Applied Classical Electrodynamics

Volume 2: Nonlinear Optics

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Preface

This is the second book of a two volume work on the classical physics of optical interactions. The two volumes deal with linear and nonlinear optics, respectively. These volumes are teaching texts developed for the academic program at the Optical Sciences Center of the University of Arizona. They are designed to give a background on issues in optical physics that relate to material science and laser applications. The emphasis on classical physics reflects the fact that most practical applications involve the classical limit. Matter obeys quantum mechanics and the interaction of radiation with matter cannot be developed with complete consistency from a model of electrons on springs developed in Volume I. Nonetheless, once the conceptual difficulties resolved by quantum mechanics are dealt with, nearly all remaining cases in applied optics can be modeled classically. Classical mechanics leads to a useful phenomenology of considerable breadth of application.

The design of the volumes is modular, and there is considerable flexibility in the order in which the text can be read or taught. In the Table of Contents, the chapters, and in some cases the sections, contain, in parentheses, a reference back to the basic material needed for the chapter. Most basic material is in the first three chapters. Some supplemental techniques are discussed in Chapter 10. Otherwise one can read the book in almost arbitrary order. In addition, each topic is developed starting from a fairly elementary level. One need not master all of the background material if all one is interested in is an overview of the ideas. The problem sets are designed to tie the book together into a coherent whole, and deal with conceptual issues that might otherwise become diversionary. Those who wish to do the problems should be aware that the degree of difficulty varies substantially. Each problem has been tried at least once by one of the authors (the first author has answered all problems to within factors of ε_0).

The second volume draws heavily on the concepts in Volume I, but not necessarily on the algebra. For practical applications, Volume II can be used without Volume I. Only in dealing with the Kerr nonlinearity and stimulated Brillouin scattering is there so much algebra involved that we refer extensively to material in the first volume. We have found it to be difficult to organize and present the field of nonlinear optics in its entirety in a simple way. The basic problem is that there is a great richness in nonlinear phenomena, and new areas are developed every year. In addition, there are a large number of different principles that must be integrated into any one particular case, and each case calls for a different combination of principles. Since we have no crystal ball that will clarify what will become important in the future, we take an approach that emphasizes those aspects of nonlinear optics that have proved to be of most value over the past two decades, and are therefore likely to continue to be of importance in the future. We try, however, to maintain contact with areas of current interest, and areas that have been of interest in the past, but which have been temporarily abandoned for lack of suitable lasers or nonlinear materials.

The books are designed to allow a reader with a background in classical mechanics and electromagnetism to learn enough to continue on with texts that specialize in the specific topics. Thus each topic is not covered in great detail, and we have tried to judge which areas are adequately covered in existing texts to avoid duplication. This volume assumes that the interested reader is willing to use recent books by Shen (general discussion of relevant experimental techniques), Yariv (stimulated scattering and general background), and Zernike and Midwinter (parametric oscillators, materials, and optimization of nonlinear interactions).

This book is printed in camera ready form from a personal computer. The software package we use is still under development by M. Sargent III and colleagues. The experimental software imposes some defects and constraints.

We gratefully acknowlege the useful suggestions of G. Salamo, E. van Stryland, and A. Smirl who have been using preliminary versions of these volumes in their courses.

Tucson Arizona
October 5, 1985

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[1] The number in parenthases refers to earlier chapter or section needed as background.

13
Introduction to Nonlinear Optics

One of the benefits of mastering the classical physics of optics comes from its application to nonlinear optics. Quantum mechanics is needed primarily to provide noise sources to seed the growth of stimulated scattering; otherwise most nonlinear phenomena are classical. Most of nonlinear optics is a generalization of electro- and acousto-optics. Indeed there is no precise dividing line that distinguishes linear from nonlinear optics. Similarly, there is no precise division between nonlinear optics and electrical engineering.

Most basic principles that govern nonlinear optics, especially crystal optics (Chapter 7) and slowly varying techniques (Chapter 10), have been covered in detail in Volume I. This volume is largely self-contained insofar as the relevant formulae and nomenclature are summarized in Appendix C of this volume for quick reference. For the beginner, it is best to master at least some of Volume I before trying this one.

