

Solids under intense ultrafast excitations:
a time-dependent Bethe-Salpeter approach

Andrea Marini

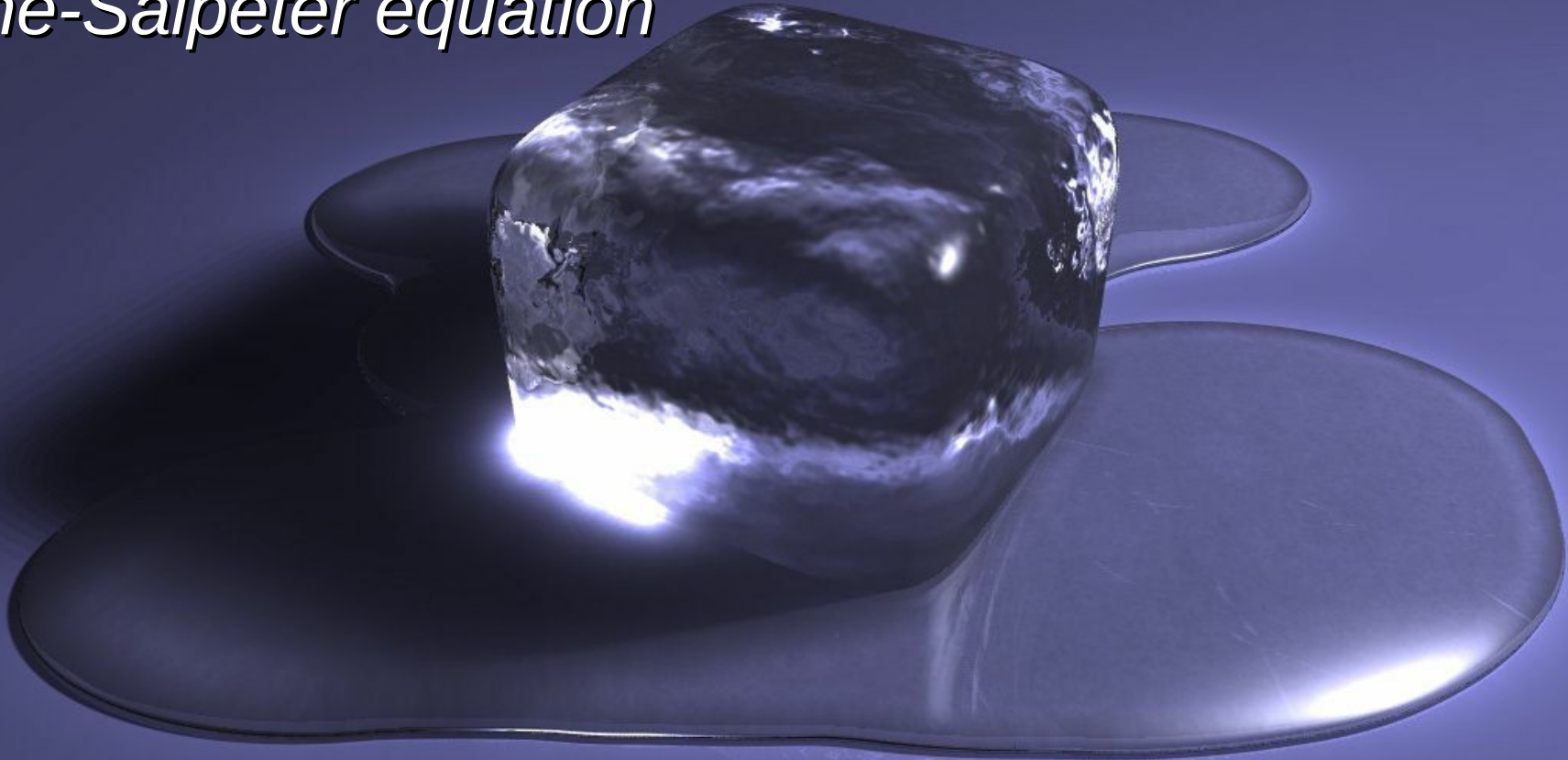


<http://www.yambo-code.org/andrea>

1+1≠2

Motivations

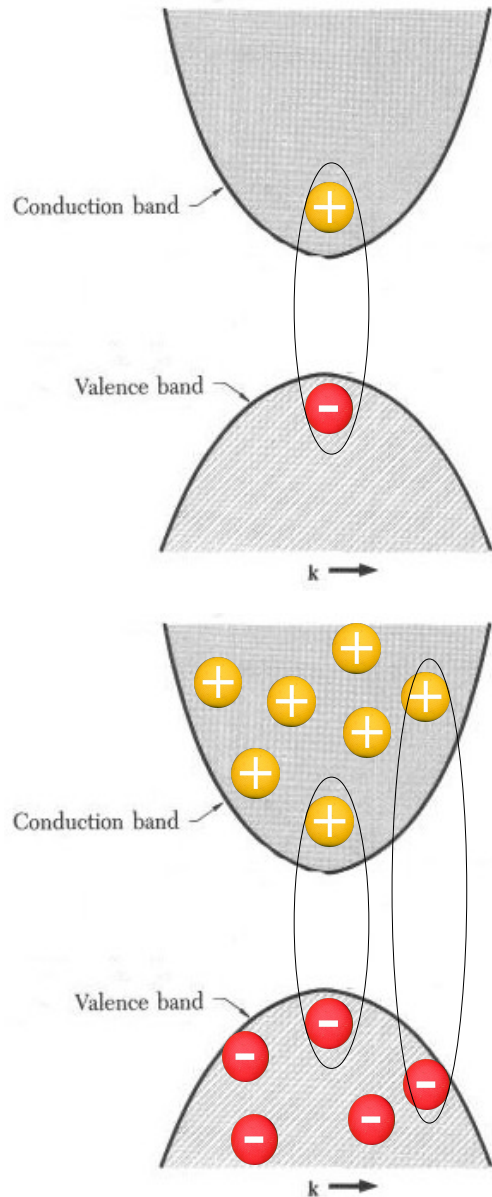
*The Time-dependent
Bethe-Salpeter equation*



