**Abstract**

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|  | Raman spectroscopy has revolutionized molecular spectroscopy especially coming to the fields of biology, material science and analytical chemistry. Stimulated Raman techniques overcome the limitations that exist in spontaneous Raman spectroscopy where the time bandwidth relationship of Raman excitation pulse hinders tracking of structural events in ultrafast time scale. Impulsive Stimulated Raman Spectroscopy (ISRS) is one such technique that is used to track nuclear dynamics with femtosecond accuracy. The vibrational wavepacket evolving in both excited and ground state of small molecules is studied using ISRS to learn more about the vibronic photophysics of chemical systems. The technique helps to observe nuclear dynamics of Raman active vibrational modes from terahertz to larger wavenumbers with considerable sensitivity. It also suppresses background signals easily which has plagued ultrafast vibrational spectroscopy for decades and also resonantly enhances the signal-noise ratio of Raman spectra. In ISRS, time domain signal raw data is truncated and Fourier transformed to get information on the normal modes of vibration. We use sub-femtosecond pulses to spectrally disperse the ISRS data in the visible region due to complexity in generation and management of temporarily short pulse in UV/IR region. methylene blue, nile blue and 4-dimethylamino-4'-nitrostilbene (DNS) molecule were selected as potential candidates keeping in mind the presence of low frequency modes which becomes difficult to observe using already well established frequency resolved FSRS due to Rayleigh scattering . DNS complex is utilized to study Twisted Intermolecular charge transfer (TICT). TICT is defined as a to dual fluorescence phenomenon in which torsional change takes place in excited state from a planar locally excited conformer to a perpendicular CT conformer, following electronic excitation. The coupled vibrational modes of TICT molecules are analyzed to describe twist in excited electronic state. We employ computational calculations using DFT and TD-DFT level of theory to observe nature of potential energy surface and to calculate the vibrational spectra in both ground and excited state on other known TICT molecules. Using both impulsive stimulated spectroscopy and ab-initio calculations we try to create a clearer picture on how chemical system evolves. |