Toward Complete *Ab Initio* Spectroscopy of Small Molecules

Attila G. Császár

Institute of Chemistry, Eötvös University,
P.O. Box 32, H-1518 Budapest 112, Hungary

Viktor Szalay

Crystal Physics Laboratory, Research Institute for Solid State Physics and Optics,
Hungarian Academy of Sciences, P. O. Box 49, H-1525 Budapest, Hungary

Jonathan Tennyson

Department of Physics and Astronomy, University College London, Gower Street,
London WC1E 6BT, United Kingdom

For the accurate first-principles computation of complete rovibrational spectra one needs to solve both the quantum mechanical electronic structure and nuclear motion problems with exceedingly high accuracy. Results from our own research, obtained in collaboration with a number of colleagues, including W. D. Allen, G. Czakó, T. Furtenbacher, T. Lynas-Gray, O. L. Polyansky, S. V. Shirin, and N. F. Zobov (in alphabetical order), in both of these areas are presented during the talk.

State-of-the-art *ab initio* electronic structure computations can result in highly accurate potential energy (PES) and dipole moment (DMS) surfaces. Our recent results are reviewed, highlighting the hierarchy of the physical effects to be considered. The emphasis is on the prototypical triatomic molecule H_2O.

Variational strategies for solving the nuclear motion problem are discussed next. Emphasis is put either on the simplicity of the approach or on the utility of the solution strategies in handling singularities in the rovibrational Hamiltonian. Representative numerical results and interesting chemical applications are presented for selected systems.