Absorption in strong pulsed fields: GaAs

Exciton collapse caused by Pauli blocking: h-BN

Motivations (I): Non-linear phenomena



Linear Optics:

- Weak perturbations
- Low carriers density

Especially in the linear regime, there are no populations of electrons, holes or excitons at all—that is, the system is unexcited—and a probe beam merely tests the transition possibilities of the system. (...) Clearly, in the linear case this cannot have any relation to the possible existence of exciton populations. [S.W. Koch, Nat. Mater. 5, 523 (2006)]

$$P(t) = \chi^1 E(t) + \chi^2 E^2(t) + \dots$$

Non-linear phenomena:

- Strong perturbations (comparable to the electric field inside the atom)
- High carriers density ($\approx 10^{22} \text{ cm}^{-3}$)

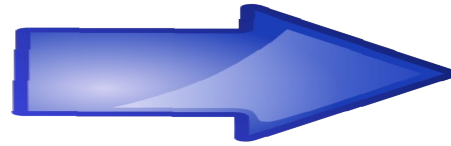
If the series does not converge we have Non Perturbative effects.

1+1≠2

Motivations (II): Non-linear phenomena

Non-linear phenomena

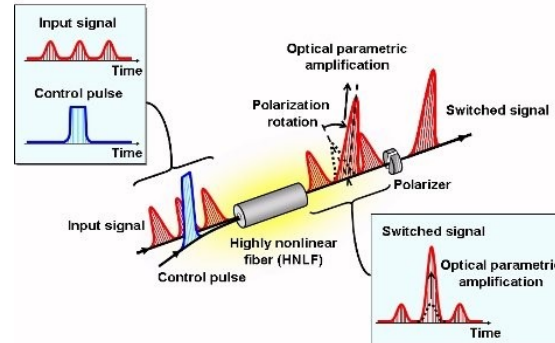
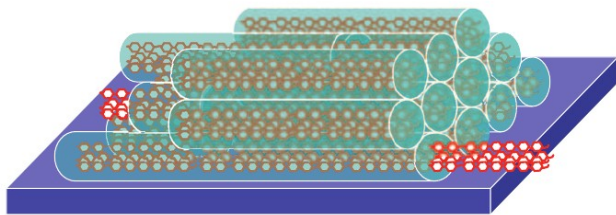
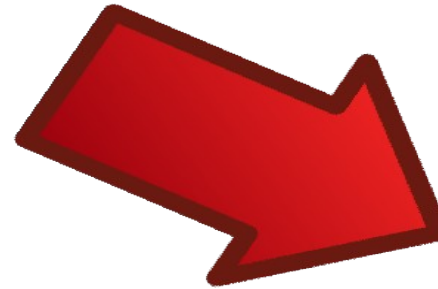
$$1 + 1 \neq 2$$



FLASH II.

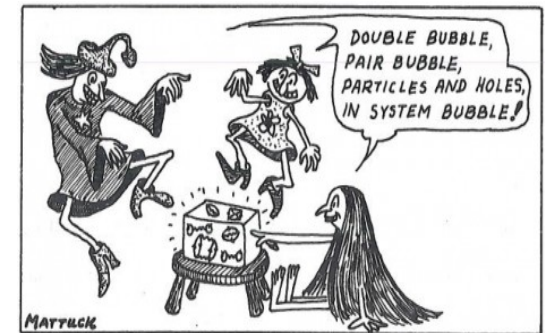
Saturation Phenomena

X-ray induced transparency in Al
(Nagler. Nature, 2009)



High-gain materials
Amplified light emission in
aligned polymers (I.B. Martini.
Nature, 2007)

Nano optical devices
Single-molecule (Hwang. Nature, 2009) and
Carbon Nanotube (Tans, Nature, 1998)
optical transistors



Severe
testing-ground for
Many-Body theories

$$1 + 1 \neq 2$$

Non-equilibrium Green's function Theory in a DFT framework

Any observable is a functional of the Green's functions ($G^<$)



Kadanoff & Baym Equations, 1962

DFT (Ab-Initio)

Perturbations

NEGF theory

$$i \frac{\partial}{\partial t} G_{nmk}^<(t) = [H_k + U_k(t), G_k^<(t)]_{nm} + I_{nmk}^<(t)$$

Kadanoff-Baym "Statistical Mechanics" (1994)

DFT+NEGF \rightarrow **AINEGF**

- ✓ Parameter free, predictive and accurate
- ✓ Valid out-of-equilibrium
- ✓ Valid beyond the linear response

$$N_c(t) = -i \sum_{nk} G_{nmk}^<(t)$$

Marker of potential non-perturbative effects

$$P(t) \propto \sum_{mnk} r_{mnk} G_{mnk}^<(t)$$

$$\chi(\omega) = \frac{P(\omega)}{E(\omega)}$$

Absorption in the Linear Response limit and Probe absorption in Pump&Probe experiments

