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Pump and Probe experiments from first-principles in bulk silicon:

ultra–fast carrier relaxation and transient reflectivity

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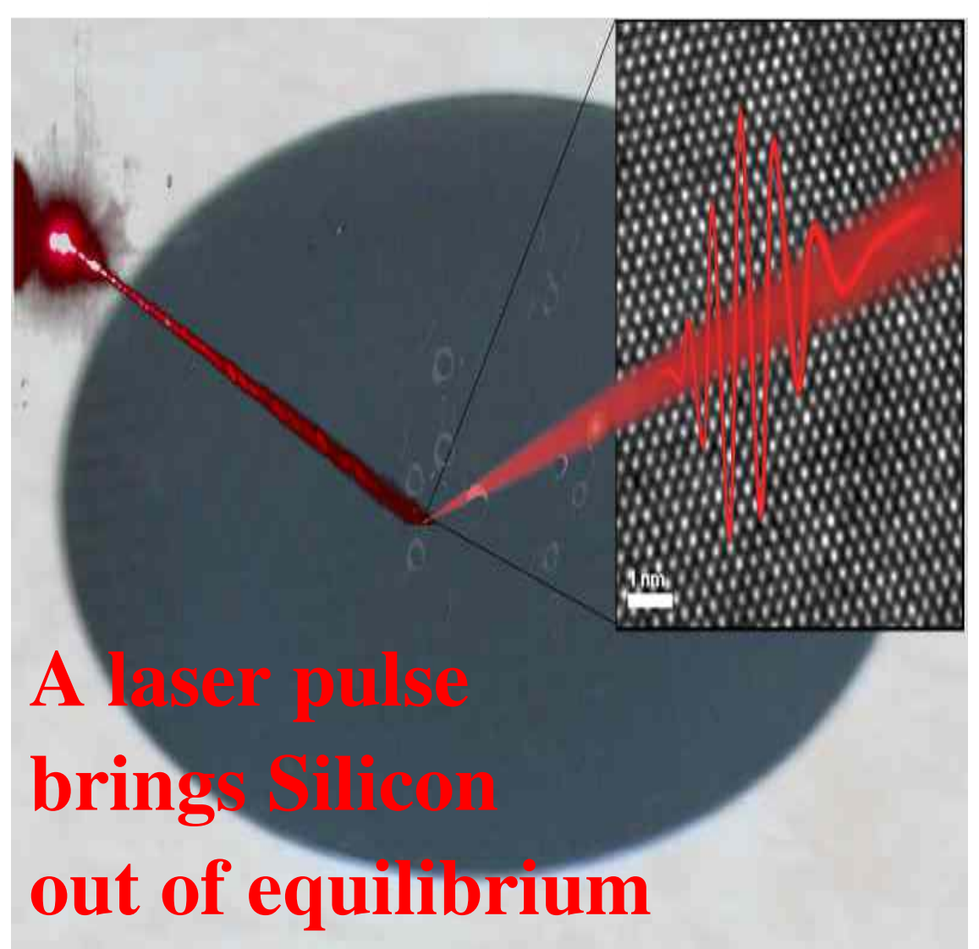


A laser pulse brings Silicon out of equilibrium

Abstract

The development of ultra-short laser pulses has opened the opportunity to investigate the dynamics of electrons on the fs time-scale (10^-15 s). After the photo-excitation with such lasers pulses, electrons are in a regime which is highly out-of-equilibrium. Here we present a novel numerical approach, based on the merging of the out-of-equilibrium Green's function method with Density-Functional-Theory, to describe this regime in semiconductors. Silicon is used as reference material to show the physical process involved. The simulations are also compared with recent two photon photo-emission and transient-reflectivity measurements. In the 2PPE experiment we show that different processes take place: (i) scattering between degenerate states, activated by the pump-pulse induced symmetry breaking, (ii) L -> X inter-valley scattering, and, finally, (iii) the relaxation towards the thermal equilibrium. In the TR experiment we underline the key role of optical-gap renormalization induced by the pump pulse, combined with bleaching, needed to explain the experimental signal. Moreover we discuss how the same approach is able to capture coherent excitons in materials with strong electron-hole interaction. This ensures a correct description of the interaction with the pump pulse. Finally we consider a model system with strong electron-hole interaction and discuss how the approach should be extended to capture the formation of non-coherent excitons populations and describe time resolved photoemission in this case.

Non-equilibrium Green functions[1-3]

Fully ab-initio approach:

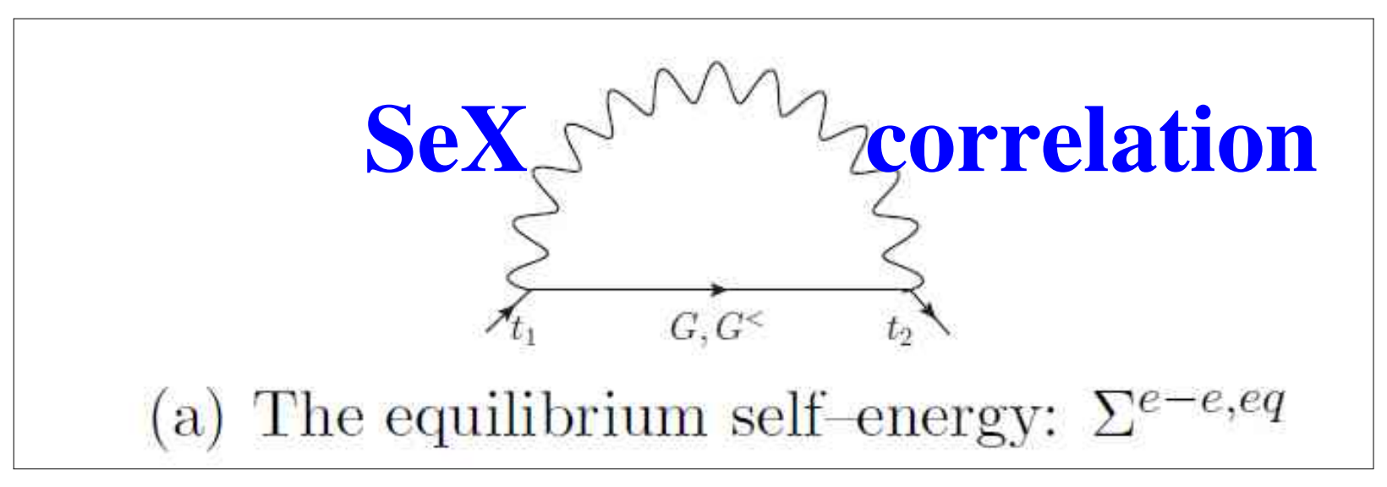
Yambo [4]

$$i\partial_t G^< - \left[H^0 + V^H + U^{ext} + \Sigma_s, G^<\right] = \{\Sigma^>, G^<\} - \{\Sigma^<, G^>\}$$

$$f^{(e/h)}_{nk}(t) \approx \mp i\,\Delta G^<_{nnk}(t) \qquad P(t) = -e\sum_{nmk} r_{nmk}\,\Delta G^<_{nmk}(t)$$

microscopic version of the optical Maxwell-Bloch equations for f(t) and P(t)

