CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE

ÉCOLE POLYTECHNIQUE

Femtosecond probed Excited States in Condensed Matter:

What can the theory calculate and predict from first principles?

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COMMISSARIAT À L'ÉNERGIE ATOMIQUE

nanophase

Outline

- LSI Theory Group presentation
- Lifetime of Excitations:
  - Theory
  - Numerical Calculation
  - Preliminary Results for Silicon and Diamond
- Comparison with the Experiment? Possible future input coming from *Slicing sur Soleil*

LSI Theory Group

Permanents:	Olivier Hardouin-Duparc	CNRS	
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	Jean-Claude Toledano	Ecole	
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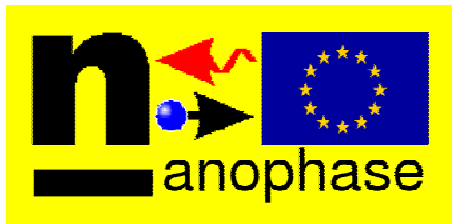
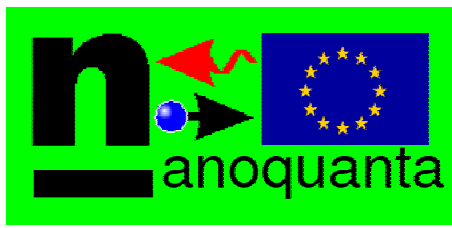
PhD Students:	Fabien Bruneval	CEA	
	Virginie Quequet	CEA	
Visitors:	Jorge Serrano	EU Nanophase	
	Wojtec Welnic	EU Marie Curie TS	
	Hans-Christian Weissker	EU Marie Curie TS	
SystemManager:	Andrea Cucca	CNRS	

EU Links

- 2000-2004: EU RTN (Research Training Network) **NANOPHASE**



- 2004-2008: EU NoE (**Network of Excellence**) **NANOQUANTA**

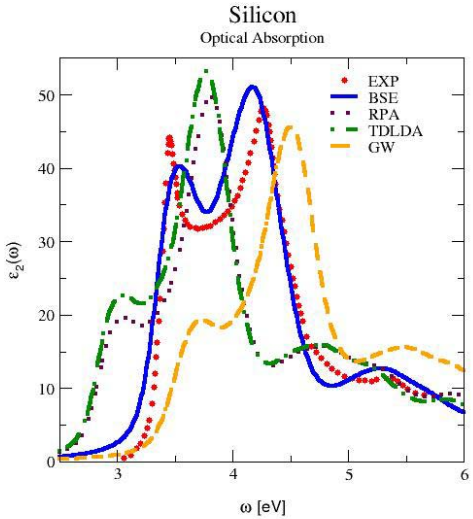


- EU Marie Curie Training Site

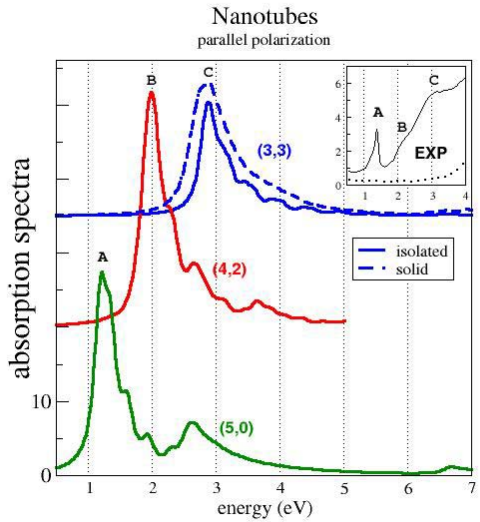
We

- Develop **Analytically** the **Theory** and the **Approximations**;
- Implement the Theory in **Algorithms** and in **Codes** (sometime GNU/GPL Open Sources);
- Calculate **Numerically** a wide range of Properties for a wide range of Systems, both **Empirically** and **Ab Initio**;
- Compare with the **Experiment** (when available).