1+1≠2

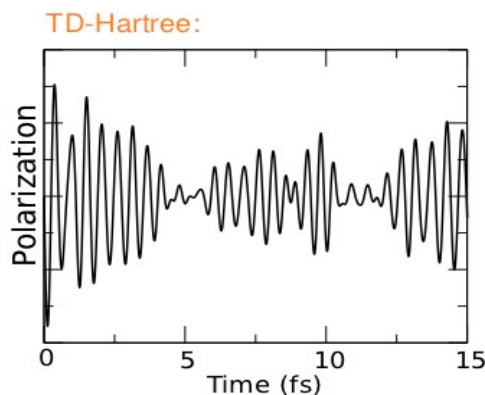
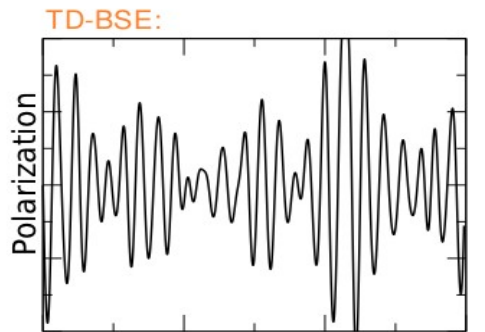
The Time-dependent Bethe-Salpeter Equation

$$i \frac{\partial}{\partial t} G_{nmk}^<(t) = [h_k^{DFT} + U_k(t) + \underline{V_k^H(t)} - \underline{V_k^H(t)} + \Sigma_k^{COHSEX}(t) - \underline{\Sigma_k^{COHSEX}(t)}, G_k^<(t)]_{nm}$$

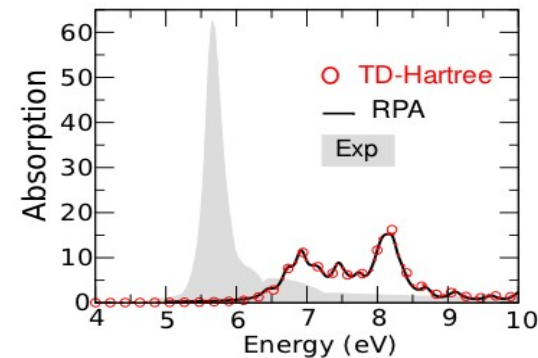
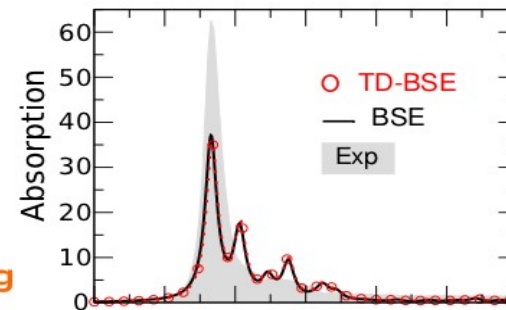
- ✓ By subtracting the unperturbed (underlined) Hartree and XC it reduces to the standard DFT+GW+BSE approach in the linear regime
- ✓ It leads to a "controllable" dynamics in the high-order Green's function.

