The Abdus Salam International Centre for Theoretical Physics

2440-1

16th International Workshop on Computational Physics and Materials Science: Total Energy and Force Methods

10 - 12 January 2013

Competition between the electronic and phonon-mediated scattering channels in the out-of-equilibrium carrier dynamics of semiconductors: an ab-initio approach

Andrea Marini
ISM, CNR
Italy

Strada Costiera, 11 - 34151 - Trieste - Italy • Tel. +39 0402240111 • Fax. +39 040224163 • sci_info@ictp.it • www.ictp.it
ICTP is governed by UNESCO, IAEA, and Italy, and it is a UNESCO Category 1 Institute

Competition between the electronic and phonon-mediated scattering channels in the out-of-equilibrium carrier dynamics of semiconductors: an ab-initio approach

XVI international workshop on computational physics and materials science: total energy and force methods. 10-12 January 2013, Trieste (Italy).

Andrea Marini

Material Science Institute
National Research Council
(Monterotondo Stazione, Italy)

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

Outline

Motivations and experimental evidences

The AiNEGF approach: solving the Dyam-Kadanoff equations in a Kohn-Sham basis

Out-of-equilibrium electron-phonon scattering

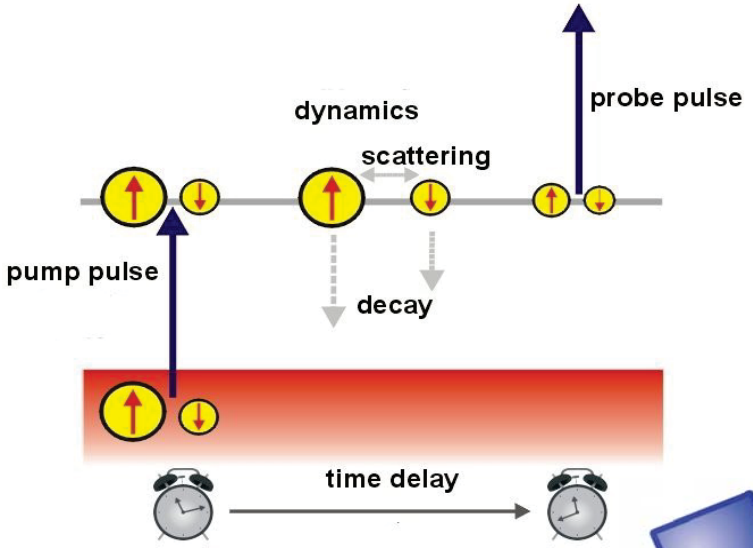
Out-of-equilibrium electron-electron scattering

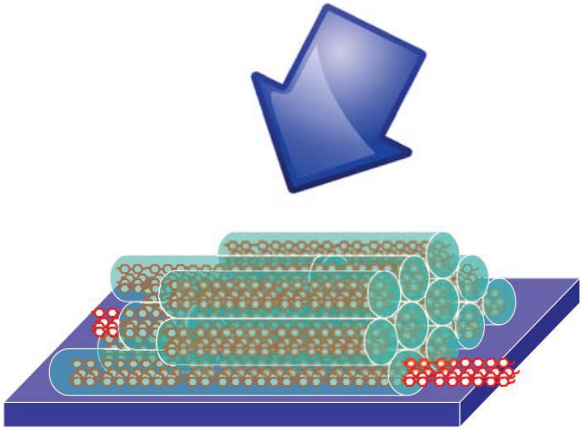
Silicon: intravally scattering of photo-generated electrons

Conclusions...

Motivations and experimental evidences

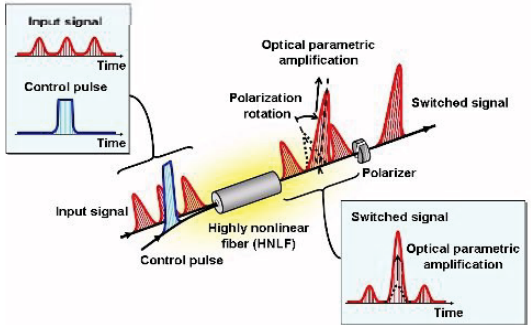
Physics on an ultra-short/ultra-strong scale





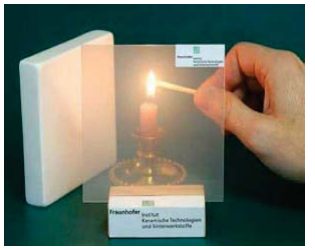
High-gain materials

Amplified light emission in aligned polymers (I.B. Martini. Nature, 2007)