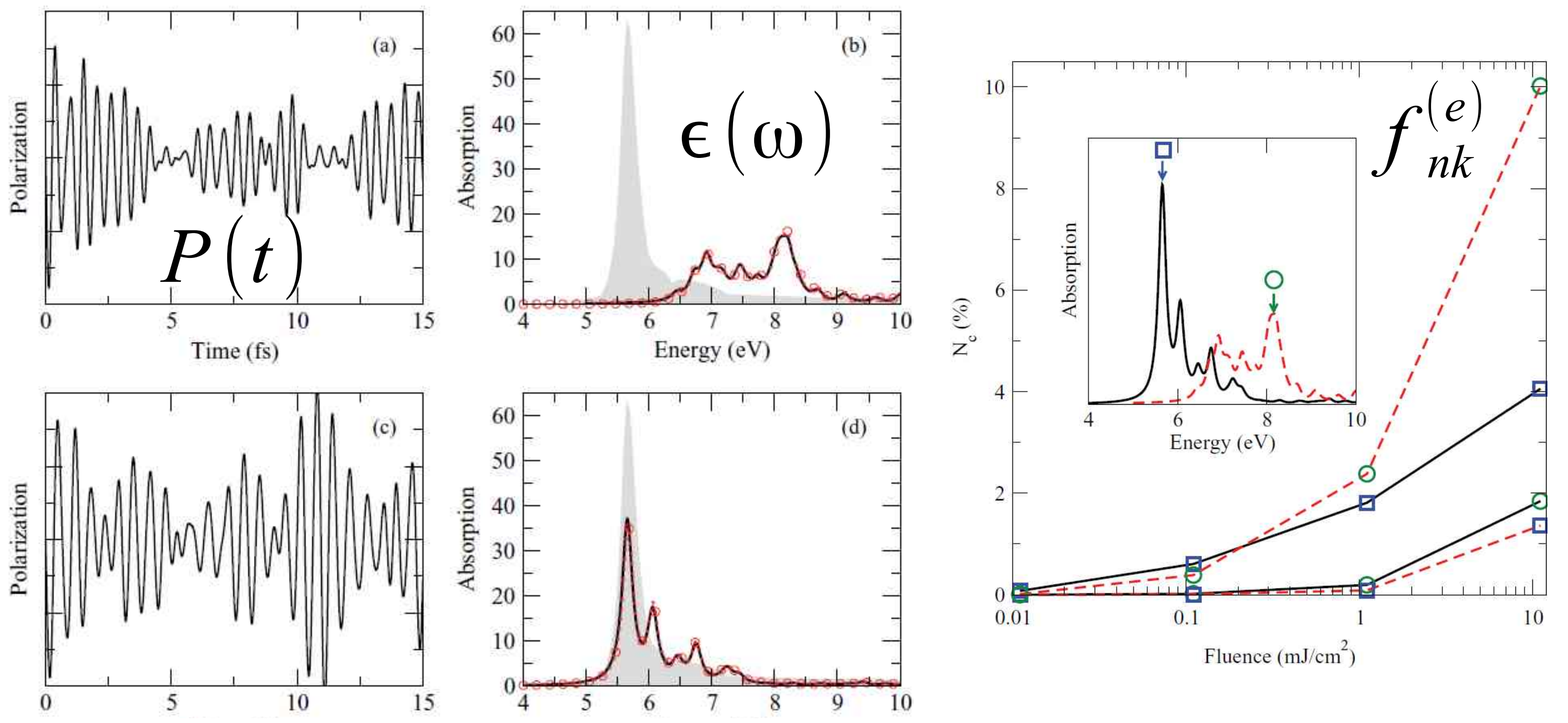
Coherent excitons: absorption of the pump-pulse [1]



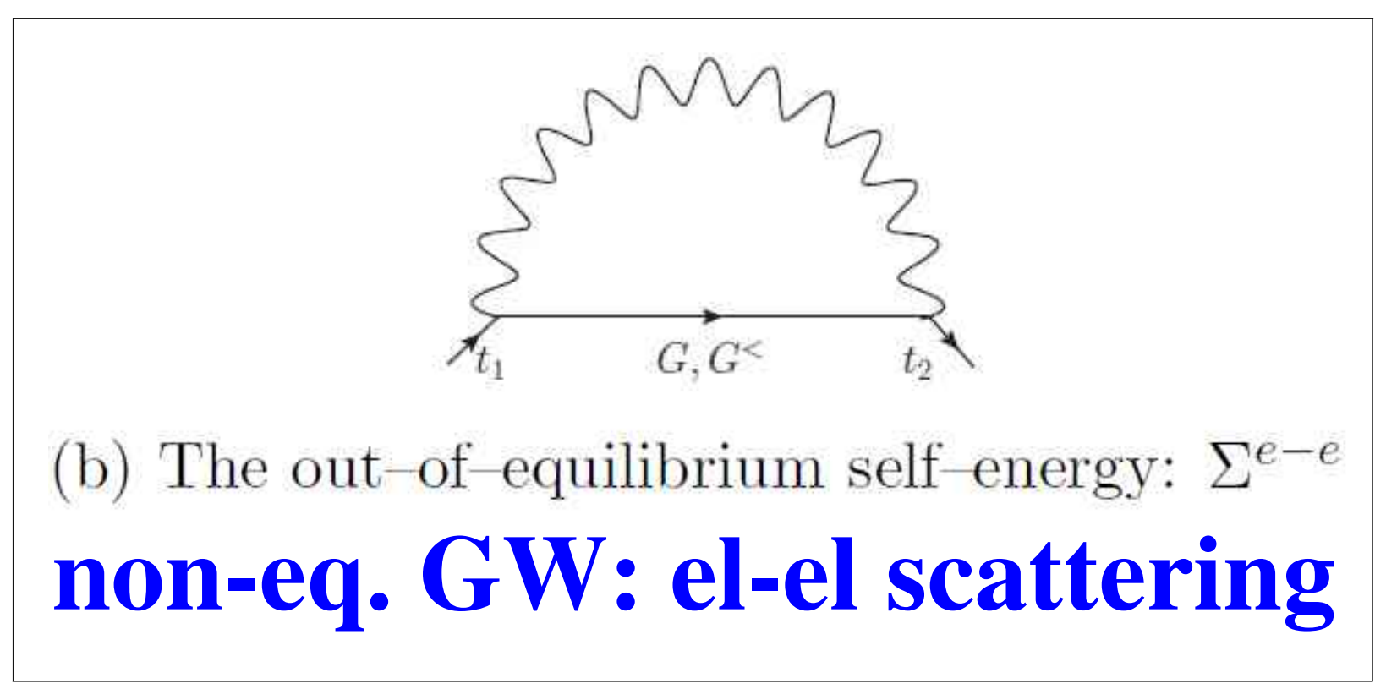
(a) The equilibrium self–energy: $\Sigma^{e-e,eq}$

Carriers generation:

- Absorption of the pump includes electron-hole interaction.
- Carriers are created according to the exciton distribution

