Applied Nonlinear Optics

G. H. C. New and J. W. Haus

The recent fiftieth anniversary celebrations marking the invention of the laser and the birth of modern nonlinear optics were major historical milestones. Theodore Maiman's observation of laser action in ruby in May 1960 [1] provided the essential tool that enabled Peter Franken's team at the University of Michigan to perform their legendary 1961 experiment in which they saw optical second harmonic generation for the first time [2]. From this small beginning, nonlinear optics has grown into the vast and vibrant field that it is today.

The Optical Society Centennial provides an opportunity to reflect on developments in nonlinear optics in the intervening years and, specifically, to focus on some of the highlights in the development of the field between 1975 and 1990. The theoretical foundations of optical frequency mixing were laid by Nicolaas Bloembergen's Harvard team in a seminal 1962 paper [3], which was prescient for introducing innovative ideas that strongly influenced later developments in the field; some specific examples will be mentioned later. In 1979, Nicolaas Bloembergen (see Fig. 1) was awarded The Optical Society's Ives Medal, the society's highest award. He won a quarter share of the 1981 Nobel Prize "*for his contribution to the development of laser spectroscopy*," in addition to his pioneering work on nonlinear optics.

By the early 1970s, many of the conceptual foundations of nonlinear optics had been laid, and a remarkable number of crude experimental demonstrations of techniques that are now routine had been performed. Progress over the ensuing decades was often prompted by advances in laser technology and, crucially, in materials fabrication. Suddenly it would become possible to implement an experiment so much more effectively than previously that it would soon become an established laboratory technique, or might even form the basis of a new commercial product.

A major achievement of the period was the fabrication of layered crystalline structures in which phase-matching is determined by the periodicity of the layers. Remarkably, this "quasi-phase-matching" (QPM) technique was originally suggested in the 1962 Harvard paper mentioned earlier [3], and it is a prime example of a principle that took more than two decades of gestation between original inspiration and final fruition.

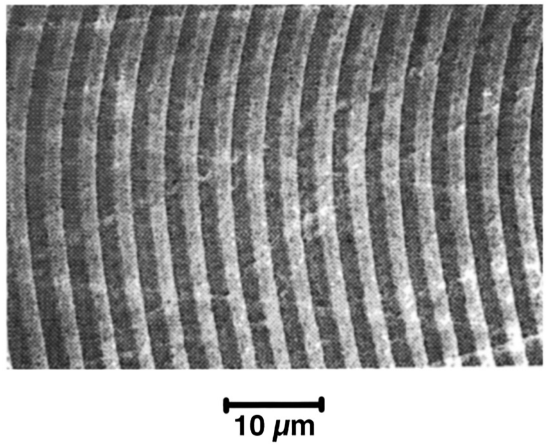
Quasi-phase-matching materials have periodically reversed domains, each one coherence length thick. The finished product is like a loaf of sliced bread in which alternate slices (of anisotropic crystal) are inverted (see Fig. 2). The problem is that each "slice" has to be only a few micrometers thick, so it would be a little thin for one's breakfast toast! It took more than two decades to develop the sophisticated crystal growth techniques needed to fabricate media with such thin layers. Today, QPM is routine; indeed many researchers have abandoned traditional birefringent phase-matching altogether. The most well-known QPM medium is perhaps periodically poled lithium niobate (abbreviated PPLN and pronounced "piplin"), and practical devices of high conversion efficiency are commercially available. In 1998, Robert Byer (see Fig. 1) and Martin Fejer were awarded The Optical Society's R. W. Wood Prize "*for seminal contributions to quasi-phase matching and its application to nonlinear optics*." More recently, in 2009, Robert Byer received the Ives Medal, The Optical Society's most prestigious award.

The need for tunable coherent light sources to replace tunable dye lasers drove the development of solid-state devices; these are not subject to messy chemical spills, and the tuning ranges achievable in a single medium can extend from the ultraviolet to the mid-wave infrared (3–5-μm) regimes.

▲ **Fig. 1.** Images of five scientists who have made major breakthroughs in the development of nonlinear optics. From top left to bottom right they are: Nicolaas Bloembergen, Robert L. Byer, Chuangtian Chen, Linn F. Mollenauer, and Stephen E. Harris. (Bloembergen, Mollenauer, and Harris photographs courtesy of AIP Emilio Segre Visual Archives, Physics Today Collection; Byer photograph courtesy of AIP Emilio Segre Visual Archives, Gallery of Member Society Presidents; Chen photograph courtesy of Professor Chen Chuangtian.)

