

NONLINEAR OPTICS

Edited by

**P. G. HARPER
and
B. S. WHERRETT**

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Nonlinear Optics

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Edited by

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and

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Director's Preface

The Sixteenth Scottish Universities Summer School in Physics was held on the Riccarton Campus of Heriot-Watt University, from July 28th to 15th August 1975.

The subject of the School, Nonlinear Optics, is still of course very young (dating from the early 1960s) but has expanded so rapidly and successfully that our interpretation of the subject was extended to include a number of topics in Laser Spectroscopy. The notion of Nonlinear Optics deals with phenomena that occur in the high radiation intensities obtainable in laser beams and encompasses interaction with material systems varying from high temperature plasmas at very high density through conventional solid state material to very isolated atomic and molecular systems at low pressure. The prevailing common factor is of course that the interactions between light and matter can be expressed in terms of various orders of nonlinear susceptibilities.

Our lecturers were all well-known for recent contributions to current research on this fast moving subject. The demonstrations of their mastery, in a variety of styles, led to a very successful School attended by nearly 100 students varying from those just beginning research to those returning to research in retirement! At the end of the School we were all convinced that the topic of Nonlinear Optics continues to provide a useful unifying theme and I hope that this volume will appropriately confirm this feeling. The major organization of the School was capably undertaken by the Secretary, Dr. M. J. Colles; he reinforced this notion in some concluding remarks in which he showed that we did consider all non-linear susceptibilities up to seventh order.

It is a pleasure to acknowledge contributions made by many others: Dr. W. Taylor, as Treasurer; Dr. R. B. Dennis and Mr. I. Grieve, as Social Secretaries; Professor P. G. Harper and Dr. B. S. Wherrett, as editors, Miss Myrtle Lee and her staff in the University Residences; Mrs. J. McGregor

and all the Secretaries in the Physics Department and many others whom space does not permit to mention individually.

The intense interest and time spent by the above contributed greatly to the professional, social and sporting success of the School.

S. D. Smith
Heriot-Watt University
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Editors' Note

Most of the articles in this volume have been prepared by the authors and follow closely the lectures presented at the Summer School. Exceptions are the synopsis of Professor Walther's lectures, and Professor Stoicheff's introductory lecture which serves here as a summary.

The boundaries separating the various topics are largely due to the specialization demanded by the rapid growth of Nonlinear Optics. It seemed appropriate in this volume to point out connections between apparently separate developments and wherever possible draw them together. The prefatory article by B. S. Wherrett attempts to provide a framework for such an integrated view.

We are grateful to Stella Sharpe for secretarial assistance in preparing the final version of the manuscripts.

P. G. Harper
B. S. Wherrett

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From Linear Dispersion to Self-Induced Transparency: An Introduction to Nonlinear Optics

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A vast amount of new information has been amassed in the field of Quantum Electronics over the past fifteen years, since the development of lasers. It is the aim of this introduction to give an overall view of that section of quantum electronics which we have chosen to regard as Nonlinear Optics, for the purpose of the Summer School. The idea is to provide a framework in which may be placed the following contributions from the nine invited lecturers.

The underlying cause of all linear and nonlinear optical effects is of course the interaction of electromagnetic radiation with matter. Thus, each contributor dealt with one or more aspects of the *response* of matter to radiation. The effect on the radiation itself provides the detectable evidence of the interaction, in most instances, and in turn provides information on the reaction of the material system and on the physical characteristics of the material. Alternatively one could choose to look upon the effect on the field as being the important consequence of the interaction; for example as the provision of a new source of radiation (chapters 2, 4, 5 and 7). We have chosen to order the chapters of this proceedings by considering the "degree of alteration" of the material system appropriate in each. Thus in chapters 1 and 2 we are concerned primarily with phenomena in which the material plays (over a period of time) a passive role. Spontaneous processes in which the fractional excitation of the material is negligible are discussed in chapters 3 and 4. Stimulated absorption, emission and Raman scattering appear in chapters 2-7; for such processes the material is excited, ultimately to saturation. The populations of the material states can actually become reversed (on an appropriate time scale) for the coherent mechanisms described in chapter 8. Finally the role of the radiation is, in chapter 9, to change completely the state of the material.