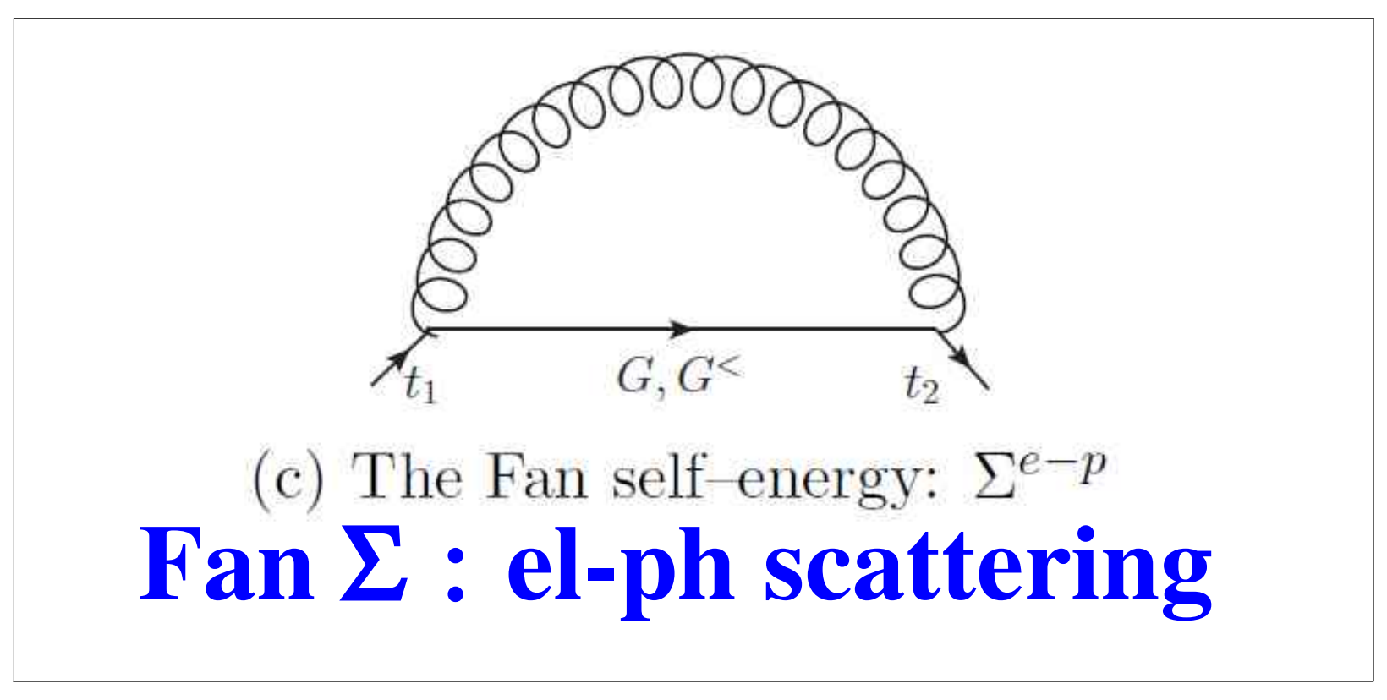


Carrier dynamics: scattering processes[2-3]



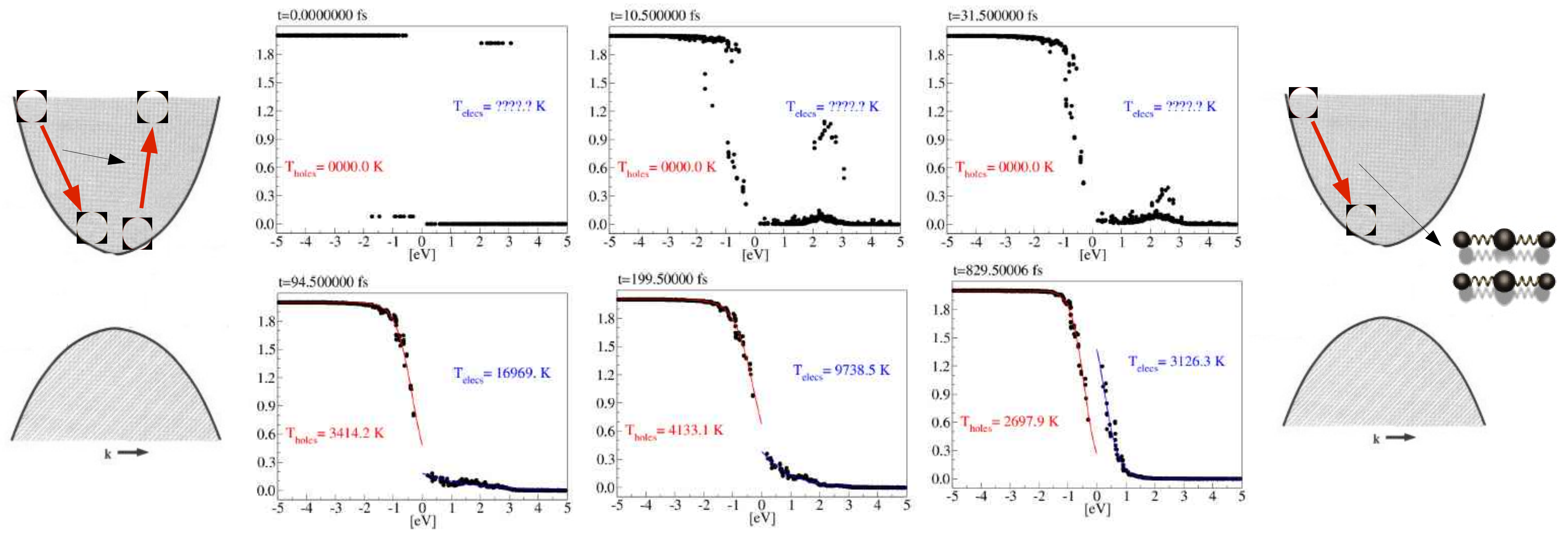
(b) The out–of–equilibrium self–energy: Σ^{e-e}