An important nonlinear optical process for creating a wideband coherent light source is optical parametric generation. This is essentially sum-frequency generation (the generalized version of second harmonic generation) running in reverse. A high-frequency "pump" wave drives two waves of lower frequency, known as the "signal" and the "idler"; in photon language, the pump photon divides its energy between the signal and idler photons. Without a seed to define a particular frequency band, the signal and idler grow from noise, with frequencies determined by the phase-matching conditions. An optical parametric amplifier is a device of this kind with a signal or idler seed to fix the operating frequency. If the gain is high, the conversion efficiency can be quite large, even for a single-pass system. However, the efficiency can be greatly improved by placing the nonlinear medium within a well-designed cavity, creating an optical parametric oscillator (or OPO).

The first OPO was demonstrated by Giordmaine and Miller as early as 1965, but subsequent progress was slow, largely because nonlinear crystals of the necessary high quality were not available. Indeed, the OPO is another example of a device where technological capability lagged seriously behind concept. By the late 1980s, however, the introduction of new nonlinear materials coupled with progress in laser technology made it possible to realize low-threshold OPOs. Synchronous pumping can be employed, in which case the OPO is driven by a train of short pulses with the repetition rate matched to the round-trip time of the cavity. OPOs are now standard devices in the well-found laser lab.



▲ **Fig. 2.** SEM image of a periodically poled lithium niobate wafer. (Reproduced with permission from [4]. Copyright 1990, AIP Publishing LLC.)

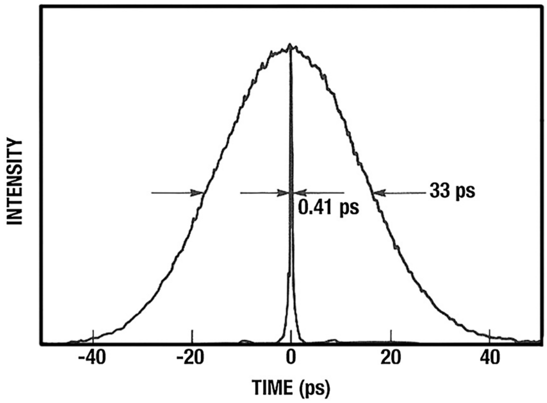
A number of new nonlinear materials that are now household names were developed in the 1980s. Using theoretical tools as a guide, C.-T. Chen (see Fig. 1) and co-workers discovered nonlinear materials such as BaB_2O_4 (beta barium borate, or BBO) and LiB_3O_5 (lithium borate, or LBO), both of which are widely used today. Other materials studied since that time include orientational-patterned III-V semiconductors, ZGP (zinc germanium phosphide) and DAST (4-dimethylamino-N-methyl-4-stilbazolium). Using a range of different nonlinear optical interactions, these have played an increasingly important role in extending the range of tunable coherent sources to the long-wave infrared (8–12 μm) and beyond to the terahertz regime. In recognition of the central role of materials technology, The Optical Society sponsored a 1988 conference entitled "Nonlinear Optical Properties of Materials," and key results were published in a special issue of the *Journal of The Optical Society of America B* [5].

The nonlinear interactions mentioned so far are all second order, which also means that they involve the interaction of three waves. Third-order processes lead to a wide range of four-wave phenomena, which include third harmonic generation, self-phase modulation via the optical Kerr effect (nonlinear refraction), optical phase conjugation, and optical bistability, to name just a few. They also form the basis of much of nonlinear spectroscopy, and quantum optical effects too.

Many important applications are based on nonlinear refraction. In combination with diffraction, it is the essential ingredient in the formation of spatial solitons, while with group velocity dispersion, it is crucial in the control of temporal pulse profiles. The 1970s and 1980s saw rapid progress in the understanding of optical pulse propagation and the development of nonlinear pulse compression techniques. Most of the techniques involve judicious combinations of self-phase modulation (SPM) and group velocity dispersion (GVD). Both of these processes cause a pulse to acquire a carrier frequency sweep (or "chirp"), but the overall effect depends on whether the two processes work with or against each other and whether they occur simultaneously or in succession. If they act simultaneously and in opposition, pulse propagation is governed by the nonlinear Schrödinger equation, which supports optical solitons.