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We will discuss firstly a few ideas which are common throughout most of nonlinear optics.

Just as the radiation-matter interaction is the physical basis of nonlinear optics so is the interaction-Hamiltonian, which is discussed formally in chapter 4, the basis for the mathematical description of NLO phenomena. One uses this Hamiltonian to determine the redistribution of charge in the material, on a *microscopic* scale, due to the radiation field. This redistribution corresponds, on a *macroscopic* scale, to a polarization. The complete radiation field must then be determined, in a self-consistent manner, by including this polarization in the electromagnetic wave equations. In practice the vast majority of NLO processes are described adequately by applying the "electric-dipole approximation" to the Hamiltonian. In order to use this approximation the interaction is considered firstly within a "polarizable unit" of the material system. This is a volume in which the electromagnetic field can be assumed to be uniform at any given time. For a gas this volume encloses a single atom or molecule, the dimensions of which are small compared to the radiation wavelength. In a solid the unit is a volume large by comparison to atomic dimensions but again small compared to the wavelength. The total polarization set up in the medium is then the summation over all polarizable units, each term in the sum containing a spatial factor appropriate to the average radiation field in that unit ($\mathbf{E}(t)$). As the field is uniform within a unit the radiation interaction looks formally like that of an electric-dipole in a constant field. For example in atomic systems it is appropriate to use the form:

$$H_{\text{interaction}} = -e\mathbf{r} \cdot \mathbf{E}(t).$$

The realm of nonlinear optics, as far as the radiation frequencies are concerned, is precisely that in which the electron-dipole approximation is relevant; that is the spectral region from the far-infrared to the ultraviolet. For shorter wavelengths \mathbf{E} can no longer be assumed to be uniform over atomic dimensions whilst in the microwave and longer wavelength regions the "magnetic-dipole approximation" is more relevant.

It is because of the applicability of the electric-dipole approximation that one is able to frame much of nonlinear optics in terms of *susceptibilities*. That is, the macroscopic polarization set up in the material can be expanded in terms that are directly proportional (spatially) to some power of the field amplitude.

$$\mathbf{P}(\mathbf{r}, t) = \chi^{(1)}\mathbf{E}(\mathbf{r}, t) + \chi^{(2)}\mathbf{E}(\mathbf{r}, t)\mathbf{E}(\mathbf{r}, t) + \dots$$

The susceptibilities, $\chi^{(n)}$, are functions only of the radiation frequencies and of the material characteristics; they are independent of position when the electric-dipole approximation holds. The usefulness of such an expansion

also relies on the dominance of the lower order terms. This dominance occurs in all but the high saturation cases (chapter 8). In the above formulation of H_{int} and of \mathbf{P} , \mathbf{E} and \mathbf{P} are treated as classical parameters; it is essential to quantize the material states however in order to obtain expressions for the susceptibilities $\chi^{(n)}$. This “semi-classical” treatment of the interacting radiation-matter system is appropriate to nearly all of those processes which we have chosen to include in NLO; the exceptions are spontaneous emission and Raman scattering.

The common denominators in the description of most of the NLO phenomena with which our lecturers were concerned are thus: (i) the usefulness of the electric-dipole approximation and of the semi-classical treatment for the radiation-matter interaction, (ii) the consequent polarization expansion in which specific, low-order terms dominate and in which nonlinear susceptibilities are meaningful. We will now turn to discussion of the individual phenomena.

There are many independent parameters in nonlinear optics that can suitably be formulated in the dimension of time. In any particular experiment the relative values of these time scales determines the nature of the NLO process and, to a large extent, the mathematical technique that can most efficiently be used to describe the process. For this reason much of the remainder of this introduction will deal with time scales.

In order to run through the physical processes of NLO in some sort of logical manner it is convenient to begin with *passive* or “*frequency mixing*” processes, in which the material acts essentially as a catalyst. In all of NLO one approximate time restriction is that the period over which measurements are made be long compared to the radiation oscillation periods, ω^{-1} . Hence energy (or frequency) conservation is applicable.

