Coherent Nonlinear Optics

Recent Advances

Edited by M. S. Feld and V. S. Letokhov

With Contributions by
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With 134 Figures

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1. Coherent Nonlinear Optics

M. S. Feld and V. S. Letokhov

The gold rush phenomenon—an intense period of rapid discovery and exploitation—is a well-known phase experienced by many fields of the natural sciences at certain points in their evolution. Optical physics, stimulated by major advances in laser research and technology, is currently in the midst of such a period. Everywhere new veins of gold are being uncovered and mined by thousands of prospectors, most of whom have crossed over into this exciting research area from a diversity of other disciplines. Their principal tool is the laser and its associated analytical, spectroscopic and dynamical techniques. The purpose of this volume is to make available to our co-workers in optical physics in-depth reports on the current status of a set of important topics in this rapidly changing field.

The laws of optical physics were formulted 50 to 100 years ago. They remain true for laser light and, in fact, form the basis of operation for the laser itself. However, *coherence* and *nonlinearity* are relatively new concepts which have become central to describing the interactions of laser light with matter. These ideas are fundamental to the many new techniques becoming available for studying atoms and molecules. Coherent nonlinear optics is the theme of this book, and the editors have brought together a selected set of specifically prepared reviews of current topics of active interest. Three major areas—coherent resonance effects, multiphoton resonant processes and coherent Raman processes—are covered.

1.1 Introductory Comments

The coherent nature of the interaction of laser light with matter manifests itself in different ways, with interesting applications. About twenty-five years ago DICKE [1.1] pointed out that the spontaneous emission from an ensemble of excited quantum systems can occur at a greatly accelerated rate, via a mechanism he termed *superradiant emission*. In this process the particles respond cooperatively because of their mutual interaction with the common radiation field, giving rise to coherent radiation. DICKE's original discussion considered both optical and microwave emission, but it emphasized the latter regime, where the sample is small compared to the wavelength λ of the emitted radiation. Four years later the laser was

proposed [1.2] as a means of extending the maser principle of generating coherent microwave emission into the optical spectral range. The production of coherent radiation by lasers and masers is based on the principle of *stimulated emission* from excited particles into a small number of modes of an optical resonator (one mode in the ideal case), a process which is distinct from superradiant emission. The laser principle is, of course, now widely used to generate coherent light. Nevertheless, the production of coherent optical radiation via cooperative spontaneous emission from an ensemble of excited particles outside the resonator is of great physical interest. However, the theoretical results developed for long wavelength superradiance are not directly applicable to the optical regime, since the sample volume is much larger than λ^3. This was recognized in the first demonstration of superradiant emission [1.3], and much recent attention, both experimental and theoretical, has been given to the exploration of this interesting effect. The present state of our understanding of superradiance is reviewed in Chap.2 by Feld and MacGillivray.

High-resolution laser spectroscopy is based on the ability of laser radiation to induced nonlinear behavior and phase coherence in atomic and molecular systems. These principles have led to a set of new methods for producing extremely narrow "Doppler-free" spectral resonances in two-level and multilevel systems. The concept of saturating an atomic transition by means of monochromatic laser radiation was first worked out by LAMB [1.4] in his 1974 analysis of a gas laser, and narrow laser saturation resonances were observed shortly thereafter [1.5]. The extension of these ideas to three-level systems soon followed [1.6]. These initial developments have led to a series of techniques which are now standard tools for precision studies of atoms and molecules [1.7,8]. One of the most important recent developments in this area is the extension of the *method of separated fields* to the optical regime [1.7,8]. The separated fields technique is based on the fact that when a beam of atoms traverses a region of electromagnetic field, phase memory of the resonant interaction is preserved. Constructive interference can then occur as the atoms pass through a second field. This property is fundamental for producing very narrow absorption resonances in separated microwave fields [1.9]. Unfortunately, it cannot be directly applied in the optical range because the separation between the fields is large compared to λ. Indeed, because of the divergence of the atomic beam, the atoms traversing the first light beam at a given point, and hence acquiring an optical polarization of a given phase, intersect the second light beam at a range of points, thus giving rise to destructive interference. CHEBOTAYEV and co-workers [1.10] showed that this obstacle can be overcome by using two-photon transitions or, in the case of single-photon transitions, by using three optical fields. These techniques, which are closely connected with the photon echo effect [1.11], are now among the most powerful and elegant tools in the field of high-resolution laser spectroscopy. Coherence in high-resolution laser spectroscopy is reviewed in Chap. 3 by Chebotayev.