In the early 1970s, Hasegawa and Tappert had suggested that optical fibers offered the ideal environment for solitons, but it was not until 1980 that Mollenauer, Stolen, and Gordon at what was then still Bell Telephone Labs actually observed optical soliton propagation in a fiber. Later, in 1988, Mollenauer and Smith demonstrated the transmission of 55-ps pulses over 400 km by supplying Raman gain at 42-km intervals. The possible use of solitons in optical communications was vigorously pursued in the 1990s but has rarely been implemented commercially. Nevertheless, research on solitons (both temporal and spatial) had a significant impact on nonlinear optics and indeed on laser technology as well. Linn Mollenauer (see Fig. 1) was awarded The Optical Society's Charles Hard Townes award in 1997 for his work on optical solitons and their applications to data transmission. Earlier, in 1982, he had received the R. W. Wood Prize for his work on color-center lasers, which played a vital role in early soliton experiments.

The race to achieve ever shorter optical pulses began on the day Maiman demonstrated the first laser and is likely to run for as long as laser research continues. Its hallmark has always been the strong and highly productive synergy between nonlinear optics and laser development. On the one hand, nonlinear interactions are strengthened by the high peak power of short laser pulses, but nonlinear optical processes are themselves exploited in advanced laser systems to promote the generation of shorter pulses.



▲ **Fig. 3.** Eighty times compression of a pulse. (Reproduced with permission from [6]. Copyright 1984, AIP Publishing LLC.)

▲ **Fig. 4.** Supercontinuum generation using a prism to disperse the colors in the pulse. (Image courtesy of [7]. © 2008 SPIE, image credit: E. Goulielmakis, reprint_permission@spie.org.)

The basic principle of pulse compression involves the application of SPM and GVD in opposition (as for solitons), but in succession rather than simultaneously. The idea, which originated in the late 1960s, is to start by imposing SPM to broaden the pulse spectrum and create the bandwidth required to support a shorter pulse. The wideband signal is then compressed by using a dispersive delay line, usually based on a pair of diffraction gratings in a Z-shaped configuration, which has a similar effect to that of negative GVD. An attractive option is to introduce the SPM in an optical fiber since, for non-trivial reasons, the simultaneous effect of SPM and *positive* GVD produces stretched profiles that are ideal for efficient compression.

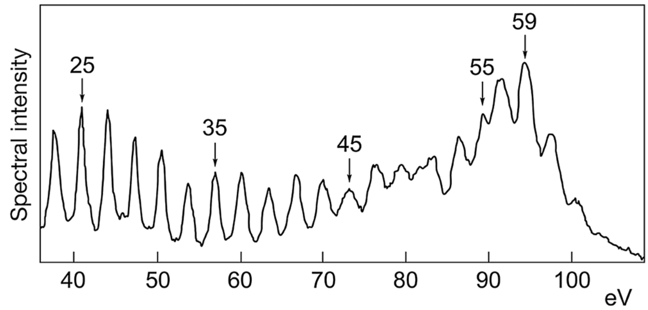
Fiber-grating compressors were first demonstrated in 1981, and there followed a series of record-breaking experiments that included the remarkable 1984 demonstration by Johnson, Stolen, and Simpson of a compression factor of ×80 (from 33 ps to 410 fs, Fig. 3). The culmination of this effort was the famous achievement of a 6-fs pulse by Fork, Shank, and Ippen in 1987, a result that held the world record for the shortest optical pulse for many years thereafter.

Other important nonlinear effects occur when pulses are launched in a fiber. Under suitable conditions, the combination of SPM and stimulated Raman scattering (SRS) creates a signal that extends over more than an octave in frequency bandwidth. A broadband signal of this kind is called a supercontinuum and has valuable applications in metrology and spectroscopy (Fig. 4).

Most developments in nonlinear optics in the 1960s involved solid media, especially crystals, although liquids also featured in experiments on the optical Kerr effect. By contrast, the 1970s and 1980s saw the beginning of work on the nonlinear optics of atoms and molecules in the gas phase that would come to full fruition in the 1990s and 2000s in effects such as high harmonic generation (Fig. 5), attosecond pulse generation, electromagnetically induced transparency, and slow light.

