INTRODUCTION TO NONLINEAR OPTICS: A SELECTED OVERVIEW

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INTRODUCTION

Historical Perspective

Nonlinear optics (NLO) has enjoyed great success as a discipline for over 30 years now. Although it was a relative newcomer to nonlinear wave sciences, other examples being nonlinear fluid dynamics, acoustics, plasmas etc, it has contributed many new phenomena. Since its inception in 1962, nonlinear optics has passed through many phases and different topics have been “hot” at any given time.\textsuperscript{1,2} One of the fascinating features of the nonlinear optics field is its regenerative power to develop new topics over the years. The first ten years witnessed demonstration of many of the fundamental interactions such as second harmonic generation (SHG), sum and difference frequency generation, stimulated Raman, Brillouin and Rayleigh scattering, self-focusing etc. And many more interesting phenomena were predicted theoretically, some having to wait two decades before experimental confirmation was forthcoming. This “novelty” trend continued into the second decade with the development of multiple nonlinear spectroscopies and their applications to materials science, phase conjugation, bistability leading to concepts of all-optical signal processing, the beginnings of nonlinear optics in fibers, etc. The third phase, from about the mid 1980s to the present has had its own highlights such as the development of nonlinear guided wave optics, especially in fibers where a whole spectrum of new propagation effects and light induced non-centrosymmetric effects were found, the exciting development of efficient, widely tunable sources through optical parametric oscillators, temporal solitons and their potential for long-haul communications, a surprising variety of spatial solitons, terahertz sources, femtosecond pulses, generation of tens of higher harmonics in gases etc. One of the exciting recent developments is the blurring of the roles of second and third order nonlinear optics, namely the creation of second order nonlinear effects via third order nonlinearities, and the use of second order effects to mimic third order phenomena. Many of these topics will be discussed in this book.

The key to applications of nonlinear optics is, has been and always will be the availability of appropriate materials. The initial stages of the field which focused on demonstrating and understanding new effects utilized the materials available at that time. For
example, for second harmonic generation, materials developed for piezoelectric applications which also require non-centrosymmetric media were used first. In the case of third order, much of the early work was done with liquids. Ultimately, the search for better materials was driven by the realization that in order for any applications to be practical, nonlinear optics had to move forward from the era in which high power lasers were almost exclusively needed to observe nonlinear phenomena. Compact semiconductor lasers with 100s of mW power levels drove the need for sub-watt nonlinear optics. The search for better materials gained momentum in the mid to late 1970s and continues unabated to the present day. In the case of second order materials, there have been multiple goals including doubling into the UV region of the spectrum, widely tunable sources via parametric interactions, inexpensive sources in the blue, etc. The exciting concept of all-optical processing has fueled third order nonlinear optics for many years.

**Formalism**

Traditionally nonlinear optics has been discussed in terms of the nonlinear polarization induced in a nonlinear medium by the mixing of one or more intense electromagnetic waves. Typically multiple beams with different frequencies are incident onto a nonlinear medium, either modifying the linear optical properties of the medium or leading to the generation of new waves at new frequencies. As a matter of notation, the incident fields $\mathbf{E}(r,t)$ of frequency $\omega_i$ and wavevector $k_i$ for propagation along the z-axis can be written in the form:

\[
E(r,t) = \frac{1}{2} E(r,\omega_i) \exp[i(\omega t - k r)] + \text{c.c.}
\]

For plane waves, $\mathbf{e}_i$ is the electric field unit vector, $f_i(x,y) = 1$ and $a_i(z)$, the slowly varying (complex) amplitude, is normalized so that $|a_i(z)|^2$ is the intensity in units of W/cm². When the interacting beams are of finite extent, for example in a waveguide, the $f_i(xy)$ describe the transverse field profiles. The nonlinear polarization

\[
P^{NL}(r,t) = \frac{1}{2} P^{NL}(r,\omega) \exp[i(\omega t - k \cdot r)] + \text{c.c.}
\]

induced by the mixing of the optical fields has the general form:

\[
P^{NL}(r,t) = \int dr_1 dr_2 dt_1 dt_2 \chi^{(2)}(r-r_1,t-t_1;r-r_2,t-t_2) E(r_1,t_1) E(r_2,t_2) + \]

\[
\int dr_1 dr_2 dr_3 dt_1 dt_2 dt_3 \chi^{(3)}(r-r_1,t-t_1;r-r_2,t-t_2;r-r_3,t-t_3) E(r_1,t_1) E(r_2,t_2) E(r_3,t_3)
\]

where $E(r,t)$ is the total electric field in the medium and $\chi^{(n)}$ is the n’th order nonlinearity. Here the $r - r_1$ allow for a spatially non-local response, e.g. carrier diffusion and $t - t_1$ allow for a polarization field at time t to be generated by fields at an earlier time $t_1$, for example due to a finite carrier recombination time. For a total field of the form $E(r,t) = \sum \frac{1}{2} E(r,\omega_i) \exp[i(\omega_i t - k_i r)] + \text{c.c.}$, i.e. an expansion in terms of its Fourier components, the induced polarization can be written as a Taylor’s expansion in the Fourier components of the mixing fields.