The **theoretical spectroscopy** group at LSI develops theoretical and numerical approaches for the calculation of electronic properties, in particular for the interpretation and prediction of many-body effects in excitation spectra measured by various spectroscopies. For more information please visit: https://etsf.polytechnique.fr/. Contact person: Vitaly Gorelov vitaly.gorelov@polytechnique.edu. In collaboration with Matteo Gatti and Lucia Reining.

Quantum many-body effects in the design of materials for photovoltaics.

The behavior of charge excitations in materials is a key process for optoelectronics technology, that raises numerous fundamental questions. In the field of photovoltaics (PV), semiconductor models based on the independent-particle picture can give precious insight but depend on adjustable parameters and are tailored to specific classes of materials. This limits their capability to make reliable predictions and to profit from the opportunities offered by many-body phenomena to achieve a breakthrough in the improvement of device efficiency. Electron-hole coupling, hot electrons coupled to phonons, and multiple exciton generation, are all challenging effects of the electronic interactions that can be turned into novel pathways for improved solar cells.
Most often, a three-step picture is used to describe a PV device, where the **creation of an electron-hole pair**, its **propagation**, and the **separation of charges** are described separately.
There are challenging questions for each of the three steps:

1. **What makes a material an optimal light absorber?**
   Here, the main goal is to understand how to exploit electronic interactions to increase the light absorption of materials. We will use and develop theoretical and numerical methods based on state-of-the-art Green's function theory [1] to investigate a class of materials, recently termed metavalent [2], that display extraordinarily large light absorption.
   [1] R. M. Martin, L. Reining, D. M. Ceperley, Interacting Electrons: Theory and Computational Approaches (Cambridge University Press)
   [2] B.J. Kooi, M. Wuttig, Chalcogenides by design: functionality through metavalent bonding and confinement, Advanced Materials 32 (21), 1908302 (2020).

2. **How to simulate efficiently the propagation of charges in complex materials?**
   Once created by absorption of light, the electronic charges propagate in the material. The main challenge here is to compute charge dynamics efficiently in realistic materials that are inherently complex. We will extend to the question of charge dynamics a computational scheme, named Connector Theory (COT) [3], that has been recently introduced by our group for efficient calculations of materials properties importing knowledge from models.
   [3] M. Vanzini, et al. Connector theory for reusing model results to determine materials properties. npj Comput Mater 8, 98 (2022)

3. **How do electron-hole pairs split?**
   In the PV device electron-hole pairs need to be separated into free holes and electrons to form a current. To elucidate the process of electron-hole separation, we will develop analytical models and solve the Bethe-Salpeter equation (BSE) within many-body perturbation theory for studying electron-hole excitations in realistic

materials. Our group pioneered the development of the BSE, which is now routinely used to understand the electron-hole coupling effects in the optical spectra of complex materials [4].

[4] V. Gorelov, et al. Delocalization of dark and bright excitons in flat-band materials and the optical properties of V2O5. npj Comput Mater 8, 94 (2022).

The aforementioned questions are interconnected: interested candidates will contribute to at least one of them.