article. (E)

52. "Nonlinear optics and solid-state lasers: 2000," Robert L. Byer, IEEE J. Sel. Top. Quantum Electron. **6**, 911–930 (2000). Contains a large section on OPOs, told from the personal story of research at Stanford, put in the context of their other work. (E)

53. "Ultrafast optical parametric amplifiers," G. Cerullo and S. De Silvestri, Rev. Sci. Instrum. **74**, 1–18 (2003). Explains how OPO systems are put together and perform. (E-I)

54. "Few-optical-cycle pulses tunable from the visible to the mid-infrared by optical parametric amplifiers," D. Brida, C. Manzoni, G. Cirmi, M. Marangoni, S. Bonora, P. Villoresi, S. De Silvestri, and G. Cerullo, J. Opt. A, Pure Appl. Opt. **12**, 013001-013014 (2010). A nice review article that includes both theory and experiments. (E)

B. Third-order and higher nonlinearities in transparent media

Third-order nonlinearities arise from expanding the nonlinear susceptibility to third order in the electric field. These nonlinearities do not require a center of inversion symmetry, so they occur in all materials: gases, liquids, glasses, and crystals. Third-order nonlinearities create both the third-harmonic and an intensity-dependent change in the refractive index. The extension from second- to third-harmonic is relatively straightforward, at least in concept. The major use of these higher-order harmonics is to reach ultraviolet wavelengths, where there are no lasers.

55. "3rd-order optical susceptibilities of liquids and solids," R. W. Hellwarth, Prog. Quantum Electron. **5**, 1–68 (1977). An extensive analysis of the effects created by the third-order nonlinearity. (I-A)

56. "Optical third-harmonic generation in alkali-metal vapors," R. B. Miles and S. E. Harris, IEEE J. Quantum Electron. **QE-9**, 470–484 (1973). Theoretical only, this paper provides a comprehensive introduction to thinking about higher-order nonlinearities. (A)

1. High harmonic generation (HHG) and extreme nonlinear optics (ENO)

Since third-order nonlinearities are small, the efficiency of conversion to third-harmonic is usually quite small unless the laser intensity is very high. In this case, even higher-order harmonics are readily seen. When the electric field strength of the light is high enough, optical nonlinearities can generate multiple orders with light having frequencies much greater than the original (typically 100–1000 times greater). This is done using femtosecond pulses; proper phasing of the harmonics can lead to extremely short extremely intense pulses, approaching attoseconds long (10^{-18} s). The Taylor expansion that initially defined orders of nonlinearity breaks down, resulting in the regime of extreme nonlinear optics. All of the following papers are recommended to learn about this field, which physics students may find very exciting:

57. "High-order harmonic generation and other intense optical field–matter interactions: Review of recent experimental and theoretical advances," J. G. Eden, Prog. Quantum Electron. **28**, 197–246 (2004). Includes historical background and progresses from elementary to advanced. (E-A)

58. "The propagation of powerful femtosecond laser pulses in optical media: Physics, applications, and new challenges," S. L. Chin, S. A. Hosseini, W. Liu, Q. Luo, F. Theberge, N. Akozbek, A. Becker, V. P. Kandidov, O. G. Kosareva, and H. Schroeder, Can. J. Phys. **83**, 863–905 (2005). Special Einstein review paper. (E)

59. "Extreme nonlinear optics: Coherent X rays from lasers," H. C. Kapteyn, M. M. Murnane, and I. R. Christov, Phys. Today **58** (3), 39–46 (2005). (E)

60. "Intense few-cycle laser fields: Frontiers of nonlinear optics," T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545–591 (2000). (E)

61. "The physics of attosecond light pulses," P. Agostini and L. F. DiMauro, Rep. Prog. Phys. **67**, 813–855 (2004). (E-I)

62. "Optics in the relativistic regime," G. A. Mourou, T. Tajima, and S. V. Bulanov, Rev. Mod. Phys. **78**, 309–371 (2006). (E)

C. Nonlinear refractive index

The third-order nonlinearity produces a term that provides a nonlinear refractive index. This is sometimes called the optical Kerr effect (OKE) because the square of the light's electric field causes the change in refractive index. (The ordinary Kerr effect is a refractive index change that depends quadratically on *applied* electric field.) The first two papers describe some general principles and the rest describe nonlinear refractive index in particular media.

