Multiple-photon excitation and dissociation of polyatomic molecules

V. N. Sazonov

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Multiple-Photon Excitation and Dissociation of Polyatomic Molecules, Ed. C. D. Cantrell. Springer-Verlag, Berlin; Heidelberg; New York; London; Paris; Tokyo, 1986, pp. 288 (Topics in Current Physics. V. 35)

The book is devoted to the interaction of powerful infrared radiation, the source of which is a pulsed CO_2-laser, with polyatomic molecules (the number of atoms is in the range of 5–10). The IR radiation excites these molecules to high vibrational levels, and under certain conditions leads not only to excitation but also to dissociation of the molecules. This phenomenon, discovered more than 15 years ago, continues to attract the interest of many investigators since, first of all, technological applications are possible, for example, in the field of isotope separation or nonthermal stimulation of chemical reactions; and secondly, the physics of the phenomenon is very interesting and is not entirely understood even now.

The book under review is a collection of articles. It consists of the first chapter, which contains a relatively brief popular preface by the editor, and of seven subsequent independent chapters the authors of which[1)] describe their investigations addressing themselves to specialists working in the same field.

In the first chapter the editor, in particular, writes that during the last 15 years molecular physics is undergoing a renaissance associated with the appearance of powerful IR lasers. A completely new interesting field of molecular physics has emerged a part of whose new directions is represented in this book.

The second chapter contains extensive experimental material regarding excitation and dissociation of 44 different molecules under conditions when the molecules are in the gas phase in a cell and are subjected to single-frequency irradiation by one CO_2-laser. A review of the literature is given with 294 references. The authors note that different molecules under the action of powerful pulsed IR radiation exhibit approximately the same properties.

In the third chapter experiments are described on single-frequency irradiation of molecules in molecular beams. This completely eliminates the possible effect of collisions on excitation and dissociation. These experiments show that the redistribution of energy from the vibrational mode being excited to all other vibrational modes occurs only at very high vibrational levels near the dissociation threshold.

The fourth chapter is devoted to two-frequency experiments when IR radiation from the first laser excites the molecules to low-lying vibrational levels, and then a more powerful IR radiation from the second laser or UV radiation produces dissociation of the excited molecules. Such an arrangement has many advantages, in particular, the resonance IR radiation from the first laser can be relatively weak, and this makes it possible to produce dissociation of even those molecules whose absorption bands lie at some distance from the frequencies of the powerful CO_2-lasers. The two-frequency arrangement is used for the dissociation of molecules containing heavy elements and having a small isotope shift.

The fifth chapter describes experimental studies of narrow resonances in the spectrum of the multiple-photon absorption of SF_6 molecules cooled by the gasdynamic method to a temperature of approximately 30 K. The existence of such resonance once again indicates that there is no appreciable redistribution of vibrational energy at low-lying levels.

In the sixth chapter some theoretical models are developed which include the vibrational mode being excited and a thermostat consisting of the other vibrational modes. A detailed discussion is given of the transition from the coherent description, which takes into account the nondiagonal elements of the density matrix, to the simpler noncoherent description which takes into account only the level populations, i.e., only the diagonal elements. A review is given of the theoretical papers which contains 154 references.

The seventh chapter is devoted to a theoretical study of the adiabatic inversion in polyatomic molecules. Adiabatic inversion allows a complete depopulation of the lowest level and the transfer of the entire population to upper levels. This process, which was studied for the first time in connection with nuclear magnetic resonance, can be realized in the optical range by means of a slow variation of the intensity of laser radiation at a constant frequency. The great selectivity of adiabatic inversion makes it useful for isotope separation.

In the eighth chapter results are presented of a numerical calculation of the propagation of IR radiation taking into account its possible self-focusing and self-defocusing. At the same time the superfluorescence of a three-level system is discussed. This chapter is not very strongly related to the other chapters.

The book does not have a review article which would bring together the results of the separate chapters. Some interesting questions, for example, the stochastic nature of the intramolecular dynamics are almost not examined. However, all the topics that form the subject of the book are presented in very great detail and at a high level. The book is of considerable interest for specialists.

[1]The authors of the several chapters are: I—C.D. Cantrell (Center for applied optics, University of Texas at Dallas); II—J. L. Lyman, G. P. Quigley, and O. P. Judd; III—A. S. Sudbo, P. A. Schulz, B. R. Shen, and Y. T. Lee; IV—R. V. Ambartsumyan; V—S. S. Alimpiev, N. V. Karlov, E. M. Khokhlov, S. M. Nikiforov, A. M. Prokhorov, V. G. Sartakov, and A. L. Shtarkov; VI—M. F. Goodman, J. Stone, and E. Thiele; VII—G. L. Peterson and C. D. Cantrell- VIII—F. P. Mattar, P. R. Berman, A. W. Matos, Y. Claude, C. Goutier, and C. M. Bowden.