13.1 NONLINEAR POLARIZATION

The nonlinear polarization is formally written as

$$\mathbf{P}(\mathbf{r},t) = \varepsilon_0 \boldsymbol{\chi}^{(2)} : \mathbf{E}(\mathbf{r},t)^2 + \varepsilon_0 \boldsymbol{\chi}^{(3)} \vdots\, \mathbf{E}(\mathbf{r},t)^3 + \cdots \tag{13.1}$$

where terms of fourth order and higher in **E** have been dropped since they normally play no role in nonlinear optics. The two nonlinear terms are referred to as quadratic and cubic, and in all cases discussed in this volume, $\mathbf{P}(\mathbf{r},t)$ is a weak polarization so that the methods of Chapter 10 can be used to calculate radiated fields. The notation in nonlinear optics can be confusing, especially in the usage of $\boldsymbol{\chi}^{(3)}$, and we caution the reader to carefully compare the conventions used between any two sources before comparing any results. We have used what we believe to be current notation.

Equation (13.1) is not to be taken as literal truth. It is to be understood in the same spirit as the linear susceptibility in that the χ's are to be associated with the amplitudes of plane-wave fields.

13.1.1 Three-Wave Mixing

The general nonlinear process using the quadratic nonlinearity involves three-wave mixing in which two fields are combined to produce a third. We then take $\mathbf{E}(\mathbf{r},t)$ to be written as

$$\mathbf{E}(\mathbf{r},t) = \frac{1}{2}\,[\boldsymbol{E}_1(\mathbf{k}_1,\omega_1)e^{i(\mathbf{k}_1\cdot\mathbf{r}-\omega_1 t)} + \boldsymbol{E}_2(\mathbf{k}_2,\omega_2)e^{i(\mathbf{k}_2\cdot\mathbf{r}-\omega_2 t)} + \mathrm{cc}]. \quad (13.2)$$

The two fields are written explicitly as eigenvectors of the medium. The square of the field is then really a dyad product and reads

$$\mathbf{EE} = \frac{1}{4}\,[\boldsymbol{E}_1\boldsymbol{E}_1 e^{i(2\mathbf{k}_1\cdot\mathbf{r}-2\omega_1 t)} + 2\boldsymbol{E}_1\boldsymbol{E}_2 e^{i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{r}-i(\omega_1+\omega_2)t}$$
$$+ \boldsymbol{E}_1\boldsymbol{E}_1^* + 2\boldsymbol{E}_1\boldsymbol{E}_2^* e^{i(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r}-i(\omega_1-\omega_2)t} + \cdots + \mathrm{cc}]. \quad (13.3)$$

The terms omitted from Eq. (13.3) duplicate effects that are already included in the terms that are shown explicitly. The frequencies ω_p and wave vectors $\mathbf{k}_p$ of the nonlinear polarization are written as

$$\omega_p = \omega_i \pm \omega_j \qquad i,j = 1,2$$
$$\mathbf{k}_p = \mathbf{k}_i \pm \mathbf{k}_j. \quad (13.4)$$

The four possible combinations and degeneracies in Eq. (13.3) give rise to the phenomena of second harmonic generation, sum frequency generation, optical rectification, and difference frequency generation. Except for the rectification signal, which plays no important role in nonlinear optics, each of these terms gives rise to a phased array of dipoles that can radiate as discussed in Section 10.4. The radiated field must also be an eigenvector. Each triplet of eigenvalues corresponds to a possible three-wave interaction which, under appropriate conditions of phase matching, can yield efficient generation of the frequency ω_p. The three waves are denoted a,b,c such that

$$\omega_c > \omega_b > \omega_a > 0, \quad (13.5)$$

$$\omega_c = \omega_a + \omega_b, \quad (13.6)$$

and the phase match condition (see below) is written in terms of a wave vector mismatch term $\Delta\mathbf{k}$ defined as

$$\Delta\mathbf{k} = \mathbf{k}_a + \mathbf{k}_b - \mathbf{k}_c. \quad (13.7)$$

The wave triplets defined by Eqs. (13.5) and (13.6) are collectively referred to as a three-wave interaction. As we show later, the three-wave interaction is not merely a convenience. Each triplet is tied together through its own set of Manley-Rowe relations. Manley-Rowe relations are touched on briefly in Chapter 11. They play a major role as the primary conservation laws in nonlinear optics. A

Manly-Rowe relation states that waves interchange energy in units of flux over frequency. When divided by Planck's constant, they are interpretable as quanta. These quanta are classical objects. Fractional photons can be interchanged. Because quantum terminology is standard, it is used throughout this volume. The reader should understand that no quantum mechanics is implied by the terms.