63. "Dispersion of bound electronic nonlinear refraction in solids," M. Sheik-Bahae, D. C. Hutchings, D. J. Hagan, and E. W. Van Stryland, IEEE J. Quantum Electron. **27**, 1296–1309 (1991). (E-I)

64. "Order-of-magnitude estimates of the nonlinear optical susceptibility," R. W. Boyd, J. Mod. Opt. **46**, 367–378 (1999). (E)

65. "Band-edge nonlinearities in direct-gap semiconductors and their application to optical bistability and optical computing," S. W. Koch, N. Peyghambarian, and H. M. Gibbs, J. Appl. Phys. **63**, R1–R11 (1988). (E)

66. "Optical nonlinearity in photonic glasses," K. Tanaka, J. Mater. Sci.: Mater. Electron. **16**, 633–643 (2005). Excellent paper from first principles; includes bulk glasses. (E)

67. "Resonant optical nonlinearities in semiconductors," E.

Garmire, IEEE J. Sel. Top. Quantum Electron. **6**, 1094–1110 (2000). (E)

68. "Third-order nonlinear optical organic materials for photonic switching," B. Luther-Davies and M. Samoc, Curr. Opin. Solid State Mater. Sci. **2**, 213–219 (1997). (E)

1. Four Wave Mixing (FWM)

Through the nonlinear refractive index, three input waves couple and produce a different output wave. In principle these waves may have any frequency, intensity, or direction, subject to conservation of energy and momentum. When the frequencies are the same, it is called *degenerate four-wave mixing* (DFWM). A common FWM geometry involves two intense pump beams interfering in a nonlinear medium to form a refractive index grating. A fourth beam results when a third beam is diffracted off this grating, offering a geometry with background-free measurements. The first two references review the concept and the next two show how it is useful for spectroscopy.

DFWM is the origin of photorefractive phase conjugation, discussed in a chapter of a dissertation on fullerenes. A section below in this Resource Letter will describe more about photorefractive effect and phase conjugation.

Four-wave mixing of pulses in fibers is the source of self-phase modulation (SPM) and frequency mixing in fibers that can confound signals in long-distance fiber telecommunications. This will be discussed in a separate section on fibers. Finally, FWM in laser amplifiers has been suggested as a means for frequency conversion, important in some telecommunication applications, as suggested in Ref. 74. This is understood by including laser gain as an imaginary term in the analysis of the nonlinear susceptibility. (The imaginary term for gain is minus the imaginary term for loss.)

69. "Four-wave mixing," Markus B. Raschke, University of Washington, accessed at ⟨http://faculty.washington.edu/mraschke/cars/fwmixing.pdf⟩.

70. "The transient grating—A holographic window to dynamic processes," J. T. Fourkas and M. D. Fayer, Acc. Chem. Res. **25**, 227–233 (1992). (E)

71. "Femtosecond transient-grating techniques: Population and coherence dynamics involving ground and excited states," E. J. Brown, Q. G. Zhang, and M. Dantus, J. Chem. Phys. **110**, 5772–5788 (1999). (I)

72. "Transient gratings, four-wave-mixing and polariton effects in nonlinear optics," J. Knoester and S. Mukamel, Phys. Rep. **205**, 1–58 (1991). Develops a fully microscopic framework for calculating four-wave mixing and analyzing transient grating spectroscopy in molecular crystals. (A)

73. "Limiting and degenerate four-wave mixing in novel fullerenes," Daniela Marciu, Ph.D. thesis, Chap. 3, Virginia Polytechnic Institute, accessed at ⟨http://scholar.lib.vt.edu/theses/available/etd-022299-083514/unrestricted/CHAPTER3.PDF⟩. Discusses how DFWM becomes the origin of photorefractive phase conjugation. (E)

74. "Frequency conversion by nearly-degenerate four-wave mixing in travelling-wave semiconductor laser amplifiers," N. Schunk, G. GroPkopf, R. Ludwig, R. Schnabel, and H. G. Weber, IEE Procceedings **137** Pt.J. , 209–214 (1990). (I)

2. Two-wave mixing (two-beam coupling)

Two-wave mixing is a form of degenerate four-wave mixing in which the grating caused by the optical nonlinearity diffracts power from one beam into the other. This can only occur if the nonlinear refractive index grating becomes displaced laterally from the intensity grating. This happens when photoinduced carriers move within the material. This occurs naturally in photorefractive materials (electro-optic crystals that have static internal fields), such as barium titanate, or when a lateral external field is applied to electro-optic semiconductors or polymers, or when one beam has a small frequency shift from the other. Two-wave mixing enables coupling of power from a strong pump wave into a weak signal wave, offering a form of amplification to the signal; the references in this section describe how it works. More is discussed in the section below on the photorefractive effect.

