# NEGF@Work: pros and cons of the solution of the BKE based on the GKBA


## Andrea Marini



Solving the Two-time Kadanoff-Baym Equations. Status and Open Problems

Physics Department, March 12, 2019

Kiel, Germany


FLASH it

Istituto di Struttura della Materia


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

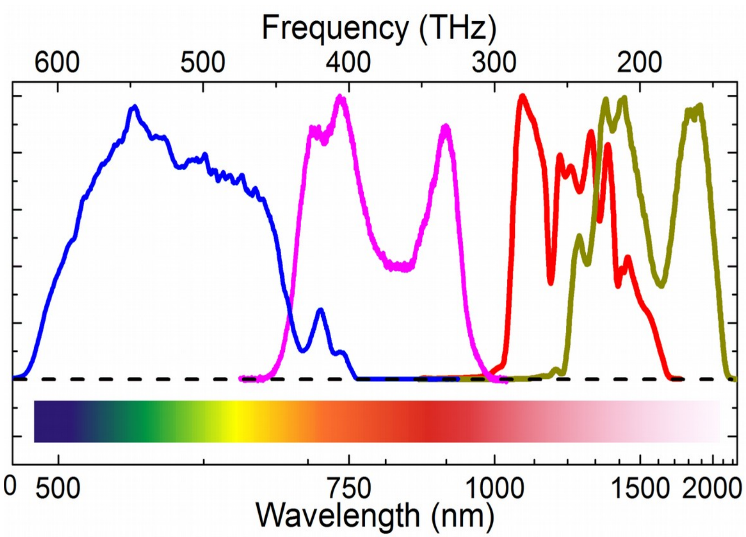
http://www.yambo-code.org/andrea

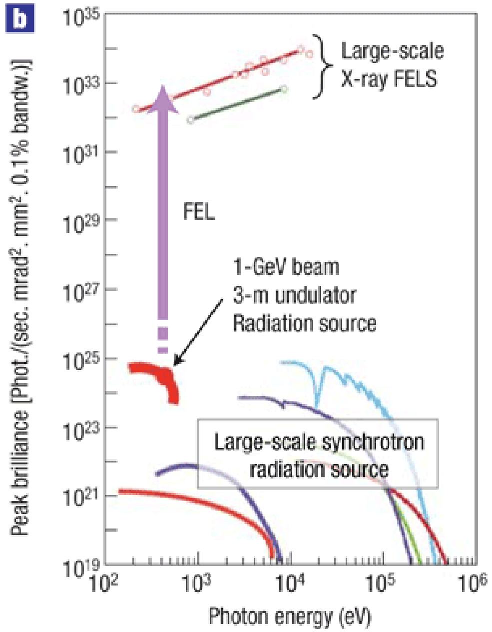
# Outline

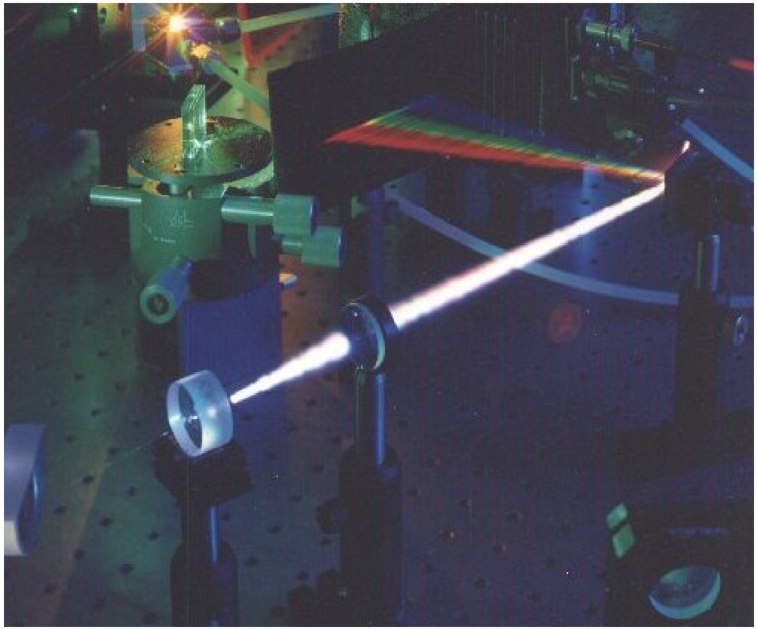
- The **Users** Perspective
- An approach based on **Large Scale** Computational Physics
- “Dream” NEGF and GKBA+approximations
- Ab–Initio NEGF (AiNEGF) at work. Pros.
- Open issues (=cons)

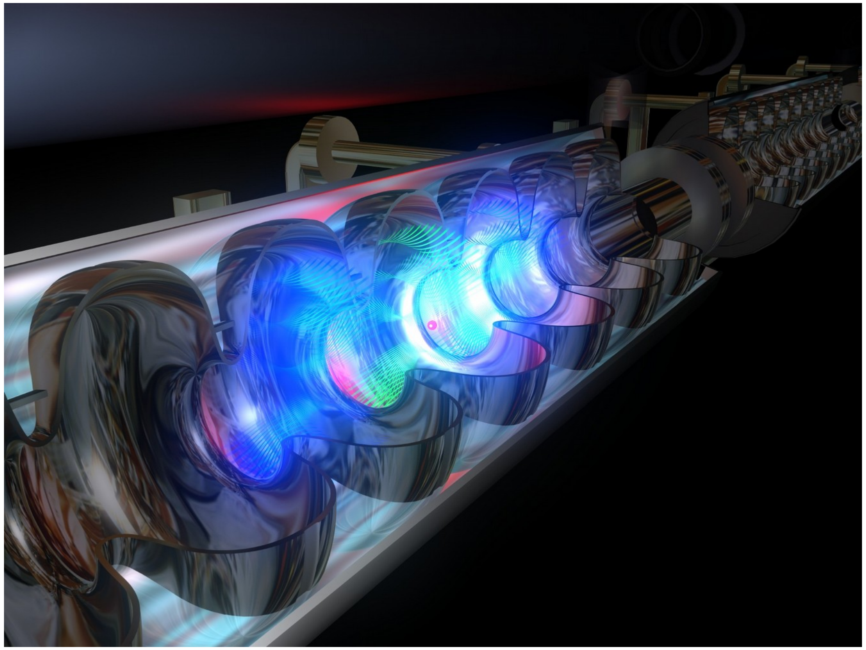

The Users Perspective

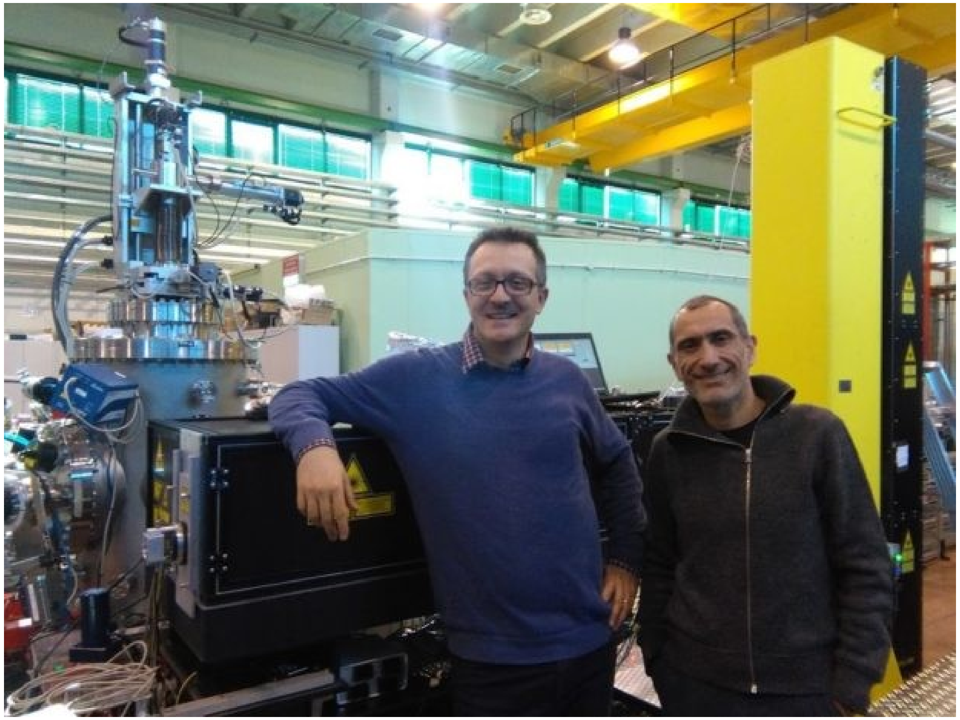
# Light sources



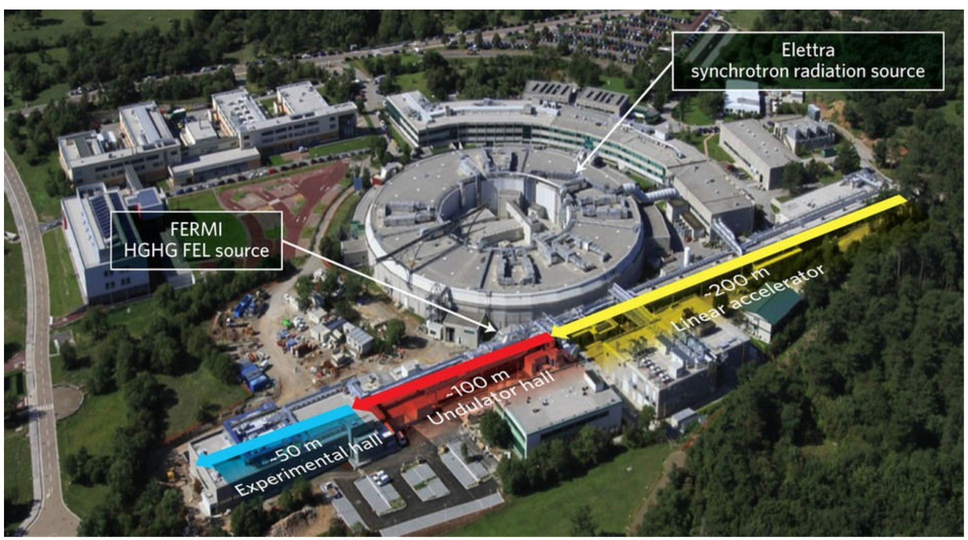





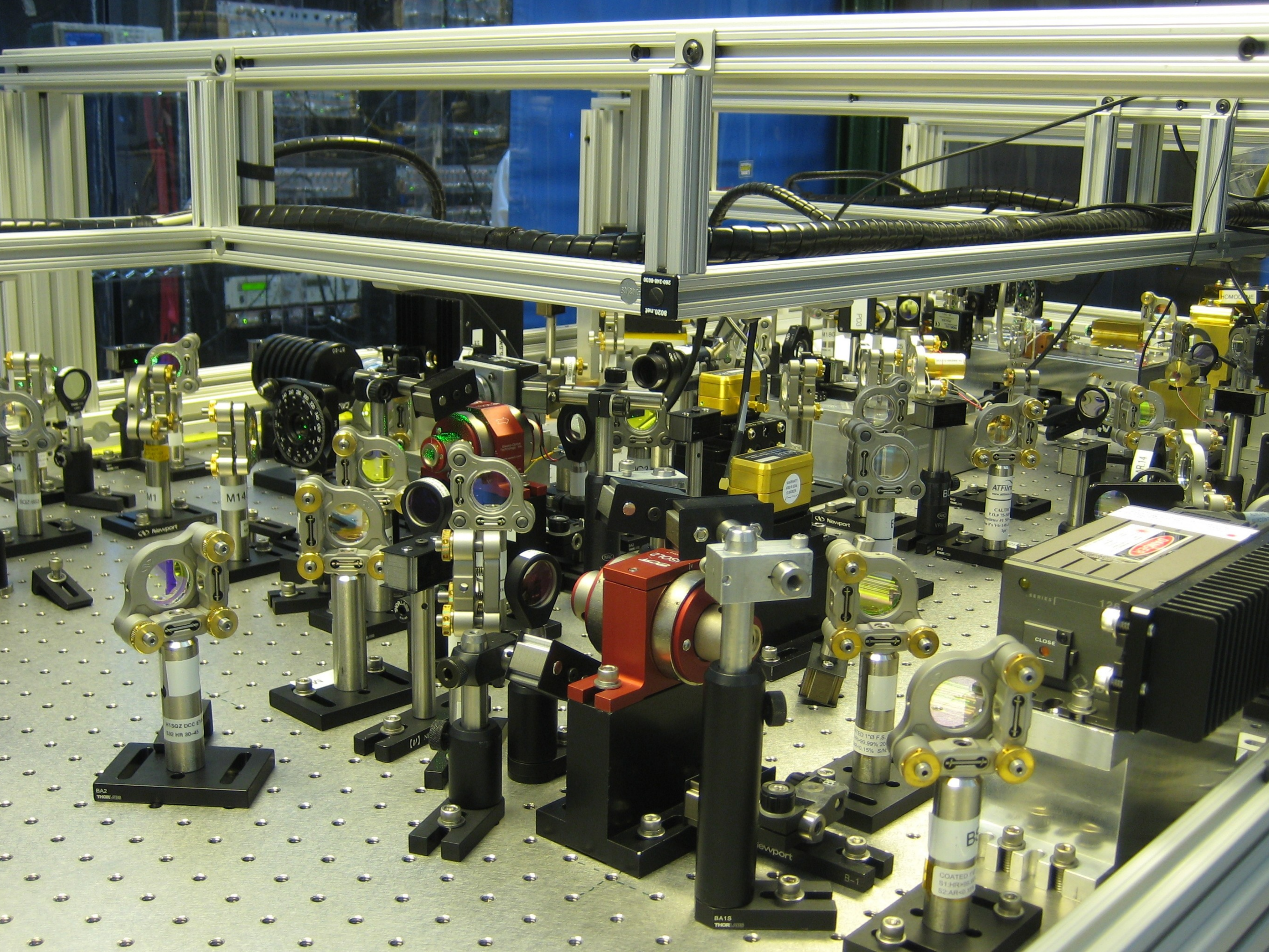


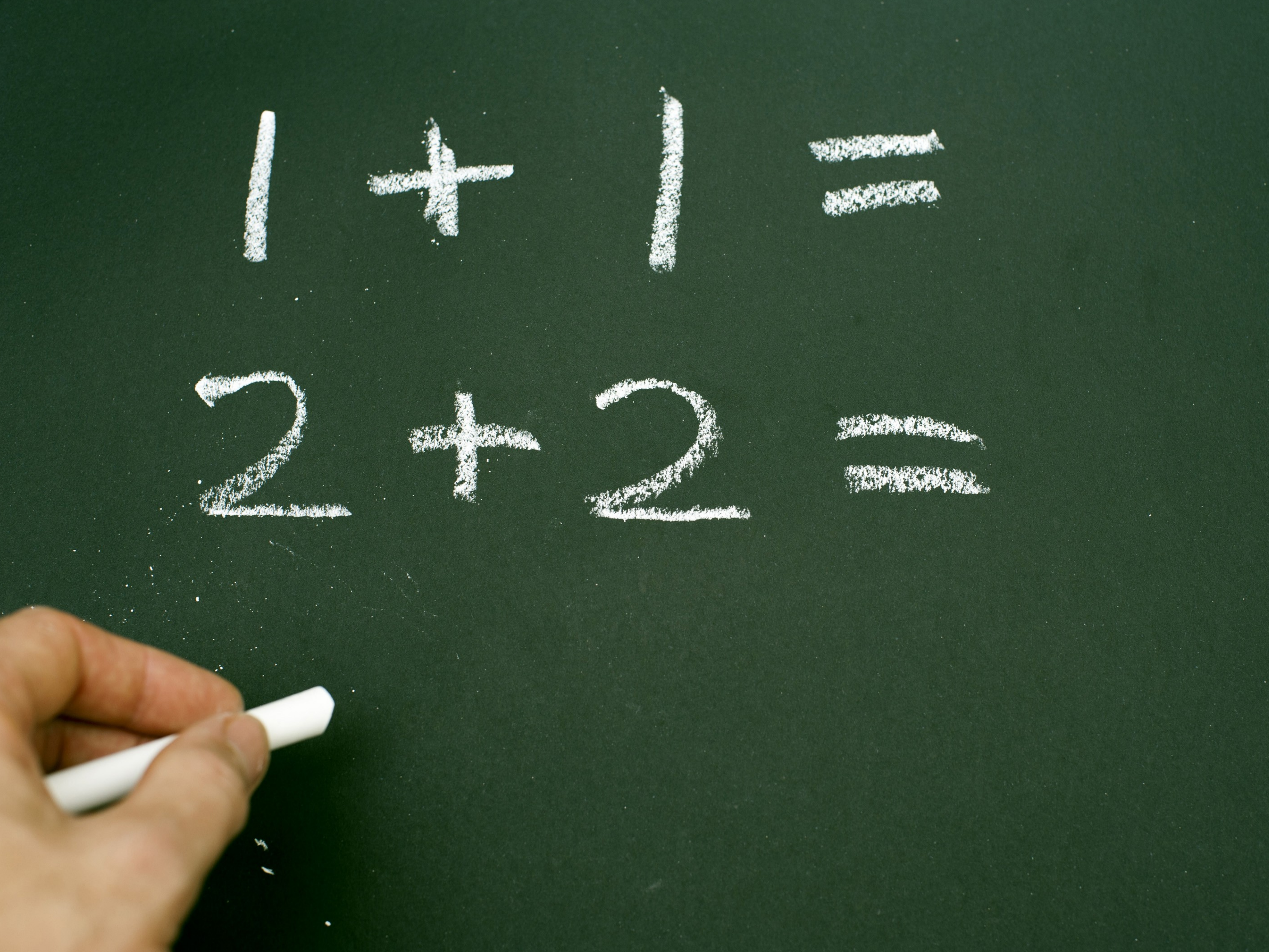
Yambo©

**EIS-YAMBO round table**
**24-25 January 2017**
**AREA Science Park, 34149 Basovizza, Trieste**

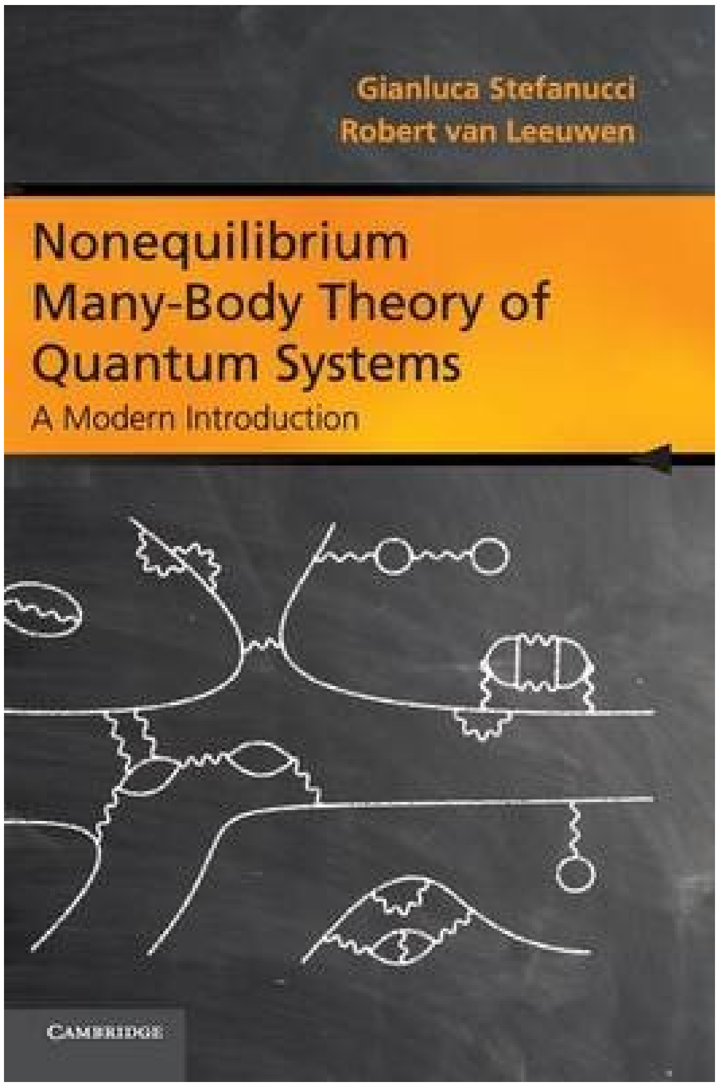



From an informal survey it turned out that three Fermi beamlines (EIS-TIMEX, EIS-TIME, Magnedyn) lack of even basic numerical tools to interpret and predict the Experimental data.


