Introduction to nonlinear laser spectroscopy

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M. D. Levenson and S. S. Kano, *Introduction to Nonlinear Laser Spectroscopy.*-Revised ed.-Academic Press, Boston; San Diego; New York; Berkeley; London; Sydney; Tokyo; Toronto, 1988, pp. 299.

The book is the second edition of the well-known monograph by Mark D. Levenson published in 1982.

Mark D. Levenson is well known for his work in the field of coherent nonlinear spectroscopy, which in many respects aided the establishment and development of this field of physics. To him belong the widely-known papers on nonlinear optics, two-photon spectroscopy, coherent four-photon spectroscopy of Raman scattering of light, and quantum optics.

In the second edition of the book errors and misprints discovered in the first edition have been corrected; for this reason "the authors are not responsible for the career of those who continue to use the first edition of the book." Moreover, the first 6 chapters have been provided with problems, new materials have been added on the calculation of the signal to noise ratio in schemes of nonlinear spectroscopy, resonance ionization spectroscopy, dynamic holography, etc.

The book is intended for graduate students and investigators working in the field of laser spectroscopy and unfamiliar with the methods of nonlinear optics. The emphasis in the presentation is on the physics of nonlinear optical processes underlying certain spectroscopic methods; correspondingly less attention is paid to the detailed interpretation of the spectra obtained. Theoretical quantum mechanical calculations in the book are based on the two-level model. For this reason the authors intentionally did not examine in their book phenomena and new results that require for their interpretation the solution of a more complicated problem taking many energy levels into account.

The book is written in a clear laconic language, not overloaded by theoretical computations. At the same time the condensed style of presentation does not prevent the authors from giving brief information on the history of the question, and on the physics of the phenomenon, so that the reader who is becoming familiar with a certain problem for the first time obtains quite a complete and deep idea concerning it.

The book consists of seven chapters. The first chapter is a collection of brief information from the theory of emission and scattering of light, the concepts are introduced of linear and nonlinear polarization of a dielectric, a description is given of the principles of operation and the construction of the main tunable lasers utilized in laser spectroscopy: dilasers, color center lasers, and parametric oscillators. The methods of tuning the wavelength of the generated radiation are discussed in detail. In conclusion of this chapter there is given a classification of nonlinear optical phenomena, the principles of the main schemes of nonlinear laser spectroscopy are briefly examined: saturation spectroscopy, coherent four-photon spectroscopy of Raman scattering of light, the spectroscopy of multi-photon absorption, non-steady-state coherent spectroscopy. On the whole the first chapter is an exceptionally clear, and saturated by carefully selected information, introduction to the field of nonlinear laser spectroscopy, from which the reader can obtain the most important information both from theory, and from practice, which considerably lightens the task of students who are only beginning independent work in the laboratory.

The second chapter contains the main information from the theory of interaction of light with matter. In sequence a discussion is given of the structure of the Hamiltonian, relaxation, vector representation of the equation of motion for the density matrix (the Bloch model), a calculation is made of the components of nonlinear polarization and the corresponding nonlinear optical susceptibility. In the concluding sections of this chapter the concept of a signal is introduced and the sources of noise in different schemes of nonlinear laser spectroscopy are analyzed in detail. This section did not appear in the first edition of the book.

In the third chapter the principles of saturation spectroscopy are presented that have become classical. At first there is a brief presentation of the theory of hole-burning leading to dips in the shape of the absorption line in the case of a Doppler broadened transition in a two-level system, the origin of additional resonances (crossover resonances) is analyzed in the case of saturation spectroscopy on close Doppler-broadened transitions, and also the special features of polarization saturation spectroscopy. Then a detailed description is given of the experimental method of saturation spectroscopy in gases. The authors present 8 schemes of laser spectrometers that have been used for implementing saturation spectroscopy, analyze the advantages and disadvantages of each of them, describe in detail the purpose of each of the components, which is particularly important for experimentalists. Then experimental results are presented obtained in the spectroscopy of gas media, a brief examination is made of the special features of multi-photon saturation spectroscopy and of spectroscopy with double resonance, saturation spectroscopy in liquids and solids. In conclusion dynamic holography, optical recording of information, bistability, and wave-front reversal are examined.