75. "2-wave mixing in nonlinear media," P. Yeh, IEEE J. Quantum Electron. **25**, 484–519 (1989). (I)

76. "Observation of high gain in a liquid-crystal panel with photoconducting polymeric layers," S. Bartkiewicz, A. Miniewicz, F. Kajzar, and M. Zagorska, Appl. Opt. **37**, 6871–6877 (1998). An applied field across the panel displaces the grating. (E)

77. "Polarization-resolved beam combination in liquid suspensions of shaped microparticles," D. Rogovin, J. Scholl, R. Pizzoferrato, M. De Spirito, U. Zammit, and M. Marinelli, Phys. Rev. A **44**, 7580–7597 (1991). (I-A)

3. Self-Phase Modulation (SPM)

Self-phase modulation is the broadening of the frequency spectrum of a pulse light when it travels through a third-order nonlinear medium. The time-dependent intensity creates a time-varying refractive index that produces a time-varying phase, the source of the frequency shift. While the effect can be seen in ordinary materials, fibers show particularly dramatic effects, and photonic crystal fibers do even more so.

78. "Supercontinuum generation in photonic crystal fiber," J. M. Dudley, G. Genty, and S. Coen, Rev. Mod. Phys. **78**, 1135–1184 (2006). Review of numerical and experimental studies. (I)

79. "Optical spectral broadening and supercontinuum generation in telecom applications," S. Smirnov, J. D. Ania-Castanon, T. J. Ellingham, S. M. Kobtsev, S. Kukarin, and S. K. Turitsyn, Opt. Fiber Technol. **12**, 122–147 (2006). (E)

80. "Ultrafast white-light continuum generation and self-focusing in transparent condensed media," A. Brodeur and S. L. Chin, J. Opt. Soc. Am. B **16**, 637–650 (1999). (E)

4. Cross-phase modulation (XPM)

XPM is a nonlinear optical effect where one wavelength of light can affect the phase of another wavelength of light through the third-order nonlinearity. This phenomenon is both a boon and a bane. It has proven useful for converting wavelengths in telecommunications, while it has also caused instabilities. It is closely related to FWM, as is the phenomenon of *cross-polarized* wave generation. Most of these phenomena are more specifically described in papers on nonlinear effects in fibers.

81. "All-optical wavelength conversion by semiconductor optical amplifiers," T. Durhuus, B. Mikkelsen, C. Joergensen, S. Lykke Danielsen, and K. E. Stubkjaer, J. Lightwave Technol. **14**, 942–954 (1996). Review paper. (I)

82. "Modulation instability induced by cross-phase modulation in optical fibers," G. P. Agrawal, P. L. Baldeck, and R. R. Alfano, Phys. Rev. A **39**, 3406–3413 (1989). (I)

83. "Cross-phase modulation in multispan WDM optical fiber systems," R. Q. Hui, K. R. Demarest, and C. T. Allen, J. Lightwave Technol. **17**, 1018–1026 (1999). (I)

5. Optical fibers

While glass is not generally considered a nonlinear medium, optical fibers are long enough that the nonlinear refractive index can build up to create many orders of π phase shift. In some cases the nonlinearities are considered a detriment to simple transmission, while in others these nonlinearities have been utilized for new interesting applications. Third-order nonlinearities are not the only nonlinear phenomena in fibers; stimulated scattering has a major impact on transmission and will be discussed below.

84. "The optical Kerr effect and quantum optics in fibers," A. Sizmann and G. Leuchs, Prog. Opt. **39**, 373–469 (1999). Introduces basic concepts and extends them into the field of quantum optics, providing a nice introduction to how optical nonlinearities affect quantum optics in fibers. (E-A)

85. "Optical nonlinearities in fibers: Review, recent examples, and systems applications," J. Toulouse, J. Lightwave Technol. **23**, 3625–3641 (2005). A well-written review of each of the nonlinear phenomena that can occur in fibers; shows how these nonlinearities affect fibers in telecommunications and sensor systems. (E)

86. "Parametric amplification and processing in optical fibers," S. Radic, Laser Photonics Rev. **2**, 498–513 (2008). Analyzes high-confinement fibers with nearly vanishing chromatic dispersion providing highly nonlinear fiber (HNLF) used for parametric amplification; shows how nonlinearities in fibers can be maximized to make them useful. (A).