M1
M14
Newport
BA2
BA1S
CLOSE
ATFilms
8020.net


1 + 1 =
2 + 2 =

# ...and the theory?!?!



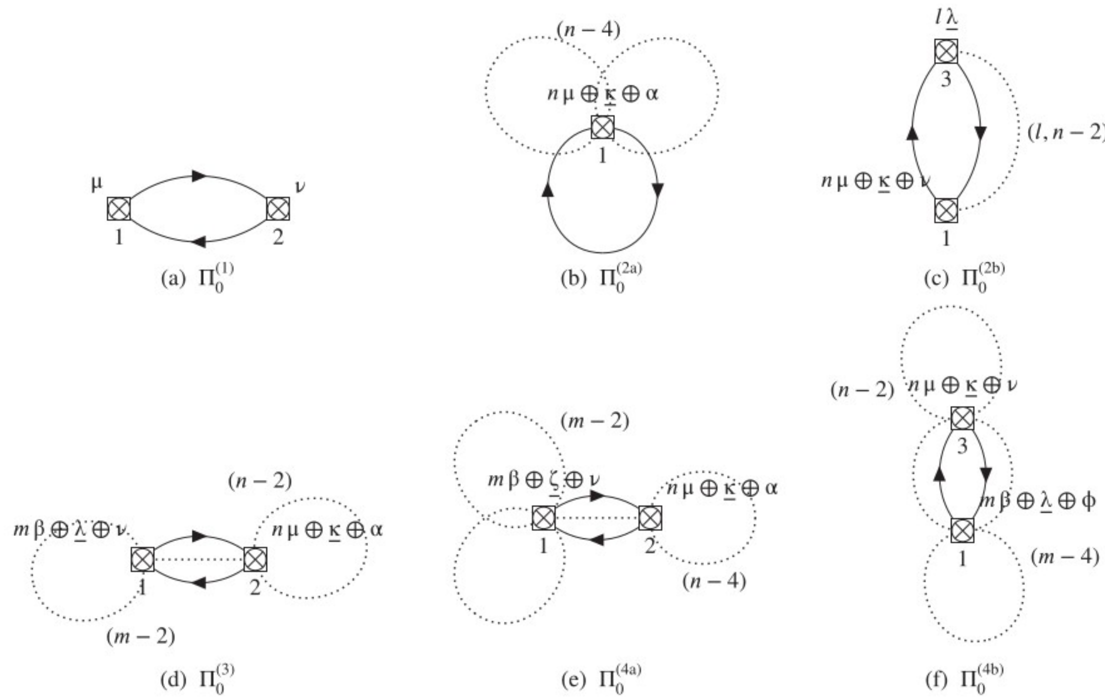
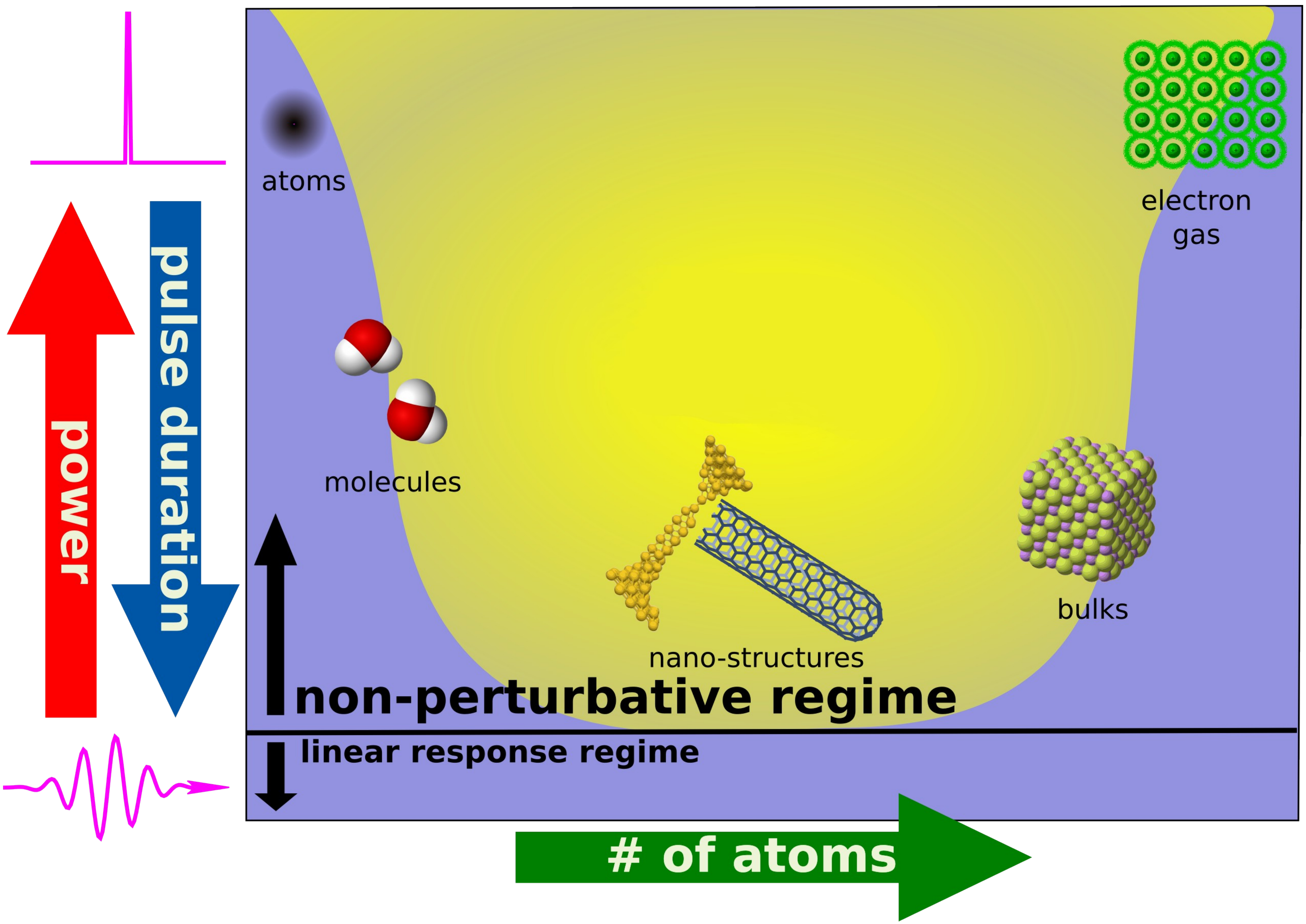



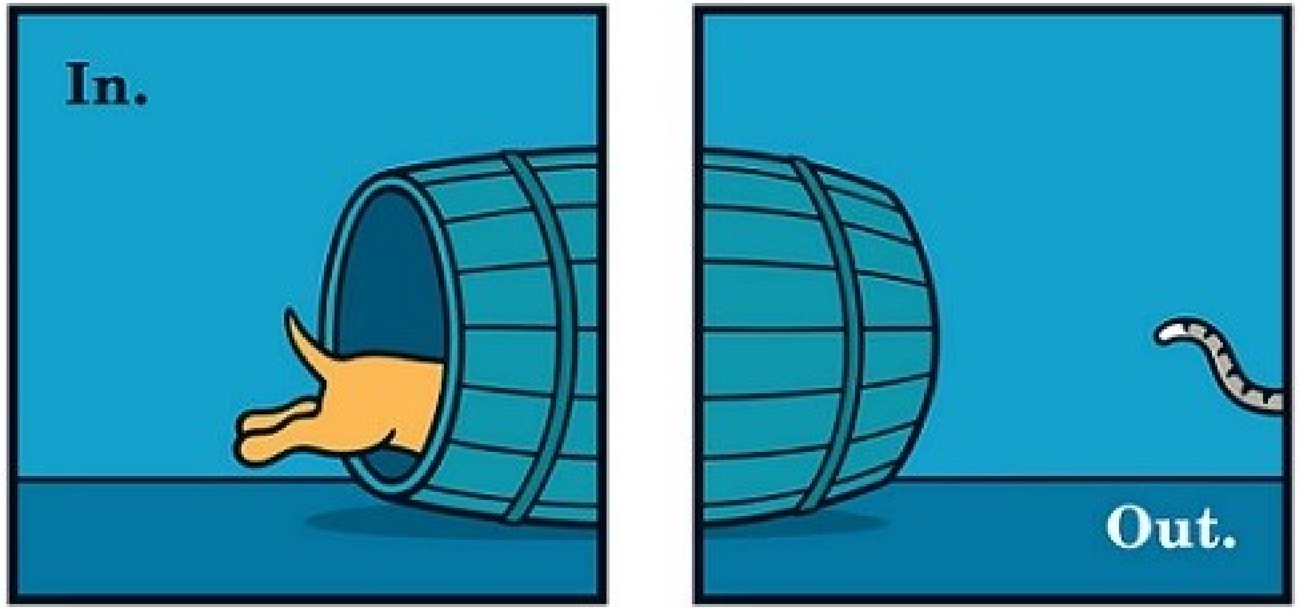

FIG. 11. Lowest-order approximation for the bosonic mass operators $\Pi_{\mu,\nu}(z_1, z_2)|_0$.

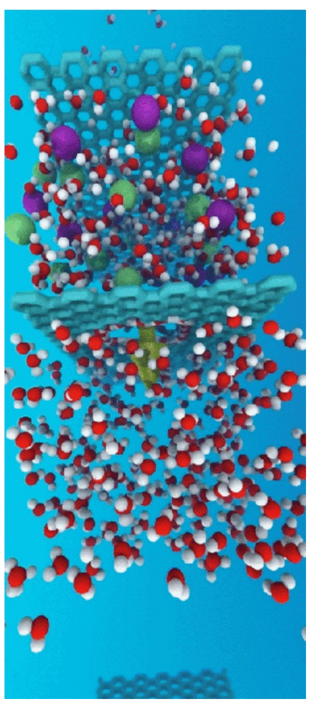
# The "Gap" (I)

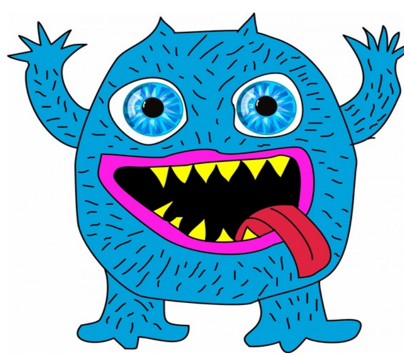


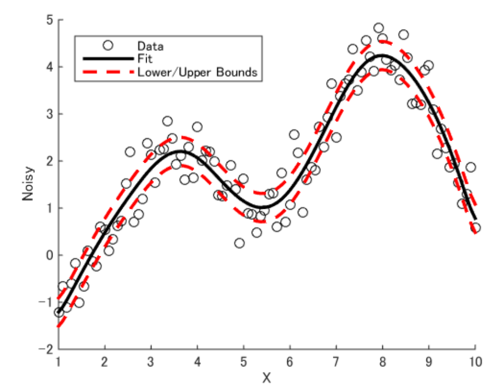
atoms
electron gas
molecules
nano-structures
bulks
non-perturbative regime
linear response regime
power
pulse duration
# of atoms

# The Ab-Initio "Way"

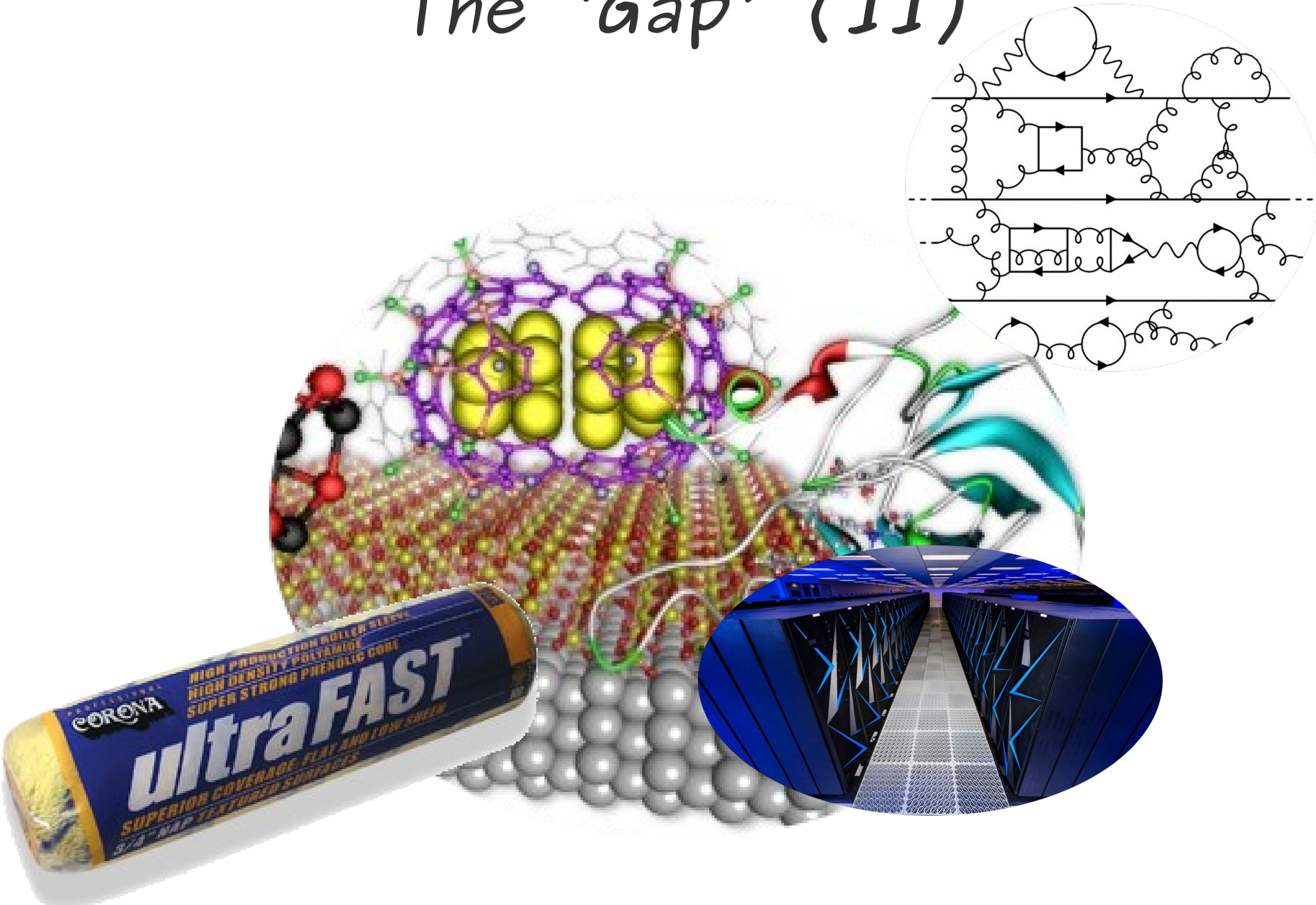


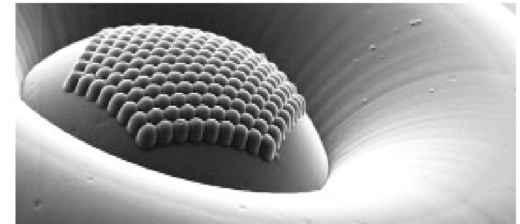



# The "Gap" (II)


CORONA
ultraFAST
HIGH PRODUCTION ROLLER SLEEVE
HIGH DENSITY POLYAMIDE
SUPER STRONG PHENOLIC CORE
SUPERIOR COVERAGE: FLAT AND LOW SHEEN


"Hi Andrea, we measured a long living coherent oscillation of the transient reflectivity in correspondance of the trion bleaching, followed by a non-radiative recombination. Can you calculate it?"

Installation 1

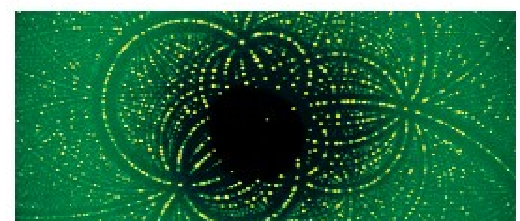
## Lithography & Patterning

Installation 2

## Growth & Synthesis

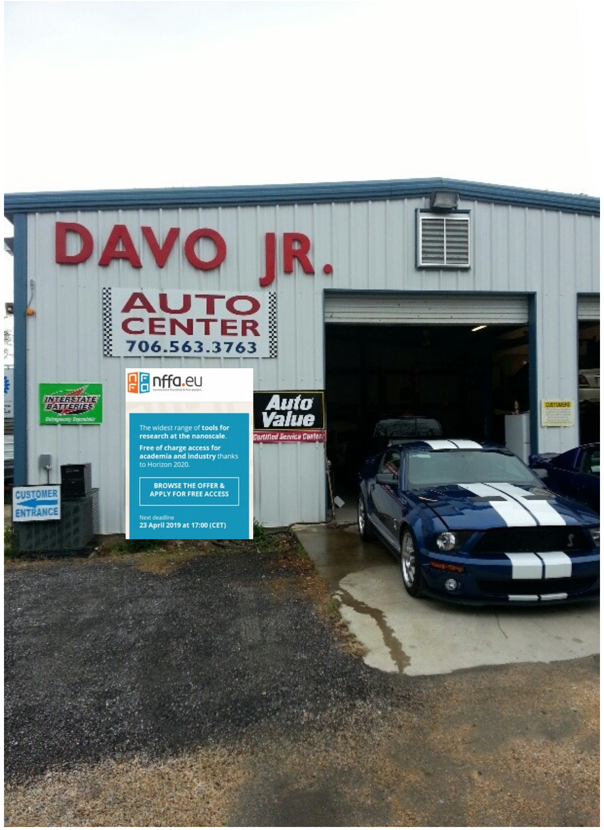
Installation 3

## Theory & Simulation



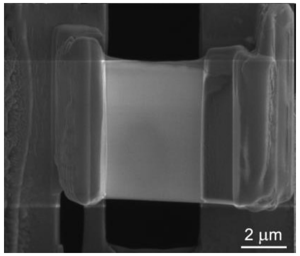
Installation 4, 5, 6

## Characterisation




Anomalous ultra-fast carriers and gap dynamics of Black Phosphorus


Ultrafast electronic dynamics across the FeRh magnetic phase transition

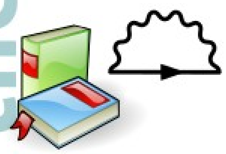



The Users Perspective
An approach based on Large Scale Computational Physics

In the heaven of a theoretical physicist there are only diagrams...