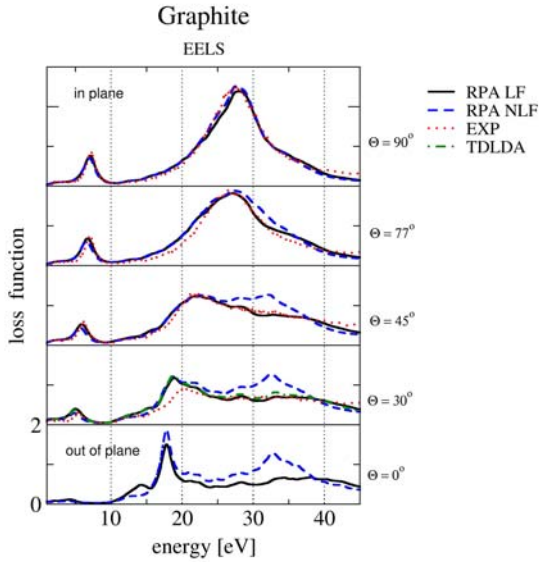
Some Results



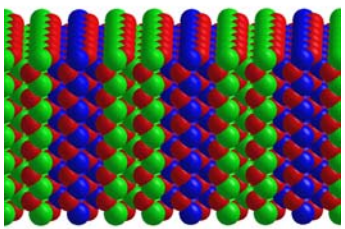
Bethe-Salpeter Equation for Optical Properties (Si)



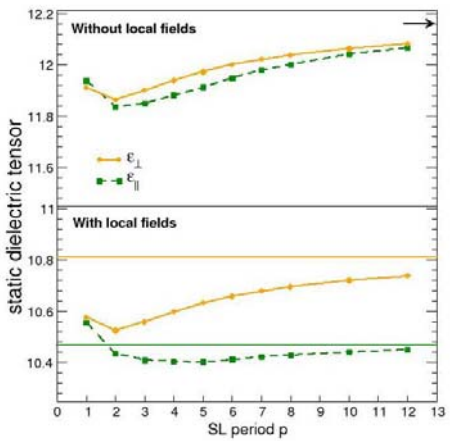
Nanotubes: Optical Properties

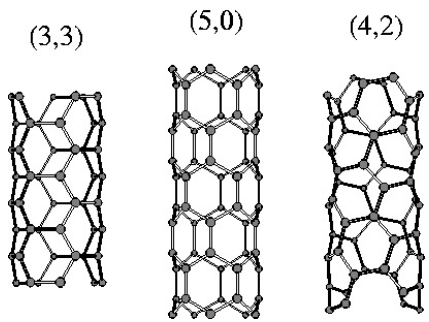


TDDFT for EELS (Graphite)