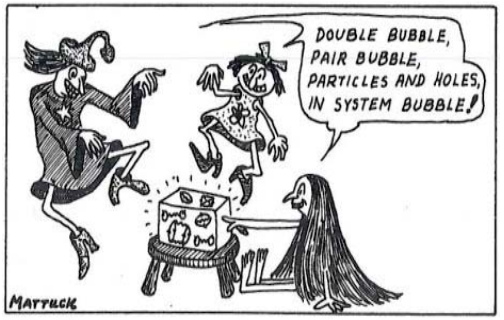
Nano optical devices

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



Saturation Phenomena

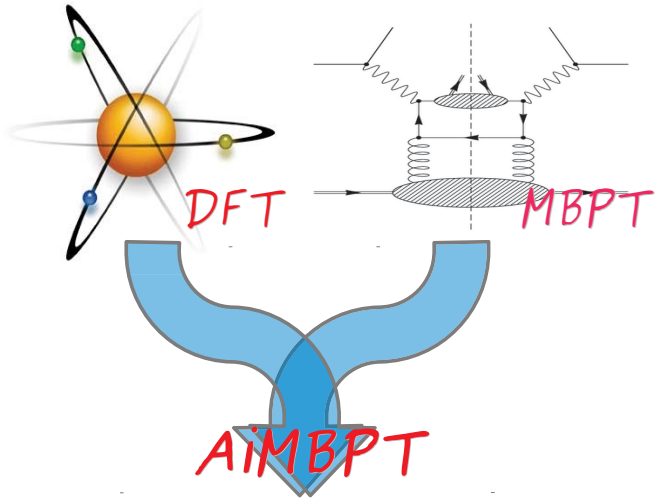
X-ray induced transparency in Al (Nagler. Nature, 2009)

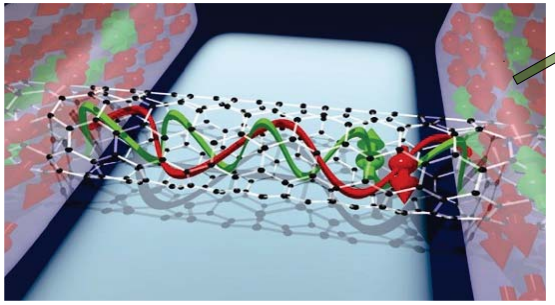


Severe testing-ground for Many-Body theories

The Ab-Initio non-equilibrium Green's function approach (AiNEGF)

The AiMBPT (Ab-Initio Many-Body Perturbation Theory)





G. Onida, Rev. Mod. Phys. 2002

AiMBPT is...

DFT

$$\left[\frac{-\nabla^2}{2}+v_s(r)\right]\psi_{nk}(r)=\epsilon_{nk}\psi_{nk}(r)$$

$$v_s(r)=v_{atoms}(r)+V_{Hxc}(r)$$

+

MBPT

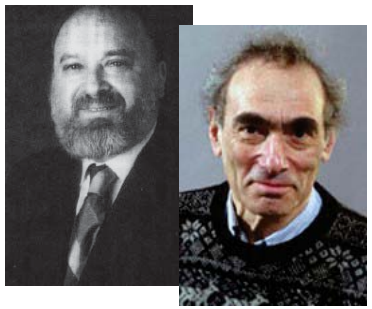
$$\Sigma = \quad + \quad$$

$$G(r_1,r_2;t)\propto\langle T\{\psi(r_1,t)\psi^+(r_2)\}\rangle$$

✓ Predictive ✓ Parameter free ✓ Universal ✓ Accurate

The Baym-Kadanoff equation in a DFT framework (I)

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



Kadanoff & Baym Equations, 1962

DFT (Ab-Initio) — Perturbations — NEGF theory

$$i\frac{\partial}{\partial t}G^<_{nmk}(t)=[H_k+U_k(t),G^<_k(t)]_{nm}+S^<_{nmk}(t)$$

Kadanoff-Baym "Statistical Mechanics" (1994)

DFT+NEGF ➡ AiNEGF

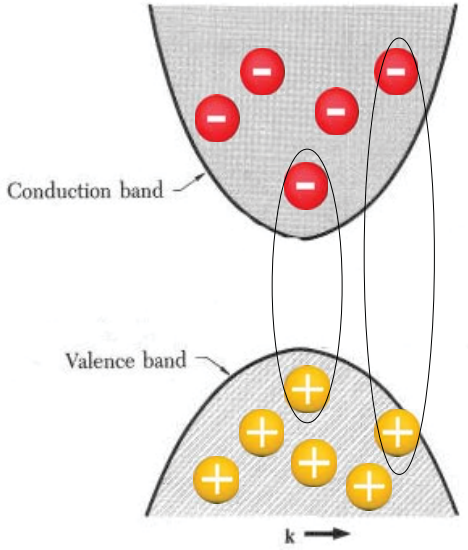
✓ Parameter free, predictive and accurate