Poster Myrta Grüning

C. Attaccalite, M. Grüning and A.M.
ArXiv:1109.2424v1



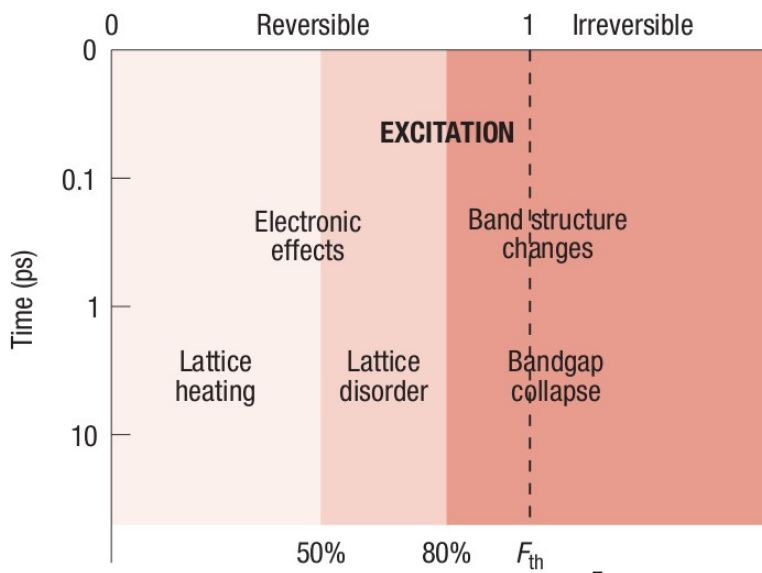
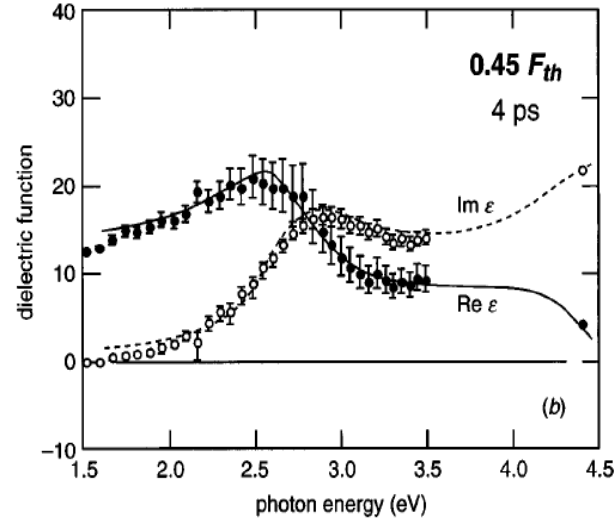
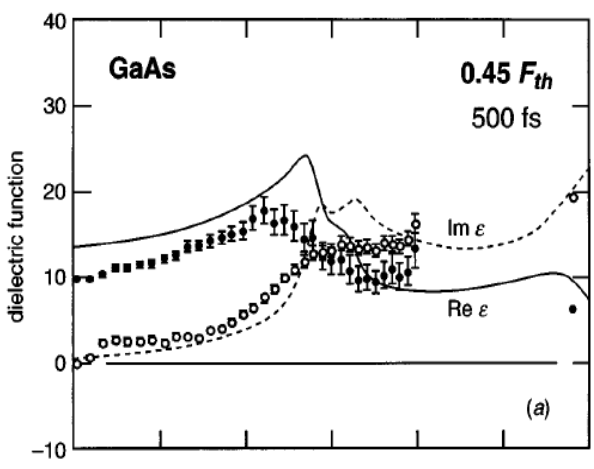
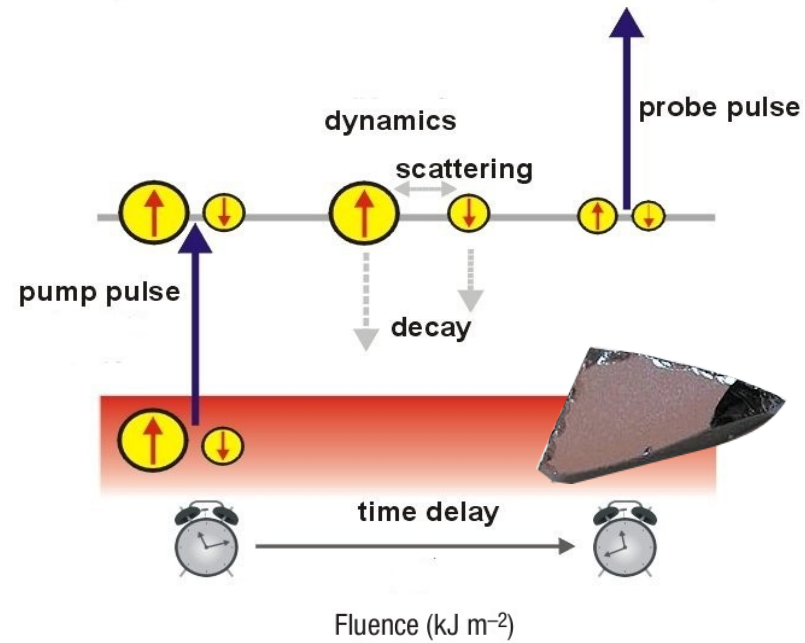
post-processing



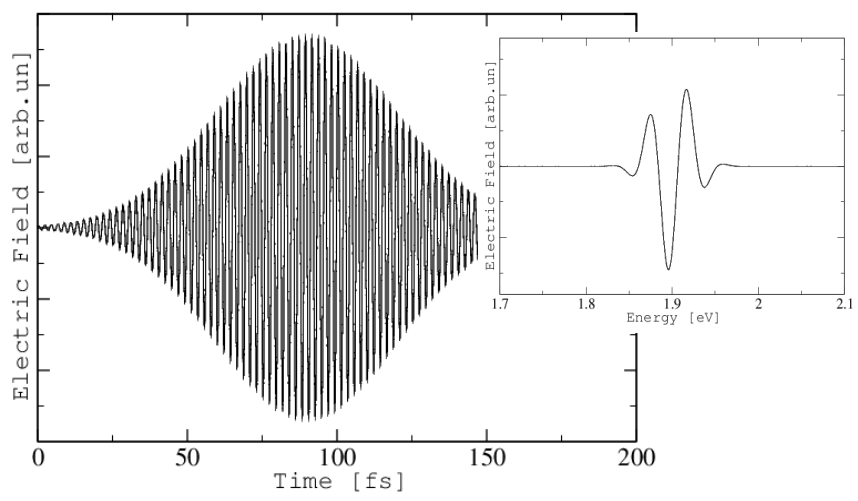
1+1≠2

Time-resolved absorption in strong pulsed fields: GaAs (I)

L. Huang, PRL 80, 185 (1997)



$$F_{th} = 1 \frac{\text{kJ}}{\text{m}^2}$$



Pump Pulse: a quasi-harmonic field 70fs long and centered at 1.9 eV (Exp. Gap is 1.52 eV)

$$F \propto \int E^2(t) dt$$

Fluence

1+1 ≠ 2

The Method [L.X. Benedict PRB 63, 075202 (2001)]

The TD-BSE

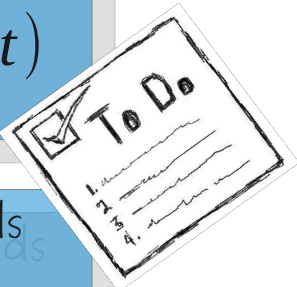
T-dependent occupations

$$f_{nk}(t) = -iG_{nk}^<(t)$$

Standard BSE (at each time)

Ansatz

$$G_{nmk}^<(t) \ll G_{nmk}^<(t)$$



Probe and Pump fields are decoupled (no sum/difference terms)

Absorption is taken in the non-thermal time window (no electron-phonon)