Multiphoton processes are one of the main sources of nonlinearity in the interaction of intense laser fields with atoms and molecules. Resonant multiphoton processes are of special interest: First, multiphoton transition probabilities are enhanced under resonance conditions, and can be observed in fields of moderate intensity. Furthermore, such processes find various applications in laser spectroscopy. Two-quantum resonant transitions in a standing wave field is an important method for eliminating Doppler broadening [1.12]. One of the techniques of high-resolution laser spectroscopy is based on this approach [1,7,8]. Resonant multistep processes permit selective photoionization of atoms. This approach is fundamental for laser methods of single-atom detection [1.13], which have recently been demonstrated experimentally [1.14]. The field of multiphoton resonant processes in atoms is reviewed in Chap.4 by Biraben, Cagnac, and Grynberg.

Over the past few years impressive progress has been made in studying multiphoton vibrational transitions in polyatomic molecules induced by intense infrared e.m. fields. The first successful demonstration of isotopically selective multiphoton excitation and dissociation by intense CO_2 laser pulses [1.15], in BCl_3, followed by similar experiments in other molecules, has triggered a torrent of experimental and theoretical activity. This wide research area, now called multiphoton (or multiple photon) infrared photochemistry, has already become the subject of special reviews [1.16,17]. Among these works are numerous papers devoted to the coherent interaction of a multilevel quantum system, the levels of which are almost equidistant, with powerful quasi-resonant radiation. Although models of this type are only simple approximations to a real polyatomic molecule, they do provide a physical basis for describing a variety of the features of such multiquantum processes, and a qualitative picture for their interpretation. The major results of this field to date are summarized in Chap.5 by Cantrell, Letokhov, and Makarov.

In a condensed medium the relaxation time of the phase memory (i.e., phase coherence) induced by an intense optical field is extremely short, in the picosecond range. Nevertheless, progress in generating intense ultrashort laser pulses [1.18] has made possible systematic studies of coherent interactions between picosecond laser pulses and molecular vibrations. The method first used for this purpose [1.19] has proved to be the most productive. In this approach the sample is simultaneously irradiated by two coherent collimated ultrashort light pulses, their frequency difference being exactly equal to the molecular vibrational frequency. This induces Raman-type excitation of the N molecules contained in a coherent interaction volume. An ultrashort pulse of variable delay then probes the state of the system as it decays. Both the intensity and direction of the scattered probe pulse can be studied. Because of the coherent nature of the interaction between the excitation and probe pulses, the interaction efficiency for short delay ($\lesssim$ phase memory time) is proportional to N^2, and depends on the relative orientation of the

wave vectors of the exciting and probe fields. As the molecular vibrations dephase, however, the interaction becomes incoherent, leading to isotropic efficiency proportional only to N. These features make it possible to separate coherent and incoherent processes occurring on a picosecond time scale. Furthermore, since the details of the dephasing process depend on the extent of inhomogeneous broadening of the vibrational transition and its internal structure, picosecond pulse techniques can be used to study these features. Recent progress in the field of coherent picosecond interactions is reviewed in Chap.6 by Laubereau and Kaiser.

The interaction of an ensemble of molecules with two laser fields offset in frequency can give rise to two-quantum Raman transitions. This nonlinear process had led to an important spectroscopic technique, coherent Raman spectroscopy. Although a comprehensive review of this field appeared in 1977 [1.20], many important advances in the basic method, coherent antistokes Raman spectroscopy (CARS), have since occurred. These new developments are presented in Chap.7 by Levenson and Song. In contrast to [1.20], Chap.7 describes the steady-state aspects of the Raman scattering process, rather than the transient ones. In this regime the pulse duration of both exciting and probe fields is long compared to the phase memory relaxation time, T_2, (or even cw), hence excitation and interrogation processes occur simultaneously. Thus, coherence must be maintained at all times by the applied fields. Taken together, Chaps.6 and 7 form a comprehensive review of coherent Raman processes under transient and steady-state conditions.

In summary, the contributions to this volume cover recent advances in three major areas of coherent nonlinear optics, coherent resonance effects (Chaps.2 and 3), multiphoton resonant processes (Chaps.4 and 5), and coherent Raman processes (Chaps.6 and 7). It is hoped that these reviews will serve as useful summaries of recent developments, and perhaps stimulate new advances in the field.

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