6. Optical solitons

A soliton is a pulse of light that propagates without spreading in time as it travels along a fiber. Its particular shape results from the time-dependence of the nonlinear refractive index overcoming the fiber dispersion. Solitons have been predicted to have important applications in fiber communication systems.

87. "Soliton communication systems," R. J. Essiambre and G. P. Agrawal, Prog. Opt. **37**, 185–256 (1997). (I)

88. "Solitons in optical communications," H. A. Haus and W. S. Wong, Rev. Mod. Phys. **68**, 423–444 (1996). (I-A)

7. Self-focusing and self-trapping

The nonlinear refractive index alters the spatial profile of a Gaussian beam, sometimes causing the beam to self-focus, sometimes forming a self-trapped beam (a spatial soliton), and sometimes causing the beam width to oscillate as the beam propagates through the nonlinear medium. When the beam self-focuses, its intensity increases to such an extent that other nonlinearities arise (stimulated Raman and Brillouin effects and self-phase modulation). The nonlinear wave equation introduced by the nonlinear refractive index is often unstable, leading to filamentation and extraordinarily rich spatial phenomena. However, in certain media the solutions are stable and generate spatial solitons, the analog of the temporal solitons discussed above. Bright solitons arise from Gaussian-like beams. Dark solitons arise from beams that have a region of minimum intensity within their Gaussian profile.

89. "Femtosecond filamentation in transparent media," A. Couairon and A. Mysyrowicz, Phys. Rep. **441**, 47–189 (2007). A review of the many nonlinear phenomena that can occur for Gaussian beams at high intensities and long transmission distances. (E)

90. "Beam nonparaxiality, filament formation, and beam breakup in the self-focusing of optical beams," M. D. Feit and J. A. Fleck, J. Opt. Soc. Am. B **5**, 633–640 (1988). Based on numerical modeling and explains the concepts very well. (I)

91. "Optical spatial solitons and their interactions: Universality and diversity," G. I. Stegeman and M. Segev, Science **286**, 1518–1523 (1999). (E)

92. "Optical solitons due to quadratic nonlinearities: From basic physics to futuristic applications of χ_2 nonlinearities," A. V. Buryak, P. Di Trapani, D. V. Skryabin, and Stefano Trillo, Phys. Rep. **370**, 63–235 (2002). (I-A)

93. "Bright and dark spatial solitons in non-Kerr media," Y. S. Kivshar, Opt. Quantum Electron. **30**, 571–614 (1998). (I)

94. "Spatiotemporal optical solitons," B. A. Malomed, D. Mihalache, F. Wise, and L. Torner, J. Opt. B: Quantum Semiclassical Opt. **7**, R53–R72 (2005). A review article. (I-A)

95. "Wave collapse in physics: Principles and applications to light and plasma waves," Luc Bergé, Phys. Rep. **303**, 259–370 (1998). A generalized discussion that will appeal to certain advanced students. (I-A)

8. Cascaded nonlinearities

An effective nonlinear refractive index can be created in a harmonic-generating material in a geometry that is far from phase-matching. The rapid oscillation between the fundamental and harmonic causes a phase shift to the initial wave, which acts as a third-order nonlinearity.

96. "X(2) cascading phenomena and their applications to all-optical signal processing, mode-locking, pulse compression and solitons," G. I. Stegeman, D. J. Hagan, and L. Torner, Opt. Quantum Electron. **28**, 1691–1740 (1996). A review paper that might not be available everywhere. (I)

97. "Coherent interactions for all-optical signal processing via quadratic nonlinearities," Gaetano Assanto, George I. Stegeman, Mansoor Sheik-Bahae, and Eric Van Stryland, IEEE J. Quantum Electron. **31**, 613–681 (1995). Research paper describing the principles and possible applications. (I)

D. Absorptive nonlinearities

Absorption of intense optical beams may excite a substantial fraction of the species from their ground state; as a result, the absorption saturates. Alternatively, for nonabsorbing atoms or molecules, intense light may increase the probability that two-photon absorption takes place. In the latter case, two photons arriving at the same time may cause energy levels spaced at twice the photon energy to absorb the laser beam. Besides two-photon absorption, three- or more-photon absorption is possible. This leads to the possibility that an initially transparent medium becomes totally absorptive if the intensity is high enough. Photoinduced absorption causes *optical limiting* and can be important in protecting sensors (and eyes!) from high intensity light beams. Multiphoton ionization is an important factor in creating optically induced plasmas, such as recorded in the micro-bubbles that define the 3D image shown in Fig. 2. The phenomena of saturable absorption and multiphoton absorption are treated separately below. First, however, review papers are offered that point out the inevitable relation between a change in absorption and a change in refractive index. Indeed, both nonlinearities often occur at the same time.