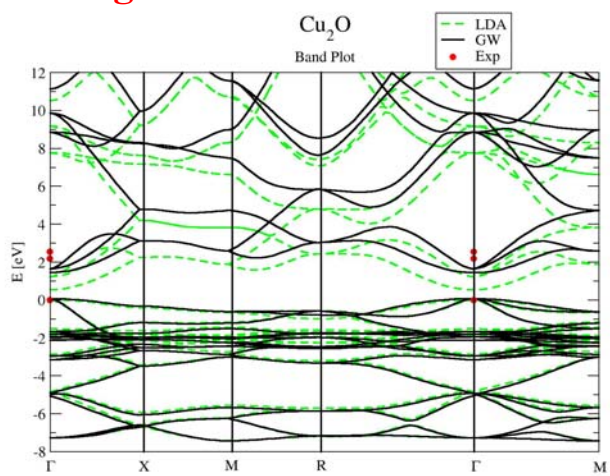


SuperLattices GaAs/AlAs





GW for Band Structures (Cu_2O) vs Angle Resolved Photoemission



http://theory.lsi.polytechnique.fr

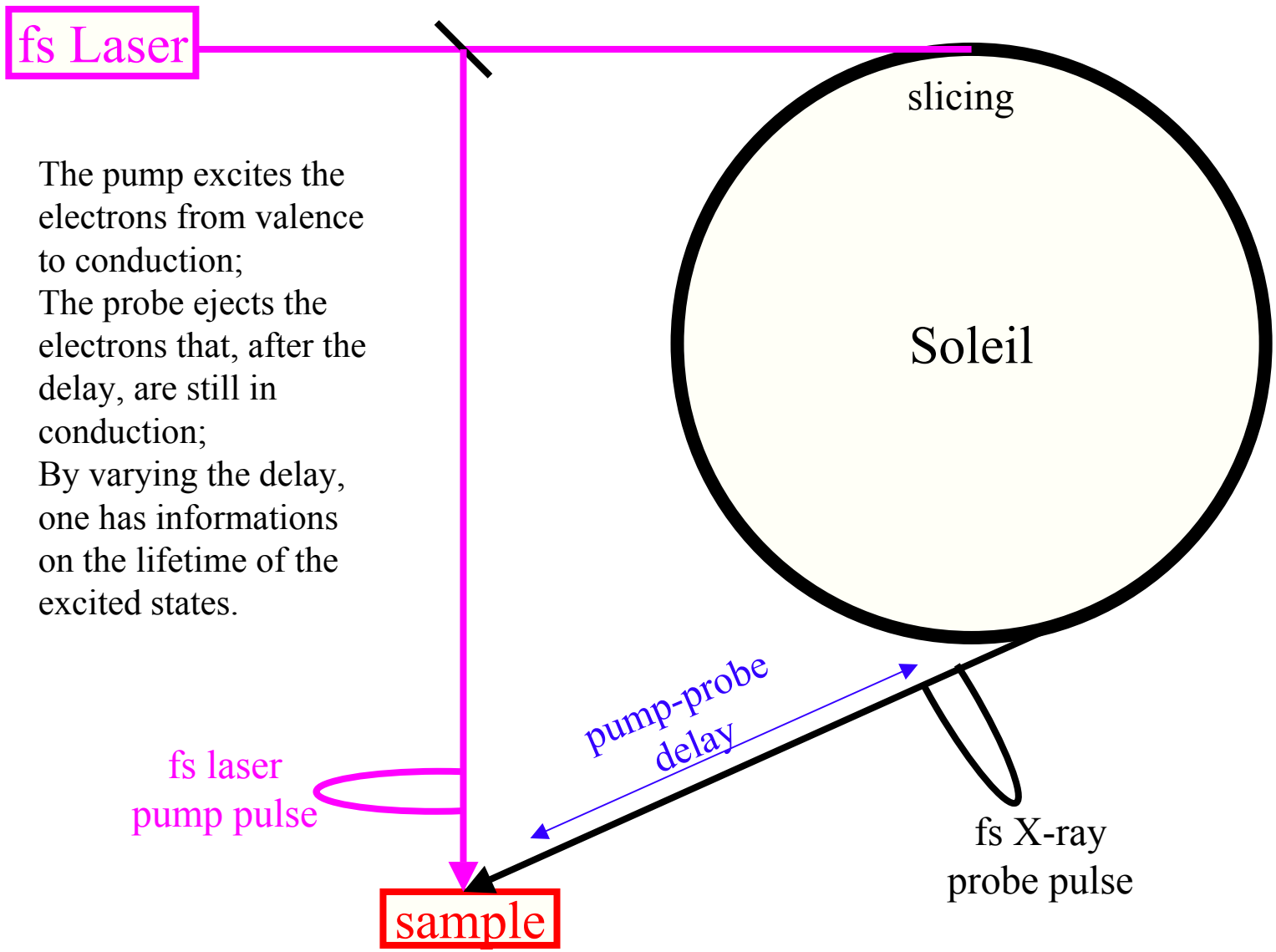
opening to

Femtosecond Probed Physics

- The **femtosecond** is the time order of excitations in matter.
- At this time resolution, we can follow the excitation in its evolution.
- Observable: **Lifetime of the Excitation**.

Pump & Probe Setup

1. The pump excites the electrons from valence to conduction;
2. The probe ejects the electrons that, after the delay, are still in conduction;
3. By varying the delay, one has informations on the lifetime of the excited states.



