AB INITIO STUDIES OF SMALL MOLECULES

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

Electronic structure calculations have become capable of predicting a large number of rovibrational band origins and other spectroscopic properties to within a wavenumber or better. Such state-of-the-art *ab initio* electronic structure computations, resulting in highly accurate potential energy (PES) and dipole moment (DMS) surfaces are reviewed highlighting the hierarchy of the physical effects to be considered. The use of high-order force fields for the representation of the PES is also discussed.

Different variational strategies for solving the nuclear motion problem are discussed next. Emphasis is put either on the simplicity of the approach, provided by the discrete variable representation (DVR) of the Hamiltonian, or on the utility of the solution strategy in handling singularities in the Hamiltonian. Generalization of the DVR approach and its accuracy is addressed. Representative numerical results are presented for triatomic systems.