98. "Kramers–Kronig relations in nonlinear optics," D. C. Hutchings, M. Sheik-Bahae, D. J. Hagan, and E. W. Van Stryland, Opt. Quantum Electron. **24**, 1–30 (1992). (I)

99. "Generalized Kramers–Kronig relations in nonlinear optical- and THz-spectroscopy," K.-E. Peiponen and J. J. Saarinen, Rep. Prog. Phys. **72**, 056401–056420 (2009). (I)

1. Saturable absorption

The first reference gives experimental results on a typical saturable absorption nonlinearity and how it is used. One application for saturable absorption is mode-locking lasers, known for many years. More recently, saturable absorbers inside a Fabry–Perot resonator provide an output mirror that has been shown to be an excellent mode-locker, as described in the last two references.

100. "Absorption, wave mixing, and phase conjugation with bacteriorhodopsin," O. Werner, B. Fischer, A. Lewis, and I. Nebenzahl, Opt. Lett. **15**, 1117–1119 (1990). (E)

101. "Theory of mode-locking with a slow saturable absorber," H. A. Haus, IEEE J. Quantum Electron. **11**, 736–746 (1975). (I)

102. "Semiconductor saturable absorber mirrors (SESAM's) for femtosecond to nanosecond pulse generation in solid-state lasers," U. Keller, K. J. Weingarten, F. X. Kartner, D. Kopf, B. Braun, I. D. Jung, R. Fluck, C. Honninger, N. Matuschek, and J. A. der Au, IEEE J. Sel. Top. Quantum Electron. **2**, 435–453 (1996). (E)

103. "Frontiers in ultrashort pulse generation: Pushing the limits in linear and nonlinear optics," G. Steinmeyer, D. H. Sutter, L. Gallmann, N. Matuschek, and U. Keller, Science **286**, 1507–1512 (1999). (E)

2. Two-photon and multiphoton absorption

The simultaneous absorption of two (or more) photons transfers their energy to a single electron that goes into an excited state at the frequency of the sum of the two photons. This process may take place in a material that was initially absorbing, causing an intensity-dependent increase in the absorption; this can limit the amount of transmitted light, producing *optical limiting*. Many photons of frequency too small to ionize atoms (or molecules) individually can act simultaneously, when suitably intense, to remove bound electrons from their orbits, leading to *multiphoton ionization*.

A particularly important application of these multiphoton absorption processes is in microscopy of biological tissues. Multiphoton microscopy is currently the preferred microscopy technique for imaging in optically scattering tissue because of the deeper penetration depth of the fundamental infrared photons and the minimized photodamage and photobleaching. Two-photon excited fluorescence and second-harmonic generation signal from living tissue provide subcellular resolution images with sufficient morphological details to be diagnostically useful.

104. "A review of optical limiting mechanisms and devices using organics, fullerenes, semiconductors and other materials," L. W. Tutt and T. F. Boggess, Prog. Quantum Electron. **17**, 299–338 (1993). (E)

105. "Organic and inorganic optical limiting materials: From fullerenes to nanoparticles," Y.-P. Sun and J. E. Riggs, Int. Rev. Phys. Chem. **18**, 43–90 (1999). (E)

106. "Multiphoton ionization of atoms," G. Mainfray and G Manus, Rep. Prog. Phys. **54**, 1333–1372 (1991). Review article. (E)

107. "Two-photon excitation fluorescence microscopy," Peter T. C. So, Chen Y. Dong, Barry R. Masters, and Keith M. Berland, Annu. Rev. Biomed. Eng. **2000.02**, 399–429 (2000). Includes history and basics. (E)