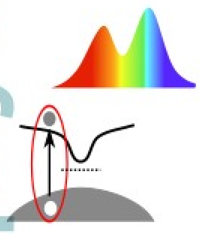
## theory

Accurate approaches from
Many-Body Perturbation-Theory &
(Time-dependent density) functional theory



## physics

To capture the interaction
of matter with
electro-magnetic radiation



## materials

0, 1, 2 ,3 D....
Through all dimensions

## applications

From traditional to
key technology areas

# Yambo©

# an ab initio tool for excited-state calculations

## interfaces

Interfaced with two popular
open-source density funtional codes:
ABINIT and PWSCF.

## parallel

Ready for the petascale era:
MPI+OpenMP structure to
run efficiently on 10K+ cores

## support

Easy shell-based user interface
Extensive online documentation
Active user forum

## team

Andrea Marini - CNR, Italy
Andrea Ferretti - CNR, Italy
Conor Hogan - CNR, Italy
Daniele Varsano - CNR, Italy
Davide Sangalli - CNR, Italy
Maurizia Palummo - Roma2, Italy
Margherita Marsili - CNR, Italy
Myrta Grüning - QUB, UK
Pedro Melo - UC, Portugal

www.yambo-code.org

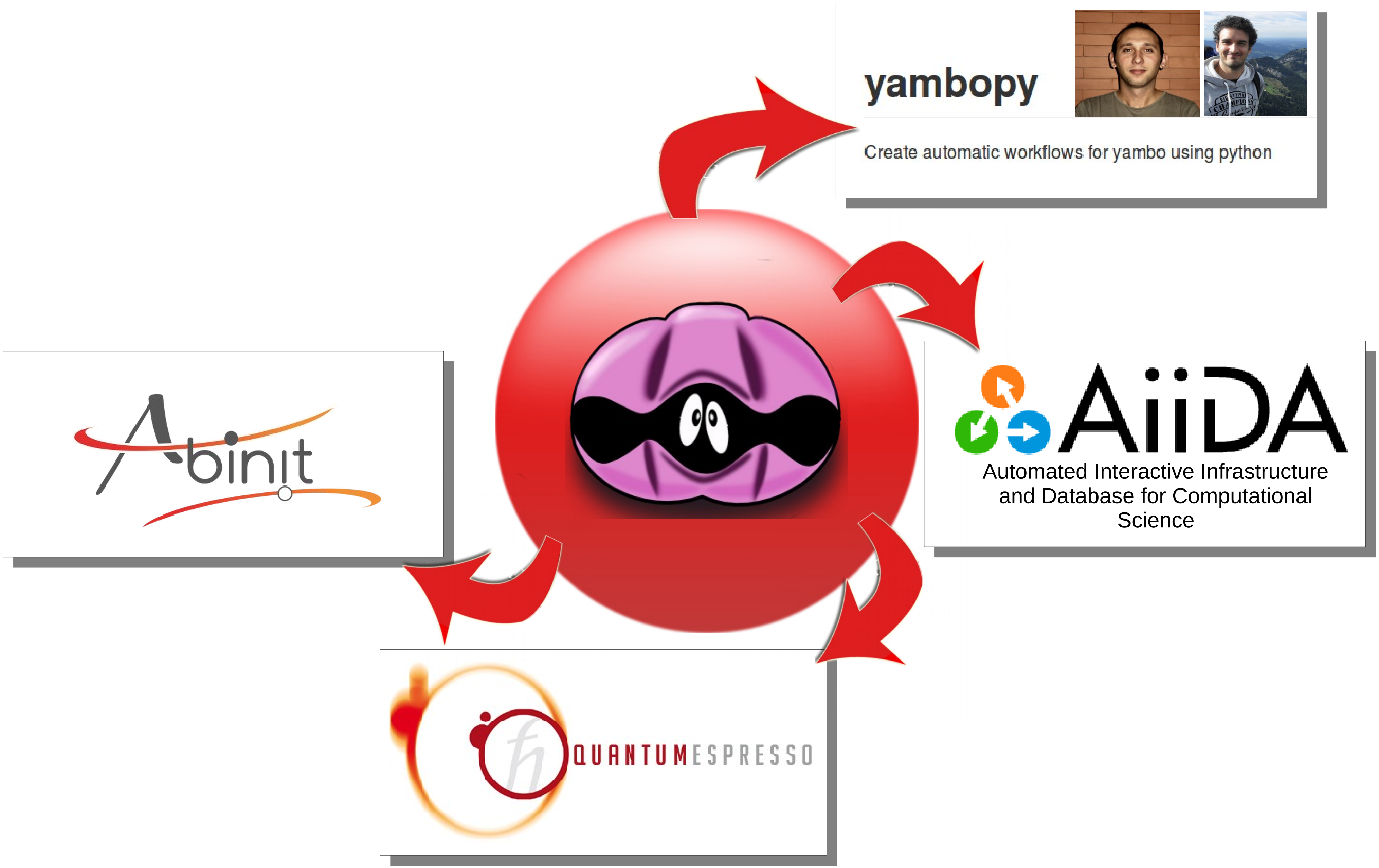
## …is collaborative

# Advanced computing of excited state properties in solids and nanostructures with Yambo

24th - 28th April 2017

CECAM-HQ-EPFL, Lausanne, Switzerland

## ...is part of communities



...is part of communities

## MATERIALS DESIGN AT THE EXASCALE

European center of excellence - a H2020 e-infrastructure

### THE CENTER

a centre of excellence aimed to disenthrall the EU leadership in materials modelling, simulations, discovery and design

### THE CHALLENGE

what if material simulations were 1000x faster and more workable?
driving the exascale transition

### THE CREW

5 research teams, 5 supercomputing centres, 1 educational institute, 2 business partners

MAX DRIVING THE EXASCALE TRANSITION

http://www.max-center.eu/

nffa.eu

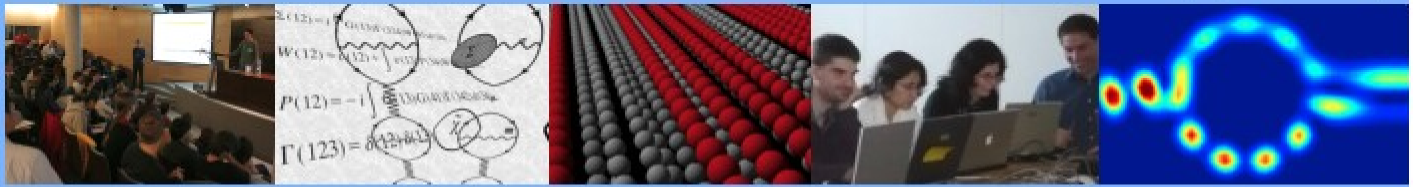
http://www.nffa.eu/

Get **free access** to the widest range of tools **for research at the nanoscale.**

an advanced distribuited infrastructure to perform growth, nano-lithography, nano-characterization, theory and simulation and fine-analysis with synchrotron, FEL and neutron radiation sources.

