Lasers and the Growth of Nonlinear Optics

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Nonlinear optical effects were seen long before the laser was invented. In 1926, Russians Sergey Vavilov and Vadim L. Levishin observed optical saturation of absorption when they focused bright microsecond pulses to power densities of kilowatts per square centimeter. Vavilov introduced the term "nonlinear optics" in 1944, and during World War II Brian O'Brien put saturation to practical use in his Icaroscope to spot Japanese bombers attacking with the sun behind them. The bright coherent light from the laser opened new possibilities.

Peter Franken (Fig. 1) realized them as he sat in packed sessions on lasers at OSA's spring meeting in early March of 1961. His mind wandered as speakers droned about applications in communications and eye surgery. Seeking something really unusual, he calculated the intensity of a 5-kW laser pulse focused onto a 10-μm spot. His answer was megawatts per square centimeter, with electric fields of 100,000 V/cm—only three or four orders of magnitude below the electric field inside an atom.

"I realized then that you could do something with it," Franken recalled in a 1985 interview [1]. Further calculations showed the fields should be able to produce detectable amounts of the second harmonic. Excited, he left the meeting and hurried back to the University of Michigan, where he and solid-state physicist Gabriel (Gaby) Weinreich began planning an experiment. He rented a ruby laser from Trion Instruments, a small Ann Arbor company that was the first to manufacture them, and got Wilbur "Pete" Peters to set up a spectrograph and camera for measurements. Weinrich told him to fire the laser into crystalline quartz, which can produce the second harmonic because it lacks a center of inversion.

They needed a long time to get usable results. Alignment requirements were demanding, and harmonic conversion was so inefficient that 3-J, 1-ms pulses containing about 10^{19} photons yielded only about 10^{11} second harmonic photons. Nonetheless, their photographic plate clearly showed the small second harmonic spot. They submitted their paper in mid-July, a little over four months after the meeting, and it appeared in the 15 August *Physical Review Letters*—without the faint second harmonic spot, which an engraver had removed because it looked like a flaw in the photo [2].

Optical harmonic generation experienced a breakthrough in 1961. "At that time, we were all thinking photons, and you can't change the frequency of a photon," recalled Franken. But working with Willis Lamb at Oxford University in 1959 had taught Franken that classical electromagnetic wave theory applied to light, so he had realized that nonlinearities might generate optical harmonics. The faint second harmonic spot that never made it into print launched modern nonlinear optics.

Franken's results caught the eye of Joe Giordmaine, who just two months earlier had begun exploring the effects of ruby laser pulses on various materials at Bell Labs. He began testing Bell's large stock of crystals left from World War II research and within a few weeks was seeing more harmonic power than Franken had. When he tested crystals of potassium dihydrogen phosphate (KDP) he was surprised to find that second harmonic emission was not just in the direction of the ruby beam, but in a ring centered on a different direction, and that the second harmonic was many times higher at some angles than others. He had discovered the importance of phase matching the fundamental and second harmonic beams. It did not work in quartz, but it did in birefringent crystals such as KDP. Bob Terhune independently discovered phase matching at the same time at the Ford Motor Co. Research Laboratory.

At Harvard, Nicolaas Bloembergen (Fig. 2) gathered John Armstrong, Peter Pershan, and Jacques Ducuing to work on nonlinear optics after he saw a preprint of Franken's paper. Armstrong and Ducuing began experiments, and all four worked on theory. Bloembergen wrote the differential equations describing harmonic generation, but solving the nonlinear problems posed a formidable task. The group spent several intense and exciting months from July 1961 to early 1962, dividing the task among themselves and working closely with Bloembergen.

