

Understanding the Role of Excitons in Nonlinear Optical Responses: The Case of Bulk Photovoltaic Effect

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The optical response of materials often reveals information about the electronic properties of a system. During the last two decades, ab initio calculations have allowed the precise characterization of optical properties in real materials, especially in semiconductors and insulators. Such a fruitful outcome has been possible thanks to an appropriate treatment of many-body interactions when describing excited states. In recent years, ab initio methods for optical properties have been pushed to calculate quantities beyond the linear regime in the external electric field.

In this talk, I will show how I structure ab initio calculations into a computational framework that allows the exploration of linear and second-order optical responses of crystals in different scenarios. An important tool in my scheme is the Optix package, which is currently under development. The aim of Optix is to be a package fully devoted to the calculation of optical properties, after interfacing with a band-structure or excitonic input description of the electronic excitations. Thus, Optix is neither an electronic-structure nor a Bethe-Salpeter code per se, but a suite to implement calculations of optical properties in a unified way once the excited-state spectrum is available. The main benefit is the flexibility and transferability of the code across different workflows regarding: (i) different basis sets such as Wannier orbitals or plane waves, and (ii) interfacing with different exciton-related codes (including ab initio or effective-interaction approaches).

As an example of the methodology, I will discuss some ongoing studies on the bulk photovoltaic effect. First, I will show how to calculate its associated optical response function from the second-order conductivity tensor in 2D materials, where excitonic effects are important [1,2]. Special attention will be paid to the role of the inter-exciton matrix elements of the dipole operator, a key quantity in these calculations that highlights the extra complexity with respect to the usual calculations of the macroscopic dielectric function. Next, I will present a model for excitons under strong light-matter interaction with a cavity mode in a coherent state. I will explain how the excitonic states are dressed by the cavity field and how to calculate their associated nonlinear optical response.

[1] J. J. Esteve-Paredes, M. A. García-Blázquez, A. J. Uría-Álvarez, M. Camarasa-Gómez, and J. J. Palacios, *npj Comput. Mater.* **11**, 13 (2025).

[2] A. J. Uría-Álvarez, J. J. Esteve-Paredes, M. A. García-Blázquez, and J. J. Palacios, *Comput. Phys. Commun.* **295**, 109001 (2024).