Pump driven choerence: evidence of an excitonic insulator transition or normal charge migration effects?

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Ultra-fast spectroscopy is a powerful tool for the observation of electronic and atomic dynamical processes. The initial photo-excitation creates an avalanche of phenomena that drives the system dynamics. The description of this complex dynamics is really challenging.

In a basic Pump and Probe experiment a first light pulse (the pump) resonantly triggers a photo-induced process exciting electrons from the valence to the conduction bands. In equilibrium conditions it is well–known that electron–hole attraction locks the pairs in bound excitonic states. Even if the bosonic nature of these states is highly debated [2] it is clear that two alternative pictures appear to be possible to describe the photo–excited dynamics: an excitonic versus a carriers based approach.

In this talk I will discuss two extreme cases where the two approaches seem to be definitively the most appropriate: the photo–induced band insulator to excitonic insulator transition [4] and the ultrafast rise time of the transient absorption in an MoS_2 monolayer [5].

In this talk I will first review the Ab Initio Non Equilibrium Green's function (AiNEGF) method[3], which represents a new frontier in the realm of atomistic simulations.

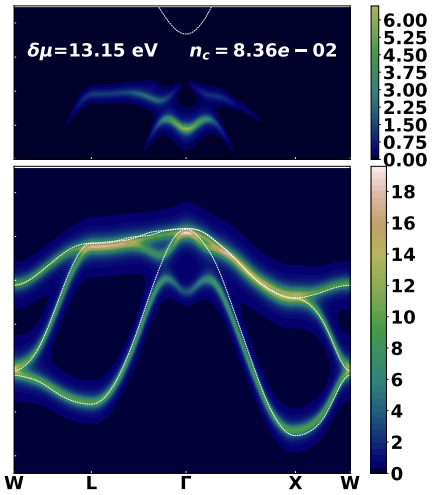

Figure 1: Occupied part of the spectral function of a bulk LiF for different values of Excitonic energy. The excitonic resonance clearly appears as a isolated band in the conduction energy region. From Ref.[4]

In the second part of the talk I will use the AiNEGF to describe a novel phenomenum: the photo–induced band insulator to excitonic insulator (NEQ-EI) transition in complex materials[4]. I will show that the NEQ-EI phase is characterized by self-sustained oscillations of the complex order parameter. I will connect the peculiar fingerprints of the NEQ-EI phase to the features observed in time-resolved angle-resolved experiments.

In the third and last part of the talk I will, instead, study another experiment [1], based on the transient reflection technique, performed on a single MoS_2 layer. The experimental data point to a rapid rise signal whose speed depends on the pump energy. This fast rise is interpreted in Ref.[1] as due to a delicate interplay of the excitonic population and polarization. Here I will, instead, demonstrate as an excellent agreement with the experiment can be obtained by using a single–particle picture and establishing a close link between the transient trasmission signal and the carriers population dynamics.

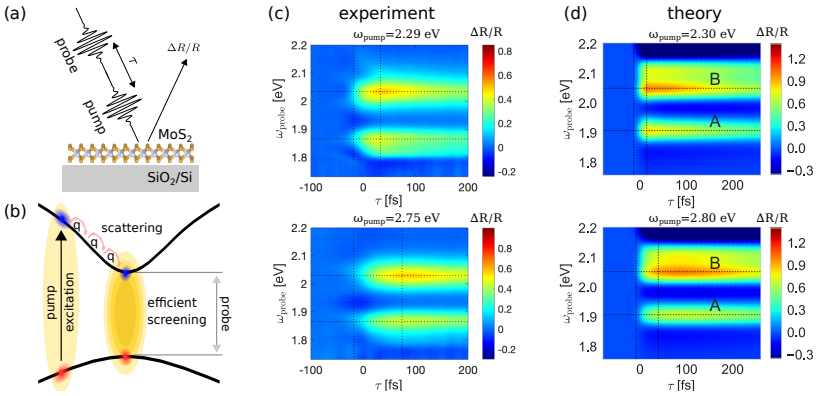

Figure 2: a) Sketch of a pump-probe scheme in reflection geometry. (b) Laser-induced dynamics: a tunable pump pulse creates electron-hole pairs that change the screening properties. As electrons and holes scatter to the band extrema, the screening becomes more efficient. (c) Experimental and (d) theoretical energy- and delay-time-dependent transient reflectivity maps for two different pump energies ω_{pump}. The theoretical data is normalized to the maximum of $\Delta R/R$. From Ref.[5]

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