VISTA Seminar

Seminar 10

9:30 – 11:00 am EST / 2:30 – 4:00 pm GMT / 3:30 pm – 5:00 pm Paris

TOC:

Can we understand surface traps in semiconductor quantum dots and how to heal them?

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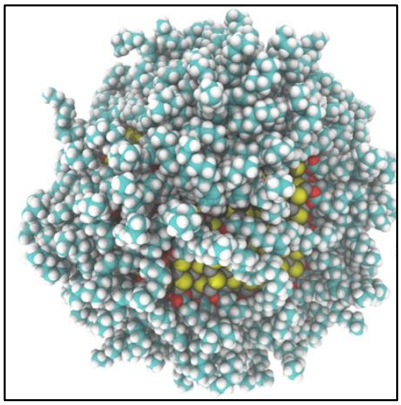

Colloidal semiconductor nanocrystals, also called quantum dots (QDs), are key players driving modern semiconductor research. QDs have size tunable optoelectronic properties and can be processed in solution. These cutting-edge materials are shaping technologies as diverse as electroluminescent and liquid crystal displays, solid-state lighting, lasers, infrared imaging and solar energy conversion. Moreover, the rapid expansion of the QD materials library and the proven possibility to form highly conductive QD superstructures warrants a bright future for QDs in many other fields.[1,2]

In these nanoscale crystallites a very high number of atoms is located on the surface and some of these atoms might introduce energy levels in the band gap that ultimataly dominate the optical and electronic properties of these materials, rendering them inefficient. The relation between surface atom coordination and electronic structure remains largely unknown, however a clearcut understanding of the atomistic origin of traps and the possibility of chemically manipulating them have capital importance to design defect-free colloidal quantum dots and make a leap forward in the development of efficient optoelectronic devices.

Recent advances in computing power established computational chemistry as a powerful tool to describe accurately complex chemical species and nowadays it became conceivable to model colloidal quantum dots with realistic sizes and shapes.[3] In this talk, I will discuss the knowledge gathered in recent experimental findings with the computation of quantum dot electronic structures. I will show the behaviour of two different systems: namely, CdSe and $CsPbBr_3$ nanocrystals as benchmark semiconductor nanocrystals showing how different types of trap states can form at their surface and how (excited state) molecular dynamics brings new insights in the dynamic formation of these traps.[4] In addition, I will suggest how computations helps in designing new experiments healing of such traps according to their chemical origin and nanocrystal composition.

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2. Kagan, C. R., Lifshitz, E., Sargent, E. H. & Talapin, D. V. Building devices from colloidal quantum dots. *Science (80-.).* **353**, (2016).
3. Houtepen, A., Hens, Z., Owen, J. & Infante, I. On the Origin of Surface Traps in Colloidal II–VI Semiconductor Nanocrystals. *Chem. Mater.* **29**, 752–761.
4. Giansante, C. & Infante, I. Surface Traps in Colloidal Quantum Dots: A Combined Experimental and Theoretical Perspective. *J. Phys. Chem. Lett.* **8**, 5209–5215 (2017).

Nonadiabatic effects within condensed-phase systems: a novel partially linearized spin-mapping approach

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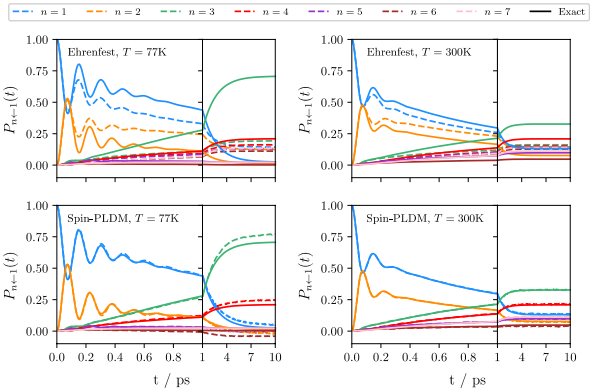

The coupled dynamics of electrons and nuclei in molecular condensed-phase systems remains a challenging problem for computer simulation; any viable approach must both be able to correctly describe nonadiabatic transitions as well as being computationally cheap. While independent trajectory-based simulations offer an inexpensive, as well as a physically motivated approach for calculating dynamical quantum-mechanical observables, many popular methods such as Ehrenfest dynamics are unable to describe certain aspects of the dynamics correctly, such as decoherence phenomena and entanglement between the electronic and nuclear subsystems.

One way of improving the accuracy of independent trajectory-based methods is to use a mapping-based approach, which describes the dynamics of the electronic subsystem as well as the nuclei using classical trajectories within a continuous mapping space. Meyer-Miller-Stock-Thoss (MMST) mapping [1, 2], where the electronic subsystem is mapped onto a set of harmonic oscillators, is one such commonly used example. However, a reformulated version of spin-mapping [3], which uses a Stratonovich-Weyl approach to describe the electronic subsystem within the spin-mapping space, has been recently obtained that appears to give rise to methods with superior accuracy compared to their MMST analogues. A further way of improving the accuracy of mapping-based methods is to use a so-called partially linearized based approach [4, 5], where the electronic dynamics associated with the forward and backward propagation paths are treated explicitly. Such methods have the appealing features that they can be rigorously derived from a path-integral formulation of the real-time correlation function and the results can also be systematically improved towards an exact solution of the quantum-classical Liouville equation (QCLE).

In this talk, I will introduce our new spin-PLDM method [6, 7], which combines the advantages of both a partially linearized based approach and spin-mapping. Dynamical observables for a range of commonly used model systems, such as the Fenna-Matthews-Olsen complex (FMO), will be presented which illustrate the methods superior accuracy in comparison to Ehrenfest and other mapping-based techniques. In particular, I will show that the method is able to obtain the relatively short-time dynamics extremely accurately, suggesting that spin-PLDM is well suited for a number of applications, such as describing nonadiabatic effects within electronic spectroscopy.

[1] H.-D. Meyer, W. H. Miller, *J. Chem. Phys.* **1979**, 71, 2156–2169.
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[5] P. Huo, D. F. Coker, *J. Chem. Phys.* **2011**, 135, 201101.
[6] J. R. Mannouch, J. O. Richardson, arXiv **2020**, 2007.05047.
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