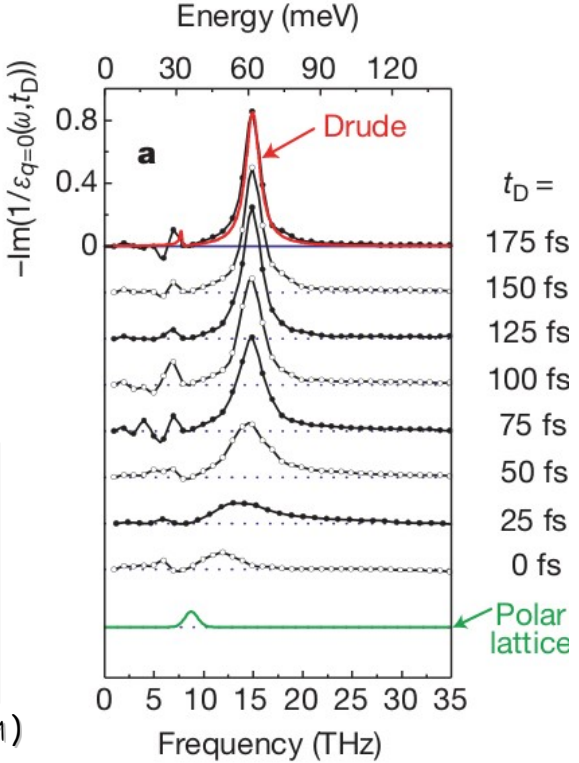
Band-gap renormalization Effects are neglected.
Band-gap collapse
 [E.N. Glazer, PRB 51, 6959]

$$H_{IJ}^{BSE}(t) = E_I \delta_{IJ} + f_I^{eh}(t)(2V_{IJ} - W_{IJ})$$

Time-resolved Probe Absorption

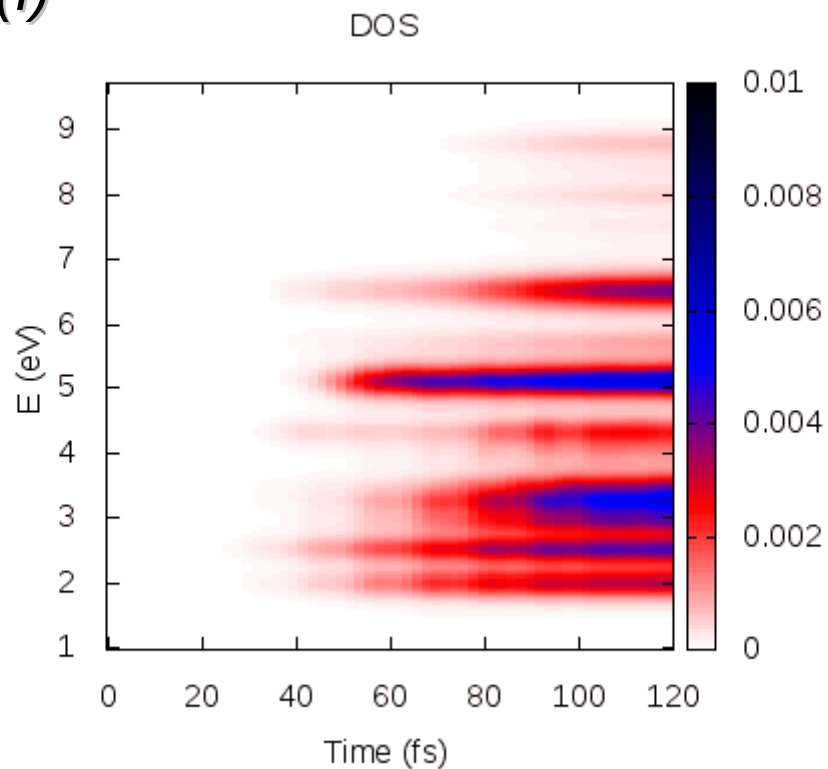
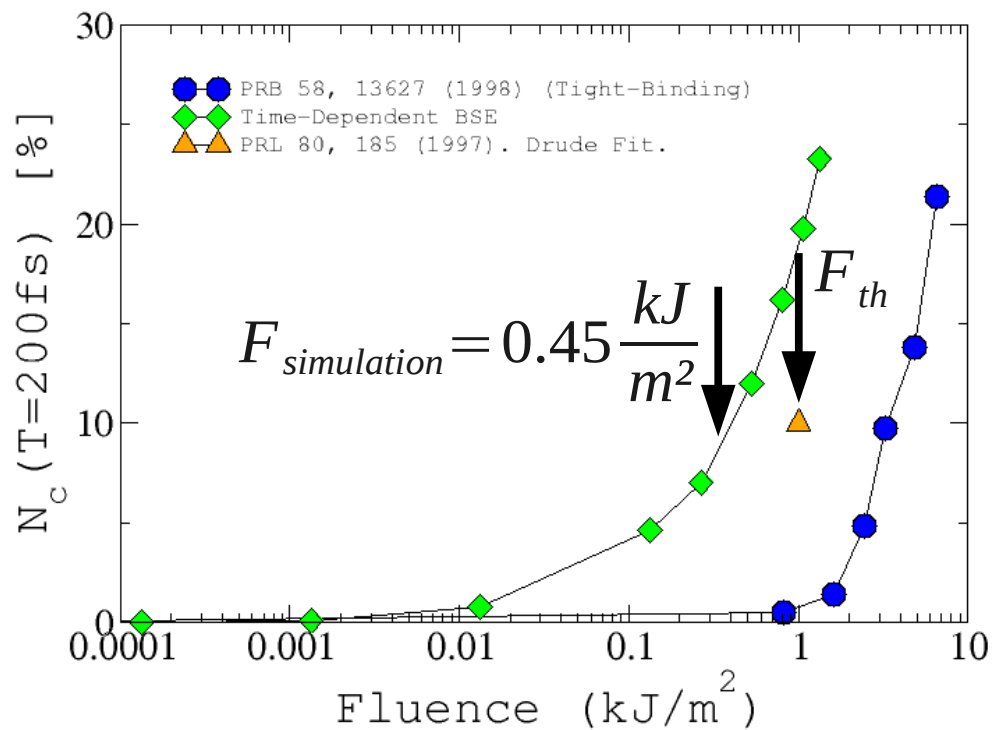
In the ultra-fast time-scale the excited electrons does not contribute to W