Quasiparticles and Lifetimes

Self-Energy

$$H^{(0)}(r)\phi_i(r) + \int dr'\, \Sigma(r, r'; \epsilon_i)\phi_i(r') = \epsilon_i \phi_i(r)$$

Quasiparticle Equation

$$\epsilon_i = E_i - i\frac{\Gamma_i}{2}$$

Quasiparticle Energy

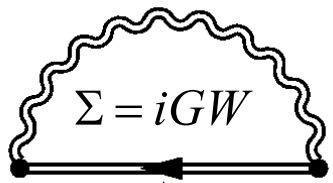
$$\tau_i^{-1} = \Gamma_i = -2\Im(\epsilon_i)$$

Lifetime

GW Approximation for the Self-Energy

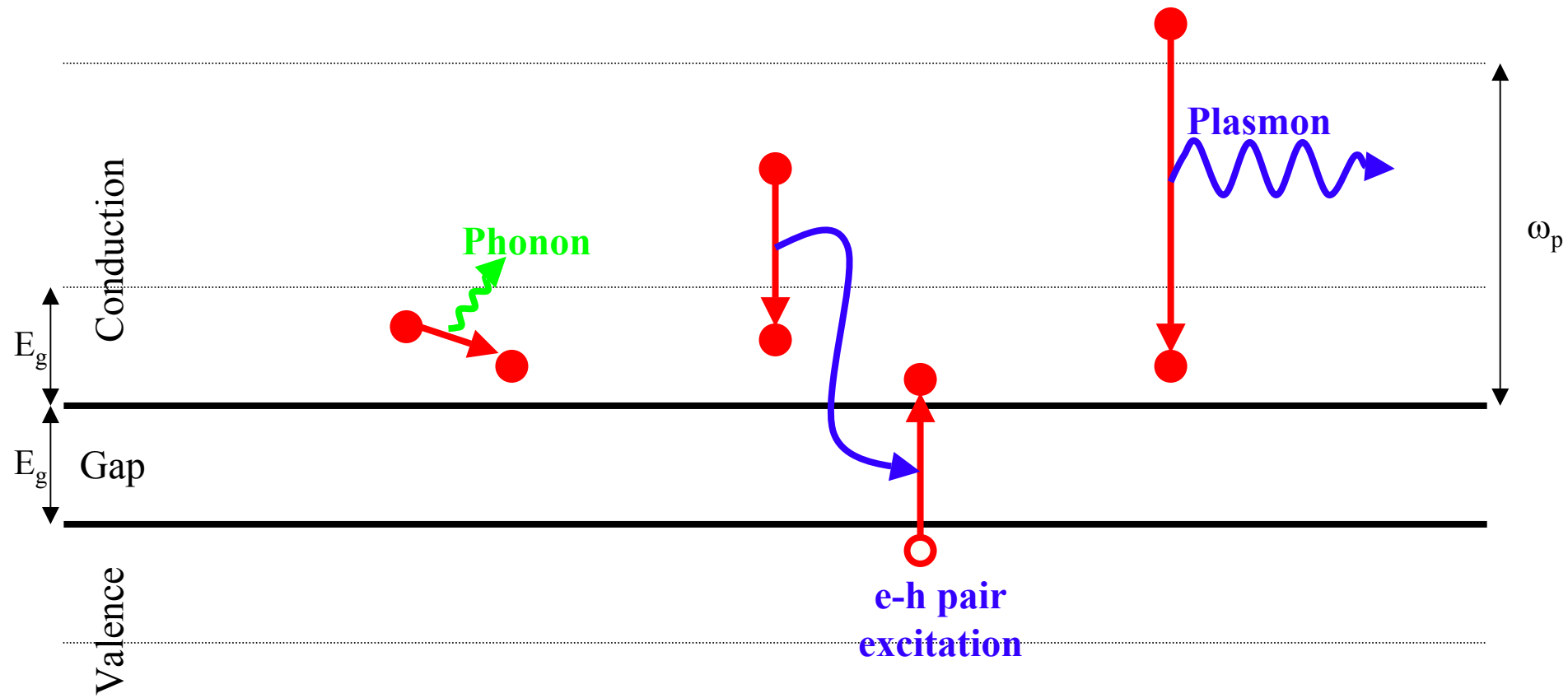
Dynamical Screened Interaction W