$$P(t)\propto\sum_{mnk} r_{mnk}\,G^<_{mnk}(t)$$

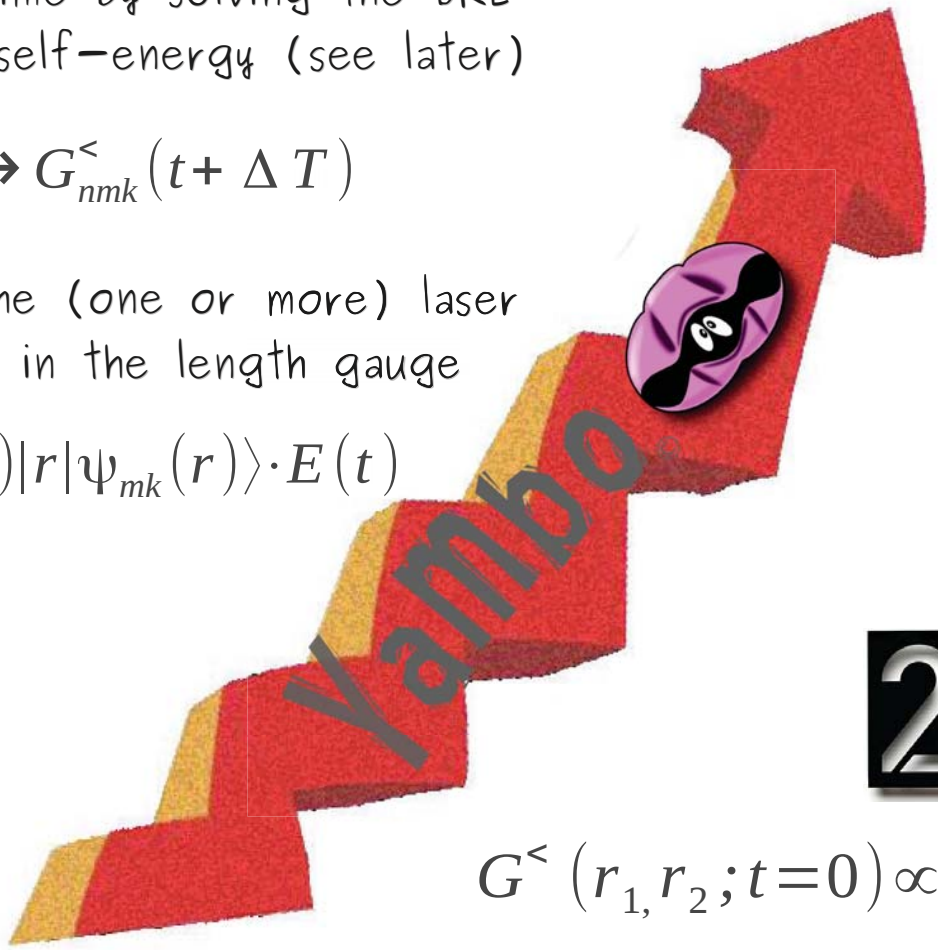
$$\chi(\omega)=\frac{P(\omega)}{E(\omega)}$$

$$N_c(t)=-i\sum_{nk\ empty} G^<_{nnk}(t)$$

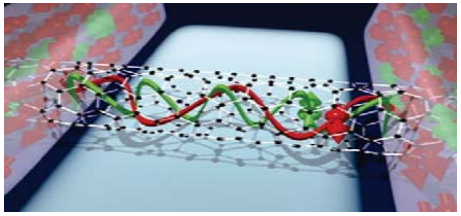
Marker of potential non-perturbative effects



The Baym-Kadanoff equation in a DFT framework (II)



1



2

$$G^{<}(r_1, r_2; t=0) \propto \langle \psi(r_1, t)\psi^{+}(r_2)\rangle$$

Boundary conditions and lesser and retarded Green's functions are written in the KS basis

3

The lesser Green's function is evolved in time by solving the BKE for a given self-energy (see later)

$$G^{<}_{nmk}(t) \rightarrow G^{<}_{nmk}(t+\Delta T)$$

The interaction with the (one or more) laser fields is introduced in the length gauge

$$U_k(t) \propto \sum_{nm} \langle \psi_{nk}(r)|r|\psi_{mk}(r)\rangle \cdot E(t)$$

4

Post-Processing

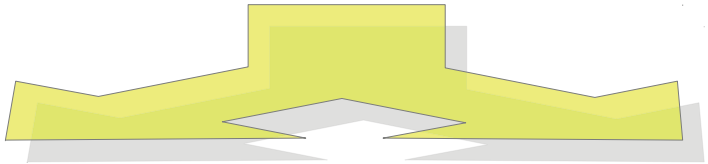
$$G^{<}_{nmk}(t)$$

$$\chi^n(\omega) \quad f_{nk}(t)$$

The out-of-equilibrium kernel

$$i\frac{\partial}{\partial t}G^{<}_{nmk}(t)=[H_k+U_k(t),G^{<}_k(t)]_{nm}+S^{<}_{nmk}(t)$$

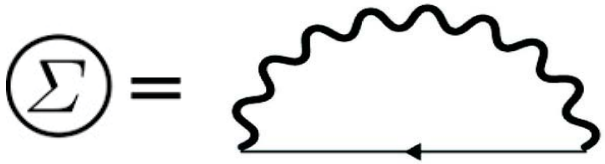
$$S(t)=\int_{-\infty}^{t}d\tau\left[\Sigma^{>}(t,\tau)G^{<}(t,\tau)+G^{<}(t,\tau)\Sigma^{>}(t,\tau)-\Sigma^{<}(t,\tau)G^{>}(t,\tau)-G^{>}(t,\tau)\Sigma^{<}(t,\tau)\right]$$



