VISTA Seminar

Seminar 23

September 16, 2021

10:00 am – 11:30 am EDT / 2:00 – 3:30 pm GMT / 4:00 pm – 5:30 pm Paris

TOC:

Non-Equilibrium Thermo Field Dynamics and Tensor-Train Approaches to Closed and Open System Evolution: Theory, Implementation and Application

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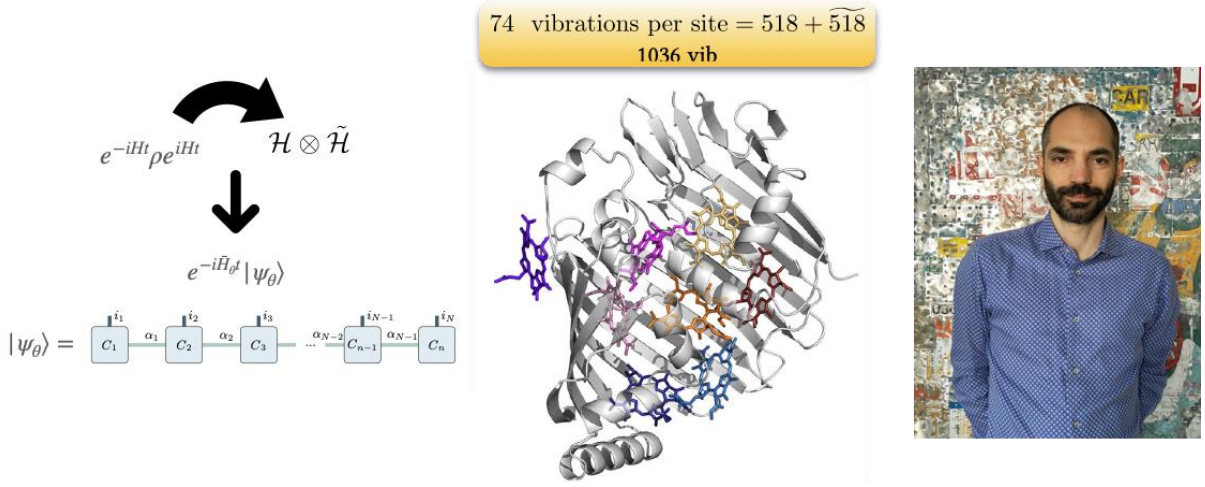

Non-Equilibrium Thermo-Field Dynamics (NETFD), is as alternative formulation of quantum statistical mechanics, completely equivalent to the commonly used density matrix approach. In this talk I will present NETFD concepts and show how they can be extended and integrated with Tensor-Train (TT) based numerical tools leading to a novel and powerful theoretical and computational framework for the study of complex quantum dynamical problems.

In addition, NETFD techniques are extended to enable the study of dissipative open systems via a new formulation of the Hierarchical Equations of Motion (HEOM) fully integrated with TT methodologies. The combination of the TFD machinery with computational advantages of TTs results in a powerful theoretical and computational framework for scrutinizing dynamics of complex multidimensional electron-vibrational systems.

The validity and the computational advantages of the developed methodologies is illustrated by applying them to the study of quantum coherence effects in the energy-transfer processes in antenna systems, to the analysis of fingerprints of vibrational modes in electron-transfer and charge-transfer processes in various model and realistic multidimensional molecular systems, as well as to simulation of other fundamental models of physical chemistry.

On the Mechanism of Alkylammonium Ligands Binding to the Surface of $CsPbBr_3$ Nanocrystals

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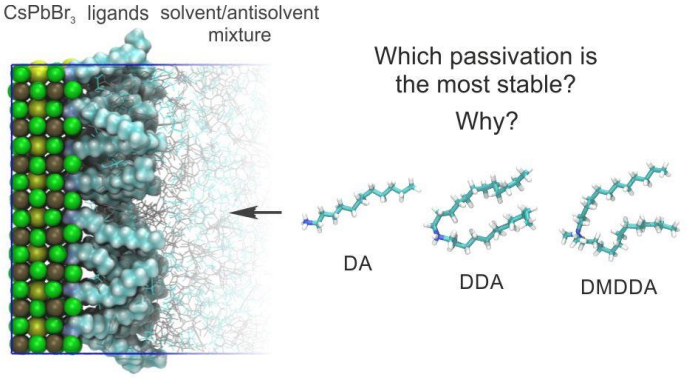

$CsPbBr_3$ nanocrystals (NCs) suffer from instabilities caused by the dynamic and labile nature of both the inorganic core and the organic-inorganic interface. Weak and dynamic binding between the NC surface and capping ligands causes rapid ligand desorption upon isolation and purification of colloids, eventually leading to a loss of structural integrity and sintering of NCs into bulk polycrystalline materials. Surface ligand engineering therefore remains an imminent research topic. Much progress in obtaining purifiable and stable colloids was achieved with a recent experimental discovery of new capping ligands, such as dimethyldidodecylammonium halides, alkylphosphonic acids, and long-chain zwitterionic ligands. However, comprehensive understanding of the NC–ligand–solvent interface and the atomistic origins of the observed differences lags behind. In this study, we use classical molecular dynamics simulations to gain insights into the inherent binding properties of three different alkylammonium ligands – primary dodecylammonium (DA), secondary didodecylammonium (DDA) and quaternary dimethyldidodecylammonium (DMDDA) – in a mixture of nonpolar (toluene) and polar (acetone) solvents, the medium which is typically encountered during purification of the NCs. Our simulations uncover three main factors that govern effective ligand–substrate interactions: (i) the ability of the head-group to penetrate into the binding pocket, (ii) the strength of head-group's interactions with the polar solvent, and (iii) higher barrier for ligand adsorption/desorption in the case of multiple alkyl chains. The interplay between these factors causes the following order of the binding free energies: DDA < DA ≈ DMDDA, while surface capping with DDA and DMDDA ligands is additionally stabilized by the kinetic barrier. These findings are in agreement with experimental observations, wherein DDA is found to loosely bind to the $CsPbBr_3$ surface, while DMDDA capping is more stable than capping with primary oleylammonium ligand. The presented mechanistic understanding of the ligand-NC interactions is then used to design new cationic ligands that are expected to make perovskite NC surfaces more robust. We anticipate that the methodology, which is used in this study, can be extended to other types of inorganic materials with a predominantly noncovalent nature of interactions with capping ligands.

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 23
Time: Sep 16, 2021 10:00 AM Eastern Time (US and Canada)

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