non-eq. GW: el-el scattering

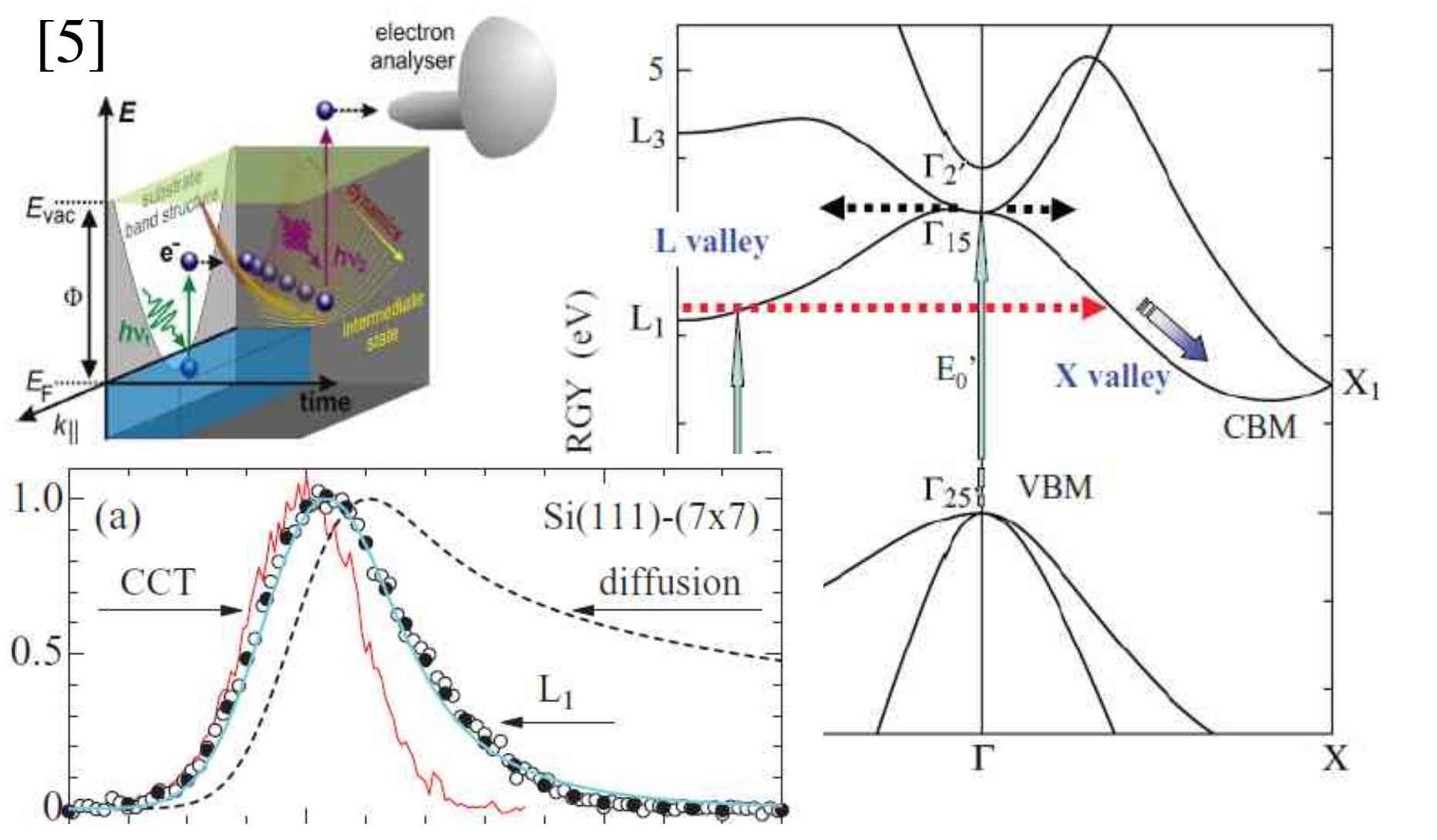


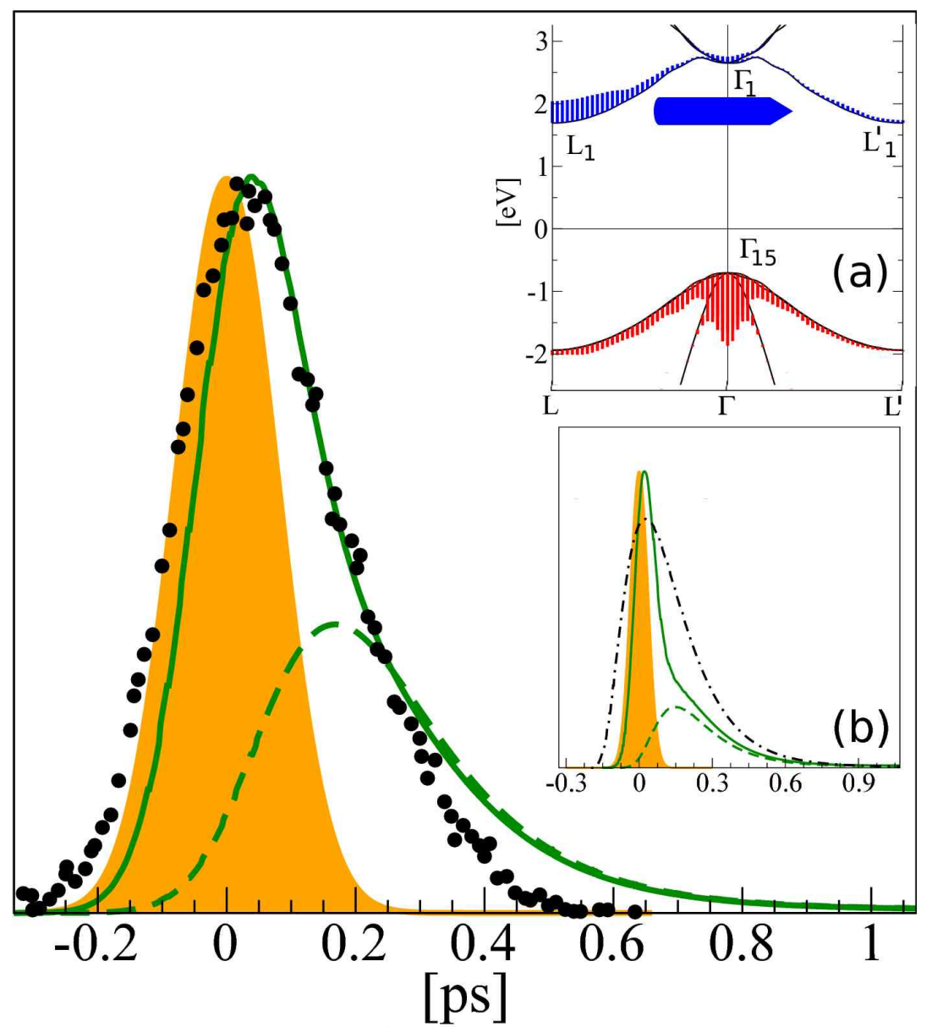
(c) The Fan self–energy: Σ^{e-p}

Fan Σ : el-ph scattering



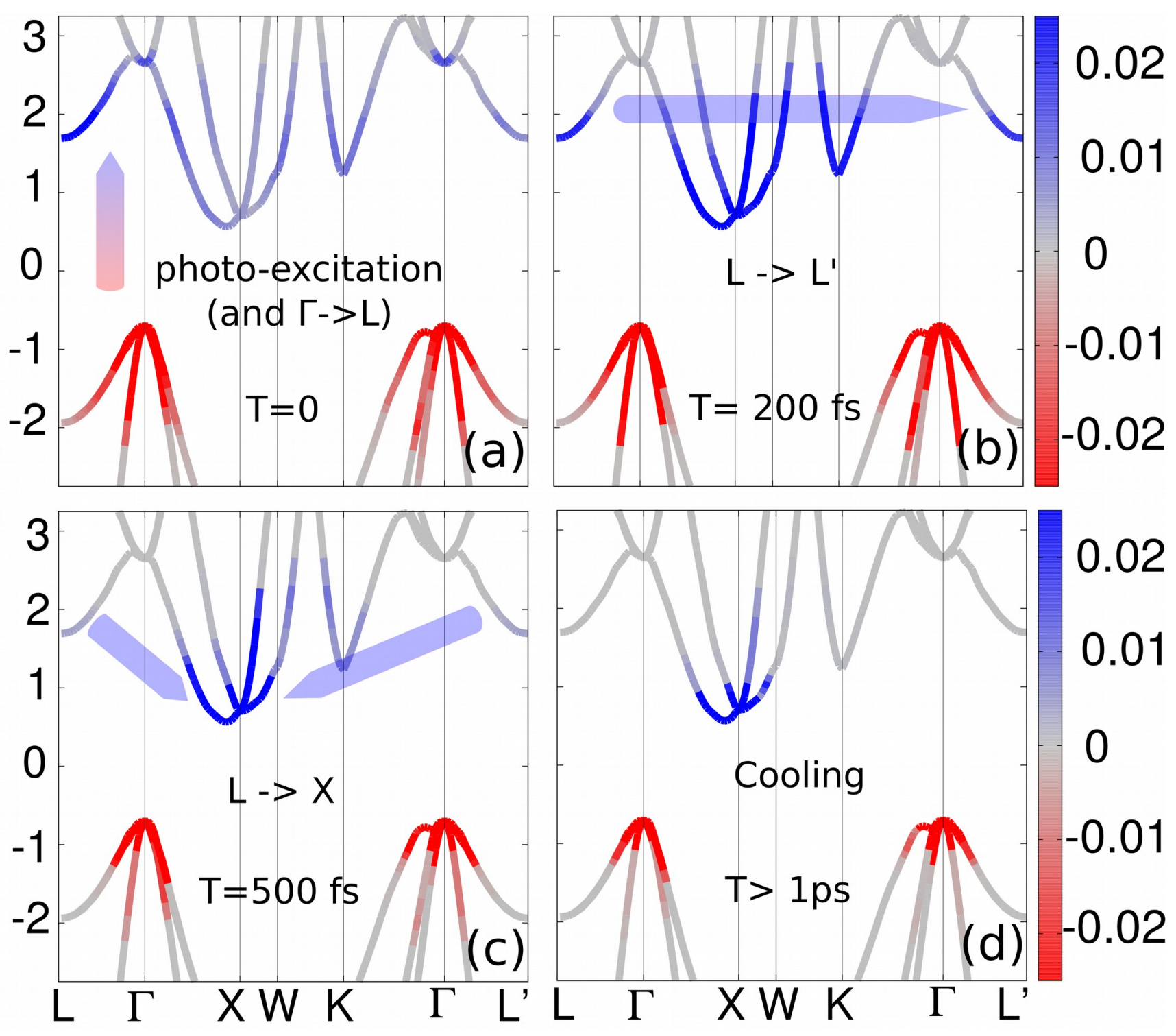
Two photons photo-emission:[6]