Phonons

Electron–Phonon scattering

E–h pairs



Electron–Electron scattering

+ Massive approximations

+ number-crunching techniques

The NEGF kernel: Electron-phonon and electron-electron scatterings from an Ab-Initio presepective

The electron–phonon out-of-equilibrium kernel (I)

arXiv:1211.0147

$$i\frac{\partial}{\partial t}G^<(t,t)=[h^{DFT}+\Sigma_s(t),G^<(t,t)]+S(t)$$

$$S(t)=\int_{-\infty}^{t}d\tau\left[\Sigma^>(t,\tau)G^<(t,\tau)+G^<(t,\tau)\Sigma^>(t,\tau)-\Sigma^<(t,\tau)G^>(t,\tau)-G^>(t,\tau)\Sigma^<(t,\tau)\right]$$

Kadanoff–Baym "Statistical Mechanics" (1994)

MBPT DFPT →

$$\Sigma^<_{nn'k}(t,\tau)=\frac{i}{N_q}\sum_{mm'q}\overline{\left(g^{q\lambda}_{nmk}\right)}\,g^{q\lambda}_{n'm'k}D^<_{q\lambda}(t,\tau)G^<_{mm'k-q}(t,\tau)$$ +

GBKA →

$$G^<(t,\tau)\approx i\left[G^r(t-\tau)G^<(\tau)-G^<(t)G^a(t-\tau)\right]$$ +

Quasi-equilibrium & Complete Collisions Approx →

$$G^<_{nmk}(T)\approx\delta_{nm}f_{nk}(T)$$ +

G^r is polaronic (no short-time correction) →

$$G^r_{nk}(T)=-ie^{-i\varepsilon_{nk}(\beta)t-\Gamma_{nk}(\beta)t}\theta(t)$$ =

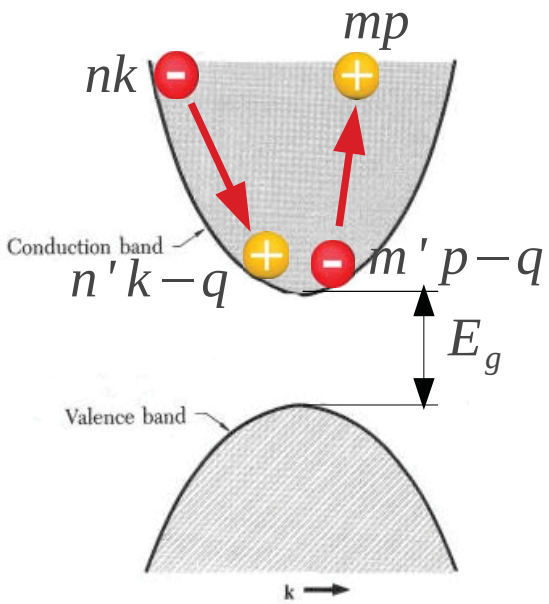
HOLE Lifetime → ELECTRON Lifetime → THERMAL Lifetime

$$\partial_t f_{nk}(T)=\gamma^{(h,e-p)}_{nk}(T)(1-f_{nk}(T))-\gamma^{(e,e-p)}_{nk}(T)f_{nk}(T)-\gamma^{(th)}_{nk}(\beta,T)$$

Electron–Electron scattering in the GW approximation

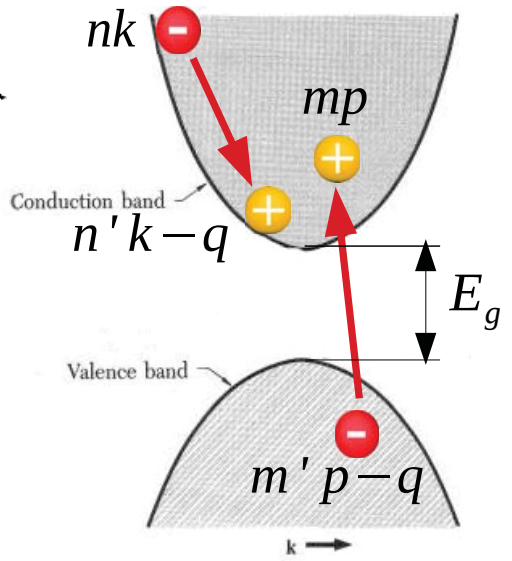
$$\partial_t f_{nk}(T) = \left(\gamma_{nk}^{(h,e-p)}(T) + \gamma_{nk}^{(h,e-e)}\right)\left(1 - f_{nk}(T)\right) - \gamma_{nk}^{(e,e-p)} + \gamma_{nk}^{(e,e-e)}(T) f_{nk}(T) - \gamma_{nk}^{(th)}(\beta,T)$$

$$\gamma_{nk}^{(e,e-e)}(T) \propto \sum_{qp} \sum_{n'mm'} \left|W^{q}_{nn'k,mm'p}\right|^2 \left[\frac{4\Gamma}{\left(\epsilon_{n'k-q} + \epsilon_{mp} - \epsilon_{m'p-q} - \epsilon_{nk}\right)^2 + 16\Gamma^2}\right] \left(1 - f_{n'k-q}(T)\right)\left(1 - f_{mp}(T)\right) f_{m'p-q}(T)$$



Intraband scattering. It takes contributions ONLY from the photo-excited electrons. Its strength goes with the carrier density.