Each three-wave interaction involves complicated dot products between the fields and $\chi^{(2)}$. These products always lead to a single scalar coefficicient χ_{eff} that characterizes the three-wave interaction. This also allows the definition of an effective polarization amplitude P_{eff} (see Section 10.4 for the definition and justification of P_{eff}) as

$$P_{eff}(\omega_c) = \varepsilon_0 \chi_{eff}^{(2)}(-\omega_c;\omega_b,\omega_a)\, E(\omega_a)\, E(\omega_b). \tag{13.8a}$$

Similarly for the polarization field generated at the frequencies ω_a and ω_b,

$$P_{eff}(\omega_a) = \varepsilon_0 \chi_{eff}^{(2)}(-\omega_a;\omega_c,-\omega_b)\, E(\omega_c)\, E^*(\omega_b) \tag{13.8b}$$

and

$$P_{eff}(\omega_b) = \varepsilon_0 \chi_{eff}^{(2)}(-\omega_b;\omega_c,-\omega_a)\, E(\omega_c)\, E^*(\omega_a). \tag{13.8c}$$

These serve to define the meaning of $\boldsymbol{\chi}^{(2)}$ in Eq. (13.1) in terms of the field amplitudes. By convention the output field frequency is always written as a negative frequency in the susceptibility. Whenever ω_a or ω_b enter the susceptibility with a minus sign, this means that the optical fields appear as the complex conjugate part of $\mathbf{E}(\omega_a)$ or $\mathbf{E}(\omega_b)$, i.e., as $E^*(\omega_a)$ or $E^*(\omega_b)$.

13.1.2 Four-Wave Mixing

The development of the cubic term in Eq. (13.1) proceeds in the same way as the development of the quadratic term. It is left as an exercise to show that if Eq. (13.2) is generalized to include four waves, then the frequencies and k-vectors are found in the combinations

$$\begin{aligned} \omega_p &= \omega_i \pm \omega_j \pm \omega_k \\ & \qquad\qquad i,j,k=1,2,3 \\ \mathbf{k}_p &= \mathbf{k}_i \pm \mathbf{k}_j \pm \mathbf{k}_k. \end{aligned} \tag{13.9}$$

The most general four-wave interaction is defined by four frequencies

$$\omega_a > \omega_b > \omega_c > \omega_d > 0. \tag{13.10}$$

There are two possible combinations of frequencies and k-vectors that can satisfy Eqs. (13.9) and (13.10). These are

$$\omega_d = \omega_a + \omega_b + \omega_c, \tag{13.11}$$

$$\Delta \mathbf{k} = \mathbf{k}_a + \mathbf{k}_b + \mathbf{k}_c - \mathbf{k}_d \tag{13.12}$$

and

$$\omega_d = -\omega_a + \omega_b + \omega_c, \tag{13.13}$$

$$\Delta \mathbf{k} = -\mathbf{k}_a + \mathbf{k}_b + \mathbf{k}_c - \mathbf{k}_d. \tag{13.14}$$

Only a few four-wave processes have been found to be useful and a standard jargon has been developed for these which are discussed on a case by case basis. The four-wave susceptibilities are defined in the same way as the three-wave susceptibilities in Eqs. (13.8a)-(13.8c).

13.2 DEGENERACY

Three- and four-wave mixings are often referred to as being degenerate. The terminology can have several meanings. The condition in which several frequencies are the same is referred to as frequency degenerate. For example, second harmonic generation is a frequency degenerate three-wave interaction in which $\omega_a = \omega_b$ and $\omega_c = 2\omega_a$. Frequency degeneracy is not necessarily a true degeneracy, which requires that two or more of the eigenvectors in the N-wave mixing be the same eigenvector. When unqualified, we limit the term degeneracy to refer to true degeneracy. The term nondegenerate refers to the absence of a true degeneracy. Deriving formulae for degenerate interactions from those describing nondegenerate interactions requires some delicacy, and we usually treat these as special cases.

