VISTA Seminar

Seminar 63

February 21, 2024

10:00 am – 11:30 am EST / 3:00 – 4:30 pm GMT London / 4:00 pm – 5:30 pm CET Paris / 11 pm CST Beijing

TOC:

Optical Properties and Electron-Phonon Interactions in Low-Dimensional Materials

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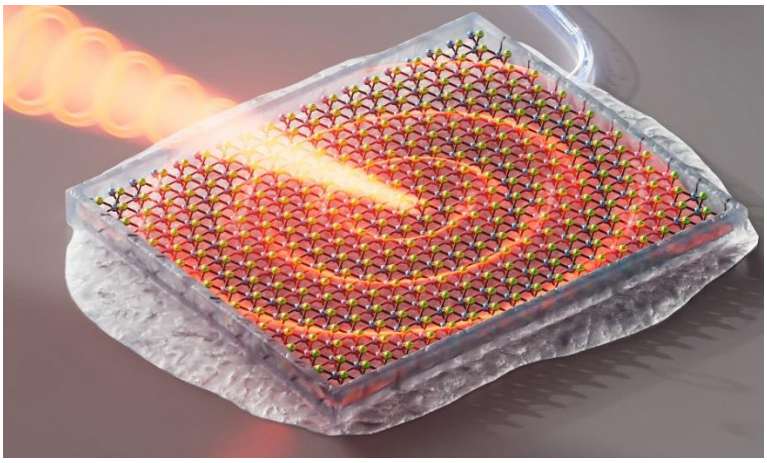

Atomically thin two-dimensional materials are direct bandgap semiconductors with a rich interplay of the valley and spin degrees of freedom, which offer the potential for electronics and optoelectronics. A strong Coulomb interaction leads to tightly bound electron-hole pairs or excitons and two-electron one-hole quasiparticles or trions. We solve the two-particle and three-particle problems for the wavefunctions for excitons and trions in the basis set of the model- Hamiltonian for single particles. The calculated linear absorptions, photoluminescence spectra, and polariton spectra as a function of doping and temperature explain the experimental data in 2D monolayers and predict novel spectroscopic features due to the many-body Coulomb interactions. I will also discuss the phonon-assisted Auger non-radiative decay mechanism of excitons in doped 2D materials, polarons in black phosphorous monolayer, and in 1D van der Waals SbPS4 material. If time allows, I will discuss the potential of graphene as a quantum phonon sensor down to a single phonon level via remote electron-phonon scattering in an adjacent boron nitride substrate. This material is based upon work supported by the Air Force Office of Scientific Research under award number FA9550-22-1-0312.

Pair-Decoupling Nuclear Degrees of Freedom in Molecular Dynamics Simulations

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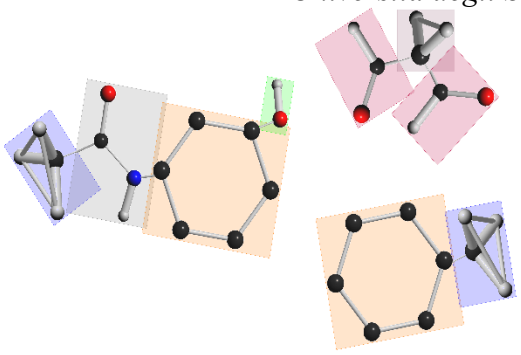

When it comes to identify the building blocks of middle-sized organic molecules, chemists mainly think in terms of the "functional group picture", in which a molecule is composed of many pieces joined together. However, is this picture truly representative of the dynamics of molecules?

We approach this question using theoretical/computational tools but with an experimentalist mindset: We want to artificially "decouple" pairs of atoms and see how the system as a whole rearranges. In practice, we run simulations of artificially modified potentials in which selected pairs of atoms (or degrees of freedom) do not perceive each other's displacement. When this artificial disturbance takes place, it affects the molecule as a whole, resulting in a lose (or gain) in energy. A measure of the average energy lost allows to understand if the decoupled pairs can or cannot be seen as belonging to approximately independent functional groups.

The formal description of pair-decoupling is based on the existence of a "pair-decoupled" Hamiltonian function $\tilde{H}$ that accounts for decoupled pairs of atoms (or pairs of groups of degrees of freedom in general). Given these definitions we may ask a more formal question: "Is the chemistry of such a system $\tilde{H}$ really different from that of the original system?" To approach this question we have developed a simulation technique called "Symplectic Explicit with Force" (SEF) integrator which can accurately integrate the equations of motion for $\tilde{H}$, allowing to compute expectation values and correlation functions, retaining the geometric properties of Hamiltonian mechanics.

Leveraging on the pair-decoupling concept and the SEF algorithm, we show that the chemistry of Salicylic Acid is rooted on its dynamical correlations. In particular, the out of plane motion of its substituents must be tightly correlated to keep the Salicylic Acid in its overall planar shape.

Furthermore, we show that the picture of organic molecules provided by the pair-decoupling concept reduces to the functional group picture for most rigid systems, but it captures quantitatively the importance of coordinated motion in flexible ones.

The pair-decoupling concept and SEF algorithm could open the route to novel mechanistic hypotheses that are embedded within complex patterns of reactions and inspire novel experiments.

References

[1] Michele Gandolfi and Michele Ceotto *JCTC* **19**, 6093-6108 (2023)

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 63
Time: Feb 21, 2024 10:00 AM Eastern Time (US and Canada)

Join Zoom Meeting
https://buffalo.zoom.us/j/93749110241?pwd=MlgraSs1ejRmdTBJanV3UVQ1NEVUQT09

Meeting ID: 937 4911 0241
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