$$\Sigma^{\mathrm{GW}}(x_1, x_2) = iG(x_1, x_2)W(x_1^+, x_2)$$



Green Function or Electron Propagator G

Some Decay Channels



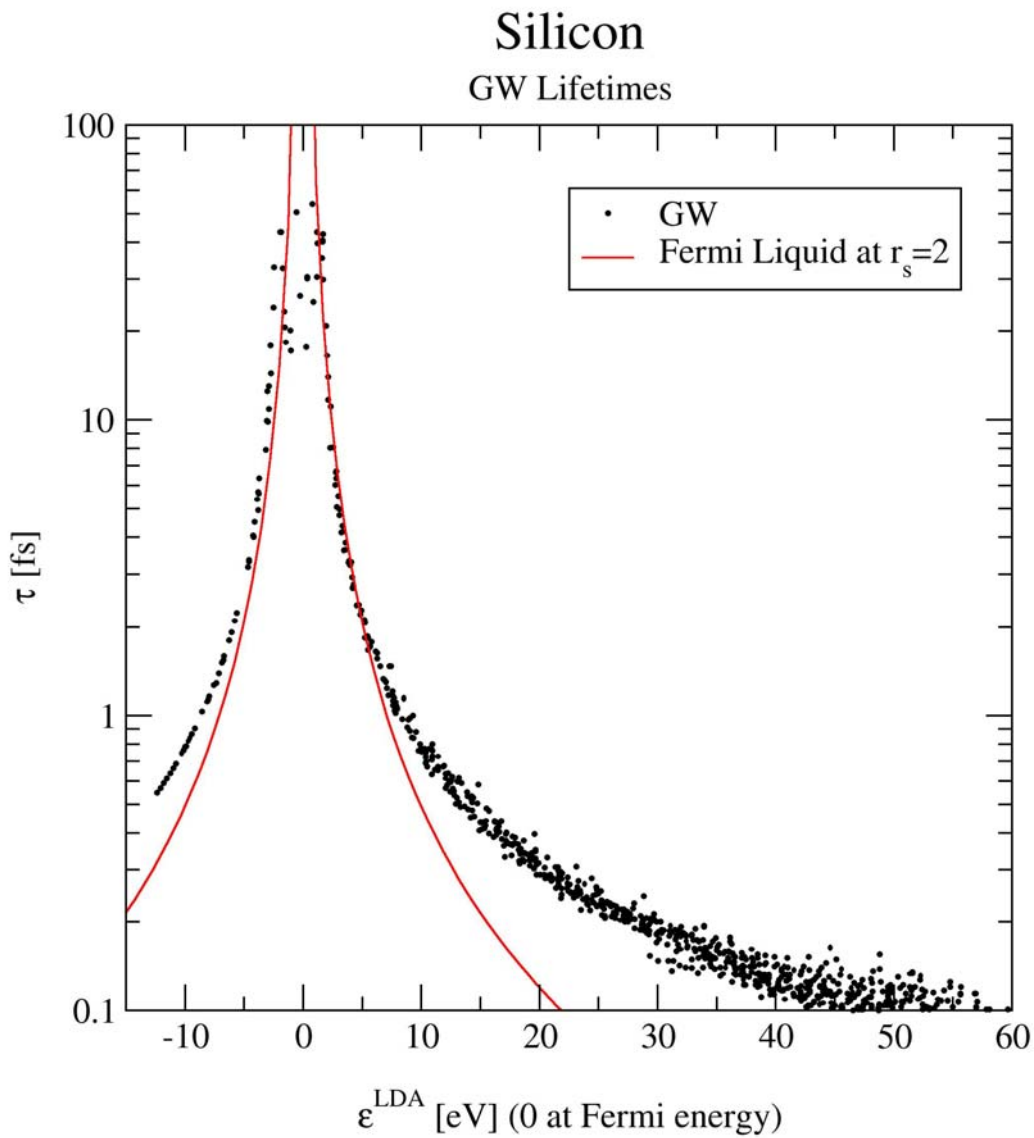
Electronic Self-Energy

- Neglected Decay Channels:
  - **Phonons**
  - **Defects**
  - **Impurities**

These can be disentangled
from the Experiment by studying at
different Temperatures (around T_D),
different Defect Density
and different Purity samples.