In the e–p case this process is ALWAYS non zero as the Debye energy is much smaller than the gap



Dominant process in the low carriers density. It takes contributions also from the unperturbed electrons. But it is zero whenever

$$\epsilon_{nk} \leq 2E_g$$

The electron-phonon out-of-equilibrium kernel (II)

HOLE Lifetime

ELECTRON Lifetime

THERMAL Lifetime

$$\partial_t f_{nk}(T) = \gamma^{(h,e-p)}_{nk}(T)\left(1-f_{nk}(T)\right) - \gamma^{(e,e-p)}_{nk}(T) f_{nk}(T) - \gamma^{(th)}_{nk}(\beta,T)$$

$$P_{ph-emit} \equiv \frac{\Gamma_{nk}+\Gamma_{mk-q}}{\left(\Gamma_{nk}+\Gamma_{mk-q}\right)^2+\left(\varepsilon_{mk-q}-\varepsilon_{nk}+\omega_{q\lambda}\right)^2} \qquad P_{ph-ab} \equiv \frac{\Gamma_{nk}+\Gamma_{mk-q}}{\left(\Gamma_{nk}+\Gamma_{mk-q}\right)^2+\left(\varepsilon_{mk-q}-\varepsilon_{nk}-\omega_{q\lambda}\right)^2}$$

$$\gamma^{(h)}(T) = \frac{\sum_{q\lambda m}\left|g^{q\lambda}_{nmk}\right|^2}{N_q} P_{ph-ab} f_{mk-q}(T)$$

$(mk-q)$

(nk)

$$\gamma^{(e)}(T) = \frac{\sum_{q\lambda m}\left|g^{q\lambda}_{nmk}\right|^2}{N_q} P_{ph-emit}\left(1-f_{mk-q}(T)\right)$$

(nk)

$(mk-q)$

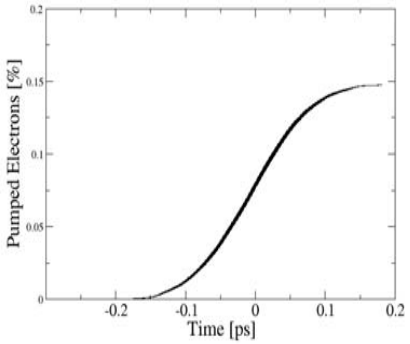
The thermal lifetime includes both kinds of scatterings weighted by the phonon occupation. It is vanishing at zero temperature and it works to create a quasi-equilibrium finite temperature state

Ultrafast Carrier Relaxation in Si: Intravalley Scattering and Energy Relaxation of photoexcited Electrons

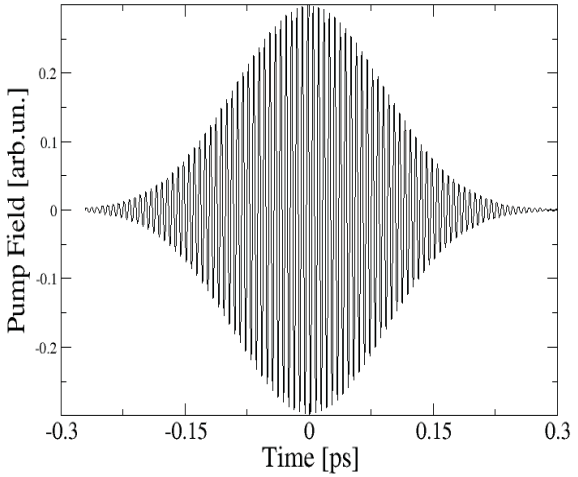
Intra-valley scattering in Bulk Silicon (I) [A. Marini, in preparation]

$$\partial_t f_{nk}(T) = \gamma_{nk}^{(h,e-p)}(T)(1-f_{nk}(T)) - \gamma_{nk}^{(e,e-p)}(T) f_{nk}(T) - \gamma_{nk}^{(th)}(\beta,T)$$