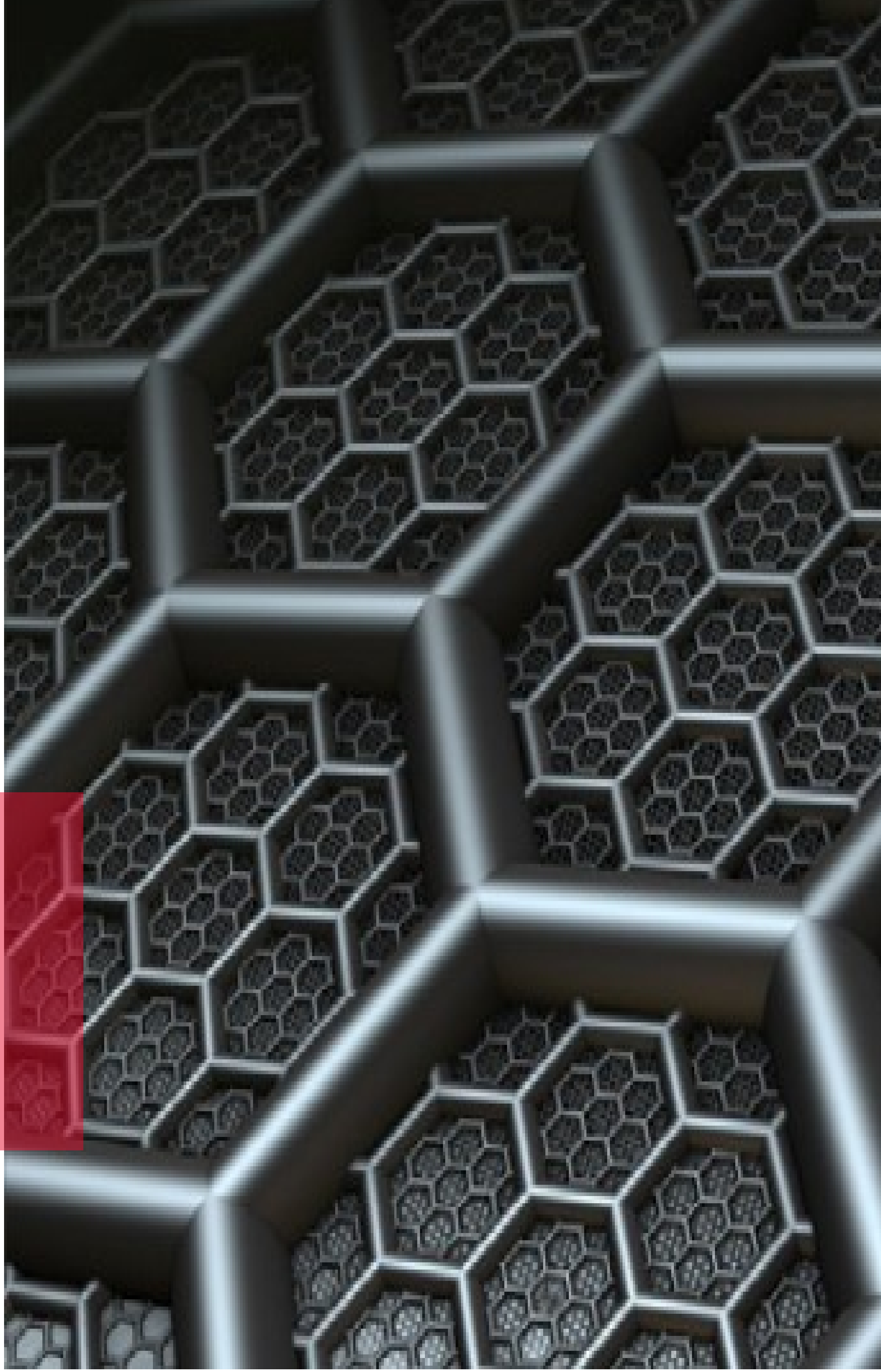
ETSF

European Theoretical Spectroscopy Facility



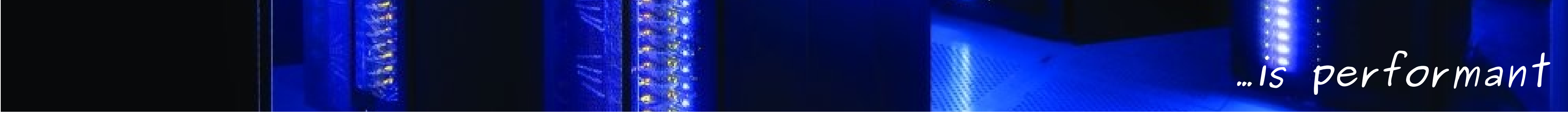
http://www.etsf.eu/

MAX DRIVING THE EXASCALE TRANSITION



# Designing materials with

# High Performance Computing



# …is performant

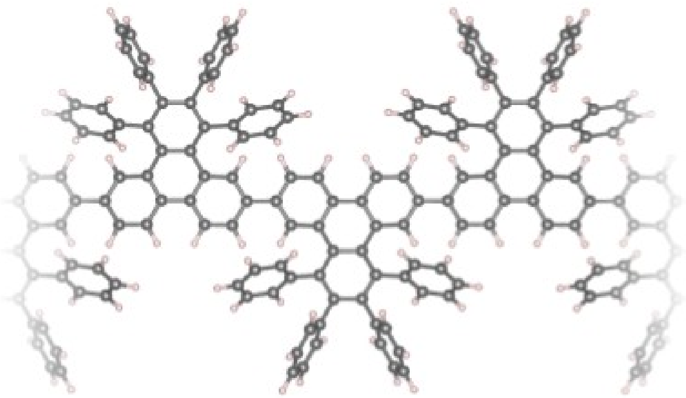
MPI

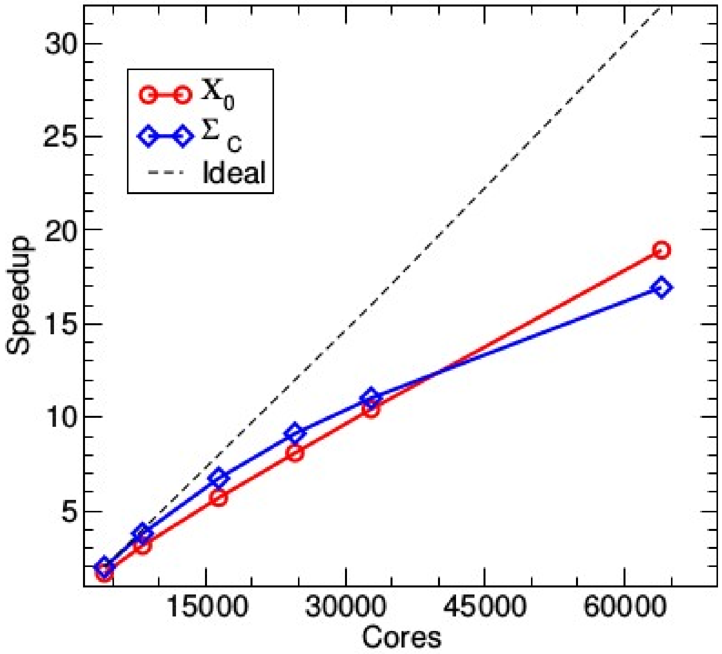
+

OpenMP

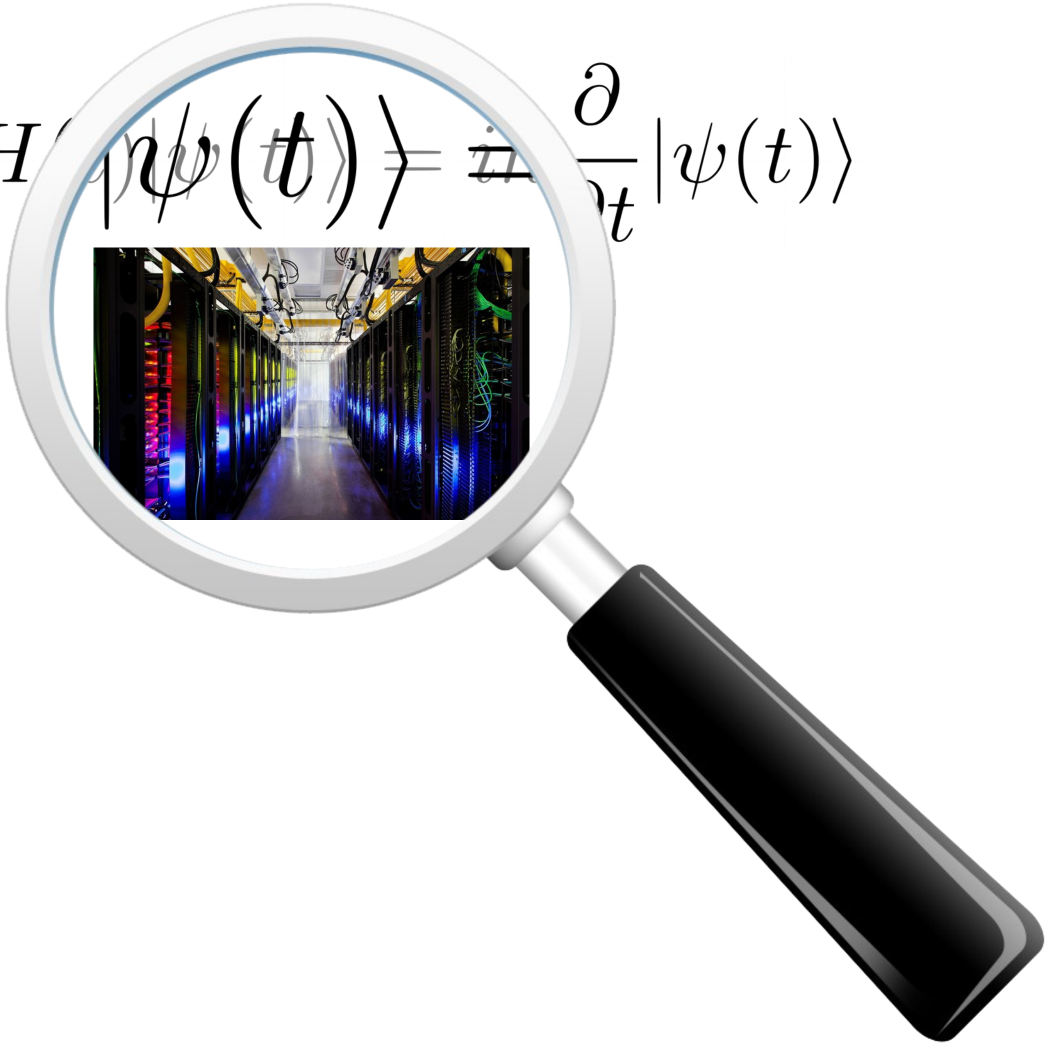
+

MAX DRIVING THE EXASCALE TRANSITION

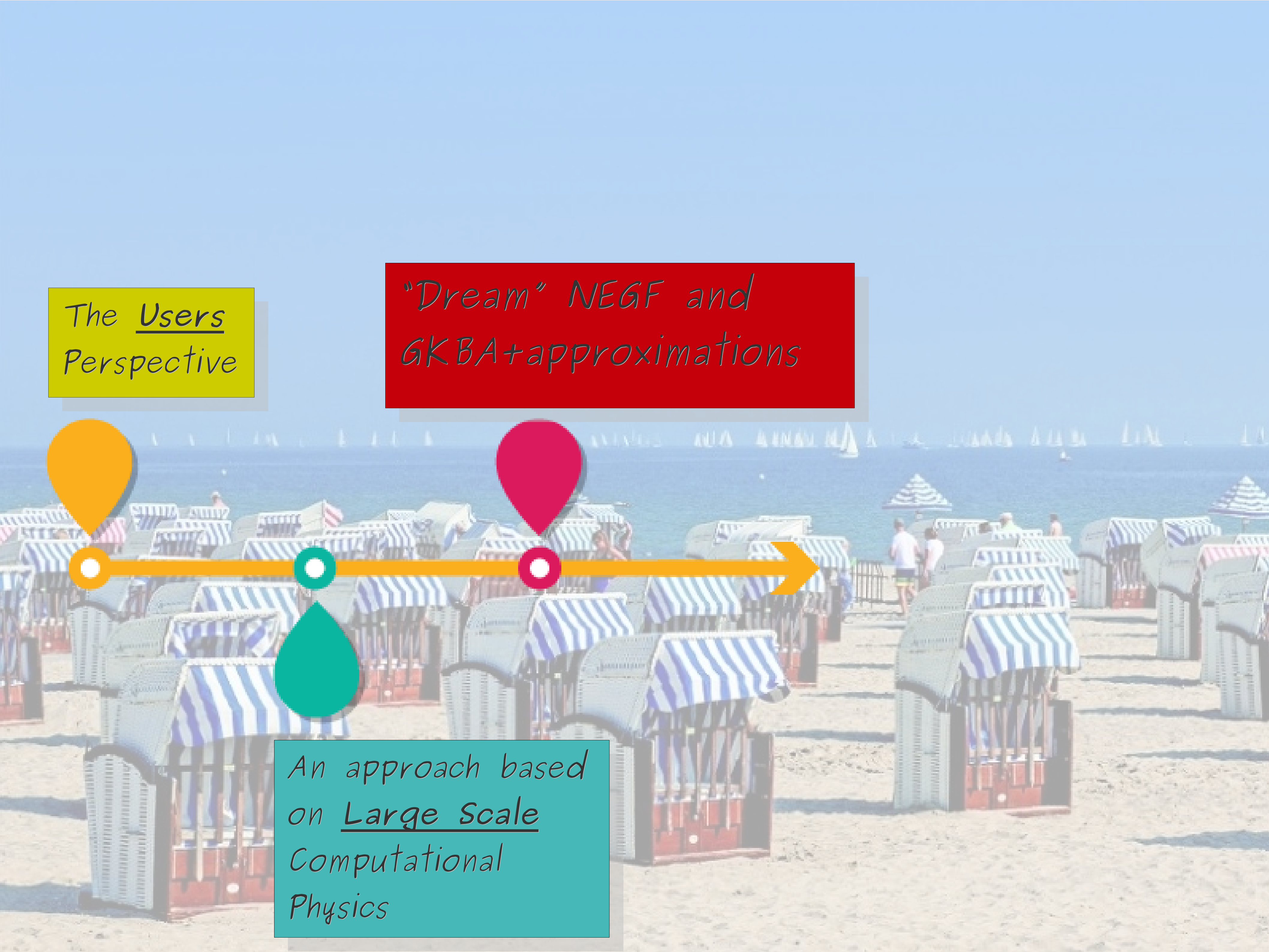





# New materials search: a paradigm shift



$$H(t)|\psi(t)\rangle = i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle$$


The Users Perspective
"Dream" NEGF and GKBA+approximations
An approach based on Large Scale Computational Physics

# The Equilibrium Hedin "Pentagons"

P. Melo, AM, PRB 93, 155102 (2016)

$$H(t) = h_0 + H_{e-\gamma}(t) + H_{e-e} + H_{e-n}$$

Quantized Electrons, PHOTONS & PHONONS

$$\hat{H}_{\text{ext}}(t) = \int \mathrm{d}^3 r\, \hat{\phi}(\mathbf{r}, t)\rho_{\text{ext}}(\mathbf{r}, t) - \int \mathrm{d}^3 r\, \hat{\mathbf{A}}(\mathbf{r}) \cdot \mathbf{J}_{\text{ext}}(\mathbf{r}, t) - \int \mathrm{d}^3 R\, \hat{V}_n(\mathbf{R}) n'(\mathbf{R}, t)$$

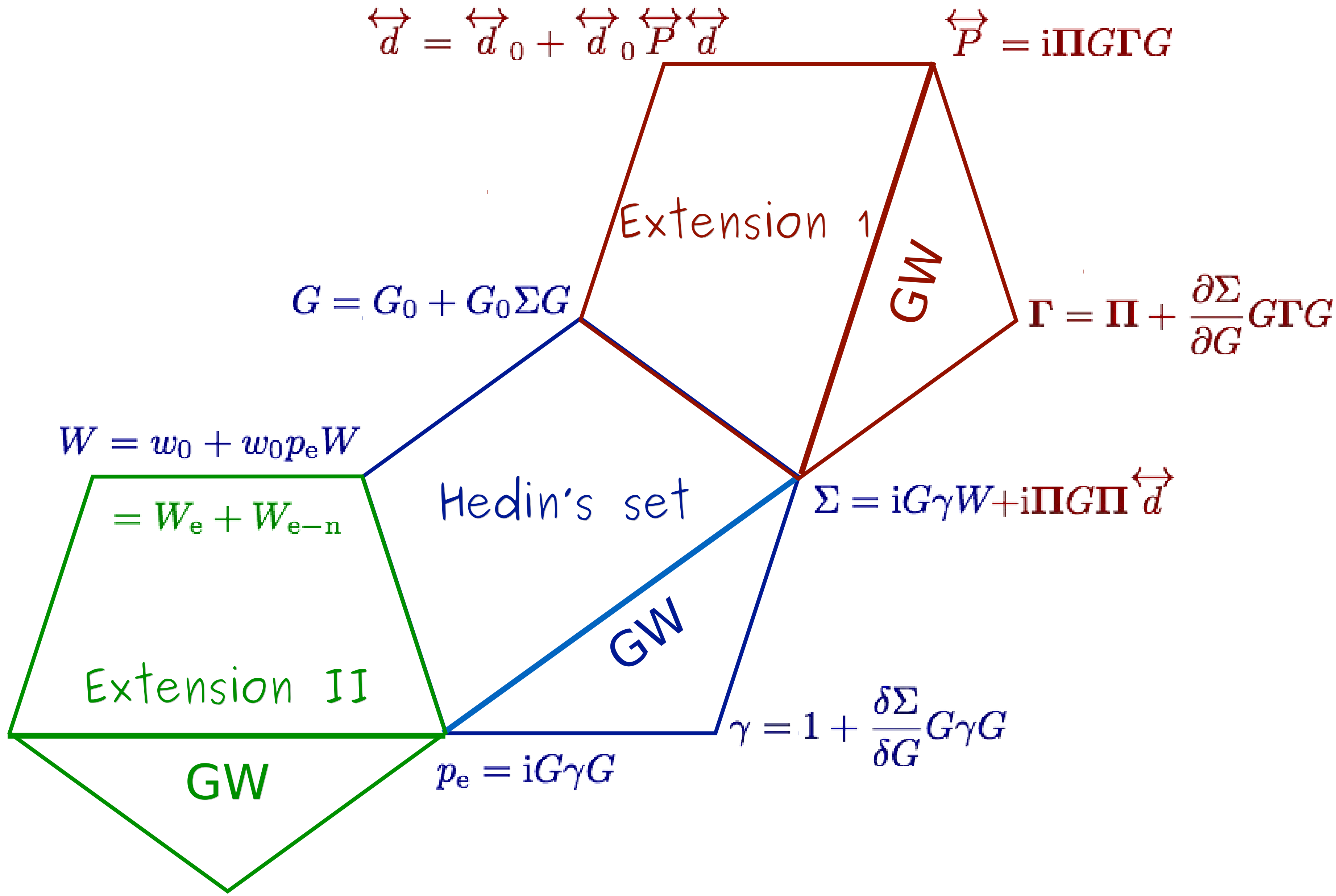
$$G(1,2) = G_0(1,2) + G_0(1,3)\Sigma(3,4)G(4,2)$$

$$\Sigma(\underline{1}, \underline{2}) = \mathrm{i} \left[ G(\underline{1}, \underline{3})\gamma(\underline{3}, \underline{2}, \underline{4})W(\underline{4}, \underline{1}^{+}) + \sum_{i,k=1}^{3} \Pi_i(\underline{1}, \underline{1}')G(\underline{1}, \underline{3})\Gamma_k(\underline{3}, \underline{2}, \underline{4})d_{k,i}(\underline{4}, \underline{1}') \right] \Bigg|_{\underline{1}'=\underline{1}}$$

P. Melo, AM, PRB 93, 155102 (2016)

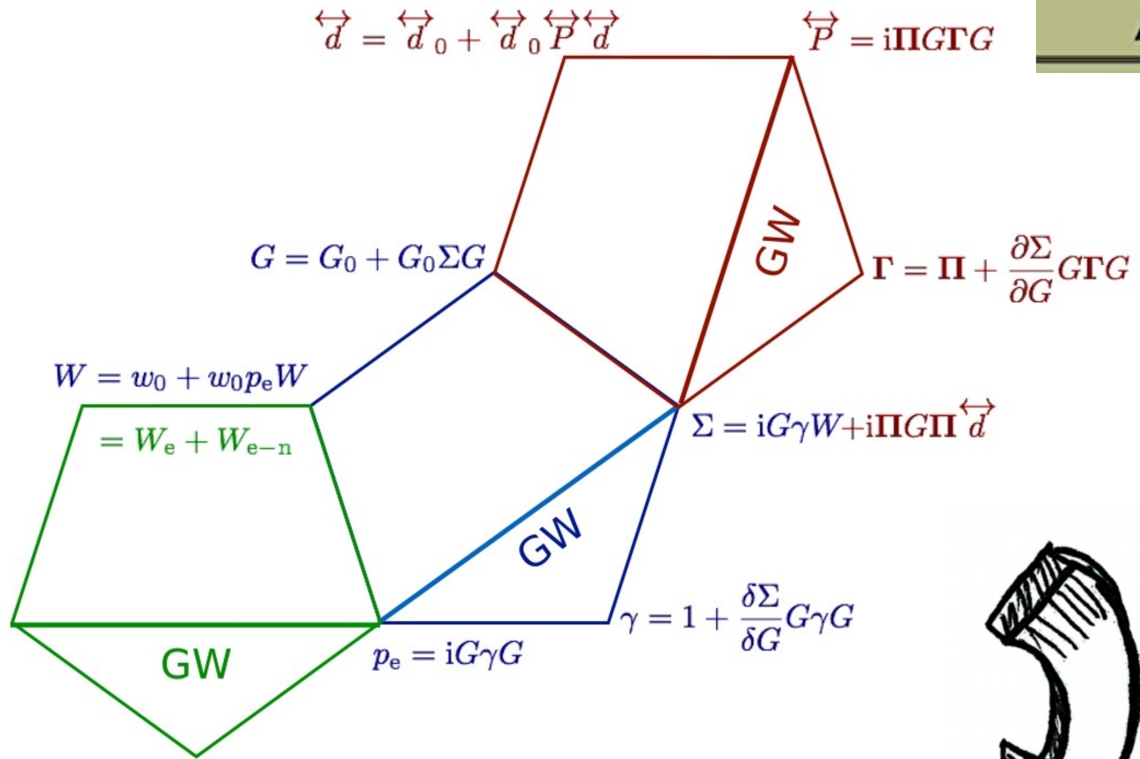
R. van Leeuwen et al, Time-Dependent Density Functional Theory, Springer Berlin Heidelberg, 2006, 33-59

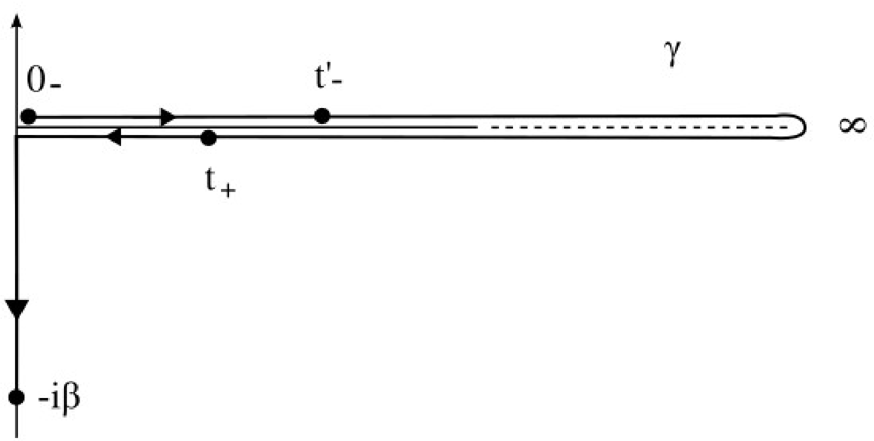
# The Equilibrium Hedin "Pentagons"



# EQ → NEQ (BKE)

Kadanoff/Baym



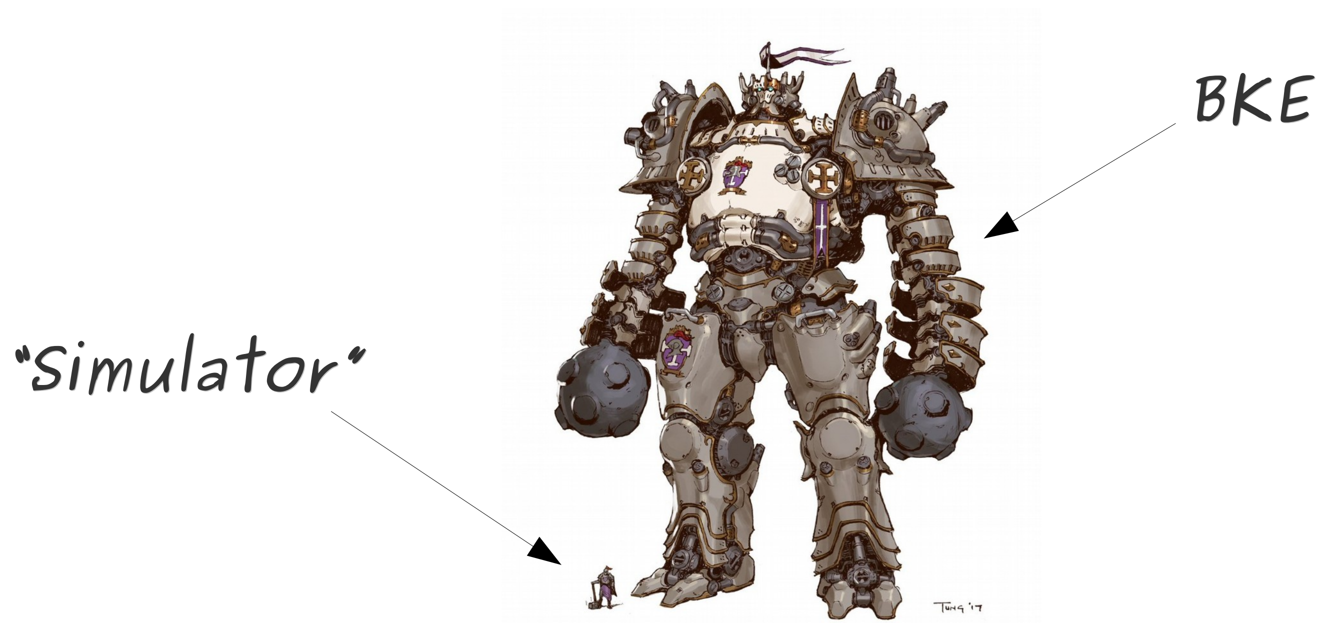




$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z'}\boldsymbol{G}(z,z') = \mathbf{1}\delta(z,z') + \boldsymbol{G}(z,z')\boldsymbol{h}(z') + \int_\gamma \mathrm{d}\bar{z}\, \boldsymbol{G}(z,\bar{z})\boldsymbol{\Sigma}(\bar{z},z')$$

$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z'}\boldsymbol{G}(z,z') = \mathbf{1}\delta(z,z') + \boldsymbol{G}(z,z')\boldsymbol{h}(z') + \int_\gamma \mathrm{d}\bar{z}\, \boldsymbol{G}(z,\bar{z})\boldsymbol{\Sigma}(\bar{z},z')$$

| Definition | $c(z,z') = \int_\gamma \mathrm{d}\bar{z}\, a(z,\bar{z})b(\bar{z},z')$ | $c(z,z') = a(z,z')b(z',z)$ |
|---|---|---|
| $k^>(t,t') = k(t_+,t'_-)$ | $c^> = a^> \cdot b^{\mathrm{A}} + a^{\mathrm{R}} \cdot b^> + a^\rceil \star b^\lceil$ | $c^> = a^> b^<$ |
| $k^<(t,t') = k(t_-,t'_+)$ | $c^< = a^< \cdot b^{\mathrm{A}} + a^{\mathrm{R}} \cdot b^< + a^\rceil \star b^\lceil$ | $c^< = a^< b^>$ |
| $k^{\mathrm{R}}(t,t') = \delta(t-t')k^\delta(t) + \theta(t-t')[k^>(t,t') - k^<(t,t')]$ | $c^{\mathrm{R}} = a^{\mathrm{R}} \cdot b^{\mathrm{R}}$ | $c^{\mathrm{R}} = \begin{cases} a^{\mathrm{R}} b^< + a^< b^{\mathrm{A}} \\ a^{\mathrm{R}} b^> + a^> b^{\mathrm{A}} \end{cases}$ |
| $k^{\mathrm{A}}(t,t') = \delta(t-t')k^\delta(t) - \theta(t'-t)[k^>(t,t') - k^<(t,t')]$ | $c^{\mathrm{A}} = a^{\mathrm{A}} \cdot b^{\mathrm{A}}$ | $c^{\mathrm{A}} = \begin{cases} a^{\mathrm{A}} b^< + a^< b^{\mathrm{R}} \\ a^{\mathrm{A}} b^> + a^> b^{\mathrm{R}} \end{cases}$ |
| $k^\rceil(t,\tau) = k(t_\pm,\tau)$ | $c^\rceil = a^{\mathrm{R}} \cdot b^\rceil + a^\rceil \star b^{\mathrm{M}}$ | $c^\rceil = a^\rceil b^\lceil$ |
| $k^\lceil(\tau,t) = k(\tau,t_\pm)$ | $c^\lceil = a^\lceil \cdot b^{\mathrm{A}} + a^{\mathrm{M}} \star b^\lceil$ | $c^\lceil = a^\lceil b^\rceil$ |
| $k^{\mathrm{M}}(\tau,\tau') = k(z=\tau, z'=\tau')$ | $c^{\mathrm{M}} = a^{\mathrm{M}} \star b^{\mathrm{M}}$ | $c^{\mathrm{M}} = a^{\mathrm{M}} b^{\mathrm{M}}$ |

$$\left[\mathrm{i}\partial_{t'} - h_{\mathrm{ext}}(t')\right] G^<(t,t') = \int d\bar{t}\left[G^r(t,\bar{t})\,\Sigma^<(\bar{t},t') + G^<(t,\bar{t})\,\Sigma^a(\bar{t},t')\right] + \mathrm{h.c.}$$