R. Huber, Nature 414, 286 (2001)

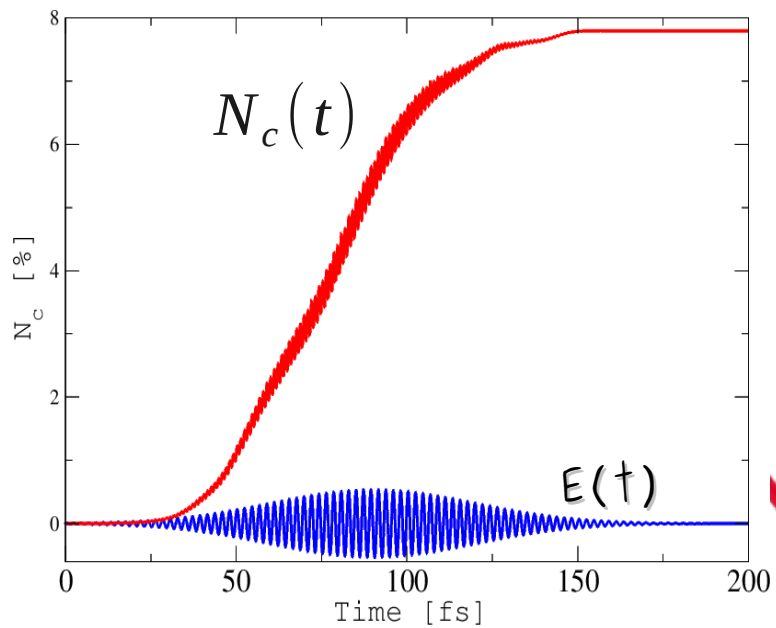


1+1≠2

Time-dependent absorption of GaAs: Results (I)

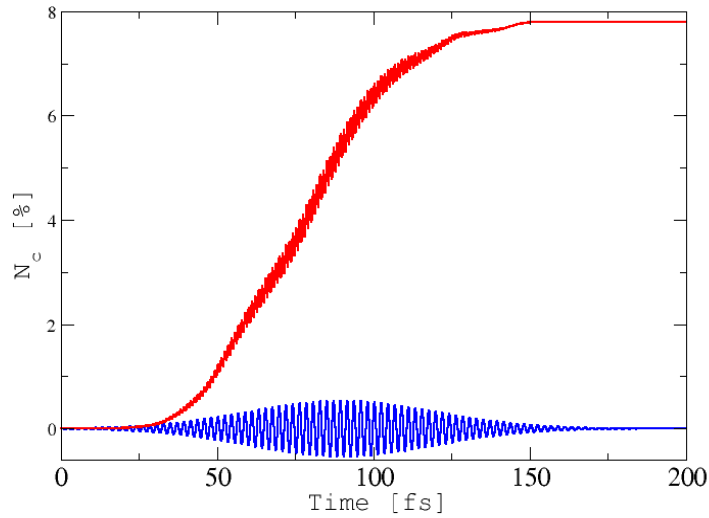


$$N_c(t) = -i \sum_{nk} G_{nk}^<(t)$$



1+1≠2

Time-dependent absorption of GaAs: Results (II)