For passive processes this conservation is amongst the frequency components of the radiation field. The lower order passive processes, all of which are described by nonlinear susceptibilities, are collated in Table I. In general a component of the polarization at frequency ω_r , ($P(\omega_r) \propto \exp -i\omega_r t$), set up by components of the radiation field at ω_a, ω_b etc., is expressed as:

$$P^{(n)}(\omega_r = \omega_a + \omega_b + \dots) = \chi^{(n)}(\omega_r; \omega_a, \omega_b \dots) E(\omega_a) E(\omega_b) \dots,$$

with suitable directional dependences (see chapter 1).

In order to discuss the time inequalities appropriate in NLO it is instructive to consider a two-level electronic system (most NLO phenomena involve electronic processes). Supposing the distributions in the pure states to be given by ψ_0 and ψ_1 then in a perturbing field one can write the eigenfunctions generally as:

$$\psi(\mathbf{r}, t) = c_0(t)\psi_0(\mathbf{r}, t) + c_1(t)\psi_1(\mathbf{r}, t)$$

TABLE I. Passive nonlinear optical phenomena. ($\omega = 0$ indicates the presence of a uniform electric field.)

Frequencies of incident fields	Frequencies of fields generated by the polarization of the medium	Susceptibility	Process
ω_1	No polarization	$0 (\epsilon = 1)$	Vacuum propagation
ω_1	ω_1	$\chi^{(1)}(\omega_1; \omega_1)$	Linear dispersion
ω_1, ω_2	$\omega_3 [\omega_3 = \omega_1 + \omega_2]$	$\chi^{(2)}(\omega_3; \omega_1, \omega_2)$	Sum mixing
ω_1	$\omega_3 [\omega_3 = 2\omega_1]$	$\chi^{(2)}(\omega_3; \omega_1, \omega_1)$	Second harmonic generation
$\omega_1, 0$	ω_1	$\chi^{(2)}(\omega_1; \omega_1, 0)$	Electro-optic linear Kerr effect
ω_1	$\omega_2, \omega_3 [\omega_1 = \omega_2 + \omega_3]$	$\chi^{(2)}(\omega_2; -\omega_3, \omega_1)$	Difference-frequency mixing
ω_1	$\omega_2 [\omega_1 = 2\omega_2]$	$\chi^{(2)}(\omega_2; -\omega_2, \omega_1)$	Degenerate difference-frequency
ω_1	0	$\chi^{(2)}(0; -\omega_1, \omega_1)$	Inverse electro-optic effect
$\omega_1, \omega_2, \omega_3$	$\omega_4 [\omega_4 = \omega_1 + \omega_2 + \omega_3]$	$\chi^{(3)}(\omega_4; \omega_1, \omega_1, \omega_1)$	Third harmonic generation
ω_1, ω_2	$\omega_3, \omega_4 [\omega_1 + \omega_2 = \omega_3 + \omega_4]$	$\chi^{(3)}(\omega_3; -\omega_4, \omega_1, \omega_2)$	Four-wave difference-frequency mixing processes
ω_1	$\omega_2, \omega_3, \omega_4 [\omega_1 = \omega_2 + \omega_3 + \omega_4]$	$\chi^{(3)}(\omega_2; -\omega_3, -\omega_4, \omega_1)$	
ω_1	ω_1	$\chi^{(3)}(\omega_1; \omega_1, -\omega_1, \omega_1)$	Intensity-dependent refractive index
$\omega_1, 0$	ω_1	$\chi^{(3)}(\omega_1; 0, 0, \omega_1)$	Quadratic Kerr effect

The major *microscopic* problem of NLO is to calculate the coefficients c_0, c_1 .

