Introduction to Nonlinear Optics

Since the early days of nonlinear optics in the 1960s, the field has expanded dramatically, and is now a vast and vibrant field with countless technological applications. Providing a gentle introduction to the principles of the subject, this textbook is ideal for graduate students starting their research in this burgeoning area.

After basic ideas have been outlined, the book offers a thorough analysis of second harmonic generation and related second-order processes, before moving on to third-order effects, the nonlinear optics of short optical pulses, and coherent effects such as electromagnetically-induced transparency. A simplified treatment of high harmonic generation is presented at the end. More advanced topics, such as the linear and nonlinear optics of crystals, the tensor nature of the nonlinear coefficients, and their quantum mechanical representation, are confined to specialist chapters so that readers can focus on basic principles before tackling these more difficult aspects of the subject.

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Preface

I set out to write this book in the firm belief that a truly introductory text on nonlinear optics was not only needed, but would also be quite easy to write. Over the years, I have frequently been asked by new graduate students to recommend an introductory book on nonlinear optics, but have found myself at a loss. There are of course a number of truly excellent books on the subject – Robert Boyd's *Nonlinear Optics*, now in its 3rd edition [1], is particularly noteworthy – but none of them seems to me to provide the gentle lead-in that the absolute beginner would appreciate.

In the event, I found it a lot harder to maintain an introductory flavour than I had expected. I quickly discovered that there are aspects of the subject that are hard to write about at all without going into depth. One of my aims at the outset was to cover as much of the subject as possible without getting bogged down in crystallography, the tensor structure of the nonlinear coefficients, and the massive perturbation theory formulae that result when one tries to calculate the coefficients quantum mechanically. This at least I largely managed to achieve in the final outcome. As far as possible, I have fenced off the 'difficult' bits of the subject, so that six of the ten chapters are virtually 'tensor-free'. Fortunately, many key aspects of nonlinear optics (the problem of phase matching, for example) can be understood without knowing how to determine the size of the associated coefficient from first principles.

In the process of writing, I also discovered several areas of the subject that I had not properly understood myself. It is of course always said that the best way to learn a subject is to teach it, so perhaps I should not have been too surprised. There were also things that I had simply forgotten. There was one 'senior moment' when, after struggling with a thorny problem for several hours, I found the answer in a book which cited a paper by Ward and New!

As to the subject itself, I have sometimes teased research students by telling them that nonlinear optics was all done in the 1960s. This statement is of course manifestly ridiculous as it stands – what have all those thousands of papers on nonlinear optics published in the last 40 years been about then? On the other hand, the statement that *all the basic principles* of nonlinear optics were established in the 1960s, does embody an element of truth. There really are very few fundamental concepts underlying the nonlinear optics of today that were not known by 1970. What has happened is not so much that new principles have been established, but rather that the old principles have been exploited in new ways, in new materials, in new combinations, in new environments, and on new time and distance scales. Technology has advanced so that many possibilities that could only be dreamt of in the 1960s can now be realised experimentally and, more than that, may even be intrinsic to commercial products. This point is elaborated in Section 1.12.

The book itself has a modular rather than a serial structure. Consequently, some chapters stand on their own feet, and most readers will probably not want to read the ten chapters in sequence. Beginners should certainly start with Chapter 1, but a reader wanting to get up to speed on crystal optics could read Chapter 3 on its own, while someone interested in dispersion could jump in at Chapter 6. The purpose of both these chapters is to support those that follow. On the other hand, readers who are not interested in crystallography should avoid Chapters 3 and 4, while those who are not concerned with the tensor nature of the nonlinear coefficients should certainly skip Chapter 4 and maybe the start of Chapter 5 too. And anyone who is allergic to quantum mechanics should be able to survive the entire book apart from Chapter 8 and parts of Chapter 9. Of course, some people may be looking for more detailed information, which they may be able to find in one of the nine appendices.

The following table shows what the reader can expect in each chapter; ticks in brackets imply a small amount of material.

Chapter	Tensors	Crystallography	Quantum Mechanics
1	×	×	×
2	×	×	×
3	(√)	√	×
4	√	√	×
5	(√)	(√)	×
6	×	×	×
7	×	×	×
8	√	(√)	√
9	(√)	×	√
10	×	×	×

As the table suggests, the chapters have a varied flavour. Some are fairly specialist in nature, while others contain mostly bookwork material. Chapter 1 serves as a gentle introduction. The polarisation is expanded in the traditional way as a power series in the electric field (treated as a scalar) with expansion coefficients that are regarded as constants. A range of potential nonlinear interactions is explored in this way. The problem of phase mismatch emerges naturally from the discussion. Although in one sense the procedure amounts to little more than cranking a mathematical handle, a surprisingly large number of important nonlinear processes can be discovered in this way. At the end of the chapter, attention is drawn to the shortcomings of the simple approach adopted, and the necessary remedies are outlined. The chapter ends with a brief overview of the entire field.

