REGULAR VS. CHAOTIC INTRAMOLECULAR DYNAMICS IN THE FIRST CH STRETCHING OVERTONE OF AROMATIC MOLECULES

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The eigenstate resolved spectra of benzene, pyrrole and triazine in the first CH stretching overtone region ($\approx 6000\ \mathrm{cm}^{-1}$), presented at the 1997 edition of this symposium (talks RG9-11), have been analyzed for signatures of regularity/chaos in the underlying vibrational dynamics. It is generally accepted that in strongly interacting systems energy level repulsion should be observed, according to the Gaussian Orthogonal Ensemble model of Dyson [J. Math. Phys **3**, 140 (1962)]. Contrary to this expectation, the tests of level spacing statistics that we have performed point to a random distribution of the energy levels, typical of more regular dynamics, both for pyrrole and for benzene, despite the different symmetry of the two molecules and the different timescales involved (100 and 20 ps respectively). For triazine the evidence is less compelling due to the sparse nature of the observed spectrum which does not allow for a robust statistical analysis. Quantum simulations by Pearman and Gruebele (next talk) show timescales and statistics similar to the experiment. The implications of these findings on the theory of molecular dynamics and vibrational energy redistribution will be discussed at the conference.

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