One introduces a dephasing time T_2 , corresponding to transitions out of the upper level ψ_1 , and assumes the perturbing field to be weak enough that $c_0(t)$ is close to unity always. For times long compared to the dephasing time $|c_1(t)|^2$ is constant:

$$|c_1(t)|^2 = \frac{|ez_{10}E/\hbar|^2}{(\omega_{10} - \omega)^2 + T_2^{-2}}$$

ez_{10} is the transition dipole-moment, $\hbar\omega_{10}$ the energy level separation and E the component of the radiation field at frequency ω . The lifetime of electrons

in the lower level, with respect to radiative excitation, is $T_2 |c_1|^{-2}$ which we shall call τ_a .

One can now demand that for passive, linear dispersion the excitation lifetime is long compared to the observation time (so that the energy absorbed from the field is small compared to the energy, $\hbar\omega_{10}$ per particle, of the material) and that this in turn is long compared to T_2 . Under these conditions $|c_1(t)|^2$ is automatically small, as required in the perturbation treatment. For nonlinear processes it is necessary that the above inequalities apply to all radiation frequencies present and to all pairs of material states. It is adequate to use standard perturbation theory to obtain the susceptibilities under two provisos. Firstly for frequencies in the vicinity of resonances one must introduce damping or dephasing times phenomenologically. Secondly one must never contravene Pauli exclusion, even between the intermediate levels involved in nonlinear processes. The latter proviso places restrictions on the summations appearing in susceptibilities. As the *density matrix* has elements $c_i^* c_j$, which are just those combinations of the c -coefficients which appear in populations and in polarization expressions, density matrix theory is a very useful method of describing NLO mechanisms. In this theory one automatically includes the initial state occupancies and the damping factors may be introduced in a relatively formal manner (chapter 1). Whichever way $\chi^{(n)}$ or $\mathbf{P}^{(n)}$ is obtained the appropriate nonlinear polarization must then be included in the wave-equations and these solved for the characteristics of the radiation field \mathbf{E} in self-consistent manner. This is the *macroscopic* problem of NLO.

For the passive processes the susceptibilities are predominantly real. As resonances are approached susceptibilities become complex and under certain conditions may be considered as totally imaginary. It is the imaginary parts of $\chi^{(1)}$ and $\chi^{(3)}$ respectively which describe *linear absorption* and *Raman scattering*. In general energy can be exchanged between the radiation and the material (over a period of time $\gg \omega^{-1}$) only for those mechanisms described by non-linear susceptibilities with "paired frequencies" (see Table II). In

TABLE II. Active nonlinear optical phenomena

Susceptibility	Process
$\chi^{(1)}(\omega_1; \omega_1)$	Linear absorption ($\omega_1 \simeq \omega_{10}$)
$\chi^{(3)}(\omega_2; \omega_1, -\omega_1, \omega_2)$	Raman scattering ($\omega_2 \simeq \omega_1 \mp \omega_{10}$)
$\chi^{(3)}(\omega_1; \omega_1, -\omega_1, \omega_1)$	Two photon absorption ($2\omega_1 \simeq \omega_{10}$) or Saturable absorption ($\omega_1 \simeq \omega_{10}$)
$\chi^{(5)}(\omega_2; \omega_1, \omega_1, -\omega_1, -\omega_1, \omega_2)$	Hyper-Raman scattering ($\omega_2 \simeq 2\omega_1 \mp \omega_{10}$)

other situations $\chi^{(n)}$ may be resonant or imaginary but will lead only to enhancement of the relevant passive process.

If one thinks of a set of two level systems in which the ground states only are occupied initially, then under equilibrium conditions an upper level population is acquired such that the rate of excitation due to "stimulated" absorption from the ground state is balanced by de-excitation via (a) stimulated emission, (b) spontaneous emission and (c) non-radiative decay mechanisms. Setting a lifetime T_1 for relaxation by processes (b) and (c), with level populations N_0, N_1 :

$$\frac{N_0 - N_1}{\tau_a} = \frac{N_1}{T_1}$$

For normal (Beer's law) absorption conditions τ_a must be large compared to T_1 . As these times become comparable one moves into the region of *saturated absorption* where for example the effective absorption coefficient becomes intensity dependent. For a Lorentzian absorption line

$$\alpha = \left[\frac{2\pi\omega N(ez_{10})^2}{n\hbar} \right] \frac{T_2^{-1}}{(\omega - \omega_{10})^2 + T_2^{-2} + \frac{2T_1}{T_2} \left(\frac{ez_{10}E}{\hbar} \right)^2}$$