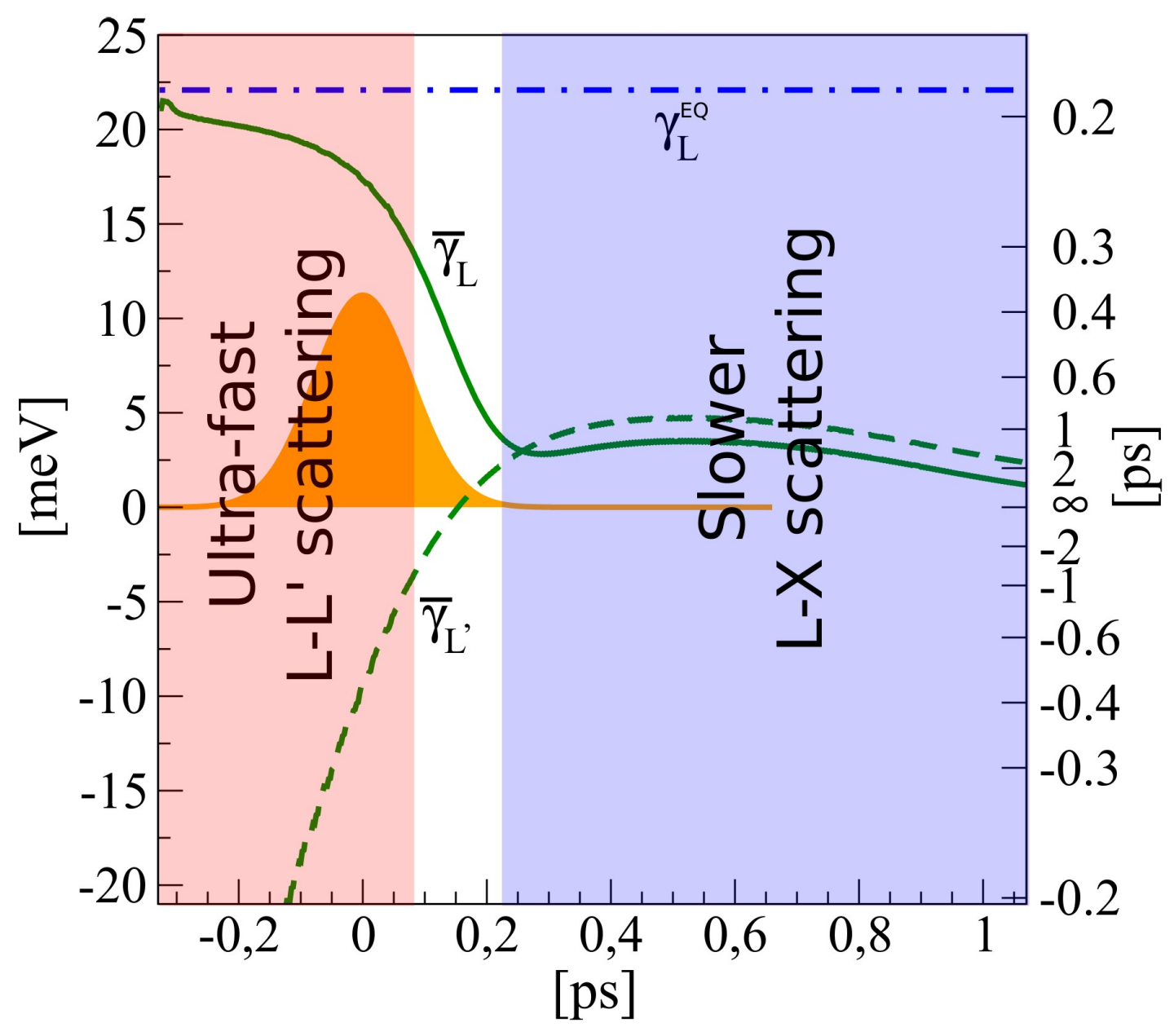




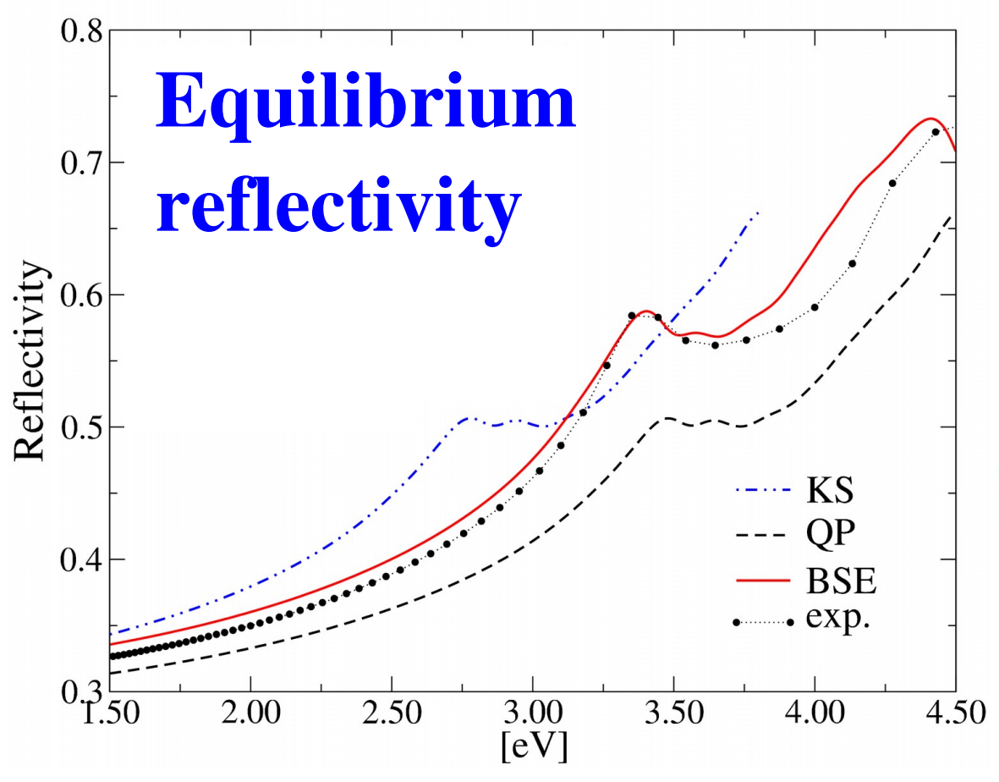
Two regimes:

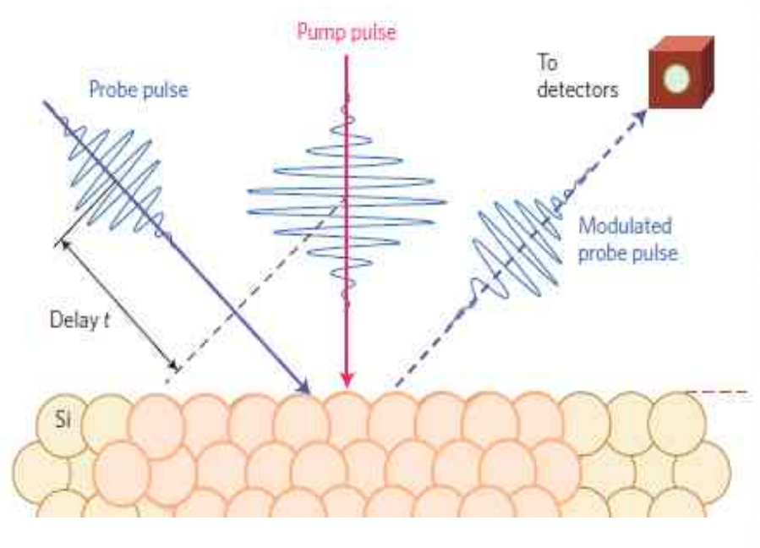
- Faster dynamics: L→L' (t<200 fs)
- Slower dynamics: L→X (t>200 fs) and thermalization (t>1 ps)

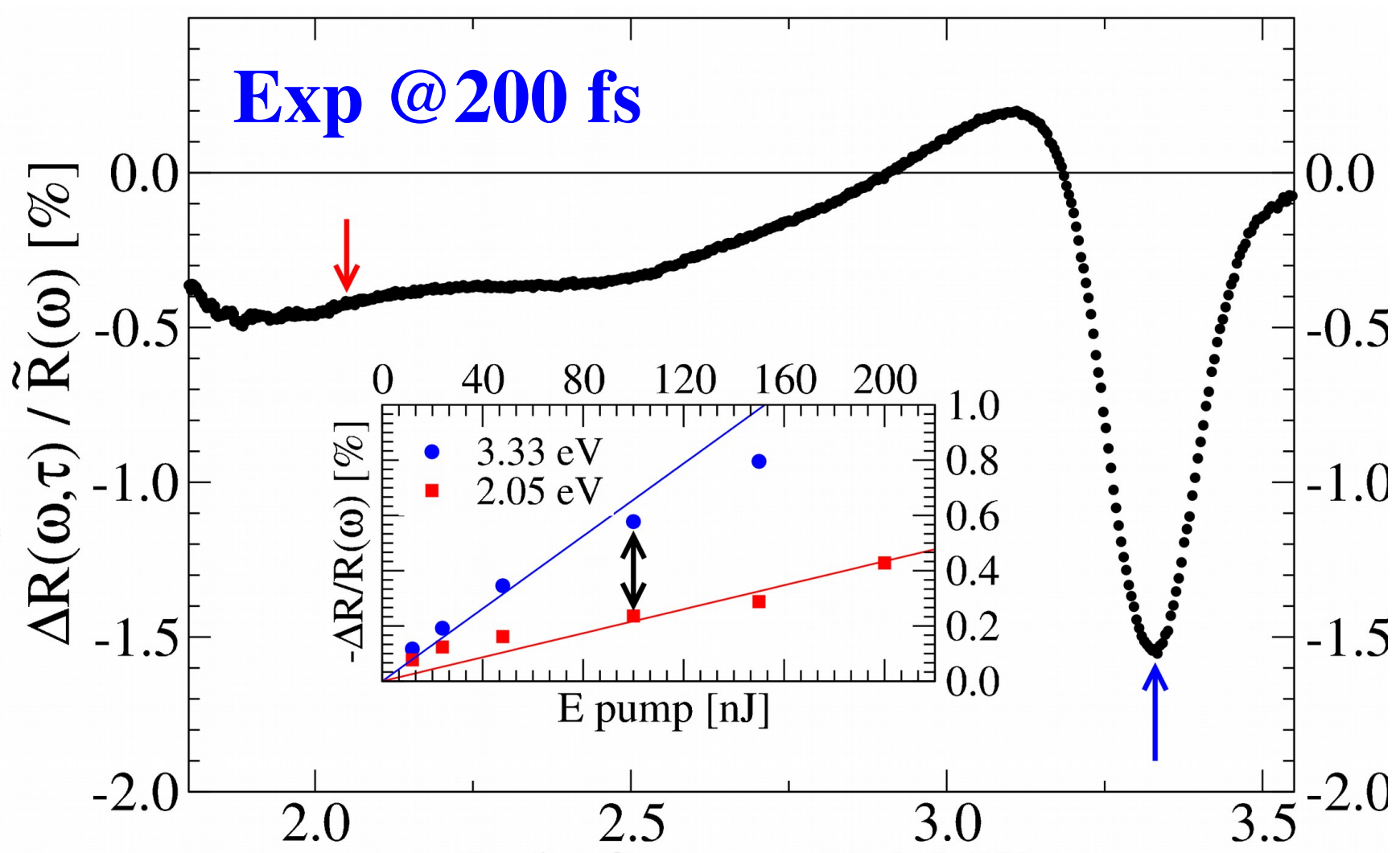




Transient reflectivity[7-8]