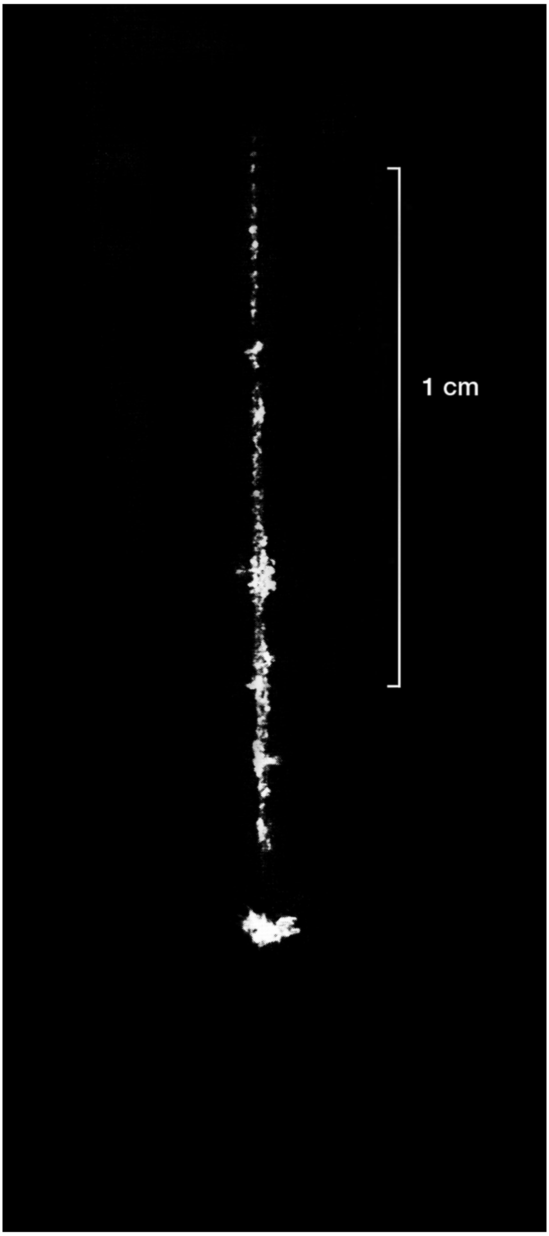
The result was a 22-page detailed analysis of light interactions in nonlinear dielectrics, published in *Physical Review* in September 1962 [3]. "It was by no means the last word, but it was a very complete first word," says Armstrong, whose name was first in alphabetical order. The codification of nonlinear interactions including harmonic generation and parametric conversion had a huge impact in the young field.

Meanwhile, experiments with high-power, single-pulse Q-switched ruby lasers at Hughes Aircraft's Aerospace group revealed an unexpected nonlinear anomaly. In early 1962, Eric Woodbury and Won Ng measured output power at several hundred megawatts, far more than expected, when they used a Kerr-cell Q-switch filled with nitrobenzene. Puzzled, they did other experiments, but the light finally dawned when measured power dropped to the expected level after they inserted narrow-pass filters centered on the 694.3-μm ruby line. Further measurements revealed unexpected light on three near-infrared lines, the strongest at 766 nm, a weaker one at 851.5 nm, and a barely detectable line at 961 nm. The increments were roughly equal in frequency units.

They reported what they thought was a new type of laser action, but it was up to Robert Hellwarth and Gisela Eckhardt of Hughes Research Labs to suggest the infrared lines were coming from stimulated Raman scattering by the nitrobenzene in the Q-switch. Experiments quickly confirmed that, and Hellwarth later developed a full theoretical model. It was a landmark discovery in nonlinear optics, showing that light interacted with molecular vibrations to stimulate scattering at Stokes-shifted wavelengths. Soon afterward, Terhune and Boris Stoicheff separately observed anti-Stokes emission.

▲ **Fig. 1.** Peter Franken. (OSA Historical Archives.)

▲ **Fig. 2.** Nicolaas Bloembergen. (Photograph by Norton Hintz, courtesy AIP Emilio Segre Visual Archives, Hintz Collection.)



▲ **Fig. 3.** Trace of damage caused by a Q-switched ruby laser pulse. (Courtesy of Courtesy of Michael Hercher.)

Charles Townes, then at MIT, analyzed Stoicheff's results and wondered whether lasers could also stimulate Brillouin scattering. In just two weeks, graduate student Ray Chiao, Townes, and Stoicheff used a ruby laser to demonstrate Brillouin scattering in a solid. Soon another student, Elsa Garmire, demonstrated Brillouin scattering in a liquid. It took years to work out the details, and in 1972 Boris Ya. Zel'dovich—the son of noted Soviet nuclear physicist Yakov B. Zel'dovich—showed that stimulated Brillouin scattering could produce phase conjugation.

Townes suggested another research direction after seeing thin filaments of optical damage in glass exposed to Q-switched megawatt pulses from a ruby laser (see Fig. 3) by Michael Hercher of the University of Rochester. Townes suspected that optical nonlinearities were self-trapping the beam and with Chiao and Garmire described how the intense beam changed the refractive index to create a waveguide. At the MIT Lincoln Laboratory, Paul Kelley developed a theory of self-focusing showing scale lengths and the effects of beam power. Unknown to U.S. researchers, Vladimir Talanov was working on the same idea in the closed Soviet city of Gorky.