The intense radiation (small τ_a) bleaches the sample by creating a large upper level population, in turn the rate of absorption must decrease, producing the "power broadened" lines indicated by the equation. This result is obtained by coupling the rate equation to the wave equation. Using the perturbation theory method one therefore appears to get (to lowest order) an effective susceptibility $\chi^{(3)}$ due to the saturation. This effective parameter differs from those discussed hitherto in that it contains T_1 . Also it is not too difficult to pass into the strong saturation region, $T_1 > \tau_a$, in which case it is insufficient to use just the lower order correction.

Stimulated emission exceeds absorption, so providing nett *gain*, in a population inverted system (corresponding to a negative value of $\text{Im } \chi^{(1)}$); stimulated Stokes scattering in a normal system also provides gain (negative $\text{Im } \chi^{(3)}$). Hence both are ripe for *laser oscillation* using cavity feedback. Oscillation occurs providing the round trip gain exceeds losses due to both transmission through cavity ends and absorption processes. Under continuous wave conditions above oscillation threshold the gain will adjust itself so as to be equal to the nett losses. Any optical process which involves exponential gain, that is where the generation of radiation of some frequency ω depends on the prior existence (due to spontaneous noise say) of radiation at ω , will in principle allow for oscillation. Such is the parametric oscillator

case (chapter 2). Conversely, for example, second harmonic production requires only the presence of the fundamental field initially, no oscillation is possible.

Everything we have said so far can apply to continuous operation, but a large number of NLO effects occur only as a consequence of *transients* or for *pulsed operation*. For example, in association with absorption there is always an anomalous dispersion (connected through the Kramers-Krönig relations). Hence associated with saturated absorption there is a nonlinear refractive index change which is proportioned to the radiation intensity. The time-dependent refractive index that will therefore be created under pulsed operation leads to pulse reshaping effects (self-steepening and self-phase modulation). If the pulse rise-time is short compared to the instantaneous value of τ_a and to T_1 then the saturation is unable to follow the pulse. The above adiabatic following process will not occur but in this non-equilibrium case the leading edge of the pulse will experience excess absorption (because the material has been unable to bleach) and vice versa. Hence again one achieves pulse reshaping.

The non-equilibrium absorption is mirrored in the gain for pulsed emission (laser). If the radiation lifetime t_c in the cavity is long then for a small transient increase in the pumping rate one can obtain a build up of radiation. The lifetime τ_a is inversely proportional to the radiation intensity so it in turn decreases. This coupling of the electron population and the radiation produces oscillations in each; in the emission these are called relaxation oscillations or spiking, having period roughly $(t_c T_1)^{1/2}$ and being damped over a time of the order of T_1 . The output of a *Q*-switched laser is essentially the initial spike of the relaxation oscillations. By decreasing the loss suddenly, rather than increasing a pumping rate (in this case sufficiently to take the gain well above threshold) then, if t_c is small by comparison to T_1 and to the pumping time, the radiation density in the cavity builds up and is emitted over a period of the order of t_c —the *Q*-switched pulse. A narrower pulse still is achieved by mode locking. This can be obtained if the gain profile of the laser is broad enough to allow several axial modes to oscillate simultaneously. By introducing, for example, a saturable absorber that bleaches only for the more intense fluctuations in the resulting cavity-radiation one achieves a series of pulses separated by the round-trip cavity period $2c/nL$ with pulse times limited by the recovery time of the absorber and by the dephasing time of the laser medium itself. Once the latter time scale is achieved coherent effects come into play causing decomposition of the pulse.

For times small compare to T_2 the perturbed state of the medium is a coherent mixture of ground and excited states. It is essential to include T_2 and T_1 in the c -coefficient equation rather than introducing T_1 in separate