13.3 PHASE MATCH CONSTRAINTS

The polarizations developed above radiate as a phased array of dipoles. This process is developed at length in Chapter 10 and the relevant formulae are given in Appendix C for quick access. Three-wave mixings are usually strongly constrained such that, except for the uninteresting rectification effect, only one three-wave interaction can occur for any one configuration of input beams and medium. Any one three-wave interaction can be solved with sufficient detail to be usefully applied to nonlinear devices. Hence we discuss three-wave interactions in great detail.

With four-wave interactions, at least two terms are always phase matched. The case where all four waves are degenerate means that $\Delta \mathbf{k} = 0$, which describes either self-focusing or two-photon absorption (see Chapter 3) depending on the circumstances. The case where $E(\omega_a)$ and $E(\omega_b)$ are degenerate, and $E(\omega_c)$ and $E(\omega_d)$ are also degenerate is also phase matched. This describes stimulated scattering. Thus four-wave mixing requires that careful attention be paid to competing processes. In all cases, it is the form of $\chi_{eff}^{(3)}$ that identifies a four-wave interaction. Four-wave interactions can also be coupled, i.e., one four-wave process can drive another one. Most of the time, phase match constraints inhibit coupling, but not always. Because of the competing processes and the couplings it is not usually worthwhile to develop the theories of four-wave mixing processes in the same detail as for three-wave mixings. Exceptions exist, but they involve special applications to particular cases that are beyond the scope of this book. Therefore, when we deal with four-wave mixing we usually emphasize the principles without going into the detail that characterizes our discussion of three-wave mixing.

13.4 NONLINEARITIES

While the field of nonlinear optics can be loosely grouped into cubic and quadratic processes, we find it to be more efficient to develop the nonlinearities according to their origins in the various force terms of Chapters 2 and 3. Those that originate in the nonlinear force terms derived from the potential given in Eq. (2.10) are constructed by methods that are analogous to those used in Sections 8.1 and 8.2. Those that come about through the Raman-active potential of Eq. (3.37) require special treatment which is given later. With this philosophy, some nonlinear phenomena such as self focusing turn out to be awkward insofar as they bridge both cases. The important phenomena that fall into this intermediate category are discussed in separate sections that emphasize this intermediate status.

There is another organization of nonlinear phenomena that we follow insofar as it saves a substantial amount of repetition. This is the division of nonlinearities into those whose function is catalytic, insofar as the medium plays no role other than to convert one light wave to another, vs. those in which role of the medium is more complex. In catalytic interactions, essentially no energy is exchanged between the optical fields and the medium and hence there is no dissipation of energy via the normal modes. In this case the Manley-Rowe relationships, which describe the interchange of energy flux in an interaction, involve the light wave alone. These types of nonlinear interactions tend to behave in similar ways, and once a few examples are mastered, others can be treated by analogy.

All major applications of quadratic nonlinearities make use of this catalytic nonlinearity. Four-wave mixings are more complicated. Generally, cubic nonlinearities are small. Large nonlinearities occur in the neighborhood of the resonances of the medium. Near resonances, energy is exchanged with the medium, and a complete energy balance requires an examination of the energy transferred to and from the medium. Such a case occurs in acousto-optics. In Chapter 11 it was shown that there is an energy defect in the scattering which goes into or out of the medium. The critical concept that we use in the case of cubic nonlinearity is the phased array of susceptibility. This conveniently organizes cubic processes so that various competing processes can be examined. It also allows an examination of when to expect coupled four-wave mixings. The phased array is also the object through which energy is exchanged.

13.5 ORGANIZATION

The characteristics described above dictate the organization of the text. It begins with a traditional discussion of three-wave mixing, concentrating on the basic nonlinearities, phase matching, and the practical aspects of nonlinear optics. Other aspects of nonlinear optics, principally third-order phenomena are covered later. The major chapters are written modularly so that they do not need to be read in order. It is useful to read at least some of Chapter 15 before reading Chapter 14. This involves deferring the details of how the effective susceptibilities are computed, but this is advantageous for the beginner. These sections cannot be understood without mastering crystal optics and the phase match discussion of Section 10.4.

The sections that deal with Raman scattering are also modular and can be read without reference to the other sections. Chapter 10 is essential to the understanding of Stokes-anti-Stokes coupling. Stimulated Brillouin is more easily understood if Chapter 11 is read first. The remaining areas within the nonlinear optics sections are handled with the goal of keeping the text as brief as possible.

ADDITIONAL READING

Hopf, F.A. and Stegeman G.I., *Applied Electrodynamics Volume I: Linear Optics*, Wiley, New York, 1985.