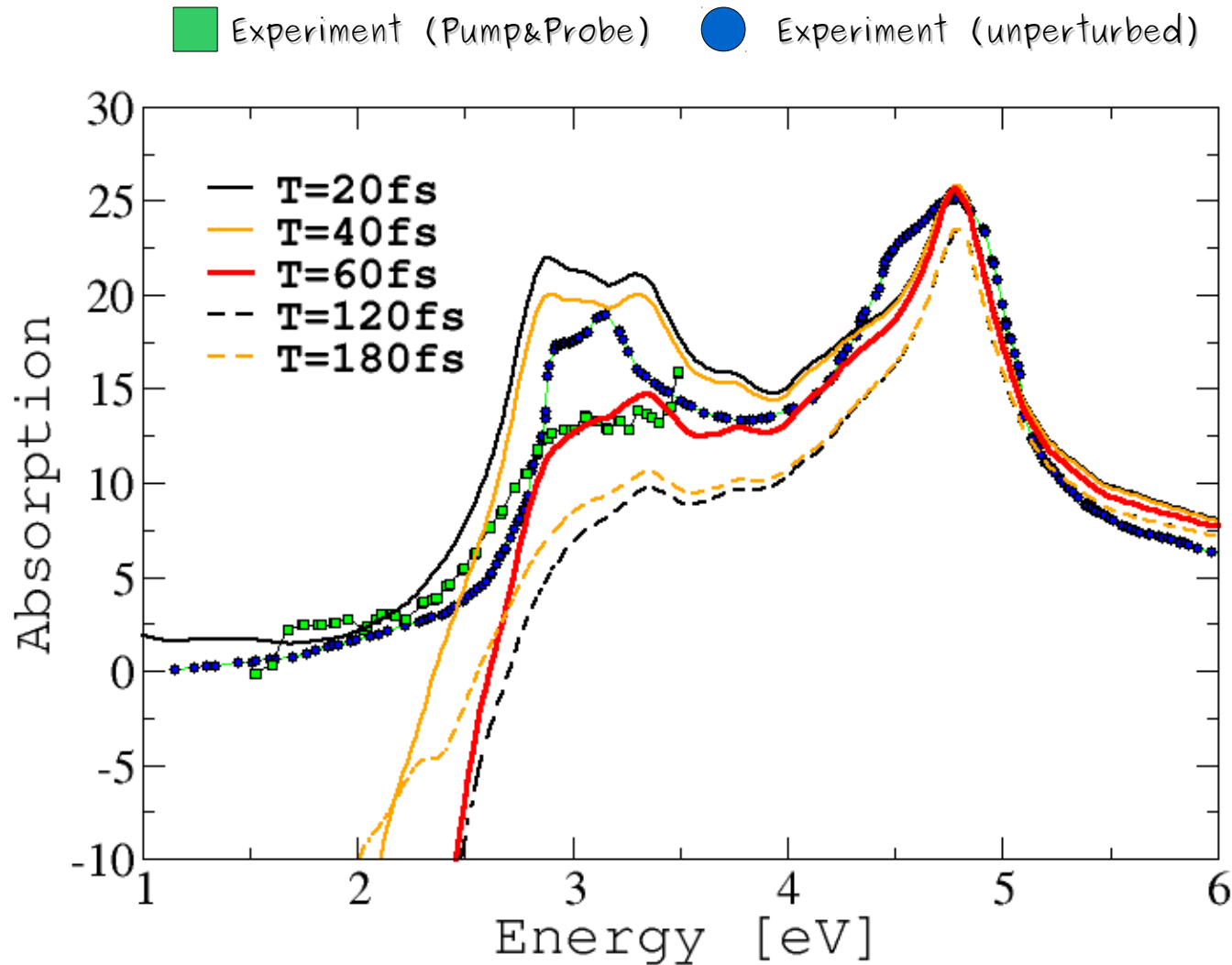
Experimental trend well reproduced



Gain region overestimated (fast electron-phonon dynamics?)