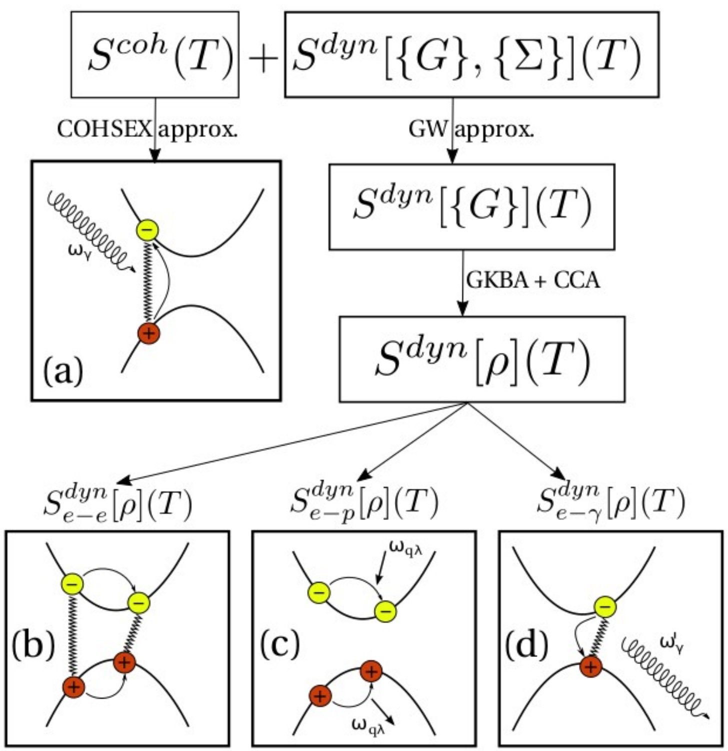


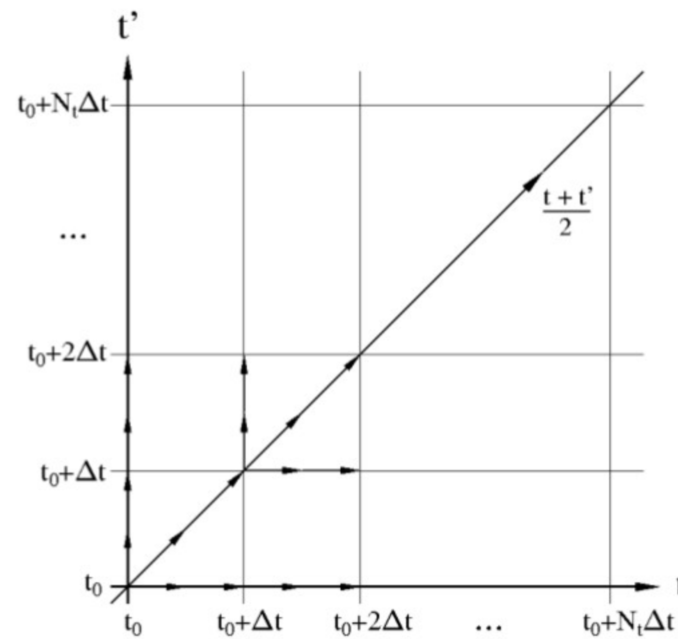
# The Generalized Baym Kadanoff Ansatz

$$\left[i\partial_{t'} - h_{\text{ext}}(t')\right] G^<(t,t') = \int d\bar{t}\, \underbrace{\left[G^r(t,\bar{t})\,\Sigma^<(\bar{t},t') + G^<(t,\bar{t})\,\Sigma^a(\bar{t},t')\right]}_{S(t,t') \approx S(T)} + \text{h.c.}$$

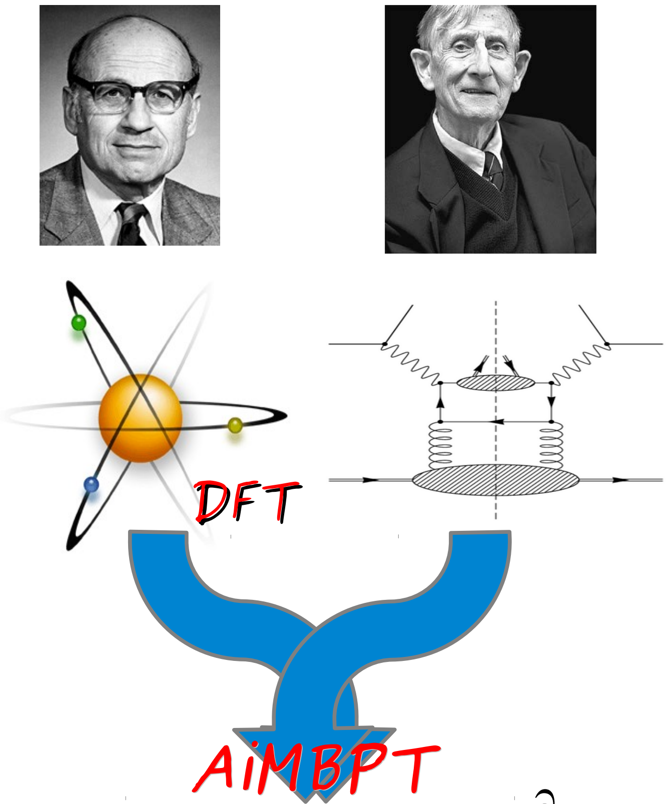
The Ansatz

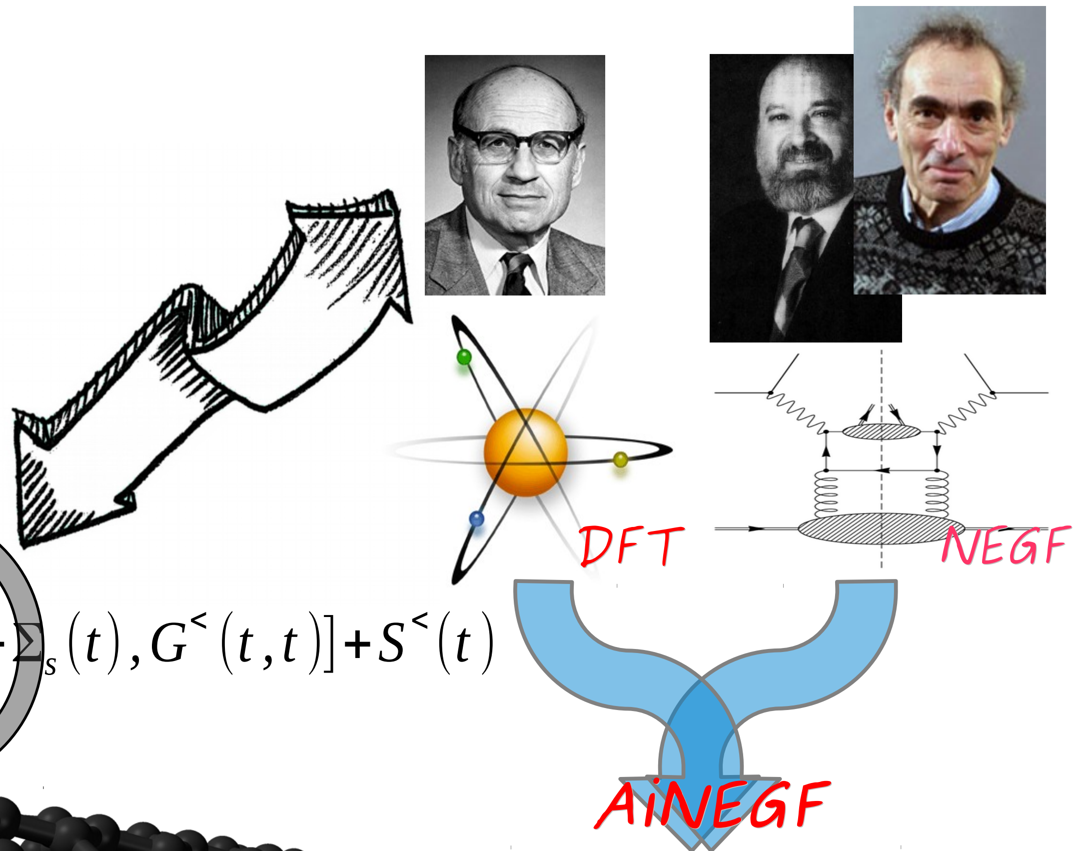
$$G^<(t,\tau) \approx i\left[G^r(t-\tau)G^<(\tau) - G^<(t)G^r(t-\tau)\right]$$



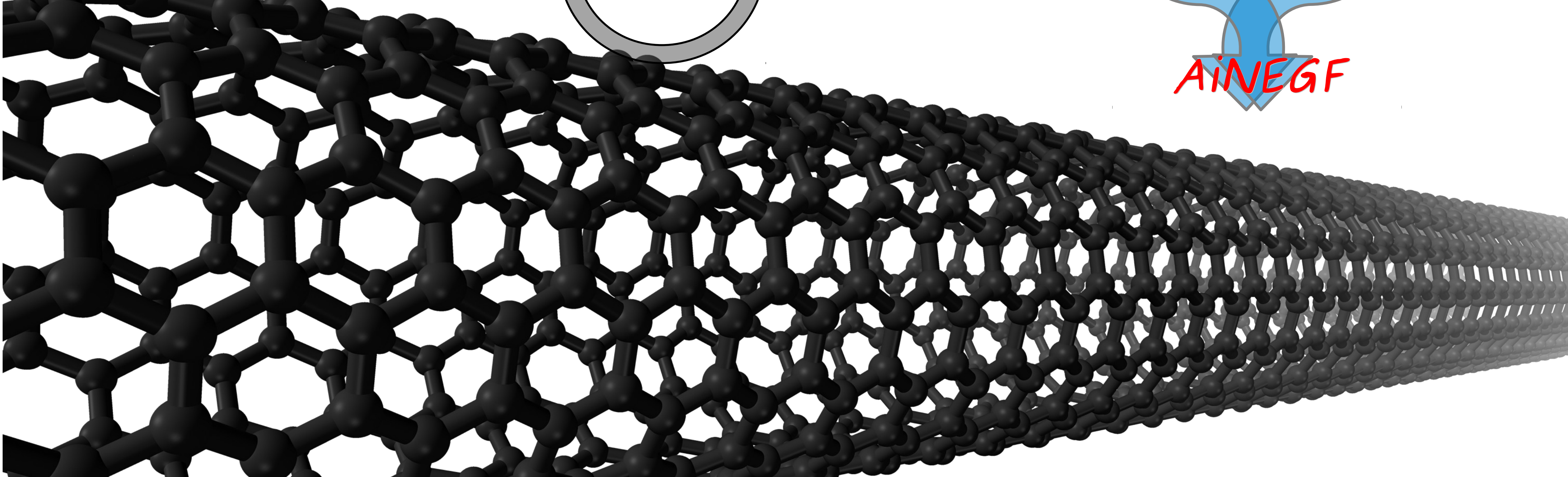



# The "Bridge"

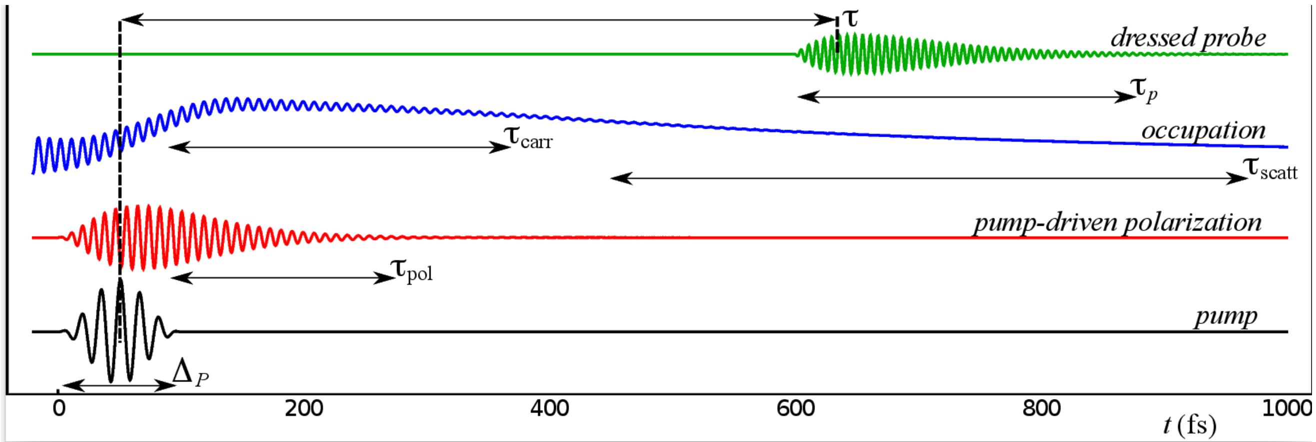



$$i\frac{\partial}{\partial t}G^{<}(t,t)=[h^{DFT}+\Sigma_s(t),G^{<}(t,t)]+S^{<}(t)$$



# The adiabatic ansatz



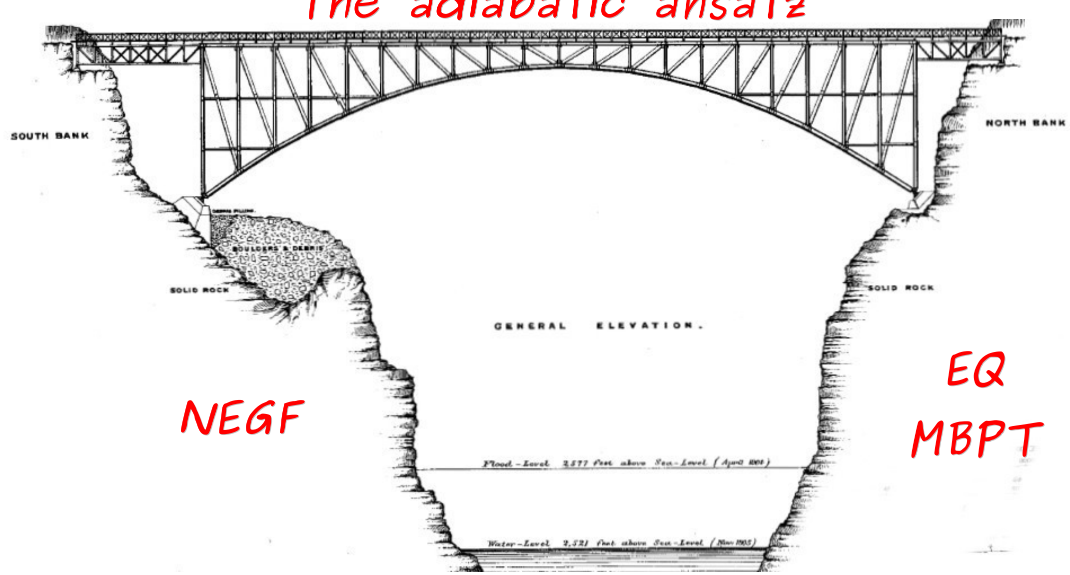

E. Perfetto, D. Sangalli, AM and G. Stefanucci
Phys. Rev. B 92, 205304 (2015)

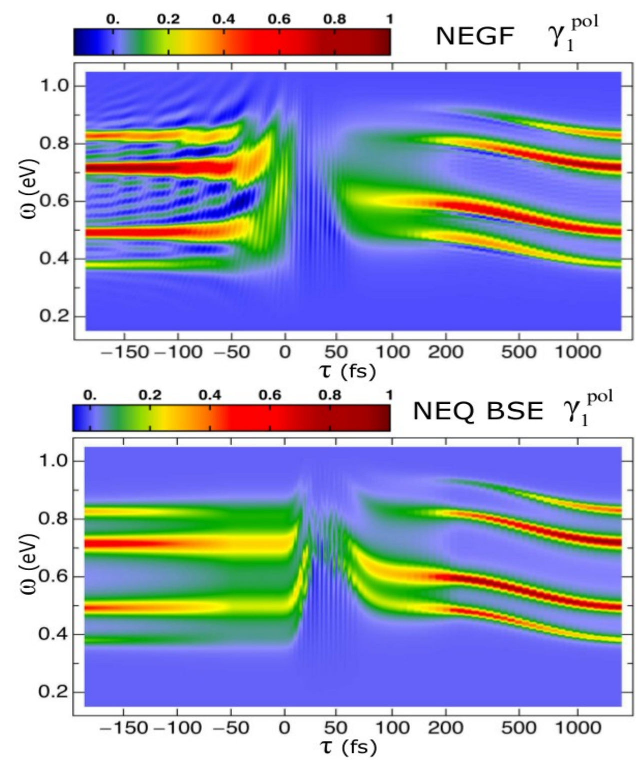
$\chi(t,t')$

The Adiabatic Regime

$\chi(t,t') \approx \chi^{\tau}(t-t')$

The adiabatic ansatz



# Some of the approximations used

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

Generalized Baym-Kadanoff ansatz

$$W^{(r/a)}_{\mathbf{G}_1,\mathbf{G}_2}(\mathbf{q},t_1,t_2;\beta)\approx W_{\mathbf{G}_1,\mathbf{G}_2}(\mathbf{q})$$

Static screening

$$G^{<}_{nn'\mathbf{k}}(T,T;\beta)\approx i\delta_{nn'}f_{n\mathbf{k}}(T;\beta)$$

Diagonal lesser GF (in the collision integral)

$$\Sigma^{\lessgtr}_{nn'\mathbf{k}}(T,T;\beta)\approx \delta_{nn'}\Sigma_{n\mathbf{k}}(T;\beta)$$