Beginning texts

Baldwin, C.G., *An Introduction to Nonlinear Optics*, Plenum, New York, 1969.

Advanced texts

Bloembergen, N., *Nonlinear Optics* Benjamin, Reading, Massachusetts, 1965.
Zernike, F. and Midwinter, J.E., *Applied Nonlinear Optics*, Wiley, New York, 1973.
Shen, Y.R., *The Principles of Nonlinear Optics*, Wiley, New York, 1984.

PROBLEMS

13.1. Verify that all four-wave mixings can be written in the form of Eqs. (13.11)-(13.14).

13.2. Identify all possible phased arrays of susceptibility that participate in a single four-wave mixing process for: (a) total nondegeneracy; (b) complete frequency degeneracy but no true degeneracy; (c) true degeneracy between the fields at ω_a and ω_b and also between the fields at ω_c and ω_d but otherwise nondegenerate; and (d) total degeneracy.
Hint: The phased arrays are driven by the squares of the electromagnetic fields. Ignore vector and tensors for this exercise.

14 Catalytic Nonlinearities

When nonlinearities arise from normal coordinates that are far from resonance, the medium does not absorb light. Light is converted from one frequency to another without interchanging energy with the medium and the medium functions as an optical catalyst. Catalytic nonlinearities are closely related to the Pockels effect and the formal development of the equations is similar. The basic physics of the nonlinearity is that a powerful electromagnetic field drives the electron far from its rest position so that the nonlinear force terms of Eqs. (2.10) and (2.14) come into play. Just as in the Pockels effect, dipolar quadratic nonlinearities cannot exist in centrosymmetric media, i.e., media with a center of symmetry. There can be quadrapolar quadratic nonlinearities even in isotropic media, but these require special circumstances to radiate. Initially we ignore quadrupoles and concentrate on media without centers of symmetry that are identified in Table C.1 (copy of Table 8.2) by a nonzero value for the number of independent components of χ_{ijk}.

The physical origins of quadratic nonlinearities lie in the internal structure and relative positions of the molecules in the lattice. The situation in most cases is complicated. One of the simpler cases, and one that is widely met in practice, is the nonlinearity in materials with tetrahedral structures. These occur in KDP and its analogues.* The origin of the nonlinearity of KDP is illustrated in Figures 14.1 and 14.2. This type of nonlinearity is frequently encountered in practice. In Figure 14.1, four atoms are arranged at the corners of a tetahedron. A strong optical field driving the motion of an electron gives the result shown in Figure 14.2.

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* The analogues of KDP are too numerous to list everywhere in this book. The first letter can read K, R, C, or A, which stands for potassium, rubidium, cesium, or ammonium. The second reads D or D*, which stands for dihydrogen or diduterium. The third letter reads P or A, which stands for phosphate or arsenate. KDP, potassium dihydrogen phosphate, has the chemical formula KH_2PO_4. These $\bar{4}2m$ materials are the most widely used in electro-optics and nonlinear optics. Any qualitative discussion of one of these materials applies to the group as a whole.

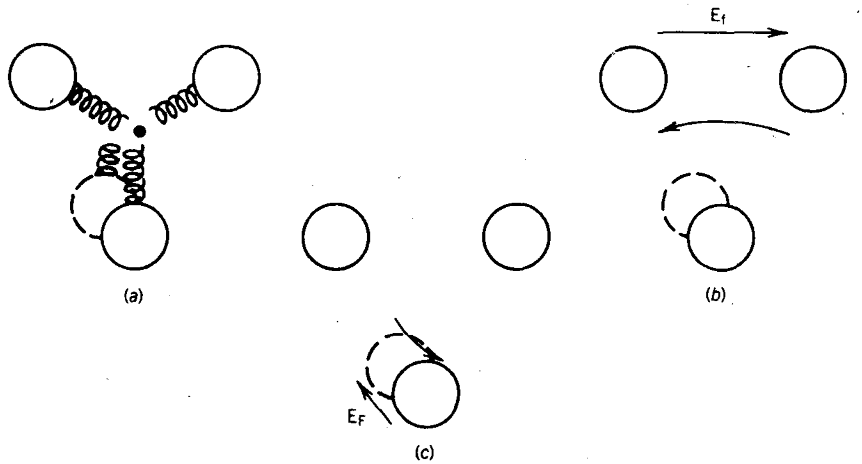

Figure 14.1. The origin of the quadratic nonlinearity in KDP. (a) Electron at rest in the absence of an incident optical field. (b) An optical field incident along the axis connecting the upper two atoms. The electron is deflected downward by the atoms. (c) If the field is along the axis connecting the lower two atoms, the electron is deflected upwards. In (b) and (c) the electron moves along the arc of a circle.

