

REAL-TIME TRACKING OF COHERENT VIBRATIONAL MOTION IN GROUND AND EXCITED ELECTRONIC STATES

SHAINA DHAMIJA, GARIMA BHUTANI, *Department of Chemical Sciences, Indian Institute of Science Education & Research, Mohali, Punjab, India*; AJAY JAYACHANDRAN, *Institut für Physikalische und Theoretische Chemie, Ludwig-Maximilians Universität Würzburg, Würzburg, Germany*; ARIJIT K DE, *Department of Chemical Sciences, Indian Institute of Science Education & Research, Mohali, Punjab, India*.

Molecular vibrations, involving both ground and excited electronic states, are at the heart of chemical transformations which necessitates understanding the origin of these vibrations. Femtosecond infrared spectroscopy and frequency-domain coherent Raman spectroscopy successfully captured full vibrational spectra but bear a few limitations like interfering lineshapes, background signals, etc. Time-domain measurement, i.e. impulsive stimulated Raman scattering employs a short Raman pump pulse, creating a nuclear wavepacket which evolves as a function of time and is interrogated using a probe pulse and Fourier transform of the temporal interferogram yields the Raman spectrum whereas the scattering background can be easily removed as a zero frequency component. However, separation of vibrational coherences in ground and excited electronic states even for small chromophores in condensed phase still remains challenging. Recently, we showed how 'spectrally dispersed' impulsive stimulated Raman spectroscopy can be employed to track time evolution of vibrational coherences in ground as well as excited states, distinctly, under non-resonant/resonant impulsive excitation [1-4]. More specifically, separation of excited-state, ground-state and solvent coherences for diatomic as well as polyatomic molecules in solution is demonstrated, which I will discuss in this presentation. The origin of spectral patterns corresponding to certain vibrational modes of the solute as well as the solvent will be presented. In addition to this, density functional theory is employed to identify the Raman active modes, which nicely correlates with the experimental observations. Details of this method as an emerging technique will be discussed.

References: [1] S. Dhamija, G. Bhutani, and A. K. De, *Asian Journal of Physics*, 255-260, 29 (3 and 4), 2020. [2] S. Dhamija, G. Bhutani, and A. K. De, *Frontiers in Optics + Laser Science* 2021, paper JW7A.70, 2021. [3] S. Dhamija, G. Bhutani, A. Jayachandran, and A. K. De, *The Journal of Physical Chemistry A*, 1019-1032, 126(7), 2022. [4] S. Dhamija, G. Bhutani, and A. K. De, *Chemical Physics Letters*, Under review (Manuscript ID: CPLETT-22-308).