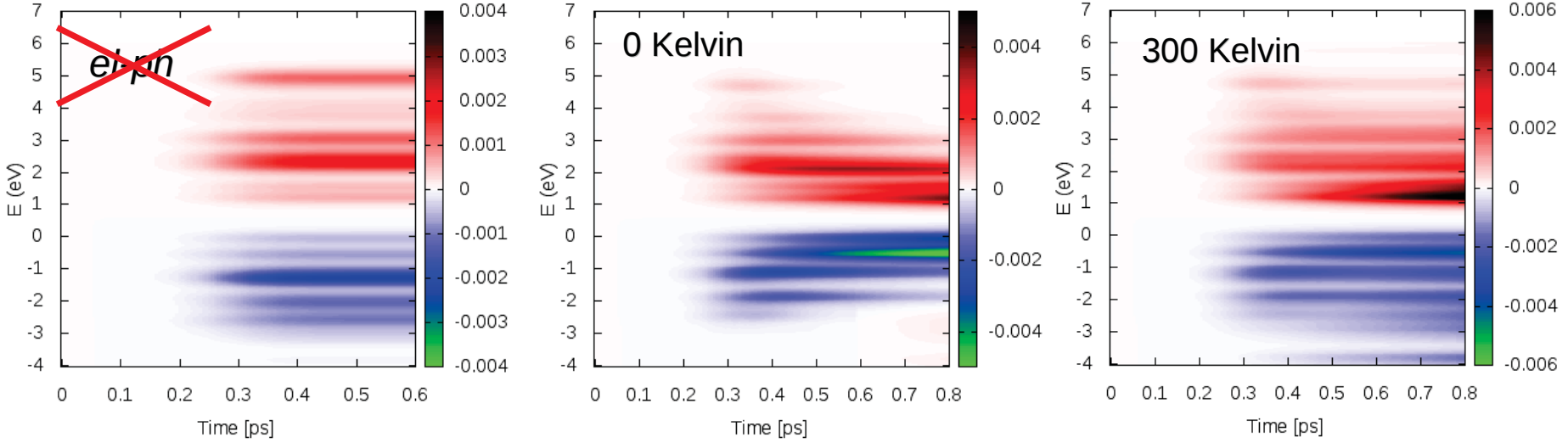
Pump field is "quasi harmonic"
with frequency 2.03 eV and fluence 0.004 $\frac{J}{cm^2}$



Pumped electrons

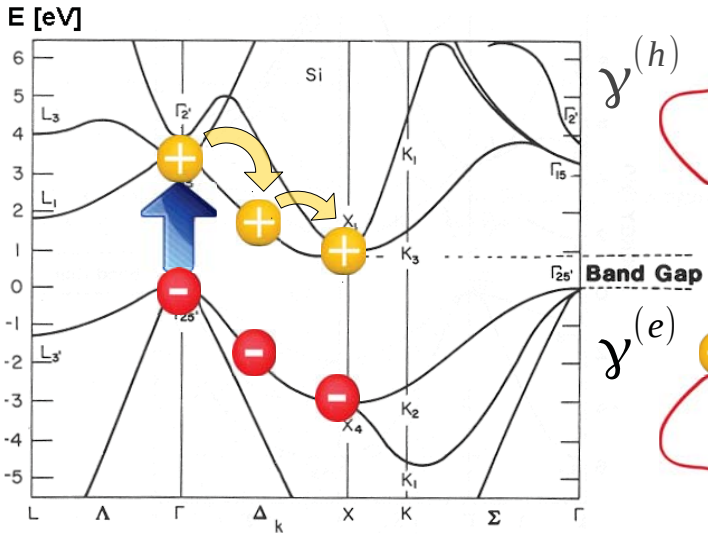


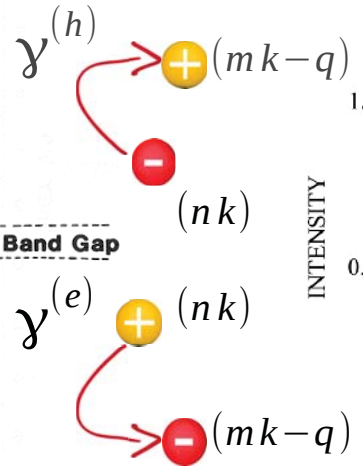
Time-dependent Density-of-states

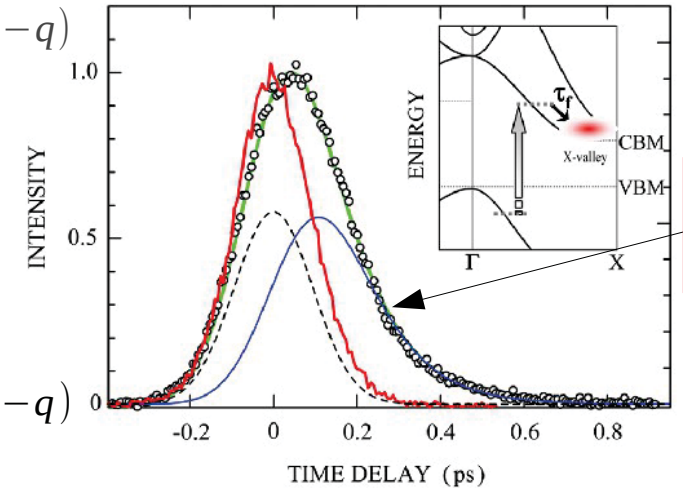


Intra-valley scattering in Bulk Silicon (II)

$$\partial_t f_{nk}(T) = \gamma_{nk}^{(h,e-p)}(T)\left(1-f_{nk}(T)\right) - \gamma_{nk}^{(e,e-p)}(T) f_{nk}(T) - \gamma_{nk}^{(th)}(\beta,T)$$



