A quantum mechanical method is presented to calculate nonlinear susceptibilities of nonabsorbing media. The nonlinear response leads to coupling among three or four light waves in the nonlinear dielectric, describing harmonic generation and frequency mixing of light. [The SCI® indicates that this paper has been cited in more than 940 publications.]

Light Waves Interact

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The realization of the ruby laser in 1960 by Maiman made it clear that the techniques of magnetic resonance and microwave spectroscopy could be extended to the visible region of the spectrum. In 1960, we had already published a paper on microwave modulation of light. In 1961, we received a preprint of the paper by P.A. Franken et al. on the second harmonic generation of light. This stimulated me to think hard about nonlinear effects in optics, building on my familiarity with nonlinear effects in magnetic resonance. Since we did not yet have a serviceable ruby laser available, our group first concentrated on theoretical concepts of nonlinear optics. I was fortunate to have several outstanding associates, and it was not hard to get them interested in the problem. One was my former student, Peter S. Pershan, who in 1961 was a postdoc about to become an assistant professor. Another postdoc was John A. Armstrong, while Jacques Ducuing was a graduate student, selected by Professor Kastler of the Ecole Normale Superieure in Paris to receive advanced training in quantum electronics at Harvard University. The quality of the team may be judged from the fact that Pershan is now a tenured professor at Harvard, interested in X-ray optics with synchrotron radiation. Armstrong is corporate vice president for science and technology at IBM. Ducuing became professor at the Ecole Polytechnique at Paris-Paris, served as director-general of CNRS, the French national research organization; and is currently assistant secretary general for scientific affairs at NATO Headquarters in Brussels.

We had daily bull sessions and discussions in the fall of 1961. The outline of the paper became clear: We would study a semiclassical perturbation approach. Nonlinear susceptibilities would be calculated quantum-mechanically for nondissipative media. The nonlinear polarization terms would act as sources to excite other electromagnetic waves with different frequencies and wave vectors. General analytic solutions of the wave equations, coupled by the nonlinearity, were worked out in detail. We were able to show that 100 percent conversion of light energy from one frequency to another was possible in principle in the case of exact phase matching. We also proposed phase correction schemes in the case of momentum mismatch. It took more than a decade before technological applications based on these ideas were realized.

The coupling of four waves with conservation of energy and momentum in a nonlinear medium is a general problem, and our solutions attracted the attention of plasma physicists and hydrodynamists as well as of laser experts. Our paper would undoubtedly have been quoted more frequently if the general acceptance of its ideas had occurred less rapidly. By 1964, I was able to write a monograph, Nonlinear Optics, in which this paper was reprinted as an appendix. Later textbooks, The Principles of Nonlinear Optics and Quantum Electronics, use the general scheme outlined in this paper as a matter of course. Nonlinear optics has grown into a substantial branch of science. Our laboratory has continued to contribute to its development with more than 200 subsequent publications.

In 1979, I received the Ives Medal of the Optical Society of America for this work. In 1981, I shared half of the Nobel Prize in physics with Arthur L. Schawlow for "contributions to laser spectroscopy." The title of my Nobel lecture was "Nonlinear optics and spectroscopy."