Diagonal self-energies

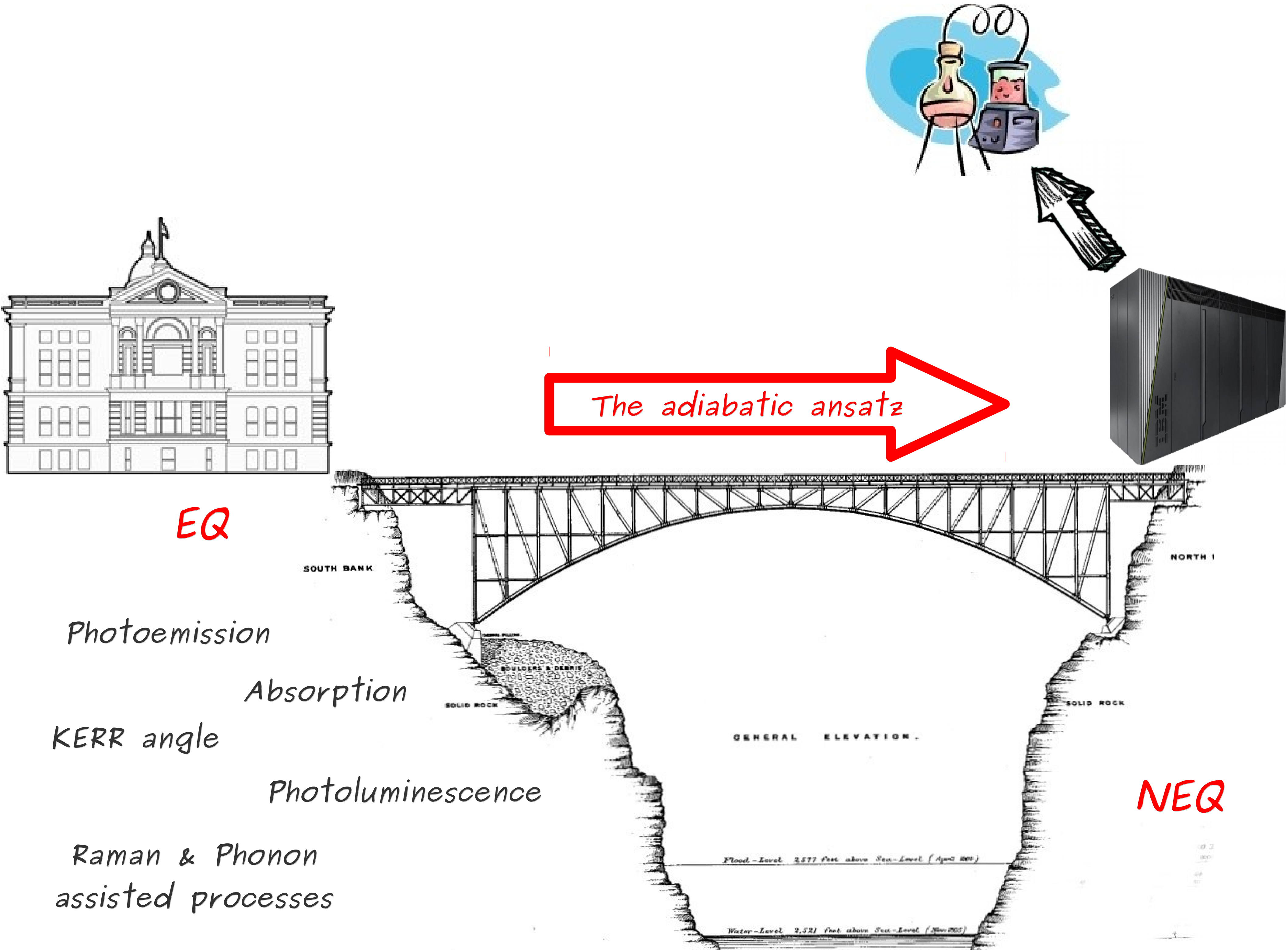
$$G^{(r)}(t,\tau)\approx G^{(r,frozen)}(t-\tau)$$

Retarded GF is not evolved on-the-fly

$$\int_{-\infty}^{t} f_{n\vec{k}}(\tau)(\ldots)\approx f_{n\vec{k}}(t)\int_{-\infty}^{t}(\ldots)$$

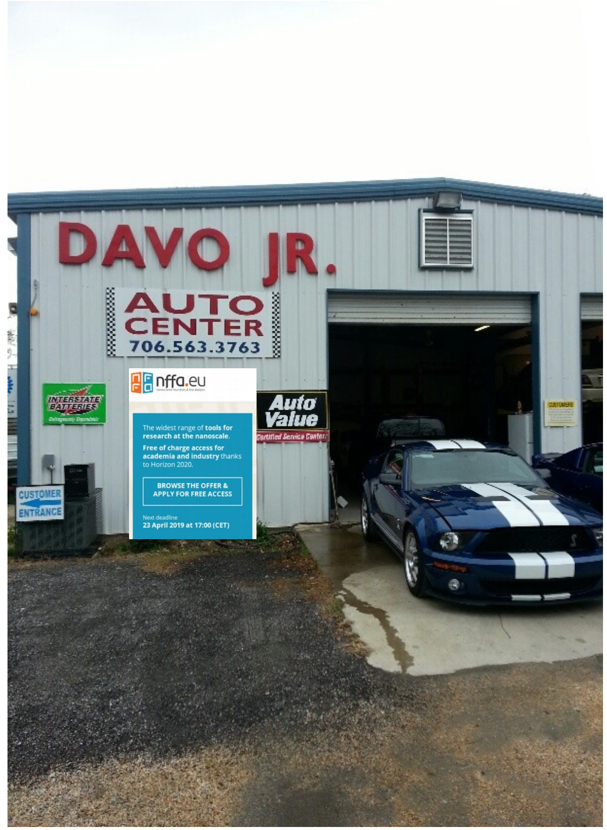
Completed Collision Approximation

A. Marini, Journal of Physics: Conference Series 427, 012003 (2013)



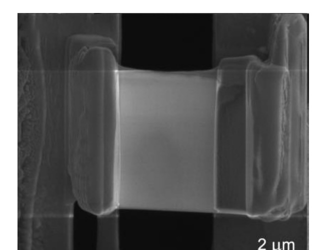
The adiabatic ansatz
EQ
NEQ
SOUTH BANK
SOLID ROCK
SOLID ROCK
GENERAL ELEVATION.
IBM
Photoemission
Absorption
KERR angle
Photoluminescence
Raman & Phonon
assisted processes


The Users Perspective
"Dream" NEGF and GKBA+approximations
An approach based on Large Scale Computational Physics
Ab-Initio NEGF (AiNEGF) at work. Pros.

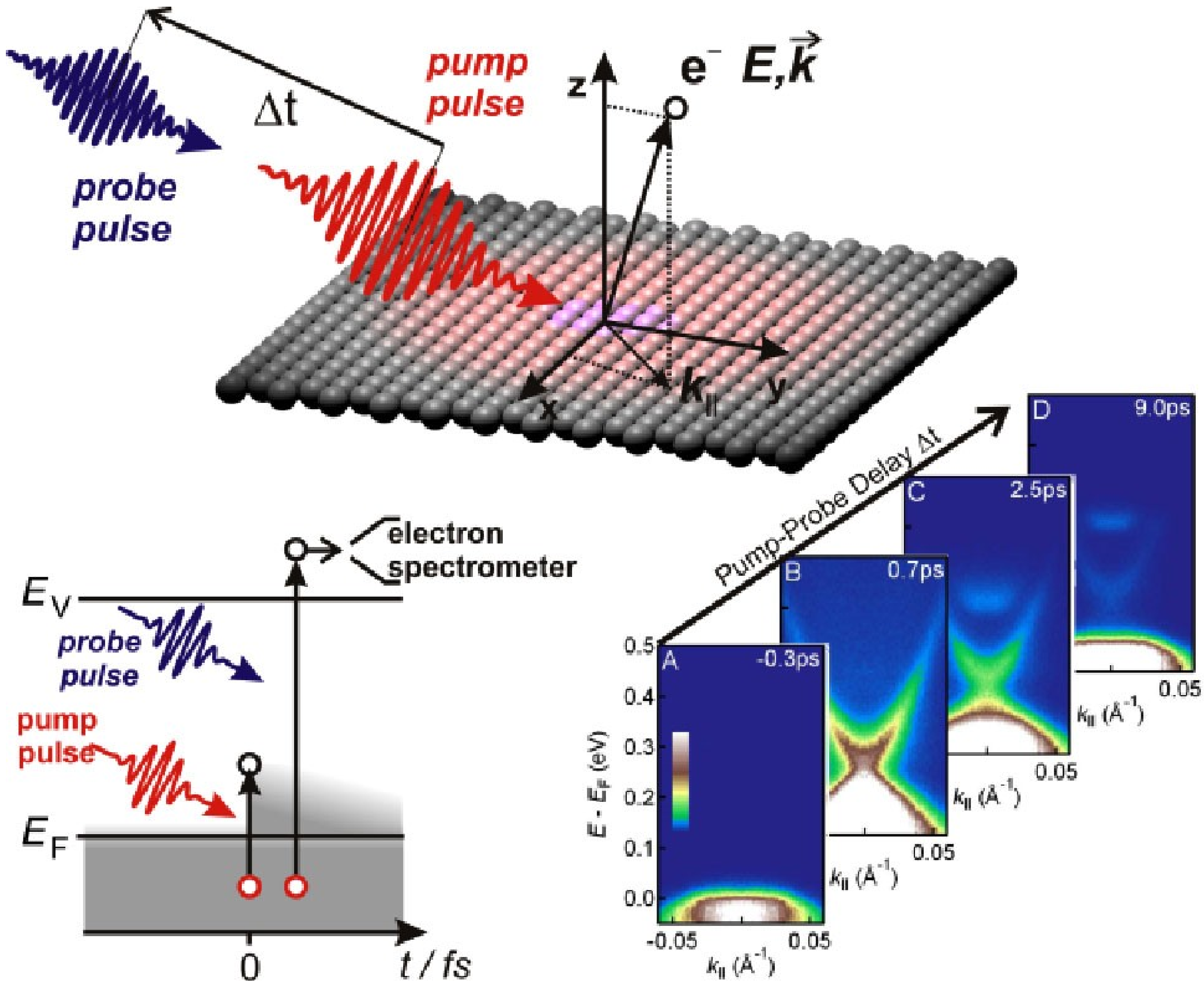



Anomalous ultra-fast carriers and gap dynamics of Black Phosphorus


Ultrafast electronic dynamics across the FeRh magnetic phase transition

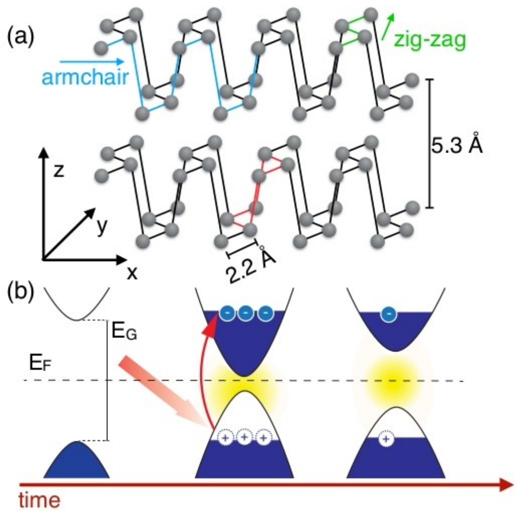


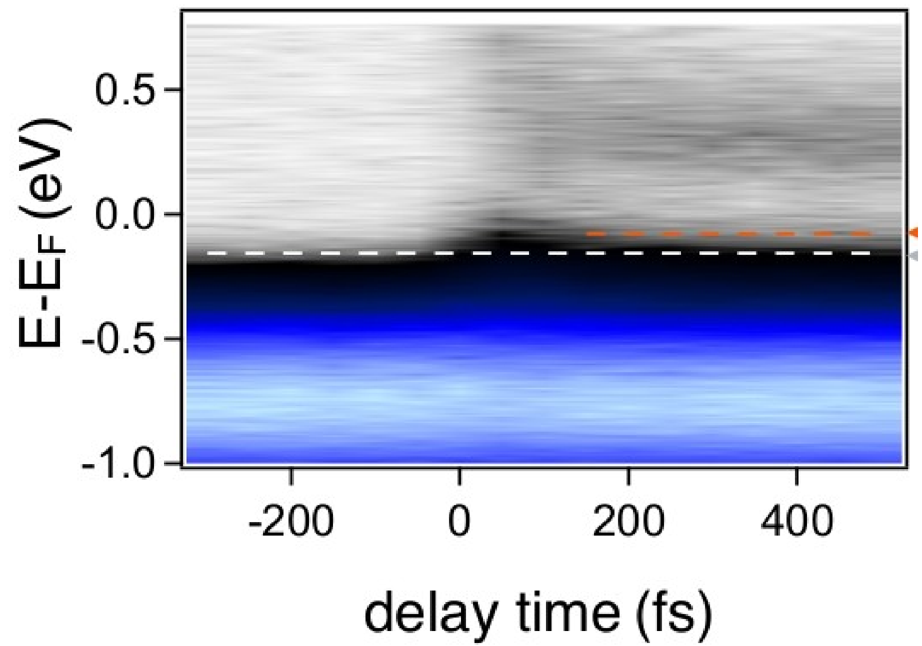
# Time Resolved Angle Resolved Photo Emission

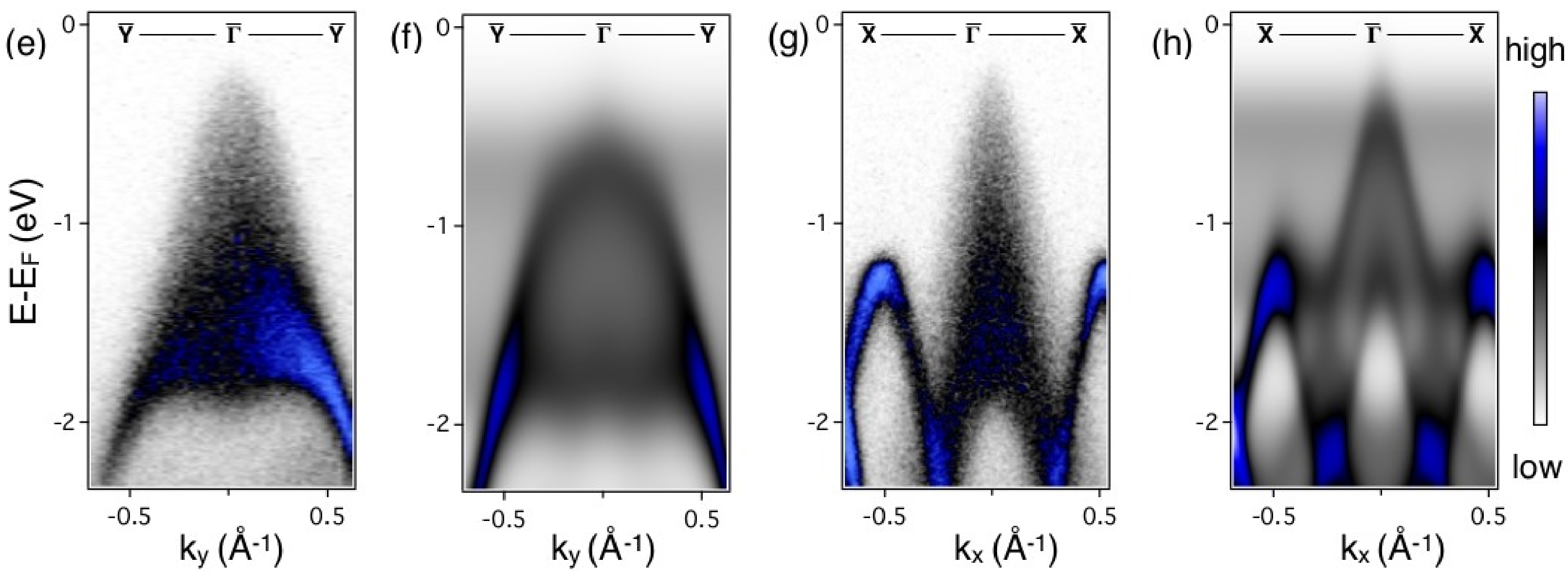


# Photocarrier-induced band-gap renormalization andultrafast charge dynamics in black phosphorus

[S. Roth,..., AM and M. Grioni, in press on 2D Materials]

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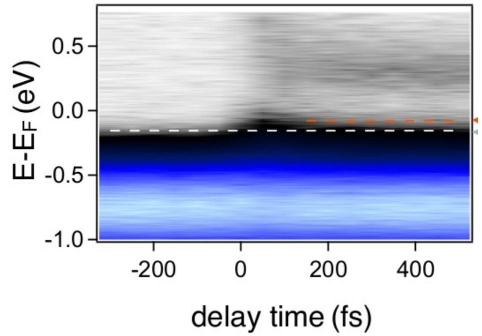