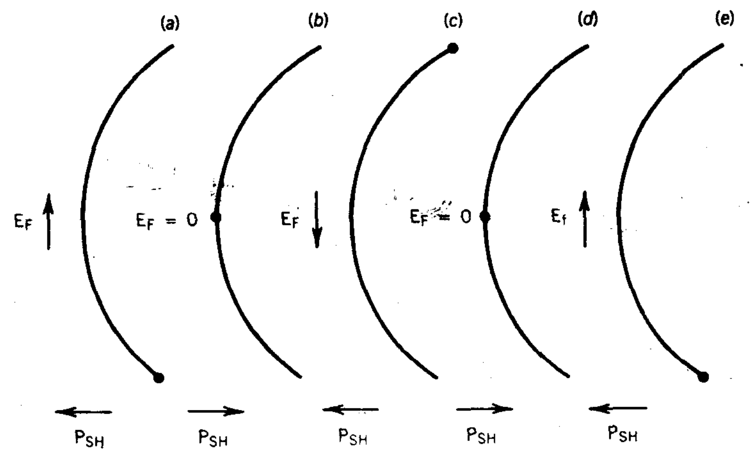

Figure 14.2. A complete cycle of the incident laser field $E_f \equiv E(\omega_f)$ results in two cycles of the second harmonic polarization. (a) E_f is maximum upwards, $P_{SH} \equiv P(2\omega_f)$ points to the left. For (b) and (d) E_f is zero and P_{SH} points to the right. (c) E_f is a maximum down and P_{SH} again points to left. (e) Begins next cycle.

When the field is parallel either to the two atoms at the top, or to the two atoms at the bottom, the electron is forced onto a path that follows an arc of a circle due to the repulsion from one of the atomic cores. Figure 14.2 shows how circular motion leads to second harmonic dipole. As the electron moves along the arc of a circle, the dipole orthogonal to the field makes two complete cycles for every cycle of the fundamental. (In the case of second harmonic generation, the historic nomenclature reads fundamental ≡ laser ≡ a,b and second harmonic ≡ c.) This second harmonic polarization radiates a second harmonic field polarized orthogonal to the incident fundamental field, a situation which utilizes skew elements (e.g., $\chi_{123} \equiv \chi_{14}$) in the nonlinear susceptibility tensor.

14.1 SECOND HARMONIC NONLINEAR TERMS

The general development of second order nonlinearities is somewhat impenetrable at first, largely because of the complexity of the notation. We start by dealing with second harmonic generation, which is notationally somewhat easier than the general case.

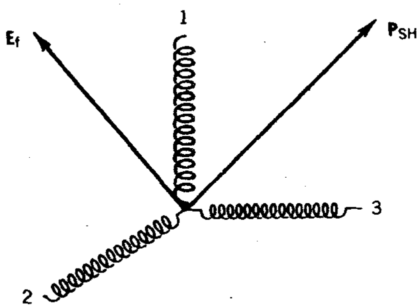

Figure 14.3. Spring system diagonal in the principal axis system. The incident laser field $E(\omega_f)$ and second harmonic polarization fields P_{SH} can point in different directions.

We confine our discussion to the second harmonic polarization. The radiated field at the second harmonic induces polarizations at the laser frequency. We assume in this section that the second harmonic field is so small that we can neglect polarizations induced by the second harmonic fields. We relax this assumption in Section 14.2.

The basic model for the light matter interaction is the electron on a spring system described in Chapters 2 and 8. It is illustrated in Figure 14.3. When a single spring constant is assumed, the formal development is not applicable to monoclinic and triclinic crystals (although the results are qualitatively the same). The laser field is not, for the moment, taken to be an eigenvector, and is written as

$$E_f(r,t) = \frac{1}{2}\,(\mathcal{E}(\omega_f,r)e^{-i\omega_f t} + cc). \qquad (14.1)$$

The equation of motion for this case is given by Eq. (8.8), namely,