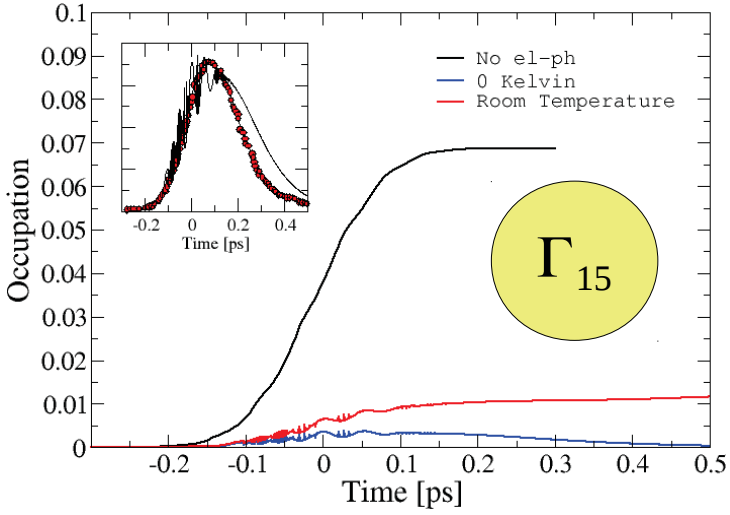


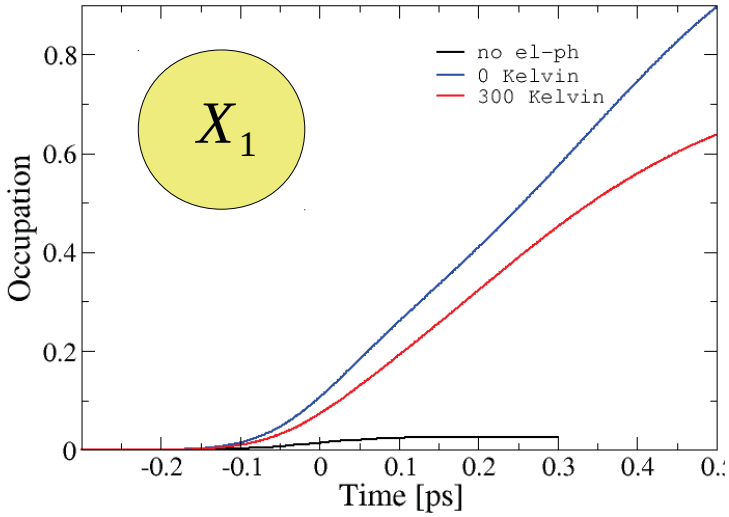


$$\tau_f^{exp} = 40 \pm 10\, fs$$

$$\tau_f^{MBPT} = 18\, fs$$

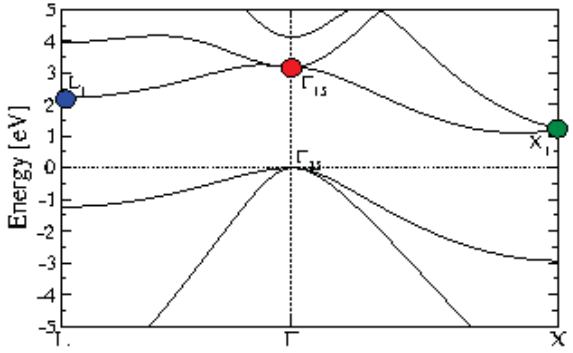
Relaxation is a **CASCADE** process. Standard MBPT does not apply!

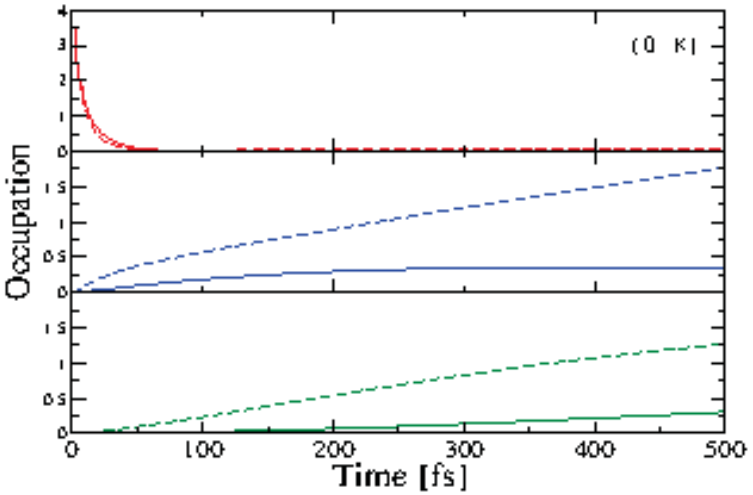


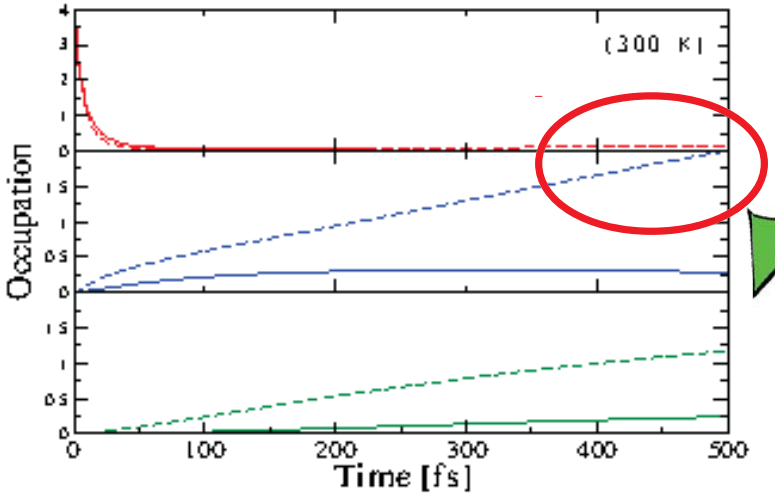


Intra-valley scattering in Bulk Silicon (III)

