Modeling out of equilibrium time-resolved ARPES in real materials with QED-TDDFT: application to WSe2

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Computer simulations that predict the light-induced change in the physical and chemical properties of complex systems, molecules, nanostructures and solids usually ignore the quantum nature of light. Recent experiments at the interface between materials science and quantum optics have uncovered situations where both the molecular system and the photon field have to be treated in detail. In this talk, we show how theoretical approaches have to be adapted to treat such coupled matter–photon problems and which effects can be anticipated. We demonstrate how the effects of treating quantum-photons can be properly included in such calculations.

We will address how periodic driving of many-body systems offers a platform to design Floquet states of matter with tunable electronic properties on ultrafast time scales. Based on the previous QED-TDDFT theoretical framework, we will introduce Floquet time-dependent density functional theory (Floquet-TDDFT) as a general and robust first principles method for predictive Floquet engineering of topological states of matter. Using this scheme we show how femtosecond laser pulses with circularly polarised light can be used to switch between Weyl semimetal, Dirac semimetal, and topological insulator states in a prototypical 3D Dirac material, Na3Bi. Our findings are general and apply to any 3D Dirac semimetal. Furthermore, we show that the concepts of Floquet analysis can be applied to monitor electron photon and electron-phonon dressing in 2D materials . This coupling leads to phonon- or photon-dressed quasi-particles (polarons or polaritons) imprinting specific signatures in the spectrum of the electronic structure that can be detected by time dependent ARPES as sidebands to the equilibrium band structure. Most strikingly, we find that the non-equilibrium electronic structure created by coherent dynamical dressing is the same for photon and phonon perturbations. We demonstrate that if time-reversal symmetry is broken by the coherent lattice perturbations a topological phase transition can be induced. The extension to spin-resolved ARPES can be used to predict asymmetric dichroic response linked to the valley selective optical excitations in monolayer transition metal dichalcogenides (TMDs). This work establishes that the recently demonstrated concept of light-induced non-equilibrium Floquet phases can also be applied when using coherent phonon modes for the dynamical control of material properties. The present results are generic for bosonic time-dependent perturbations, therefore we envision similar phenomena to be observed for example for plasmon, magnon or exciton driven materials.