Ab initio charge transport and ultrafast carrier dynamics in semiconductors and oxides

Jin-Jian Zhou, Marco Bernardi

JCAP JOINT CENTER FOR ARTIFICIAL PHOTOSYNTHESIS

Abstract:

We employ first-principles calculations to study charge transport and ultrafast carrier dynamics in semiconductors and oxides, focusing on the interactions between electrons and lattice vibrations (phonons). We developed a PERTURBO code to quantitatively predict intrinsic transport properties in materials from first-principles, which has been applied to computing mobility in complex materials, such as organic crystals, strongly anharmonic crystals, and oxides with strong electron-phonon coupling and polaron. We also carried out ab initio simulations of hot carrier relaxation in the presence of high electric field and coupled dynamics of electrons and phonons in semiconductors, which provides microscopic insight into the ultrafast processes in materials.

Introduction

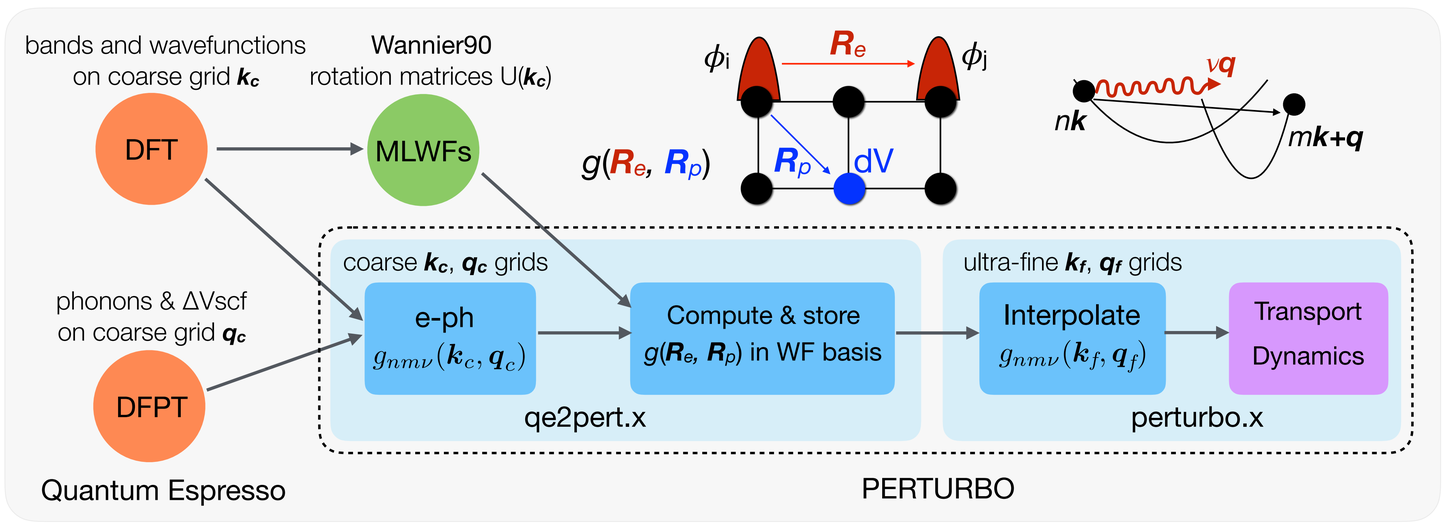
Understanding the microscopic mechanisms of light capture and conversion to fuels is crucial for designing new strategies and discovering new materials to improve the efficiency of photocatalysis.

In photocatalysis, the photo-excited carriers must be able to reach the surface/interface to participate in the desirable reactions before recombining. Hence, carrier dynamics and charge transport play a key role in photocatalytic materials.

Here, we use ab initio calculations to investigate carrier transport and relaxation processes in promising photocatalytic materials

Results, Highlights, and Accomplishments

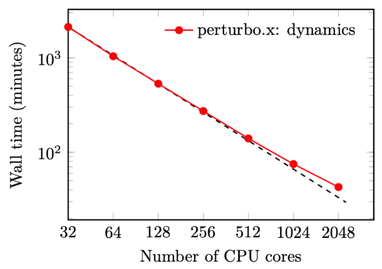
PERTURBO: computing carrier dynamics in materials from first-principles



J.-J. Zhou, *et.al.* arXiv: 2002.02045 (2020)

https://perturbo-code.github.io

Perturbo



Parallel Performance

Capabilities:

Conductivity
Carrier mobility
Seebeck coefficient
Scattering mechanisms
Hot carrier relaxation
Polaron
...

