VISTA Seminar

Seminar 25

October 14, 2021

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

TOC:

Real-time *GW*-BSE Investigations on Spin-Valley Exciton Dynamics in Monolayer Transition Metal Dichalcogenide

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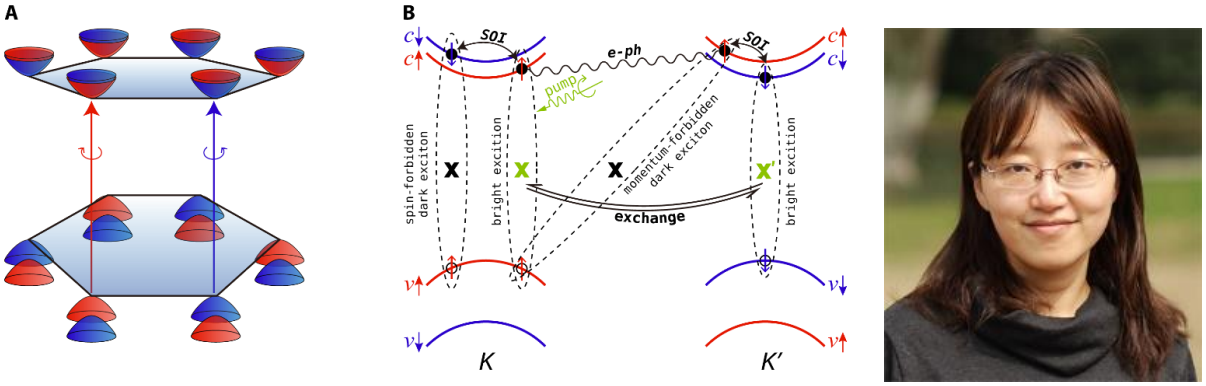

The valley exciton dynamics in two-dimensional transition metal dichalcogenides (TMDs) is crucial for the spin-valleytronics. However, a real-time *ab initio* method for the spin-resolved exciton dynamics is still missing. Here we develop an *ab initio* nonadiabatic molecular dynamics (NAMD) method based on *GW* plus real-time Bethe-Salpeter equation (*GW*+rtBSE-NAMD) for the spin-resolved exciton dynamics. From investigations on MoS_2, we provide a comprehensive picture of spin-valley exciton dynamics where the electron-phonon (*e-ph*) scattering, spin-orbit interaction (SOI) and electron-hole (*e-h*) interactions come into play collectively. Especially, we provide a direct evidence that *e-h* exchange interaction plays a dominant role in the fast valley depolarization within few picoseconds in excellent agreement with experiments. Moreover, there are bright-to-dark exciton transitions induced by *e-ph* scattering and SOI. Our study proves that *e-h* many-body effects are essential to understand the spin-valley exciton dynamics in TMDs and the newly developed *GW*+rtBSE-NAMD method provides a powerful tool for exciton dynamics in extended systems with time, space, momentum, energy and spin resolution.

Reference:

X. Jiang, Q. Zheng, Z. Lan, W. A. Saidi, X. Ren and J. Zhao* *Sci. Adv.*, **7**, eabf3759, (2021)

Time-dependent density matrix renormalization group for electron-vibration coupled problems

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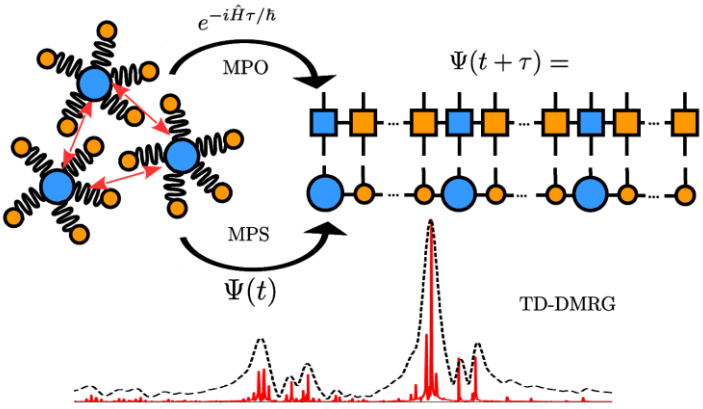

The wavefunction theory of quantum dynamics meets difficulties called exponential wall, *i.e.* the dimension of Hilbert space increases exponentially with the system size. Recently, the time-dependent density matrix renormalization group (TD-DMRG) theory emerges as a numerically exact full-quantum wavepacket method with only polynomial computational complexity, which provides an opportunity to address this long-standing difficulty. In this talk, I will introduce our recent development of TD-DMRG in the modern framework of matrix product states/operators (MPS/MPO), which is able to simulate quantum dynamics of electron-vibration coupled problems at both zero and finite temperatures.[1,2,3] For harmonic bath and linear electron-vibration coupling, we performed detailed benchmarking of TD-DMRG for spin-boson model covering delocalization and localization regimes, and for multi-mode Holstein model with more realistic molecular parameters to describe spectroscopy and exciton transfer of molecular aggregates.[1,3] Beyond that, we applied TD-DMRG to calculate the molecular nonradiative decay rate with anharmonic potential energy surface and found that the anharmonicity opens up the ability of C-H vibrations of molecule azulene to receive electronic energy, which enhances the transition rate significantly.[4,5] Finally, I will briefly introduce the open-source TD-DMRG package *RENORMALIZER*[6] developed by us and the underlying CPU-GPU heterogeneous algorithm that can improve the computational efficiency of TD-DMRG dozens of times.[3]

References:

[1] Ren, J.; Shuai, Z.*; Kin-Lic Chan, G.* *J. Chem. Theory Comput.* 2018, 14, 5027.
[2] Ren, J.*; Li, W.; Jiang, T.; Shuai, Z. *J. Chem. Phys.* 2020, 153, 084118.
[3] Li, W.; Ren, J.*; Shuai, Z. *J. Chem. Phys.* 2020, 152, 024127. (JCP Editors' Choice 2019)
[4] Wang, Y.; Ren, J.*; Shuai, Z.* *J. Chem. Phys.* 2021, 154, 214109.
[5] Ren, J.; Wang, Y.; Li, W.; Jiang, T.; Shuai, Z.* doi:10.33774/chemrxiv-2021-tnngb.
[6] https://github.com/shuaigroup/Renormalizer

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Topic: VISTA, Seminar 25
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