108. "Imaging techniques for harmonic and multiphoton absorption fluorescence microscopy," Ramón Carriles, Dawn N. Schafer, Kraig E. Sheetz, Jeffrey J. Field, Richard Cisek, Virginijus Barzda, Anne W. Sylvester, and Jeffrey A. Squier, Rev. Sci. Instrum. **80**, 081101-081122 (2009) (invited review article). (I)

109. "Multiphoton microscopy in life sciences," K. Koènig, J. Microsc. **200**, 83–104 (2000). Invited review. (E)

E. Photorefractives and nonlinearities due to carrier transport

Photorefractive materials have a nonlinearity that results from photoinduced liberation of charge carriers. If the material has refractive index (or absorption) that depends on electric field (for example, electro-optic materials) and these charges are free to move about, then their charge separation will reduce the internal electric fields, which in turn alters the refractive index (or absorption). Photoinduced carriers must live long enough to move around in an internal or applied field. This charge motion can make possible two-beam coupling (discussed above) and is particularly important because this nonlinearity can be observed at extraordinarily low optical power levels. Typically an interference grating setup inside the crystal enables observation of DFWM, as first reported in barium titanate excited by argon lasers. Reference 73 is to an on-linear chapter of a Ph.D. thesis that gives a clear introduction to photorefractive principles.

110. "Applications of photorefractive crystals," S. I. Stepanov, Rep. Prog. Phys. **57**, 39–116 (1994). Exten-

sive review of experiments and underlying theory. (E-I)

111. "Theory and applications of 4-wave mixing in photorefractive media," M. Cronin-Golomb, B. Fischer, J. O. White, and A. Yariv, IEEE J. Quantum Electron. **20**, 12–30 (1984). Early review paper. (I)

112. "A photorefractive polymer with high optical gain and diffraction efficiency near 100-percent," K. Meerholz, B. L. Volodin, Sandalphon, B. Kippelen, and N. Peyghambarian, Nature (London) **371**, 497–500 (1994). (E)

113. "Photorefractive polymers," W. E. Moerner, A. Grunnet-Jepsen, and C. L. Thompson, Annu. Rev. Mater. Sci. **27**, 585–623 (1997). (I)

114. "Optical nonlinearities due to carrier transport in semiconductors," E. Garmire, N. M. Jokerst, A. Kost, A. Danner, and P. D. Dapkus, J. Opt. Soc. Am. B **6**, 579–587 (1989). (E)

115. "Semi-insulating semiconductor heterostructures: Optoelectronic properties and applications," D. D. Nolte, J. Appl. Phys. **85**, 6259–6289 (1999). (E)

F. Optical phase conjugation

This is a particular application of DFWM in which the fourth wave is phase-conjugate to the first wave. The process can occur by any third-order nonlinearity. First observed in stimulated Brillouin scattering, it is seen in holography and in photorefractive media. The latter has become the most practical, primarily because of the low intensities at which phase conjugation can be observed. Early references are listed here because of the clarity of their presentations.

116. "Optical phase conjugation," Vladimir V. Shkunov and Boris Ya. Zel'dovich, Sci. Am. **253** (12), 54–59 (1985). Introduction for lay people. (E)

117. "Applications of optical phase conjugation," David M. Pepper, Sci. Am. **254** (1), 74–83 (1986). Introduction for lay people. (E)

118. "Holography, coherent-light amplification and optical-phase conjugation with photorefractive materials," P. Gunter, Phys. Rep. **93**, 199–299 (1982). (I)

119. "Phase conjugate optics and real-time holography," A. Yariv, IEEE J. Quantum Electron. **14**, 650–660 (1978). Also, "Author's reply," A. Yariv, IEEE J. Quantum Electron. **QE-15**, 524–525 (1979). Initial research paper. (I)

120. "Generation of time-reversed wave fronts by nonlinear refraction," W. Hellwarth, J. Opt. Soc. Am. **67**, 1–3 (1977). Initial research paper. (I)

G. Stimulated scattering

Raman scattering was well-known before lasers were invented. A small fraction of the light scattered from molecules has a frequency that is shifted from the incident frequency by the frequency of photoinduced molecular vibrations. *Stimulated* Raman scattering (SRS) occurs when light is intense enough that gain builds up at the Raman frequency. The first SRS was observed accidentally, emitted from a laser cavity that included the Raman-active liquid nitrobenzene; intense emission appeared at a wavelength longer than the laser intensity (so-called *Stokes* light). Experiments outside the cavity found that Stokes light could be amplified by SRS. Furthermore, the light-induced molecular vibrations can interact with additional incident light, causing growth of light at a frequency *greater* than the laser frequency, by an amount of the molecular vibration (so-called *anti-Stokes* light).