Applicable to:

metals,
semiconductors,
insulators,
2D materials,
...

Outlook

We have integrated all the approaches we developed into a standalone code, which is well optimized to achieve high computational efficiency and parallel efficiency.

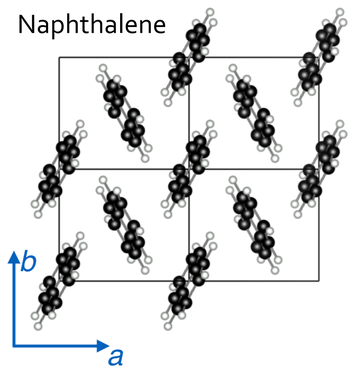
The new code can be applied to compute charge transport and simulate hot carrier cooling in complex materials with soft phonon modes and strong spin-orbit coupling, including a range of promising photocatalytic materials.

We will further explore new first-principles approaches to investigate large and small polaron transport in oxides.

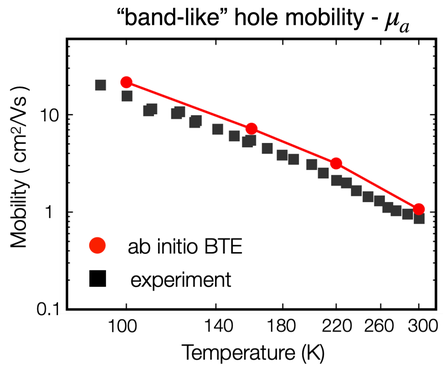
Charge transport

- Complex materials: Organic crystals

Naphthalene

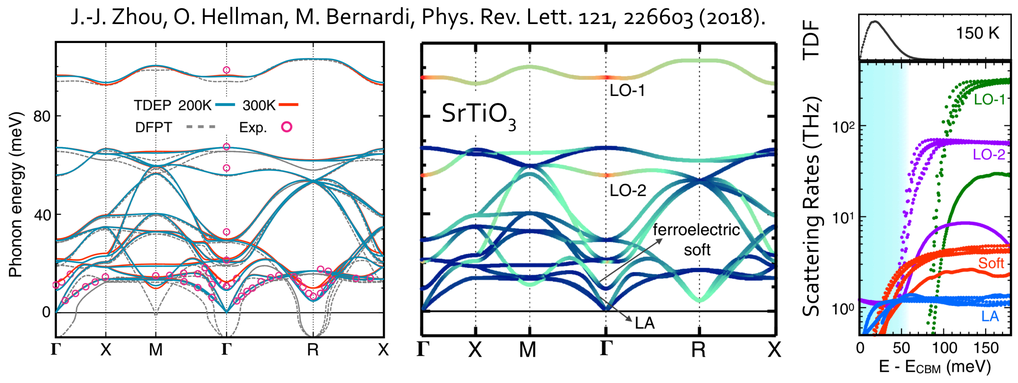


M. Bernardi *et.al* , Phys. Rev. B 97, 115203 (2018).



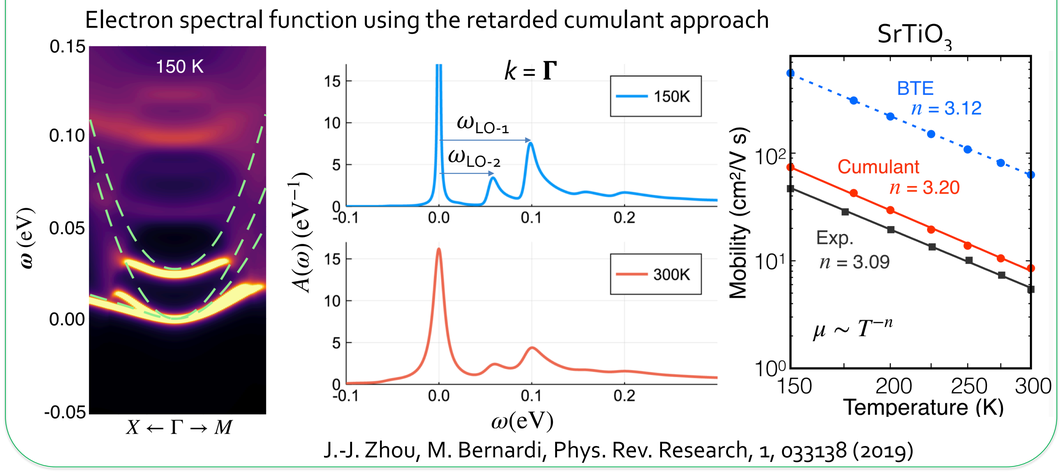
- Strongly anharmonic crystals: soft phonon modes

J.-J. Zhou, O. Hellman, M. Bernardi, Phys. Rev. Lett. 121, 226603 (2018).