The fourth chapter describes the principles of various schemes of coherent Raman spectroscopy. This area of nonlinear laser spectroscopy has since its inception in 1972 undergone particularly intensive development. M. D. Levenson was one of the first to understand that polarization spectroscopy of Raman resonances is possible, and proposed the scheme of Raman-induced Kerr Effect Spectroscopy which later became widely used. He also carried out the first experiments on optical heterodyning of a signal in this scheme of spectroscopy.

The chapter begins with a comparison of spontaneous Raman scattering and a coherent spectroscopy of the Raman scattering of light. The cubic nonlinear susceptibility tensor is introduced which describes the process of coherent

Raman scattering. Its structure, the spatial symmetry, the shape of the spectrum of the signal, the relation with the cross section for spontaneous Raman scattering are discussed. The form of the cubic nonlinear susceptibility tensor and the Raman scattering tensor for different vibrational modes for all the 32 crystal symmetry classes are presented. Here also are provided tables of the effect of nonlinearity, expressed in terms of the components of the cubic nonlinear susceptibility tensor, for different polarizations of the pump waves for all the schemes of coherent four-photon Raman spectroscopy. The systemization of these data is particularly valuable for practical use. The central place in this chapter is occupied by the section on Coherent Anti-Stokes Raman Spectroscopy (CARS). Following this the Raman-induced Kerr Effect Spectroscopy (RIKES) and the Stimulated Raman Gain and Loss Spectroscopy are examined in equal detail. The concluding sections of this chapter are devoted to applications of coherent Raman spectroscopy. Here also is given a comparative analysis of the different schemes of coherent four-photon Raman spectroscopy from the point of view of noise characteristics.

The fifth chapter of the book is devoted to multi-photon absorption spectroscopy. Here a detailed description is given of the two- and three-photon absorption spectroscopy, free from Doppler broadening, which has become one of the principal methods for the investigation of hyperfine structure and the mechanisms of line broadening of atoms in the gas phase. This chapter also examines the process of multiphoton ionization of atoms which provides the basis for the laser method of isotope separation. In conclusion a brief review is given of the applications of multi-photon absorption spectroscopy.

The sixth chapter examines coherent transient optical phenomena: free-induction decay, optical nutation, photon echo, stimulated echo, and Ramsey fringes. All these effects are explained in detail theoretically and illustrated by experimental results; the application of coherent transient optical effects for optical recording of information is examined. In this chapter the phenomenon of superradiation is not examined. And yet this effect during the 1980's has been intensively investigated theoretically and experimentally, and a number of new fundamental results were obtained.

Finally, the concluding seventh chapter is devoted to the transformation of laser frequencies by the methods of nonlinear optics for use in nonlinear laser spectroscopy.

In preparation of the second edition the contents of the book underwent slight changes. For this reason the bibliography essentially refers to the period prior to 1981. Correspondingly the book contains no reference to many new results that refer, for example, to the application of picosecond CARS spectroscopy for the diagnostics of rapid processes of relaxation in gas media, complex molecules, semiconductors, nonlinear four-photon spectroscopy of electron resonance, surface spectroscopy. Nevertheless, this circumstance does not diminish the main merits of the book, that was intended specifically as an introduction to nonlinear spectroscopy, since the principles of the methods of nonlinear laser spectroscopy, of course, have not changed during the time elapsed since the appearance of the first edition of the book. In the depth and clarity of presentation with a comparatively modest length the book has no equal. This provides all the justification to recommend the book by M. D. Levenson and S. S. Kano as an excellent aid for students of the upper years and for graduate students specializing in the field of nonlinear laser spectroscopy, and also as an excellent reference manual for specialists working in this field of physics.