Rem V. Khokhlov and Sergey A. Akhmanov founded Russia's first nonlinear optics laboratory at Moscow State University in 1962, but Cold War tensions allowed little communication with American groups. During that year, they proposed a theory to extend parametric oscillation from radio frequencies to light, offering a way to generate tunable output from fixed-wavelength lasers. Khokhlov and Akhmanov's *Problems in Nonlinear Optics* was the first book on the topic when it was published in Russian in 1964, but it did not appear in English until 1972. Bloembergen's *Nonlinear Optics* was published in 1965.

The Moscow lab soon developed efficient ways of generating second, third, fourth, and fifth harmonics. A long series of experiments with Alexander Kovrigin demonstrated an optical parametric oscillator in the spring of 1965, at nearly the same time Giordmaine (Fig. 4) and crystal expert Robert Miller demonstrated one at Bell Labs. Both pumped with the second harmonic of neodymium lasers, with the Moscow lab using KTP and Bell using lithium niobate as the nonlinear crystals. The experiments were difficult, and Bell Labs achieved only 5% conversion efficiency, but output was tunable across 70 nm, an impressive figure in 1965.

Self-focusing led to self-phase modulation. When Kelley and MIT student Ken Gustafson studied shock-wave generation in nonlinear materials, they found a phase shift that depended on the square of the field intensity. They did not make much of it at the time, but in 1967 Fujio Shimizu at the University of Toronto demonstrated that self-phase modulation in liquids could spread the spectral bandwidth of a

▶ **Fig. 4.** David Kleinman and Joe Giordmaine. (Courtesy of AT&T Archives and History Center.)

pulse [4]. In 1970 Bob Alfano and Stan Shapiro at GTE Laboratories in Bayside, New York, demonstrated more frequency spreading in glass and crystals [5]. The higher the power, the broader the bandwidth, and over the years the effect spread the spectrum enough to make white-light supercontinua.

In 1973, Akira Hasegawa and F. Tappert took another important step, extending the concept of self-trapping to describe optical temporal solitons in optical fibers [6]. Nonlinear phase modulation and dispersion interact such that pulse duration and frequency chirp increase and decrease cyclically along the length of the fiber, periodically reconstructing the original pulse. Hasegawa, Linn Mollenauer, and others later showed that solitons could transmit signals through optical fibers.

Modern nonlinear optics has come a long way from its roots, yet the fundamental groundwork remains solid. "To this day, every time I make a discovery in nonlinear optics, I look at [Bloembergen's] paper and he's done it," says Robert Boyd of Rochester. "He put the whole field together in 18 months." That feat earned Bloembergen the 1981 Nobel Prize in Physics.

Nonlinear optics is used in consumer products. Second harmonic generation turns the invisible 1.06-μm line of neodymium into a bright 532-nm green beam. "It's hard to believe you can buy these things. If you think of what's inside, it's just amazing," says Garmire. Harmonic generation also finds cutting-edge laboratory applications, generating pulses of attosecond duration or with wavelengths in the extreme ultraviolet or x-ray bands. Self-phase modulation together with mode locking produces femtosecond pulses and frequency combs. The more we try to do with optics, the more we have to think about nonlinearities. Like the laser that was essential to its birth and its applications, nonlinear optics seems to be everywhere.

Note: This chapter was adapted from [7].

References

1. Peter Franken oral history interview, http://www.aip.org/history/ohilist/4612.html
2. P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, "Generation of optical harmonics," Phys. Rev. Lett. 7, 118–120 (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. F. Shimizu, "Frequency broadening in liquids by a short light pulse," Phys. Rev. Lett. **19**, 1097–1100 (1967).
5. R. R. Alfano and S. L. Shapiro, "Observation of self-phase modulation and small-scale filaments in crystals and glasses," Phys. Rev. Lett. **24**, 592–594 (1970).
6. A. Hasegawa, "Soliton-based optical communications: an overview," IEEE J. Select. Topics Quantum Electron. **6**, 1161–1172 (2000).
7. J. Hecht, "How the laser launched nonlinear optics," Opt. Photon. News **21**(10), 34–40 (2010).