VISTA Seminar

Seminar 18

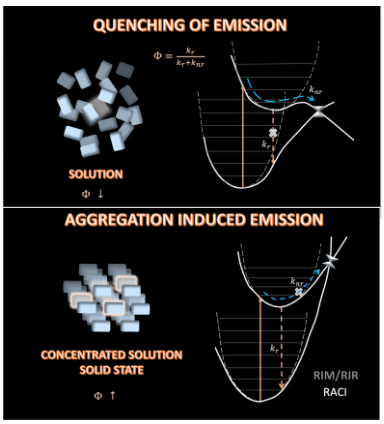
May 12, 2021

9:30 – 11:00 am EDT / 1:30 – 3:00 pm GMT / 3:30 pm – 5:00 pm Paris

TOC:

Modelling excited states in molecular crystals

Rachel Crespo-Otero
Joseph Priestley Building, Queen Mary University of London, London, UK
Email: r.crespo-otero@qmul.ac.uk



Light-emitting materials find applications in display technologies, optical communication, data storage, biological sensing and solid-state lasing. Aggregation-induced emission (AIE) offers a route for the development of luminescent technologies with high quantum efficiencies. However, maximizing fluorescence quantum efficiencies is a formidable challenge in attaining first-principles materials design, due to the interplay between the electronic structure of the chromophore and the molecular crystal. The identification of radiative and nonradiative channels, and how these are affected by aggregation, can help rationalize the emissive properties of materials and aid in the design of yet more efficient fluorophores.

In this talk, I will discuss how inter- and intramolecular processes determine the emissive properties of a series of crystals including prototypical propeller-shaped AIE, intramolecular proton transfer chromophores and aromatic molecules with applications in lasers.[1–4] The excited state mechanisms will be examined considering the competition between nonradiative and radiative pathways. The role of conical intersections will be addressed in the context of the 3D-structure of the crystals, exciton couplings and the presence of triplet states. The *fromage* (FRamewOrk for Molecular AGgregate Excitations) platform[5, 6] will be presented as a tool to assist with the investigation of photochemical processes in molecular crystals.

References

1 L. Stojanović and R. Crespo-Otero, *ChemPhotoChem*, 2019, **3**, 907–915.
2 M. Dommett, M. Rivera and R. Crespo-Otero, *J. Phys. Chem. Lett.*, 2017, **8**, 6148–6153.
3 M. Dommett, M. Rivera, M. T. H. Smith and R. Crespo-Otero, *J. Mater. Chem. C*, 2020, **8**, 2558–2568.
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5 M. Rivera, M. Dommett, A. Sidat, W. Rahim and R. Crespo-Otero, *J. Comput. Chem.*, 2020, **41**, 1045–1058.
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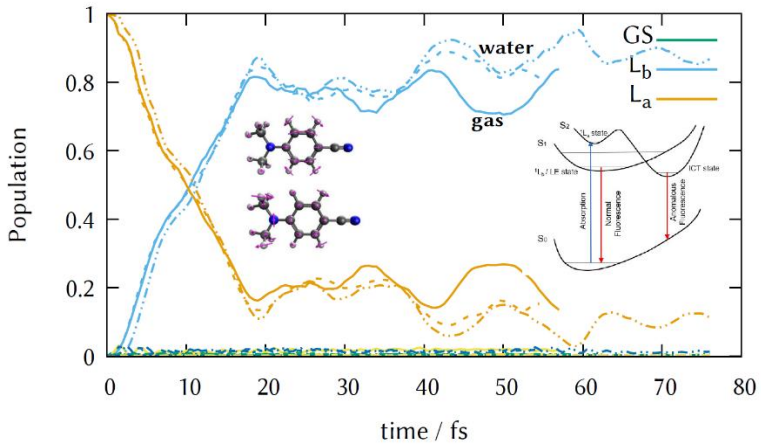
Quantum trajectories (DD-vMCG) for DMABN non-adiabatic dynamics

Sandra Gómez, Esra N. Soysal and Graham A. Worth
University College London, Department of Chemistry, 20 Gordon Street, London WC1H 0AJ, United Kingdom
Email: sandra.rodriguez@ucl.ac.uk



4-(N,N-dimethylamino)benzonitrile (DMABN) is widely known for presenting a double peak in its fluorescence spectra when measured in solution, whereas it shows a single emission peak in gas phase.(1) The last decades this has been explained by the existence of an equilibrium between two excited state minima in the S_1 surface; both with $\pi\pi^*$ character but with different orbitals involved. One configuration keeps the orbitals localized over the benzene ring (local excited or L), while the other has charge donated from the N-dimethyl side to the nitrile group. The latter, commonly called internal charge transfer (ICT) state has been object of controversy over the years and many scientific groups have argued about its structure and formation. [2,3]

In this talk, I will present the excited state dynamics method DD-vMCG[4] and apply it to describe the S_2/S_1 internal conversion of DMABN in the local excited state manifold, both in presence and absence of explicit water molecules. This internal conversion occurs shortly after UV excitation and is crucial to relax the molecule to the S_1 electronic state, where the charge transfer state is supposed to be formed after 0.7-1 ps. Experimental emission and absorption spectra measured in three different solvents will also be presented, in order to complement the dynamics simulations.

References:

1. E. Lippert, W. Lüder and H. Boos, Advances in Molecular Spectroscopy, Pergamon Press, 1962.
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4. G.W. Richings, I. Polyak, K.E. Spinlove, G.A. Worth, I. Burghardt & B. Lasorne, 2015, 34:2, 269-308.

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