The early work on third harmonic generation in the inert gases in the late 1960s, and experiments on third, fifth, and seventh harmonic generation in metal vapors in the 1970s by Harris, Reintjes, and others, all exhibited characteristics typical of the perturbative (weak-field) regime, insofar as the conversion efficiency for higher harmonics fell away sharply. The first steps on the road that would later lead to the gateway into high harmonic generation were taken in the late 1980s. By that time, laser intensities of ~100 TW/cm^2 and above were becoming available, and some remarkable results on the inert gases were recorded that marked the entry into a new strong-field regime. For the lower harmonics (up to perhaps the ninth), the conversion efficiency dropped off as before, but higher harmonics lay on a plateau on which the efficiency remained essentially constant up to a well-defined high-frequency limit. The cut-off point could be extended further into the UV by increasing the laser intensity, although a saturation intensity existed beyond which no further extension was possible. These experiments laid the foundation for work in the following decade in which harmonics in the hundreds and even the thousands were generated.

An equally dramatic line of development involved atomic systems in which the main action involved three levels linked by two separate laser fields. A number of different effects of this kind were beginning to be studied in early 1980s, most of which exploited the effect of quantum interference in one way or another. Early examples included coherent population trapping and laser-induced continuum structure, both of which were prefigured to some degree in the much earlier work of Fano and others on Fano interference.



▲ **Fig. 5.** Experimental manifestation of high harmonic generation. (Courtesy of [8]. Copyright 2011, Cambridge University Press.)

In the mid to late 1980s, the effect of lasing without inversion (LWI) caused a particular stir, probably because it contradicted a principle that most people regarded as fundamental to laser physics, namely, that population inversion was an essential prerequisite of laser action. The scheme for LWI envisaged by Harris involved three levels in a pattern roughly resembling an inverted V, or a capital Greek lambda Λ. Under normal circumstances, laser amplification on one arm of the Λ would occur only if a population inversion existed between the two levels. Crucially, however, this restriction is removed if a strong laser field is tuned to the resonance frequency of the other arm of the Λ.

The simplest explanation of how LWI works involves another quantum interference process, highlighted by Harris in 1990, called electromagnetically induced transparency (EIT). A straightforward density matrix calculation shows that the absorption and dispersion characteristics of one of the transitions of the Λ are dramatically altered in the presence of the strong coupling field tuned to the other, and indeed that the absorption goes to zero on exact resonance. Quantum interference has in effect canceled out the absorption process that normally competes with stimulated emission, thereby enabling lasing to occur in the absence of a population inversion.

Stephen Harris (See Fig. 1) received the Ives Medal in 1991 for his pioneering work in nonlinear optics. The citation specifically mentioned his work on LWI and EIT.

Given the strict word limit that we have worked within, we have naturally been forced to be highly selective in choosing the topics to cover. Literally thousands of research papers on nonlinear optics presenting the work of many hundreds of researchers were written within the time frame covered in this chapter. In view of these numbers, it is inevitable that many people will consider topics we have left out to be more important than those we have included. We extend our apologies to the majority whose work it has not been possible to mention here.

References

1. T. Maiman, "Stimulated optical radiation in ruby," Nature **187**, 493–494 (1960).
2. P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, "Generation of optical harmonics," Phys. Rev. Lett. **7**, 118–119 (1961).
3. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, "Interactions between light waves in a nonlinear dielectric," Phys. Rev. **127**, 1918–1939 (1962).
4. G. A. Magel, M. M. Fejer, and R. L. Byer, "Quasi-phase-matched second-harmonic generation of blue light in periodically poled $LiNbO_3$," Appl. Phys. Lett. **56**, 108–110 (1990).
5. C. M. Bowden and J. W. Haus, eds., "Nonlinear optical properties of materials," J. Opt. Soc. Am. B **6** (April 1989).
6. A. M. Johnson, R. H. Stolen, and W. M. Simpson, "80× single-stage compression of frequency doubled Nd:yttrium aluminum garnet laser pulses," Appl. Phys. Lett. **44**, 729–731 (1984).
7. J. Hewett, "Ultrashort pulses create ultrabroad source," historical archive, Optics.org.
8. J. W. G. Tisch, Imperial College Attosecond Laboratory; reproduced from G. H. C. New, "Introduction to nonlinear optics," Cambridge University Press 2011, with permission.