Seminar for Postgraduates (Experimental- and Solid State-Physics)

Extending Photœmission Orbital Tomography to the Time-Domain

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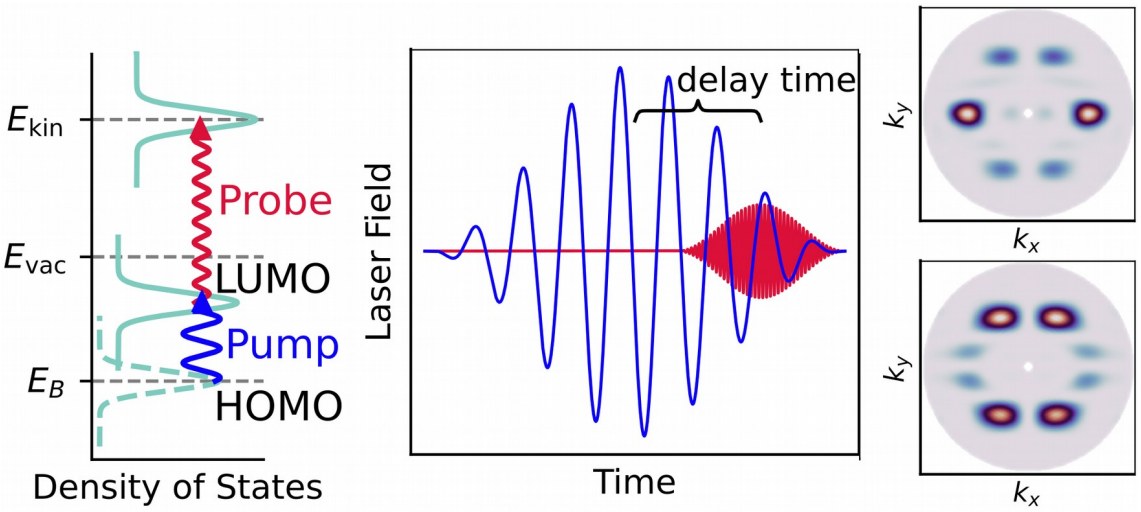
In order to gain information about the quantum nature of materials, photœmission experiments are among the most useful techniques, since they provide a direct access to the electronic structure in momentum-space. By combining these measurements with theoretical simulations, Photœmission Orbital Tomography[1] (POT) is a quantitative method to interpret the momentum-space signatures and connect these to the real-space electron distributions. This intimate connection is established by the theoretical formulation of the photœmission process with a plane-wave final-state. Although this approach is highly successful for e.g. organic molecules on (metal) surfaces,[2] it has some limitations that are important to describe.

For this purpose, we use a more sophisticated method that simulates the photœmisson process in real-time with Time-Dependent Density Functional Theory (TDDFT) and the Flux-method,[3] which allows us to get a better polarization-dependence or describe subtle effects, such as Circular Dichroism.

Moreover, by extending POT on the time-domain, we get access to time-resolved photœmission spectroscopy. Here, one can e.g. excite the system with a first photon and then probe this excited state with a second photon (pump-probe spectroscopy), such that it is possible to simulate the formations of excitons directly in time. This technique can be applied to molecules or extended systems, where we can couple the time-resolved tomography also to valley- or spin-degrees of freedom and thus get a more complete picture of quantum states of matter.



1 https://en.wikipedia.org/wiki/Photœmission_orbital_tomography

2 Puschnig et. al.: *Reconstruction of Molecular Orbital Densities from Photœmission Data,* Science ***326***, 2009

3 De Giovannini et. al., *A First-Principles Time-Dependent Density Functional Theory Framework for Spin and Time-Resolved Angular-Resolved Photœlectron Spectroscopy in Periodic Systems,* JCTC **13**, *2017*