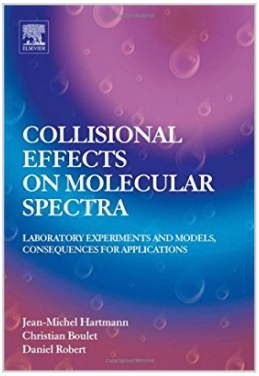

Collisional Effects on Molecular Spectra: Laboratory Experiments and Models, Consequences for Applications

By Jean-Michel Hartmann, Christian Boulet, Daniel Robert

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Gas phase molecular spectroscopy is a powerful tool for obtaining information on the geometry and internal structure of isolated molecules as well as on the interactions that they undergo. It enables the study of fundamental parameters and processes and is also used for the sounding of gas media through optical techniques. It has been facing always renewed challenges, due to the considerable improvement of experimental techniques and the increasing demand for accuracy and scope of remote sensing applications.

In practice, the radiating molecule is usually not isolated but diluted in a mixture at significant total pressure. The collisions among the molecules composing the gas can have a large influence on the spectral shape, affecting all wavelength regions through various mechanisms. These must be taken into account for the correct analysis and prediction of the resulting spectra.

This book reviews our current experimental and theoretical knowledge and the practical consequences of collisional effects on molecular spectral shapes in neutral gases. General expressions are first given. They are formal of difficult use for practical calculations often but enable discussion of the approximations leading to simplified situations. The first case examined is that of isolated transitions, with the usual pressure broadening and shifting but also refined effects due to speed dependence and collision-induced velocity changes. Collisional line-mixing, which invalidates the notion of isolated transitions and has spectral consequences when lines are closely spaced, is then discussed within the impact approximation. Regions where the contributions of many distant lines overlap, such as troughs between transitions and band wings, are considered next. For a description of these far wings the finite duration of collisions and concomitant breakdown of the impact approximation must be taken into account. Finally, for long paths or elevated pressures, the dipole or polarizability induced by intermolecular interactions can make significant contributions. Specific models for the description of these collision induced absorption and light

scattering processes are presented.

The above mentioned topics are reviewed and discussed from a threefold point of view: the various models, the available data, and the consequences for applications including heat transfer, remote sensing and optical sounding. The extensive bibliography and discussion of some remaining problems complete the text.

- State-of-the-art on the subject
- A bibliography of nearly 1,000 references
- Tools for practical calculations
- Consequences for other scientific fields
- Numerous illustrative examples
- Fulfilling a need since there is no equivalent monograph on the subject

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Editorial Review

From the Back Cover
Spectroscopy is a widely used tool for fundamental studies of the properties and dynamics of molecular systems but also for radiative heat transfer calculations and the remote sensing of the temperature, pressure, and composition of various gas media including atmospheres. In practically all cases, the effects of inter-molecular collisions on absorption spectra cannot be disregarded and precise line-shapes are required. This problem, which is “how does pressure affect the spectrum” is of considerable importance for fundamental studies (understanding the dynamics of collisions, knowledge of inter molecular interaction potential energy surfaces, etc..) and practical applications (radiative transfer in engines and atmospheres, determination of pressure/temperature/composition profiles from the inversion of infrared and Raman spectra, etc). The present book thus presents an up-to-date review, as exhaustively as possible, of the various effects of pressure on molecular spectral shapes considering theoretical and experimental aspects, as well as the consequences for remote sensing of planetary atmospheres and optical diagnostic in combustion media. Starting from isolated transitions whose broadenings and shiftings with pressure are quite well understood, more refined effects due to the finite collision duration and velocity changing and averaging effects are then discussed. Then, when lines are closely spaced, such as in manifolds and Q branches, collisional line-mixing effects, which redistribute the absorption intensity through population exchanges may be important. Then, in the window regions, such as troughs between transitions and band wings, where the contributions of many line-wings overlap, not only all preceding processes but the finite duration of collisions must be taken into account. Finally, for long paths or elevated pressures, the dipole induced during intermolecular interactions can lead to significant absorption. All these topics will be reviewed and discussed from a fourfold point of view: the various models, the experimental techniques, the available data, and the consequences for a number of practical applications. Together with a broad bibliography these elements should hopefully give the reader, if not full answers, some directions to “pursue his journey”.

About the Author
Jean-Michel HARTMANN: born in 1961, « Directeur de Recherche » for the French CNRS (Centre National de la Recherche Scientifique has been carrying research and advising PhD students in the field of the book for about twenty years. He is the director of the French Molecular Spectroscopy Network and the author of more than 100 publications in international journals.

Christian BOULET: born in 1947, Professor at Université Paris XI (Orsay) has been carrying theoretical researches in the field for more than 30 years and is the author of about 130 publications in international journals. He has been the director of the Laboratoire d’Infrarouge and of the Laboratoire de Physique Moléculaire et Applications.

Daniel Robert: born in 1940, “Emerite professor” at Franche Comté University (besançon) is also a theoretician of line-shapes who has been working in the field for more than 30 years and who is the author of about 120 publications in international journals. He has been the director of the Laboratoire de Physique Moléculaire (Besançon

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