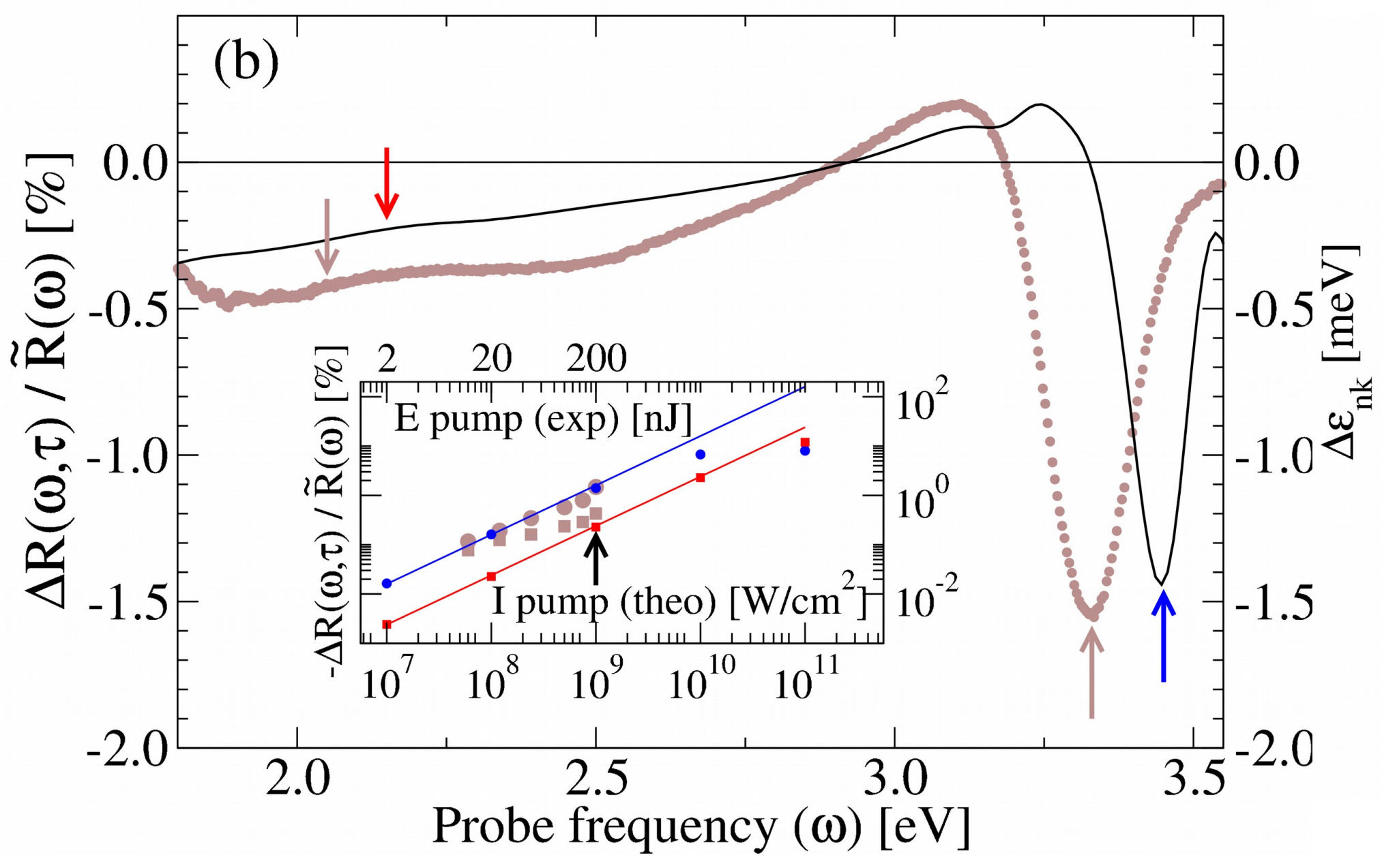


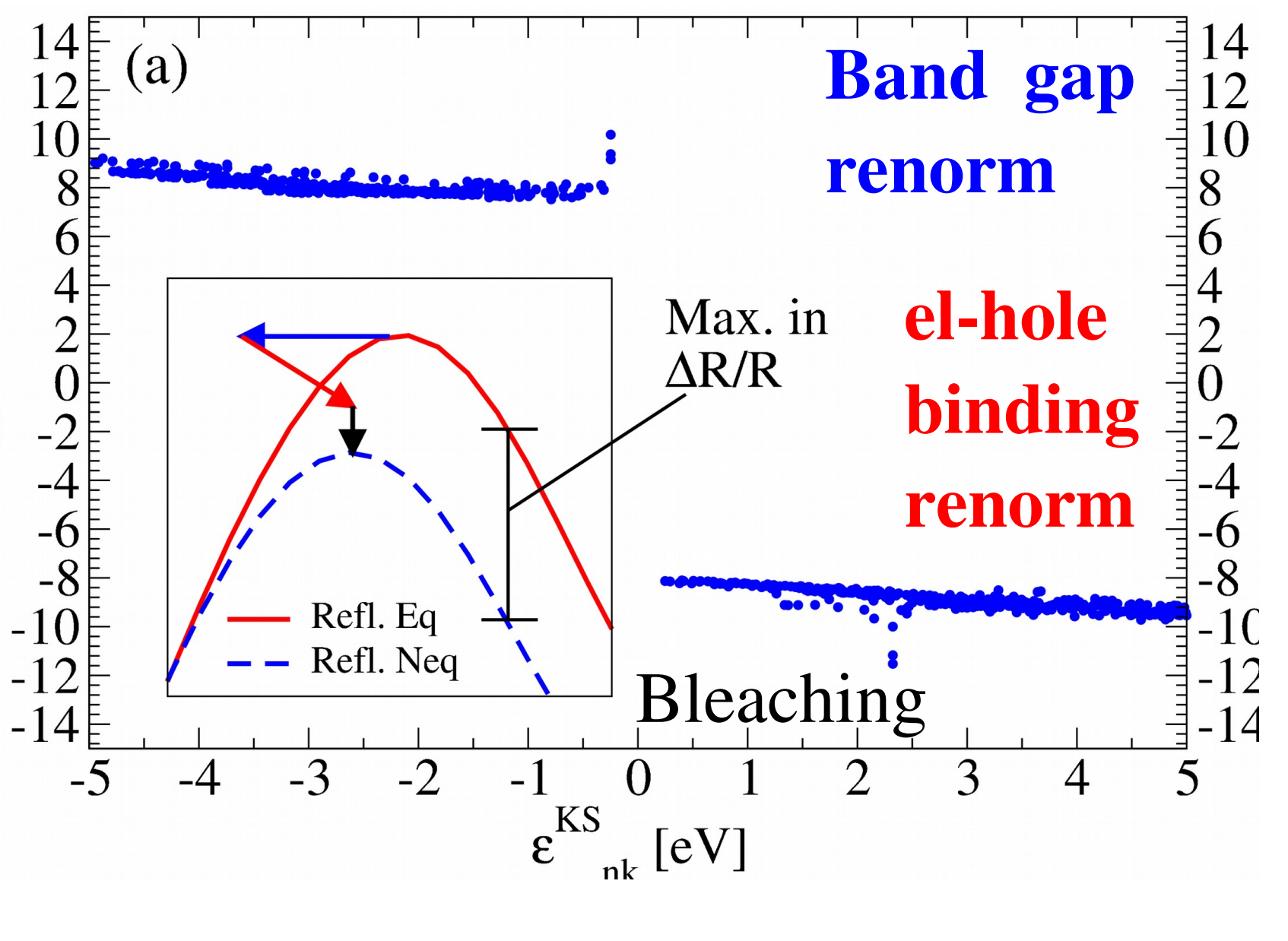




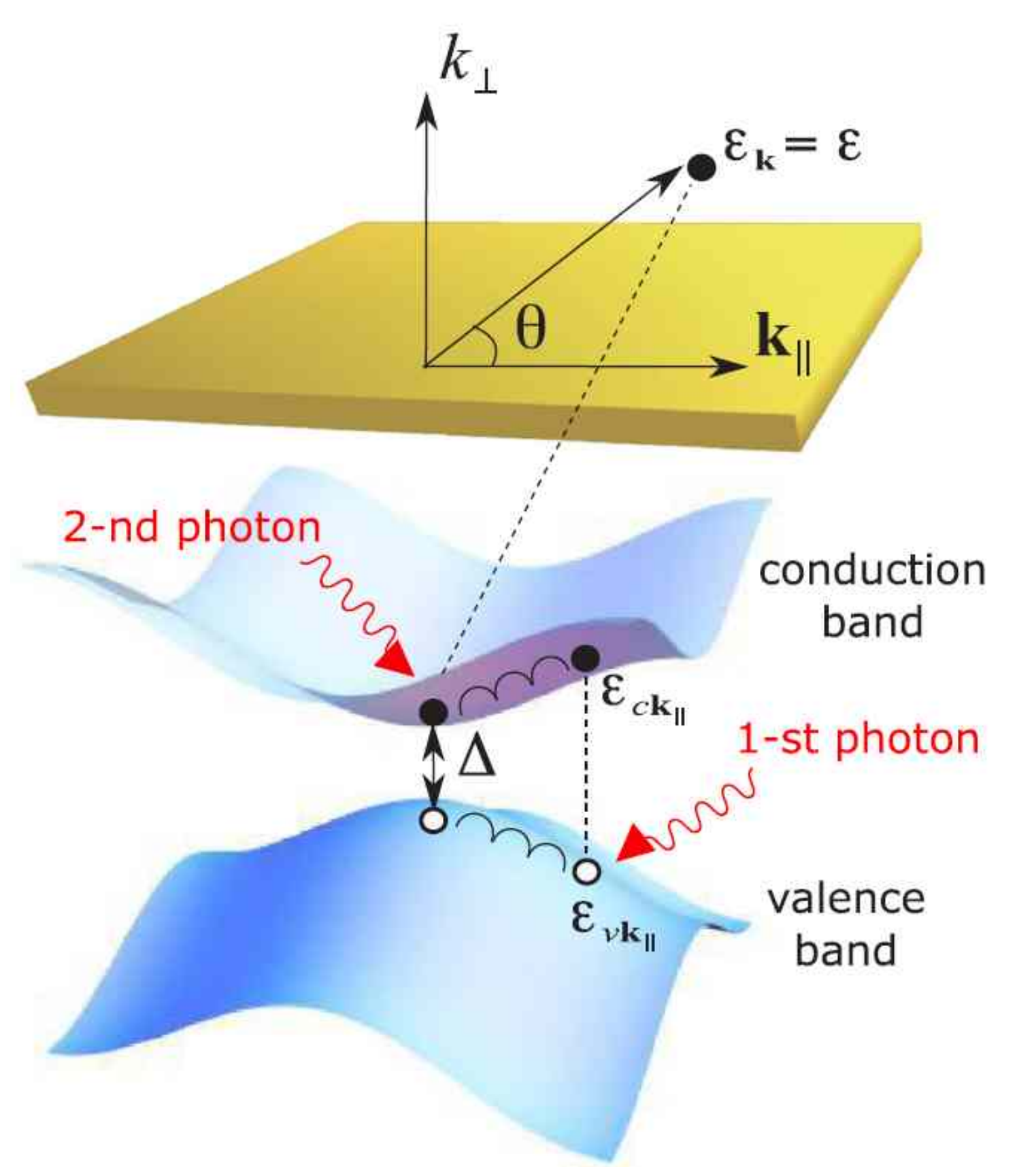
Theory vs Exp. @ 200 fs

Bleaching of the first peak + Optical gap renormalization [8]

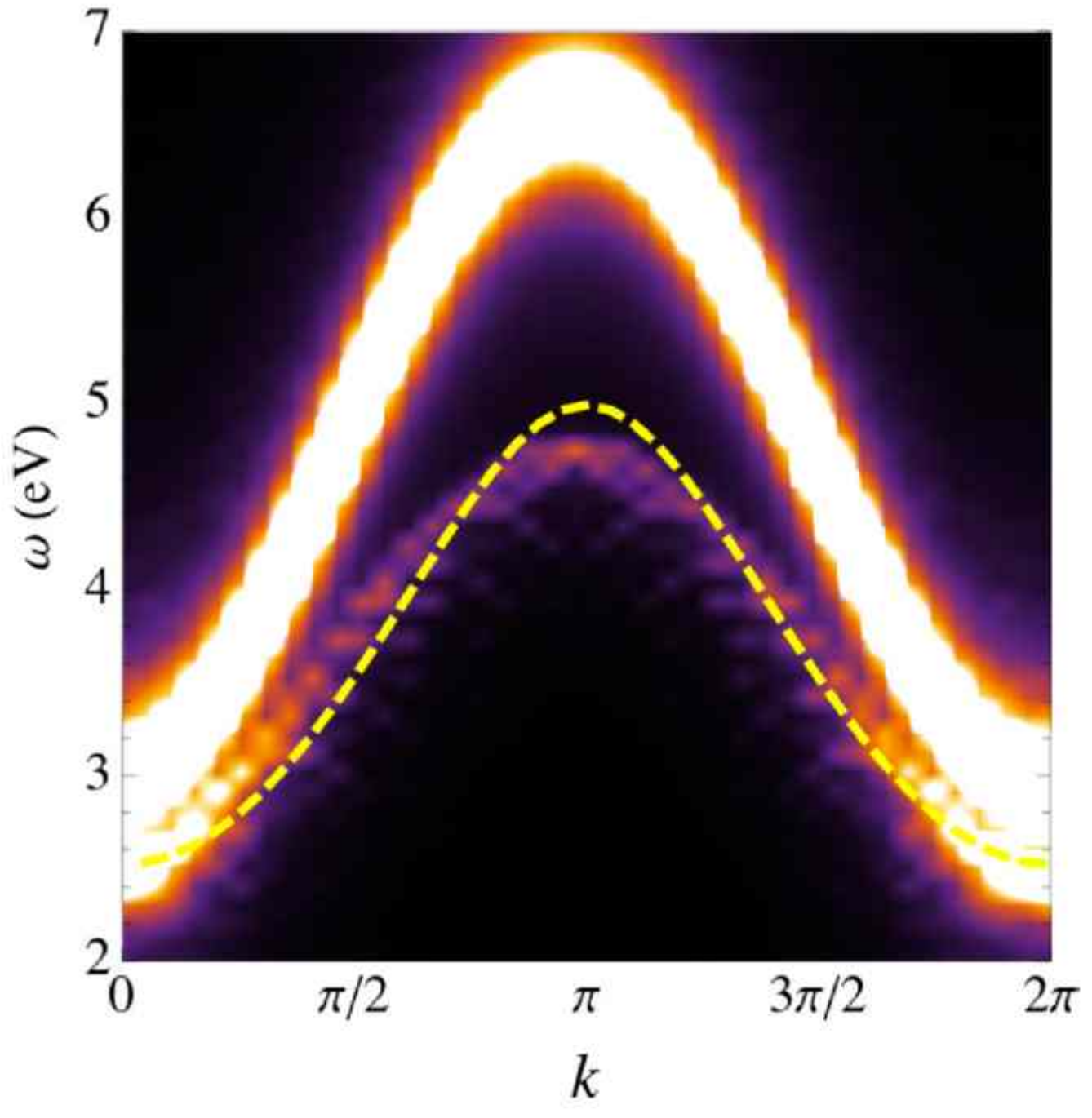




Non coherent excitons population [9]



Exciton in TR-ARPES

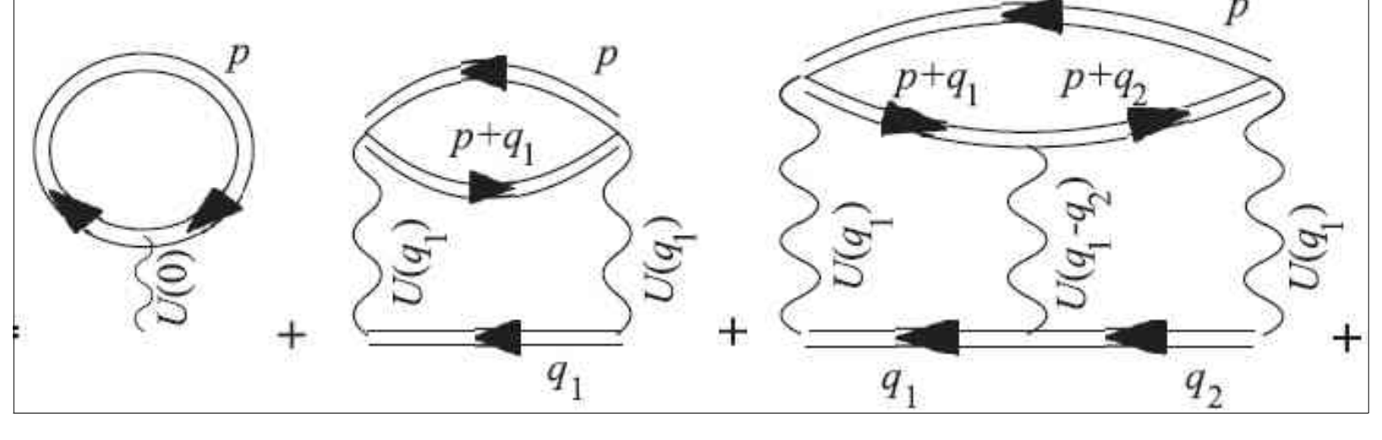


$$I(\mathbf{k}_\parallel) = -i|a_0 D_{\mathbf{k}}|^2\left[G^<_{cc,\mathbf{k}_\parallel}(\epsilon_{f\mathbf{k}} - \omega_0) + G^<_{cc,\mathbf{k}_\parallel}(\epsilon_{f\mathbf{k}} + \omega_0)\right]$$

coherent vs non-coherent excitons:

- non coherent excitons require higher order correlation
- in alternative they could be describe

T-matrix correlation



References & Acknowledgments

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