- Strong electron-phonon coupling: polaron transport

Electron spectral function using the retarded cumulant approach



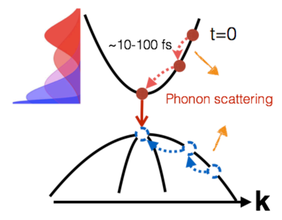
J.-J. Zhou, M. Bernardi, Phys. Rev. Research, 1, 033138 (2019)

Ultrafast carrier dynamics

- Boltzmann Transport Equation (BTE)

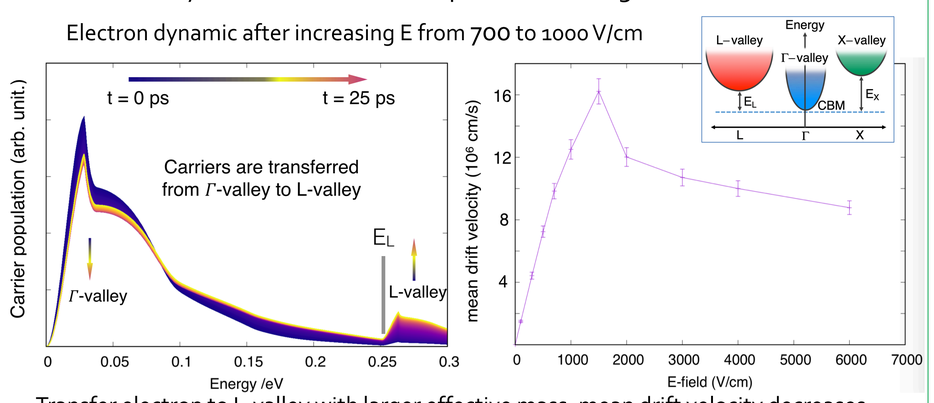
$$\frac{\partial f_{n\mathbf{k}}(t)}{\partial t} = \frac{-e\mathbf{E}}{\hbar} \cdot \frac{\partial f_{n\mathbf{k}}(t)}{\partial \mathbf{k}} + \left(\frac{\partial f_{n\mathbf{k}}}{\partial t}\right)_{\text{collisions}}$$

✓ Electron-phonon collision terms are computed completely from first-principles.
✓ Solve BTE numerically to obtain time evolution of electron occupation (and phonon occupation).



- Electron Dynamics in GaAs in the presence of High Electric-field

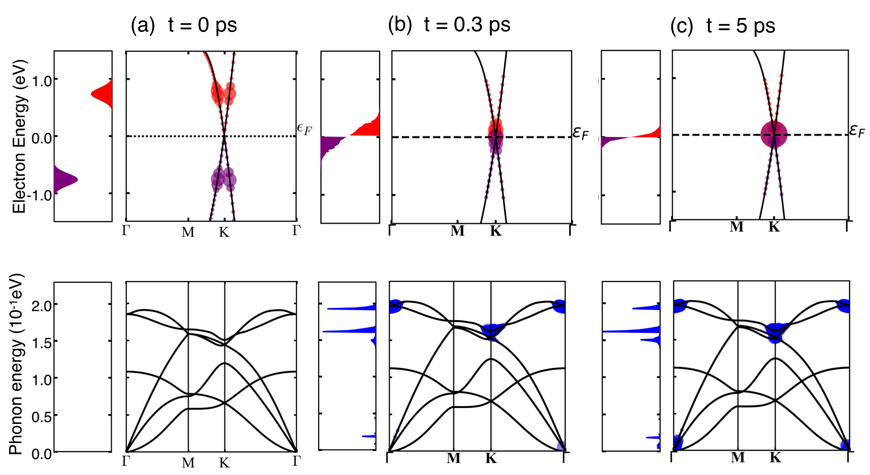
Electron dynamic after increasing E from 700 to 1000 V/cm



Transfer electron to L-valley with larger effective mass, mean drift velocity decreases.

William McCorkindale, Jin-Jian Zhou, Marco Bernardi (in preparation)

- Coupled electron and phonon dynamics in Graphene



Xiao Tong, Jin-Jian Zhou, Marco Bernardi (in preparation)

Acknowledgments

This material is based upon work performed by the Joint Center for Artificial Photosynthesis, a DOE Energy Innovation Hub, supported through the Office of Science of the U.S. Department of Energy under Award Number DE-SC0004993.