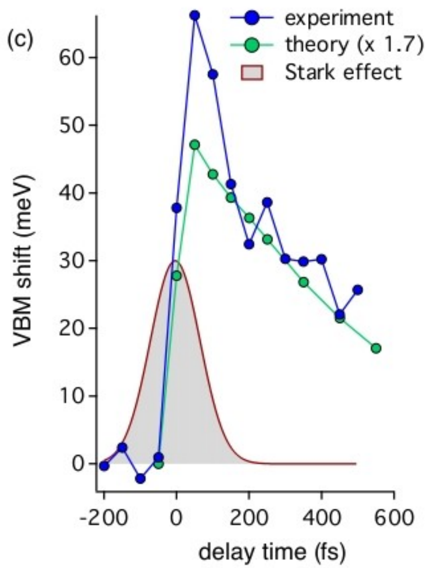


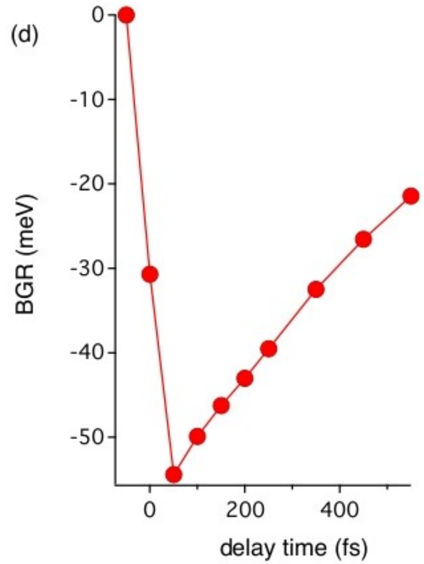


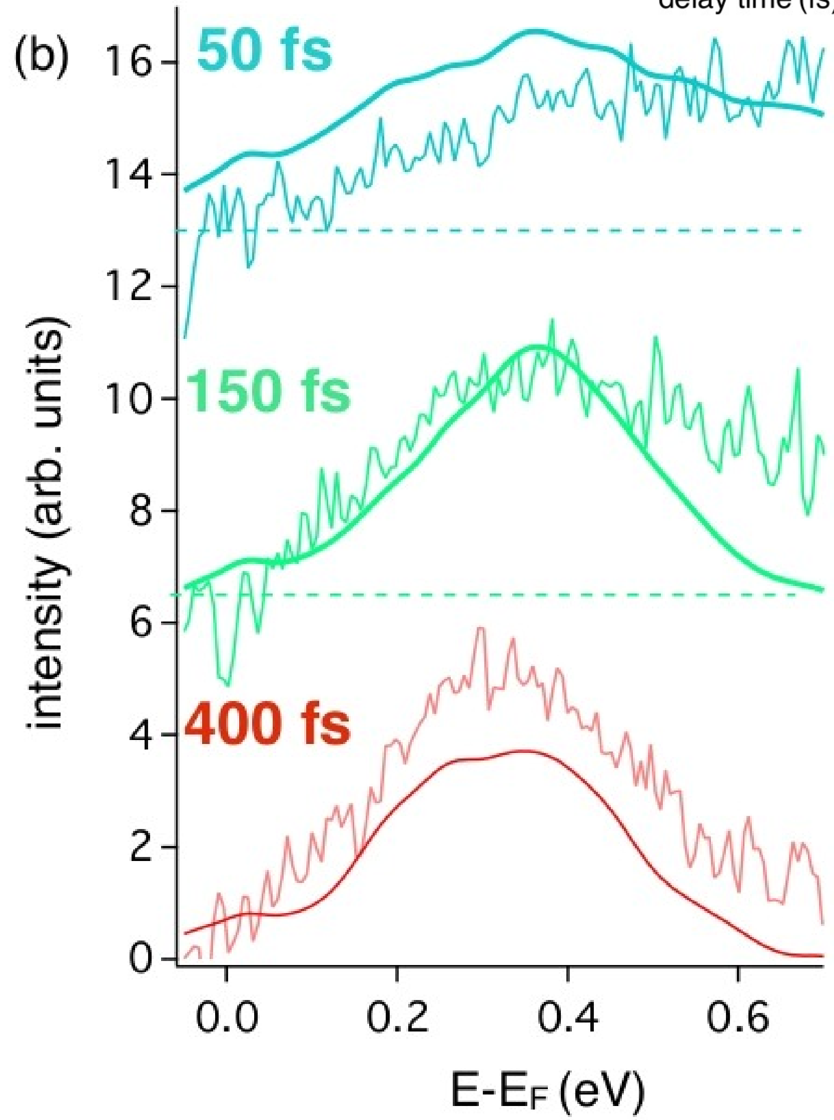


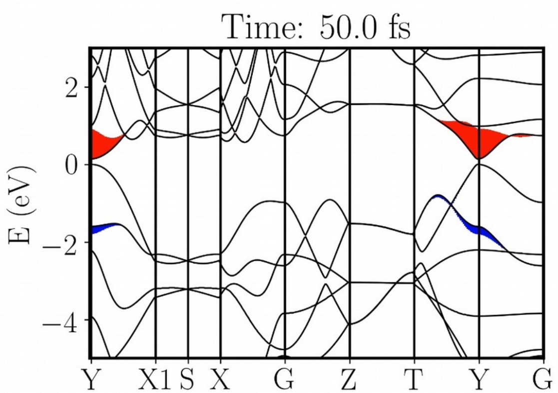


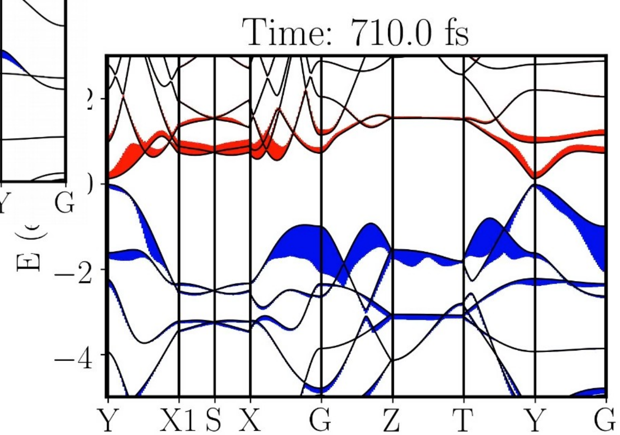

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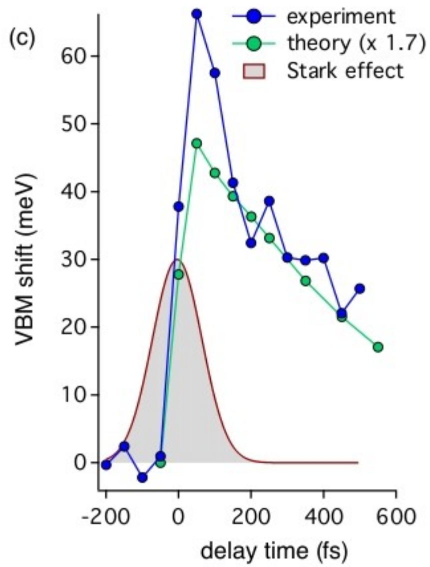
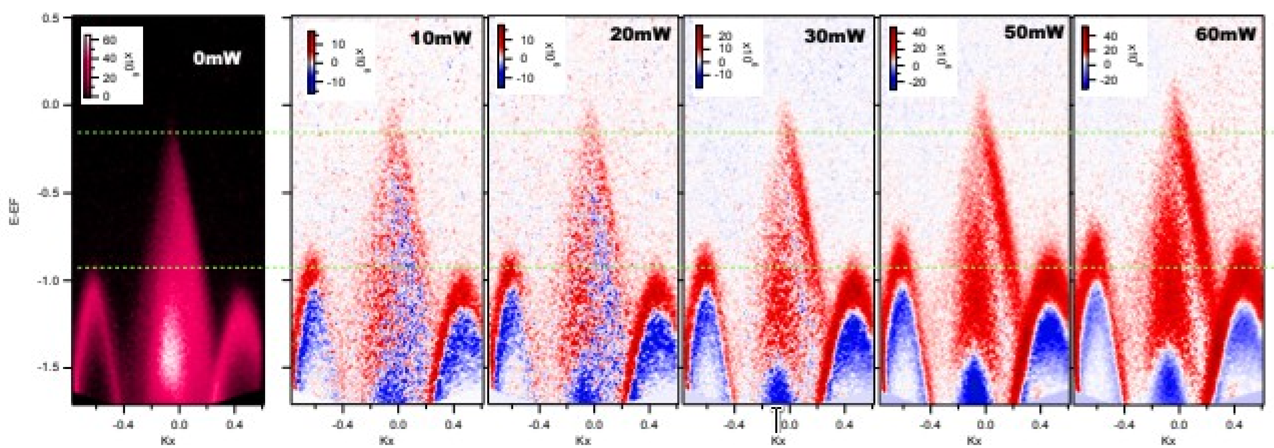


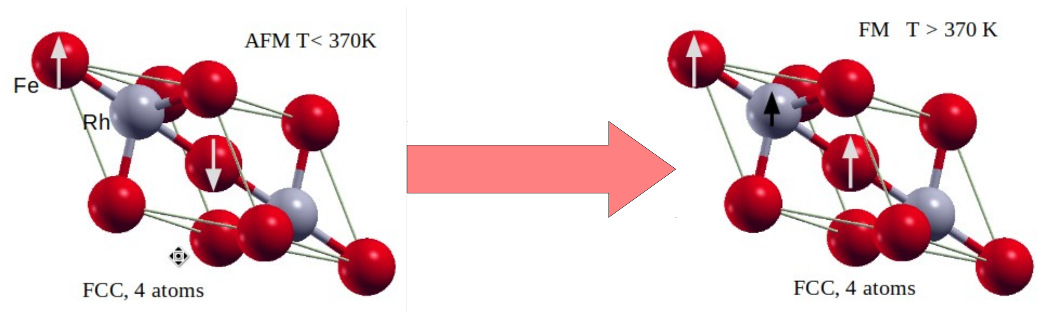

Fluence dependent shift

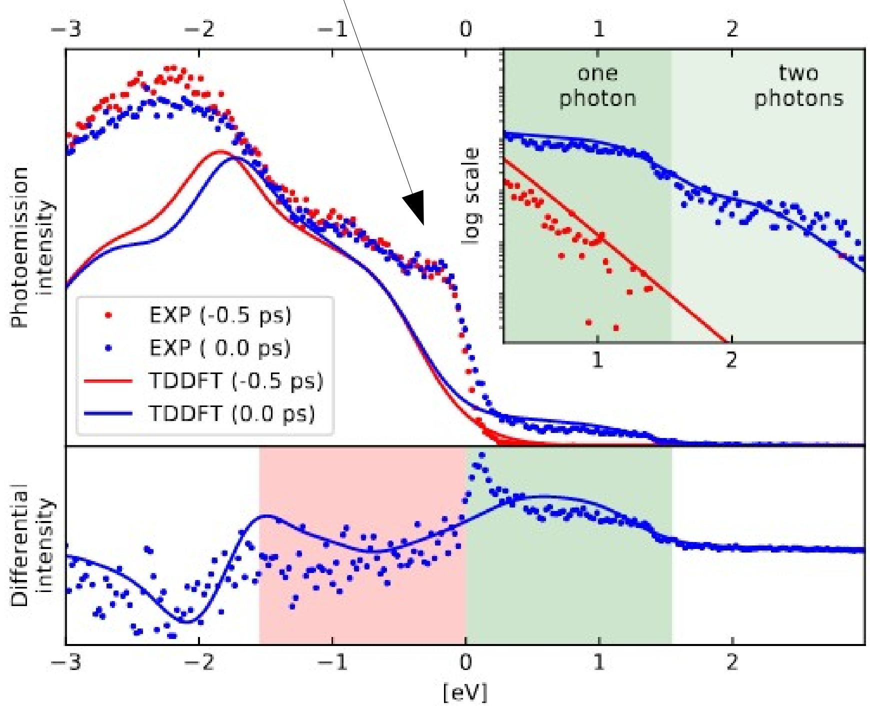


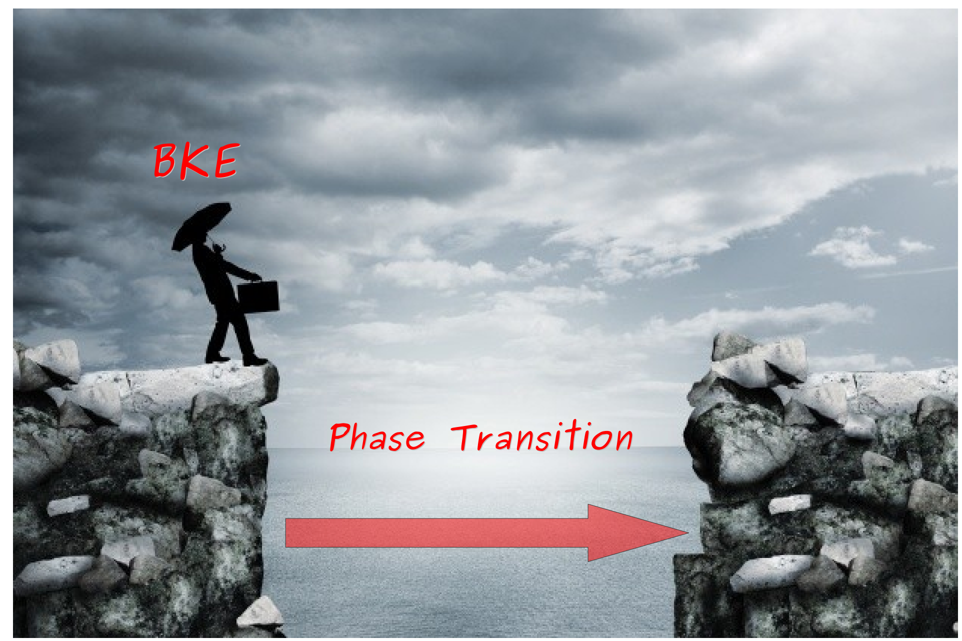

The GAP!

# Magnetization dynamics in FeRh: Experiment and First principles simulations

[Federico Pressacco, Davide Sangalli, Andrea Marini, FaustoSirotti, Matteo Gatti, Steinn Ymir Agustsson, in preparation]



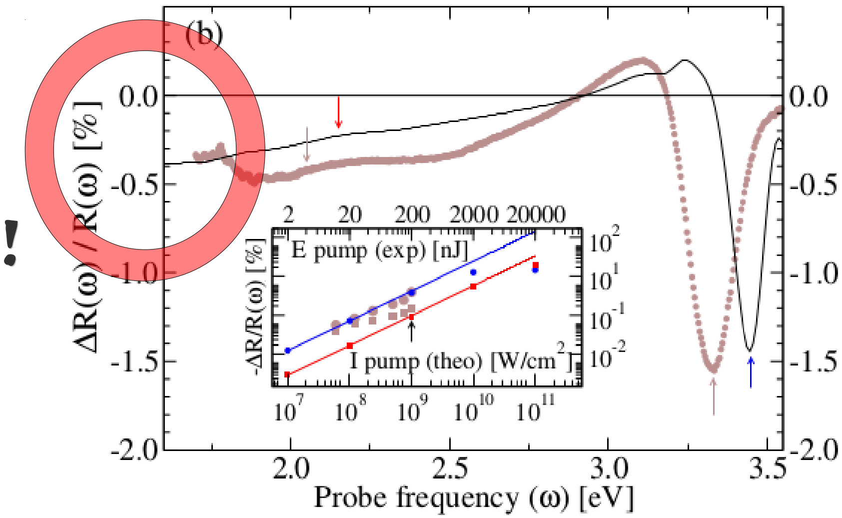




MACRO

micro

0.1%!!!!




Transient reflectivity Silicon

(D. Sangalli, S. Dal Conte, C. Manzoni, G. Cerullo and A. Marini, PRB 93, 195205 (2016))


Observables

The **Users** Perspective

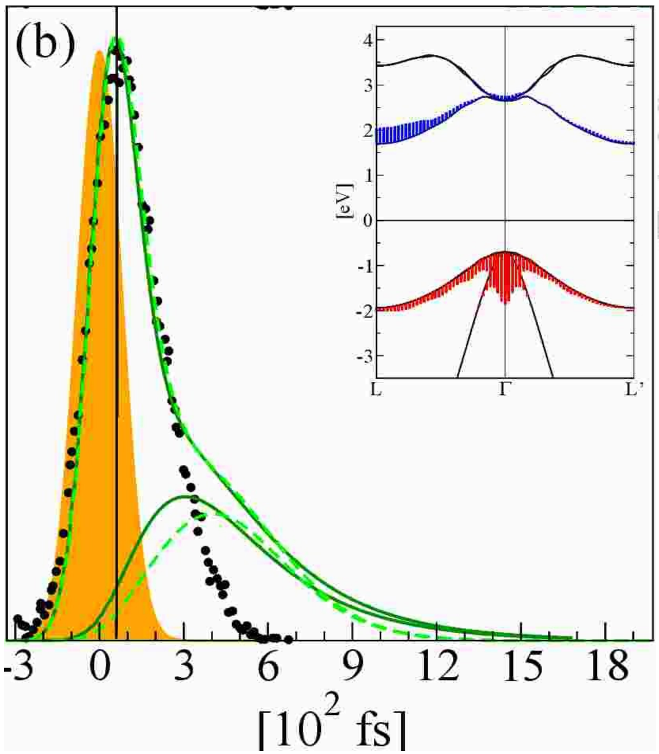
An approach based on **Large Scale** Computational Physics

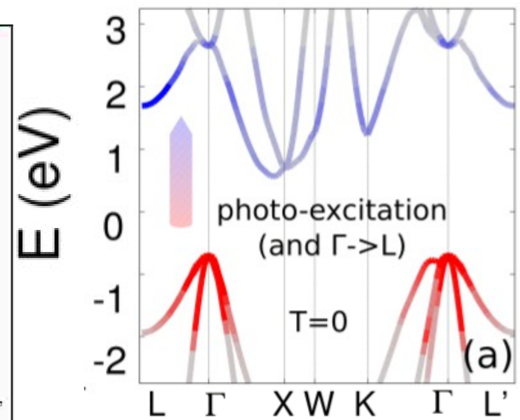
"Dream" NEGF and GKBA+approximations

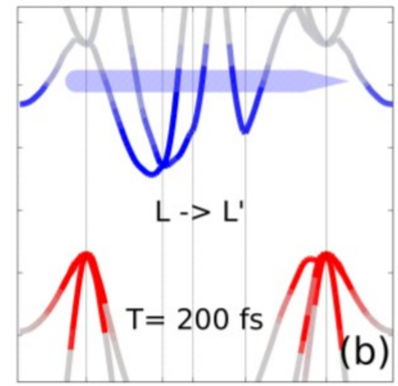
Ab-Initio NEGF (AiNEGF) at work. Pros.

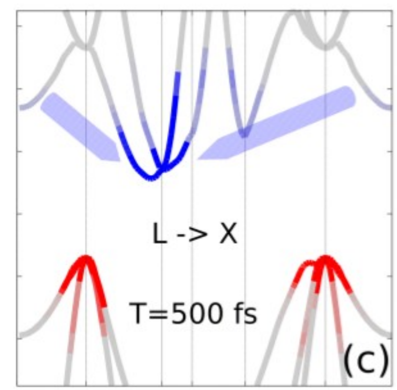
Open issues (=cons)

# EQ-MBPT extended to NEQ: wrong decay rates



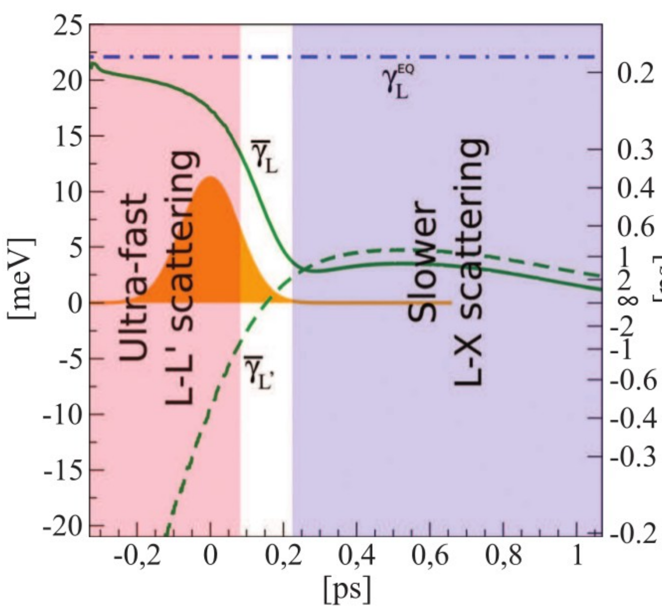



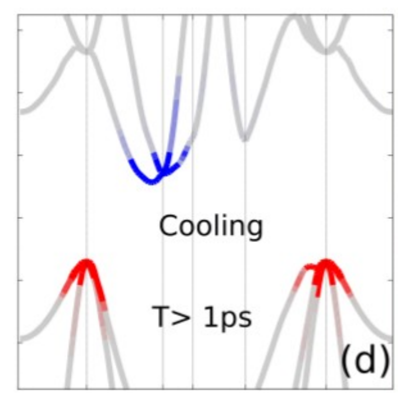

$$f(t) \neq f(0)\, e^{-\gamma_{eq} t}$$

[D. Sangalli, AM, EPL, 110 (2015) 47004]




EQ people approaching NEQ use EQ concepts! Erroneous definition of decay rates.

We do need reliable and quantitatively accurate tools OUT-of-EQ

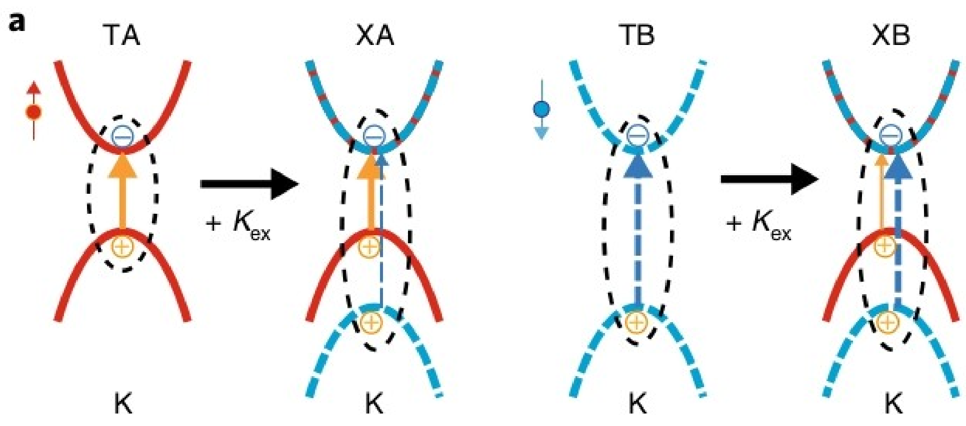


# EQ-MBPT extended to NEQ: e/h Exchange

nature physics LETTERS
https://doi.org/10.1038/s41567-018-0362-y

## Exchange-driven intravalley mixing of excitons in monolayer transition metal dichalcogenides

Liang Guo[1,2,7], Meng Wu[3,4,7], Ting Cao[3,4,7], Daniele M. Monahan[1,2,7], Yi-Hsien Lee[5], Steven G. Louie[3,4*] and Graham R. Fleming[1,2,6*]




EQ people approaching NEQ
use EQ concepts!
Excitons=BSE. Is it correct?