$$\partial_t f_{nk}(T) = \left(\gamma_{nk}^{(h,e-p)}(T) + \gamma_{nk}^{(h,e-e)}\right)\left(1 - f_{nk}(T)\right) - \gamma_{nk}^{(e,e-p)} + \gamma_{nk}^{(e,e-e)}(T) f_{nk}(T) - \gamma_{nk}^{(th)}(\beta, T)$$

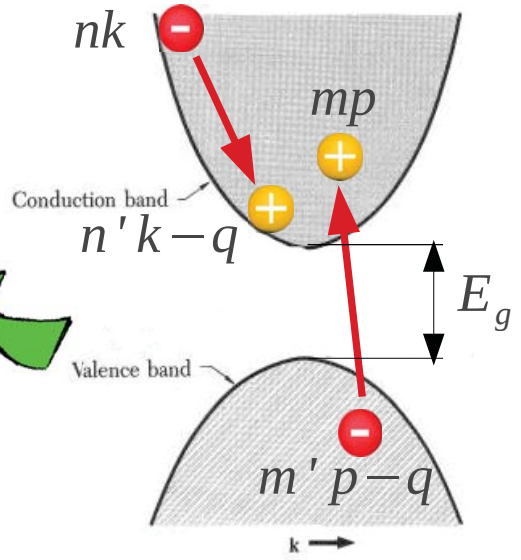






e−p

e−e+e−p



Conclusions...

The AiNEGF represents a new approach that can, potentially, extend the power of DFT-based methods in the new fields of ultra-fast phenomena (even sub-femtosecond)

Real-time approach to the optical properties of solids and nanostructures: Time-dependent Bethe-Salpeter equation
C. Attaccalite, M. Gruning, A. Marini
Phys. Rev. B 84, 245110 (2011).

Competition between the electronic and phonon-mediated scattering channels in the out-of-equilibrium carrier dynamics of semiconductors: an ab-initio approach, A. Marini, arXiv:1211.0147.

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

FLASHit 2013-2016

Unraveling ultra-fast photo-induced phenomena at the nanoscale: a joint theoretical and experimental approach

FIRB Fondo per gli Investimenti della Ricerca di Base

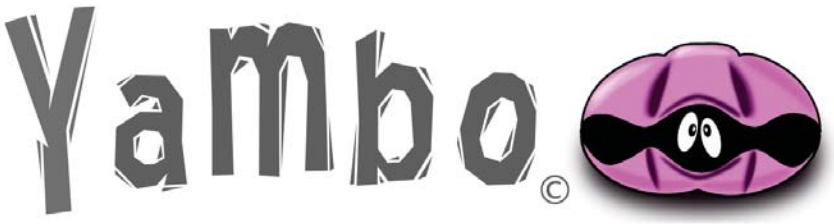
Andrea Marini. Istituto di Struttura della Materia (ISM) Consiglio Nazionale delle Ricerche (CNR). Monterotondo Stazione, Italy.

Cristian Manzoni. Istituto di fotonica e nanotecnologie (IFN) Consiglio Nazionale delle Ricerche (CNR). Politecnico di Milano, Italy.

Deborah Prezzi. Istituto di nanoscienze (NANO) Consiglio Nazionale delle Ricerche (CNR). Modena, Italy.

Gianluca Stefanucci Physics Department, University of Rome *Tor Vergata*, Italy.

http://www.yambo-code.org/flash-it



Yambo hands-on tutorial on electronic and optical excitations: from basic to advanced applications

April 8-12, 2013, CECAM headquarters in Lausanne (Switzerland)
http://www.cecam.org/workshop-0-870.html
http://www.yambo-code.org/Yambo2013/index.php

This 5-day hands-on tutorial will give participants the chance to learn both state-of-the-art approaches (GW, TD-DFT and BSE) as well as cutting edge topics and applications (electron-phonon coupling, magneto-optical effects and surface spectroscopies) as implemented in Yambo, a powerful, open-source ab-initio code interfaced with several DFT packages (including Abinit and quantum-ESPRESSO).

Lectures on the foundations of the theoretical methods will be complemented by technical ones on numerical and computational aspects. A significant part of the school will be dedicated to hands-on tutorials, where participants will be given the opportunity to carry out excited state calculations on several paradigmatic systems under the guidance of the Yambo code developers themselves.

Deadline for application is on March 1, 2013.
The number of participants will be limited to approx. 30.
The organization will partially cover the expenses of the participants.
Acceptance decisions will be made within 1 week after the deadline.