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Principles of nonlinear optical spectroscopy

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Principles of nonlinear optical spectroscopy. Shaul Mukamel (N.Y. and Oxford: Oxford University Press, 1997) p. 543

That nonlinear optical spectroscopy represents a major opportunity for the application of the laser to fundamental research was already clear back in the 60s when the laser was discovered. A huge increase in resolution — and in particular the elimination of Doppler broadening — achieved by using the nonlinear absorption saturation and two-phonon absorption effects is a remarkable accomplishment even by present day standards. By now, a resolution increase by a factor of $10^6 - 10^9$ (!) has been achieved deep inside the Doppler contour. Even though no other uses of the nonlinear effects seemed initially possible, the resolution increase was in itself quite a development in fundamental spectroscopy and quantum metrology. Suffices it to mention the relative resonance widths $\Delta\nu/\nu \simeq 10^{-13} - 10^{-14}$ on rotational-vibrational transitions in CH_4 and OsO_4, which allowed an accurate Rydberg constant measurement from the two-photon 1S–2S hydrogen transition and enabled the experimental search to be initiated for line splitting occurring in molecular enantiomers due to the weak interaction parity violation.

The advent of coherent active Raman scattering spectroscopy in two-frequency laser field marked another advance in this area. The use of nano- and picosecond laser pulses made it possible to measure low molecular concentrations and provided information, previously inaccessible, on the level population and polarization dephasing relaxation times in a condensed medium.

As frequency-change femtosecond lasers were developed and became increasingly widespread, new and more sophisticated nonlinear methods started to be developed for measuring a wide variety of diverse relaxation processes in various types of condensed medium. Thus far, however, these methods, often very impressive — especially when demonstrated at international conferences — have remained scattered in the literature, thus leaving the researcher in ignorance about the many potential applications of nonlinear optical spectroscopy.

In my view, what makes Mukamel's monograph so timely and valuable is that, based on the Green-function method and the author's own elegant correlation technique, it presents a unified treatment of the nonlinear interaction of a substance with various types of laser pulses, including multi-frequency ones and those separated in time and propagation direction.

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As a result, a unified and coherent picture of all the relevant methods emerges, including such widely used techniques as the coherent Raman scattering spectroscopy, photon echo, hole burning, pumping and probing, polarization spectroscopy, pulse effects, etc. An investigation of the table of contents below will tell more concerning the scope of the book:

1. Introduction 2. Quantum dynamics in Hilbert space. 3. Density operator and quantum dynamics in Liouville space. 4. Quantum electrodynamics, optical polarization, and nonlinear spectroscopy. 5. Nonlinear-response and optical-susceptibility functions. 6. Optical response functions of a multilevel relaxing system. 7. Semiclassical simulation of optical response functions. 8. Cumulant expansion and the multimode Brownian oscillator model. 9. Fluorescence and the spectroscopy of spontaneous and coherent Raman scattering. 10. Selective suppression of nonuniform broadening: photon echo. 11. Resonance lattices, pumping and probing, and hole burning spectroscopy. 12. Wave packet dynamics in Liouville space; Wigner representation. 13. Wave packet analysis for pulse measurements. 14. Out-of-resonance Raman scattering. 15. Polarization spectroscopy: birefringence and dichroism. 16. Nonlinear response of molecular ensembles: local field approximation. 17. Multiparticle and cooperative effects in nonlinear response.

A highly professional monograph treating such a wide range of nonlinear spectroscopy problems from a unified dynamic viewpoint is of course extremely valuable at the time when so many femtosecond experiments are in need of interpretation.

A feature of the book is that at the end of each section an excellent bibliography is given, containing references to key works on the subject.

The level of the monograph is sufficiently rigorous to make it a valuable reference book both for practicing and prospective investigators with a good background in quantum mechanics and with ambitions for state-of-the-art research.

Given that most scientists today are in good command of English I am not sure this book should be translated into Russian because its Russian market would inevitably be very limited. Speaking of translation, though, such excellent works as *Diplomacy* by H Kissinger and *Alchemy* by G Soros seem to me worth considering, which have huge readership world over and are also of great interest for us physicists. Unlike mathematicians, physicists very often encounter problems with ill-defined initial conditions and no exact solutions - which is exactly the situation inherent in diplomacy and financial matters.