Watching ultrafast processes in the light of Femtosecond Stimulated Raman Scattering

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The modern concept of atomic structure of matter goes back to more than one century ago, and the development of imaging techniques with increasing level of complexity allows nowadays to routinely deliver atomic resolution pictures. One of the most challenging perspectives of our century is to animate such pictures making molecular movies, to observe atomic motion during those elemental events underlying physical, chemical and biological processes. This would enable, for instance, tracking phase transitions, bond breaking and recombination dynamics, allosteric regulation in biology. While the transition from photography to cinematography smoothly occurred in the late 19th century, the corresponding process at the atomic scale is hindered by the simultaneous need of temporal and spatial resolution. The main routes which are currently pursued to embrace this challenge will be discussed.

Among them, time resolved Raman spectroscopy is a powerful technique to study photo-induced dynamics with structural sensitivity, whose time resolution has been improved over the years from microseconds to a few picoseconds. If a sharp spectral resolution (<15 cm-1) is to be maintained, however, no further improvement of the time resolution (<1 ps) is obtainable with a traditional two beams pump-probe layout, due to the Fourier Transform limit. In recent years, Femtosecond Stimulated Raman Spectroscopy (FSRS) has been proposed as a possible way to "circumvent" the aforementioned time-bandwidth limitation, promising simultaneously high temporal precision and spectral resolution.

Starting from a short introduction to FSRS, which we developed in our group in the last few years, I will present a few applications with emphasis on how sub-picosecond snapshots with atomic resolution can be obtained under the constraint imposed by the Heisenberg principle. These include femtomagnetism in perovskites [1] and reaction pathways and energy transport in biomolecules [2]. I will finally illustrate the making of a molecular movie featuring the first few picoseconds of a heme protein dynamics in response to photoexcitation [3].

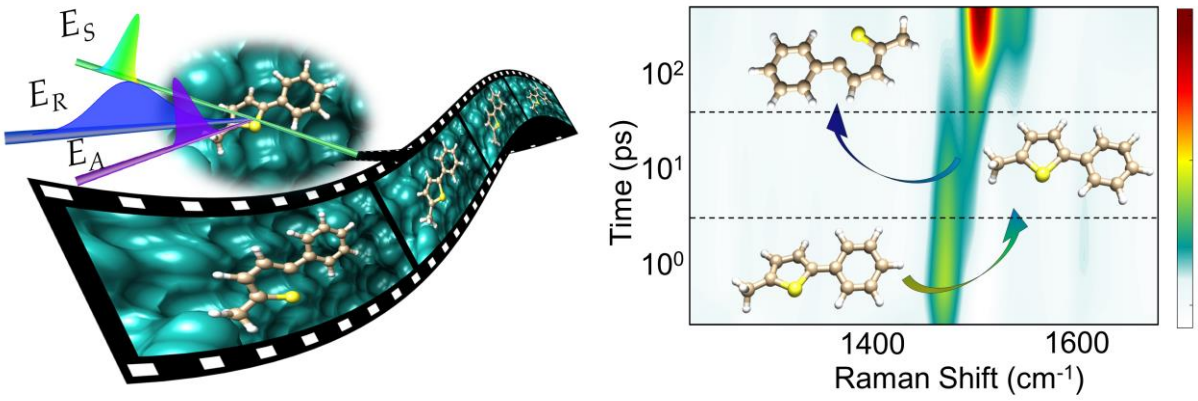

Figure: Ring opening reaction in a Diaryl Thiophene, visualized by FSRS

References

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