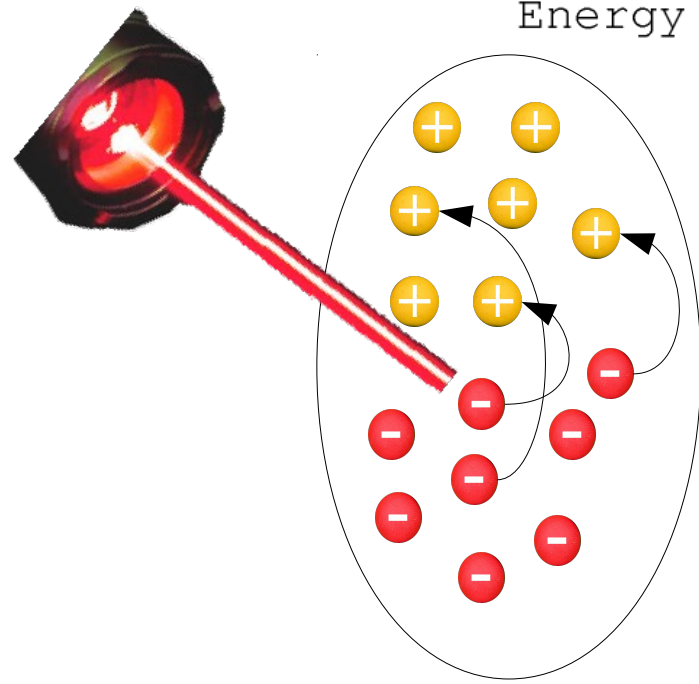
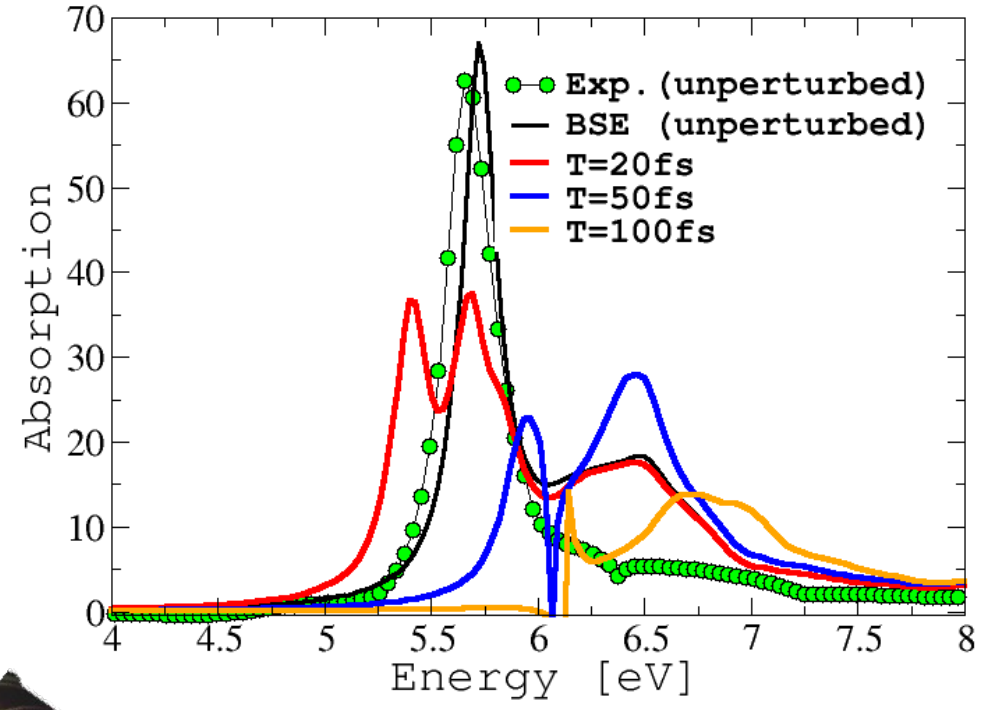
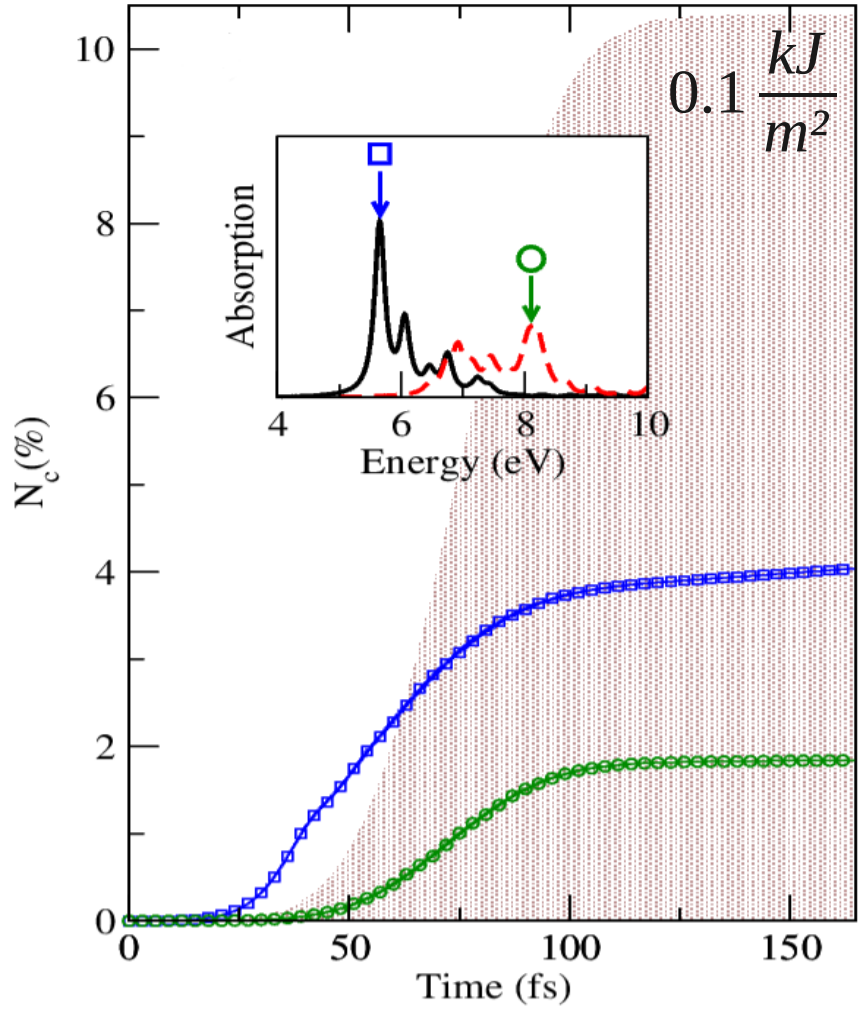
In contrast to previous studies [L.X. Benedict PRB 63, 075202 (2001)] the non-equilibrium carriers dynamics is fully Ab-Initio.



1+1≠2



Exciton collapse by Pauli blocking in hexagonal BN



The injection of carriers in the excitonic packet causes its collapse due to Pauli blocking

✓ The TD-BSE correctly enhances the carriers excitation when the pulse is resonant with the exciton

1+1≠2

Conclusions...

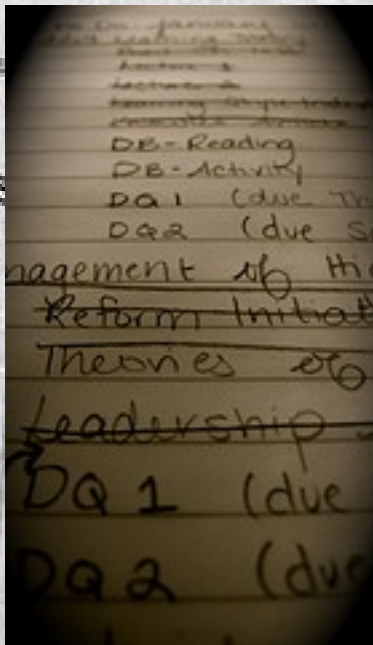
1+1≠2

Ab-Initio based out-of-the-equilibrium calculations for solids and nanostructures are now possible... with MANY possible applications...

...but...

out-of-the-equilibrium ?

TODO list





C. Attaccalite, M. Grüning and A.M.
ArXiv:1109.2424v1



Yambo: an ab initio tool for excited state calculations,
A. Marini, C. Hogan, M. Grüning, D. Varsano, *Comp. Phys. Comm.* 180, 1392 (2009).



Claudio Attaccalite



Myrta Grüning