A closely related phenomenon is Stimulated Brillouin scattering (SBS). This works in the same way as SRS, but laser light creates and scatters from *acoustic* waves (the relation between SRS and SBS is analogous to the relation between optical and acoustic phonons). The first three references describe the theory behind SRS and SBS.

Both SRS and SBS can be detrimental to transmitting high power through condensed matter, particularly optical fibers. However, both phenomena can be useful. Raman-active media placed inside optical cavities can make excellent Raman lasers, providing wavelengths not otherwise reachable. Raman lasers can be fibers or all-solid-state. Stimulated Brillouin scattering is useful because it provides phase-conjugate reflection (as do photorefractive media) and has been used for wave-front correction. Stimulated Raman scattering has considerable value in spectroscopy, which will be described in a separate section below.

121. "Theory of stimulated Raman scattering," R. W. Hellwarth, Phys. Rev. **130**, 1850–1852 (1963). (I)

122. "Stimulated Raman effect," N. Bloembergen, Am. J. Phys. **35**, 989–1023 (1967). (E-I)

123. "Theory of stimulated Brillouin and Raman scattering," Y. R. Shen and N. Bloembergen, Phys. Rev. **137**, 1787–1805 (1965). (I-A)

124. "Optical power handling capacity of low loss optical fibers as determined by stimulated Raman and Brillouin-scattering," R. G. Smith, Appl. Opt. **11**, 2489–2494 (1972). (I)

125. "Output characteristics of high-power continuous wave Raman fiber laser at 1239 nm using phosphosilicate fiber," M. Prabhu, N. S. Kim, and K. Ueda, Opt. Rev. **7**, 297–302 (2000). (E)

126. "The design and operation of solid-state Raman lasers," H. M. Pask, Prog. Quantum Electron. **27**, 3–56 (2003). (E)

127. "Design and operation of a 150-W near diffraction-limited laser-amplifier with SBS wave-front correction," C. B. Dane, L. E. Zapata, W. A. Neuman, M. A. Norton, and L. A. Hackel, IEEE J. Quantum Electron. **31**, 148–163 (1995). (E)

128. "Noise initiation of stimulated Brillouin-scattering," R. W. Boyd, K. Rzazewski, and P. Narum, Phys. Rev. A **42**, 5514–5521 (1990). (A)

129. "Generation and suppression of stimulated Brillouin-scattering in single liquid droplets," J. Z. Zhang and R. K. Chang, J. Opt. Soc. Am. B **6**, 151–153 (1989). (E)

H. Nonlinear optics and spectroscopy

Nonlinear spectroscopy uses multiple laser beams focused into a sample to create new beams because of nonlinear processes. This enables background-free and high resolution spectroscopy never seen before. Amplification of the stimulated Raman process enhances signals by many orders of magnitude. Indeed, nonlinear spectroscopy has revolutionized our understanding of atoms and molecules, with five Nobel Prizes directly related to this subject. Below are references to the excellent Nobel lectures in written form and, more recently, as videos. Also provided are review papers that highlight some of the most fruitful techniques. Numerous books and monographs have been published covering portions of these subjects and may be of specific interest to some readers. These are too numerous to list here, but are available in many libraries and can be searched online by topic.

130. "Nonlinear optics and spectroscopy," Nicolaas Bloembergen, Rev. Mod. Phys. **54**, 685–695 (1982). Nobel Prize lecture. Also online at ⟨http://nobelprize.org/nobel_prizes/physics/laureates/1981/bloembergen-lecture.pdf⟩. (E)

131. "Spectroscopy in a new light," A. L. Schawlow, Rev. Mod. Phys. **54**, 697–707 (1982). Nobel Prize lecture. Also online at ⟨http://nobelprize.org/nobel_prizes/physics/laureates/1981/schawlow-lecture.pdf⟩. (E)

132. "Femtochemistry: Atomic-scale dynamics of the chemical bond," A. H. Zewail, J. Phys. Chem. A **104**, 5660–5694 (2000). Adapted from Nobel Prize lecture with a strong chemistry orientation. Nobel lecture in written or video form online at ⟨http://nobelprize.org/nobel_prizes/chemistry/laureates/1999/zewail-lecture.html⟩. (E)