PHYSICAL REVIEW B VOLUME 47, NUMBER 23 15 JUNE 1993-I

## Exciton spin dynamics in quantum wells

M. Z. Maialle, E. A. de Andrada e Silva,* and L. J. Sham
*Department of Physics, University of California, San Diego, La Jolla, California 92093-0319*
(Received 21 December 1992; revised manuscript received 9 February 1993)

# Beyond the one-body density matrix?

PHYSICAL REVIEW B VOLUME 47, NUMBER 23 15 JUNE 1993-I

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$$\frac{d}{dt}\begin{pmatrix} N_{+2} \\ N_{+1} \\ N_{-1} \\ N_{-2} \end{pmatrix} = \begin{pmatrix} -(W_e^+ + W_h^+) & W_e^- & W_h^- & 0 \\ W_e^+ & -(\frac{1}{\tau_R} + W_{\rm ex} + W_e^- + W_h^-) & W_{\rm ex} & W_h^+ \\ W_h^+ & W_{\rm ex} & -(\frac{1}{\tau_R} + W_{\rm ex} + W_e^- + W_h^-) & W_e^+ \\ 0 & W_h^- & W_e^- & -(W_h^+ + W_e^+) \end{pmatrix} \begin{pmatrix} N_{+2} \\ N_{+1} \\ N_{-1} \\ N_{-2} \end{pmatrix},$$

"Real" Excitons Dynamics?

Eur. Phys. J. B (2018) 91: 171
https://doi.org/10.1140/epjb/e2018-90126-5

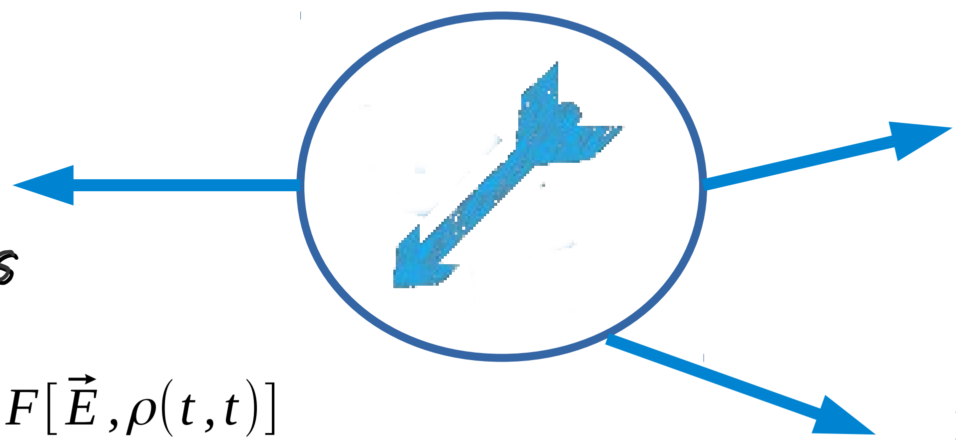
THE EUROPEAN PHYSICAL JOURNAL B

Regular Article

## An ab-initio approach to describe coherent and non-coherent exciton dynamics*

Davide Sangalli[1,a], Enrico Perfetto[1], Gianluca Stefanucci[2,3], and Andrea Marini[1]

# Laser Coherence: EOM, dissipation, dynamics ?



**Atomic displacements**

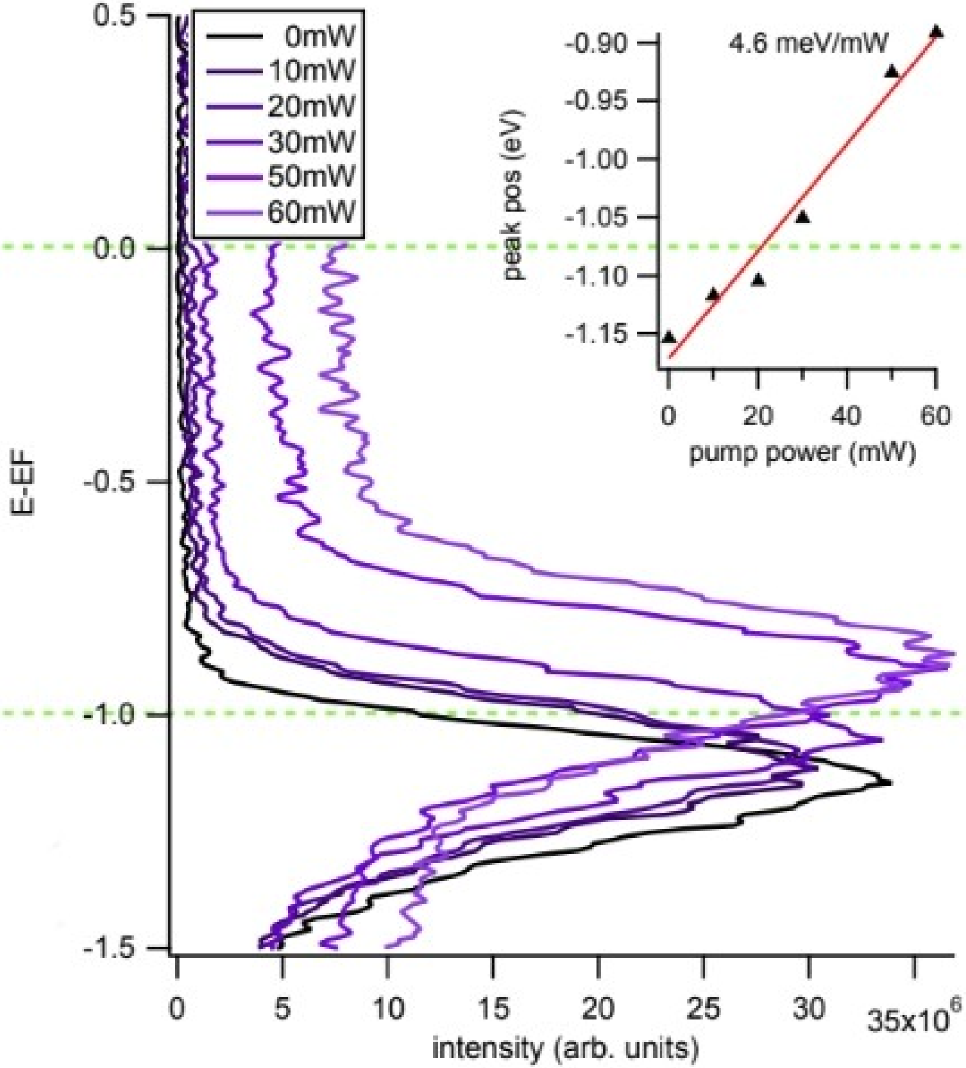
**Magnetization**

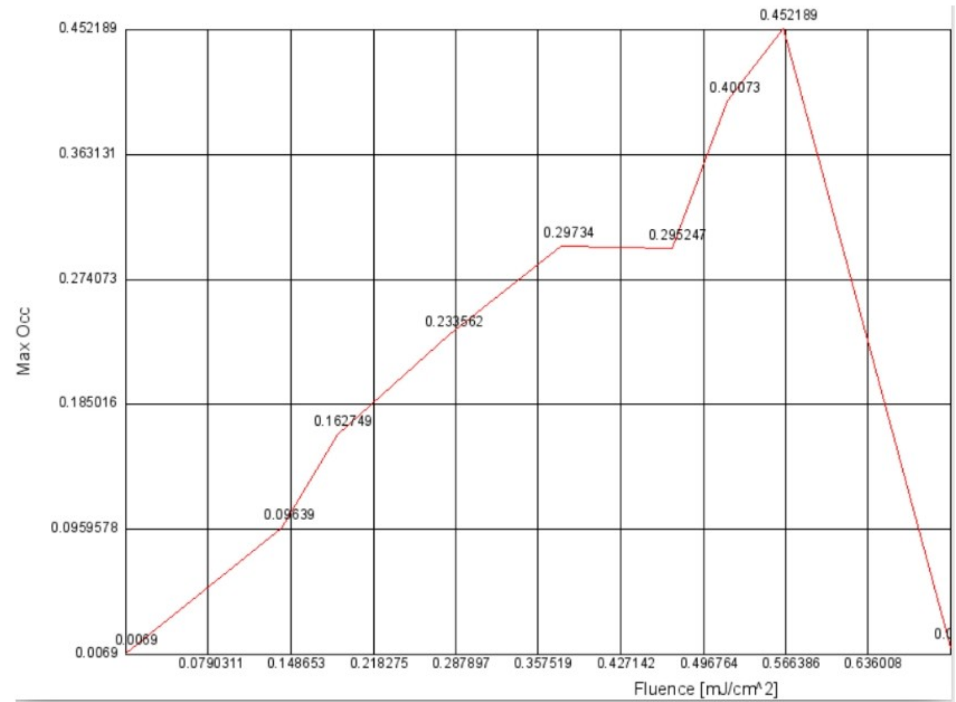
**Polarization**

$$\left(\frac{d^2}{dt^2}+\Omega^2\right)\langle b(t)+b^+(t)\rangle \sim F[\vec{E},\rho(t,t)]$$

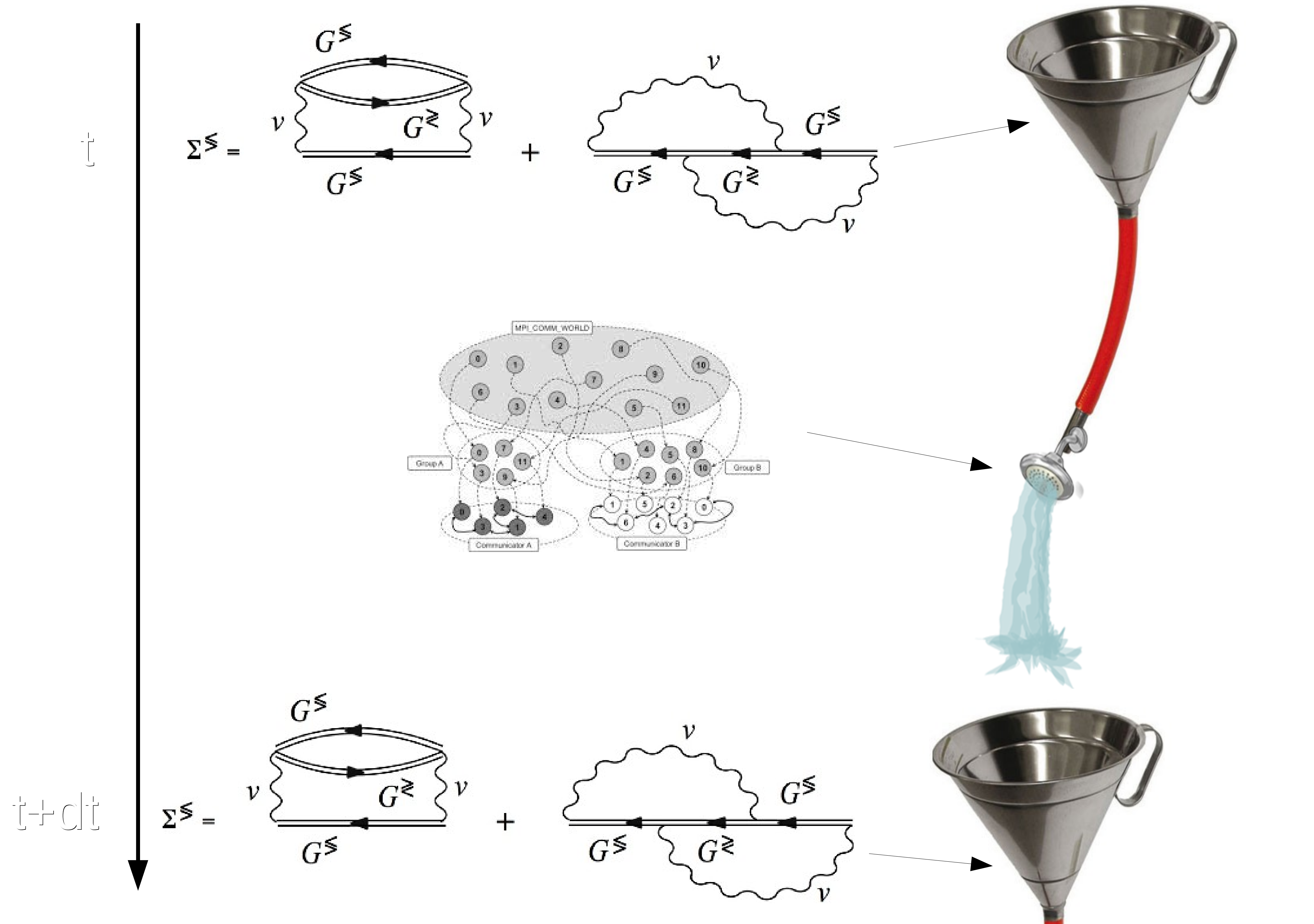
[AM, Y. Pavlyukh, PRB 98 075105]

# Transients. Example: Pump field effects.



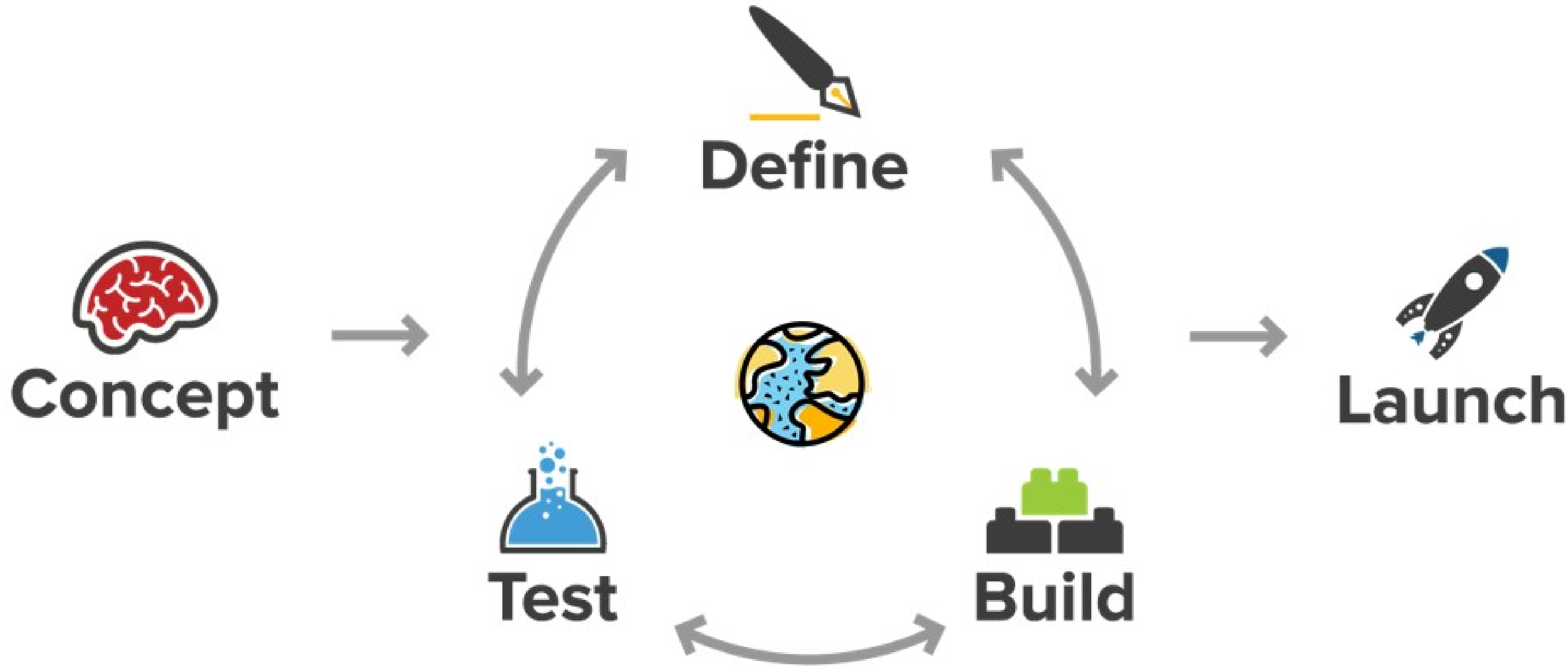



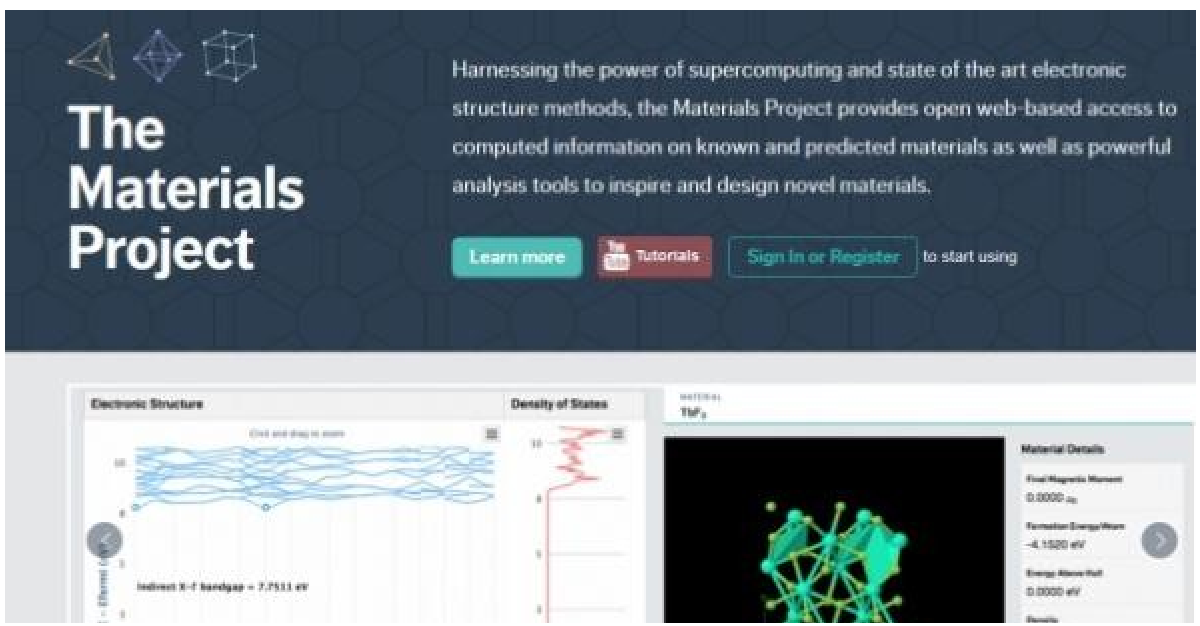
# Time evolution: intrinsically serial!



# HUGE request of theoretical support!

# Test & Validate



FLASHit

# Division of Ultrafast Processes in Materials CNR-ISM, Montelibretti, Italy

G. Stefanucci (Phys.Dept. Rome)

Istituto di Struttura della Materia

A. Marini

D. Sangalli (postDoc)

E. Perfetto (postDoc)


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