Chapter 2 starts with a detailed analysis of second harmonic generation, based on Maxwell's equations and proceeding via the coupled-wave equations. The fields and polarisations are now complex numbers, but their vectorial nature is still not taken into account. The discussion broadens out into sum and difference frequency generation, optical parametric amplification and optical parametric oscillators. The chapter ends with a treatment of harmonic generation in focused beams.

In Chapter 3, real nonlinear media are encountered for the first time. The chapter focuses on the linear optics of crystals, and contains a fairly detailed treatment of birefringence in uniaxial crystals. The principle behind quarter- and half-wave plates is outlined. A section on biaxial media is included at the end.

Chapter 4 deals with second-order nonlinear effects in crystals, and there is now no way to avoid defining the nonlinear coefficients with all their tensor trappings. Though I have tried hard to keep it simple (as much material as possible has been relegated to Appendix C), the treatment that remains is still rather indigestible. Permutation symmetries are outlined, the contracted suffix notation for the nonlinear coefficients is introduced, and the Kleinmann symmetry condition is explained. The structure of the nonlinear coefficients for the three most important crystal classes is highlighted, and examples are given to show how the results are applied in some typical nonlinear interactions.

Chapter 5 is devoted to a selection of third-order nonlinear processes. Although the coverage is restricted to amorphous media, there are still places (especially early in the chapter) where the tensor structure of the coefficients in unavoidable. The later parts of the chapter deal with stimulated Raman scattering, acousto-optic interactions, and stimulated Brillouin scattering. A section on nonlinear optical phase conjugation is included at the end.

Chapter 6 focuses on dispersion. Although basically a linear effect, dispersion plays a crucial role in many nonlinear processes, and the material covered here therefore provides essential background for Chapter 7. After a general introduction, the evolution of a Gaussian pulse in a dispersive medium is analysed in detail, and this leads on naturally to a discussion of chirping, and pulse compression. A hand-waving argument then enables a rudimentary pulse propagation equation to be deduced, which provides an opportunity to introduce the local time transformation.

Nonlinear optical interactions involving short optical pulses are treated in Chapter 7. The choice of examples is somewhat arbitrary, but the selection covers quite a wide range, and includes self-phase modulation, nonlinear pulse compression, optical solitons, second harmonic generation in dispersive media, optical parametric chirped pulse amplification (OPCPA), pulse diagnostics, and the phase stabilisation of few-cycle optical pulses.

Up to this point in the book, the strength of the nonlinear interactions has been represented by the value of the appropriate nonlinear coefficient or, more exactly, the set of numbers contained in the non-zero tensor elements for the process in question. Chapter 8 focuses on the quantum mechanical origin of the coefficients. The perturbation theory formulae that emerge are large and cumbersome and, to this extent, they may leave the reader not much the wiser! However, every effort has been made to simplify things by taking special cases, and by highlighting dominant terms in the equations, the structure of which is relatively simple. The discussion continues in Chapter 9, which deals with resonant effects including self-induced transparency (SIT) and electromagnetically-induced transparency (EIT).

No book on nonlinear optics would be complete without a mention of high harmonic generation (HHG), and the quest for attosecond pulses. The treatment of these topics in Chapter 10 is essentially classical, and certainly simple. So if the promise in the title of an introductory treatment to the subject has not always been fulfilled in some of the intermediate chapters, at least it is kept in the last chapter.

Some more detailed material is included in the appendices, the last of which contains values of useful constants in nonlinear optics.

The varied nature of the material covered in the book has affected how the reference list has been compiled. A comprehensive list of sources would be almost as long as the book itself, so I have had to adopt a selective policy on which papers to refer to. Broadly speaking, references are of three types: historical papers describing things done for the first time, books and recent journal articles where an aspect of the subject is reviewed, and (in a few cases) papers on topics of particular current interest. To find further sources, it is always worth using the Internet. The information available there may not always be of impeccable quality, but one will almost always be led to an authoritative source within a few minutes.

There are bound to be errors of one sort or another in any book of this type. I have of course made every effort to avoid them, but I doubt if anyone has ever written a physics book without getting a few things wrong. Colleagues who read the book in draft picked up quite a few glitches, but I am sure there are more. If you find something that you think is wrong, please e-mail me on g.new@imperial.ac.uk, so that I can build up a corrections file.

Geoff New
Le Buisson de Cadouin
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