133. "Passion for precision," T. W. Hänsch, Rev. Mod. Phys. **78**, 1297–1309 (2006). Nobel Prize lecture. (E) Also online as written or video at ⟨http://nobelprize.org/nobel_prizes/physics/laureates/2005/hansch-lecture.html⟩. (E)

134. "Defining and measuring optical frequencies: The optical clock opportunity—and more," John L. Hall, ChemPhysChem **7**, 2242–2258 (2006). Also online video and written lecture at ⟨http://nobelprize.org/nobel_prizes/physics/laureates/2005/hall-lecture.html⟩. (E)

135. "Optical molecular dephasing—Principles of and probings by coherent laser spectroscopy," A. H. Zewail, Acc. Chem. Res. **13**, 360–368 (1980). Explains nonlinear interactions between light and molecules very well. (I-A)

136. "Femtosecond real-time probing of reactions. 19. Nonlinear (DFWM) techniques for probing transition states of uni- and bimolecular reactions," M. Motzkus, S. Pedersen, and A. H. Zewail, J. Phys. Chem. **100**, 5620–5633 (1996). Describes the methodology of DFWM spectroscopy with ultra-short pulses very well. (E)

137. "Fundamentals of nonlinear spectroscopies," John C. Wright, ⟨http://www.chem.wisc.edu/~wright/fundamentals.html⟩. An explanation of the fundamentals, followed by a useful review of the wide variety of nonlinear spectroscopies available today. (E)

138. "Detection of trace molecular species using degenerate four-wave mixing," Roger L. Farrow and David J. Rakestraw, Science **257**, 1894–1900 (1992). Reviews and describes practical uses for gas analysis. (E)

139. "Spectral hole-burning spectroscopy in amorphous molecular-solids and proteins," R. Jankowiak, J. M. Hayes, and G. J. Small, Chem. Rev. (Washington, D.C.) **93**, 1471–1502 (1993). The earliest form of nonlinear spectroscopy is reviewed, along with applications to fundamental and applied problems. (E)

140. "Coherent anti-Stokes Raman scattering: From proof-of-the-principle experiments to femtosecond CARS and higher order wave-mixing generalizations," A. M. Zheltikov, J. Raman Spectrosc. **31**, 653–667 (2000). Reviews the growth and the development of the basic coherent anti-Stokes Raman spectroscopy from concepts through their modern implementation. This is one of the most important nonlinear spectroscopic techniques. (I)

141. "Coherent multidimensional vibrational spectroscopy," J. C. Wright, Int. Rev. Phys. Chem. **21**, 185–255 (2002). A review of techniques that combine advanced FWM and CARS, so that spectroscopists have as much versatility as with NMR. (A)

142. "Nonlinear optical spectroscopy of solid interfaces," Markus B. Raschke and Y. Ron Shen, Curr. Opin. Solid State Mater. Sci. **8**, 343–352 (2004). Available online at ⟨http://faculty.washington.edu/mraschke/mbr_pub/publications/rs_currop04.pdf⟩. Describes how the different symmetry rules enable separation of surface and volume effects with nonlinear optics. (I)

IV. CONCLUSIONS

Nonlinear optics has become an enabling phenomenon for many practical applications, both in scientific investigations, in engineering, and even in commercial products. References have been provided for the basic concepts for nonlinear optics, as well as for its use in a few cases, particularly spectroscopy. Much more could have been included, such as nonlinear materials inside a resonant cavity (e.g., optical bistability), inside waveguides, spatial instabilities, and chaos in optical systems. Short pulses exhibit a wide variety of time-dependent nonlinear phenomena, such as self-steepening, optical coherent transients, and electromagnetically induced transparency (EIT). Nonlinear optics is an important adjunct to quantum optics, including quantum information, quantum communications, and quantum computing. Nonlinear optics is used for laser cooling, trapping of neutral atoms, and Bose–Einstein condensation. Coherent control of atoms, molecules, and solid-state populations is being studied using nonlinear optical effects. I remind readers that numerous monographs and review papers have been written on most of these subjects, a list too long to evaluate here. Specific approaches to nonlinear optics and its applications can vary from quantum theory, atomic and molecular physics, condensed-matter physics, optical physics, chemistry, electrical engineering, and extend to other fields: biology, medicine, and engineering disciplines (mechanical, chemical, and aerospace). More information on books, articles, and websites pertaining to specific topics may be found using search engines on the web.

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