- Accounted Decay Channels:
  - **Electron-Hole Pairs**
  - **Plasmons**
  - **Auger processes**

Results: Lifetimes in Silicon

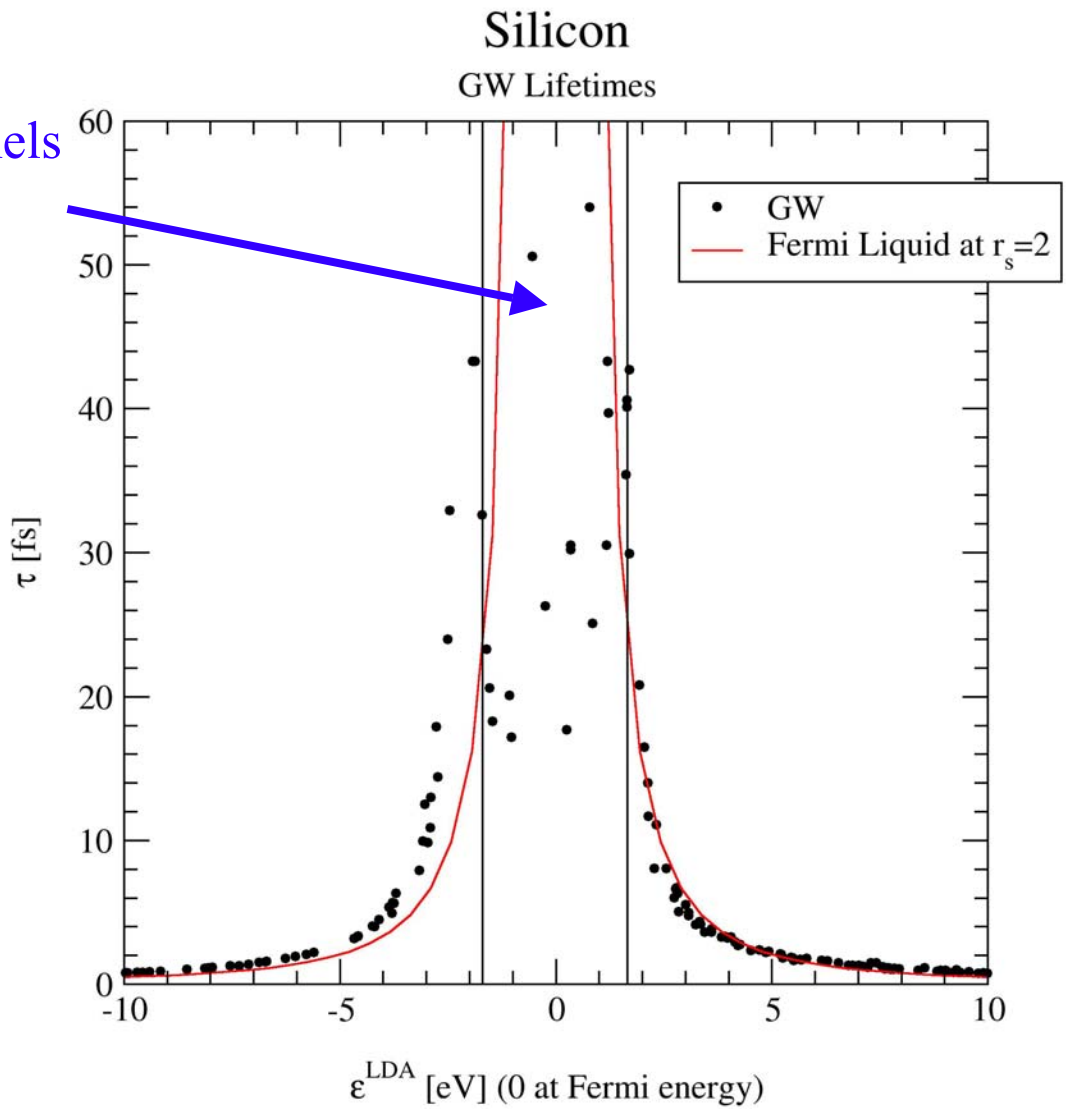


Fermi Liquid Lifetime [Quinn and Ferrel 1958]:

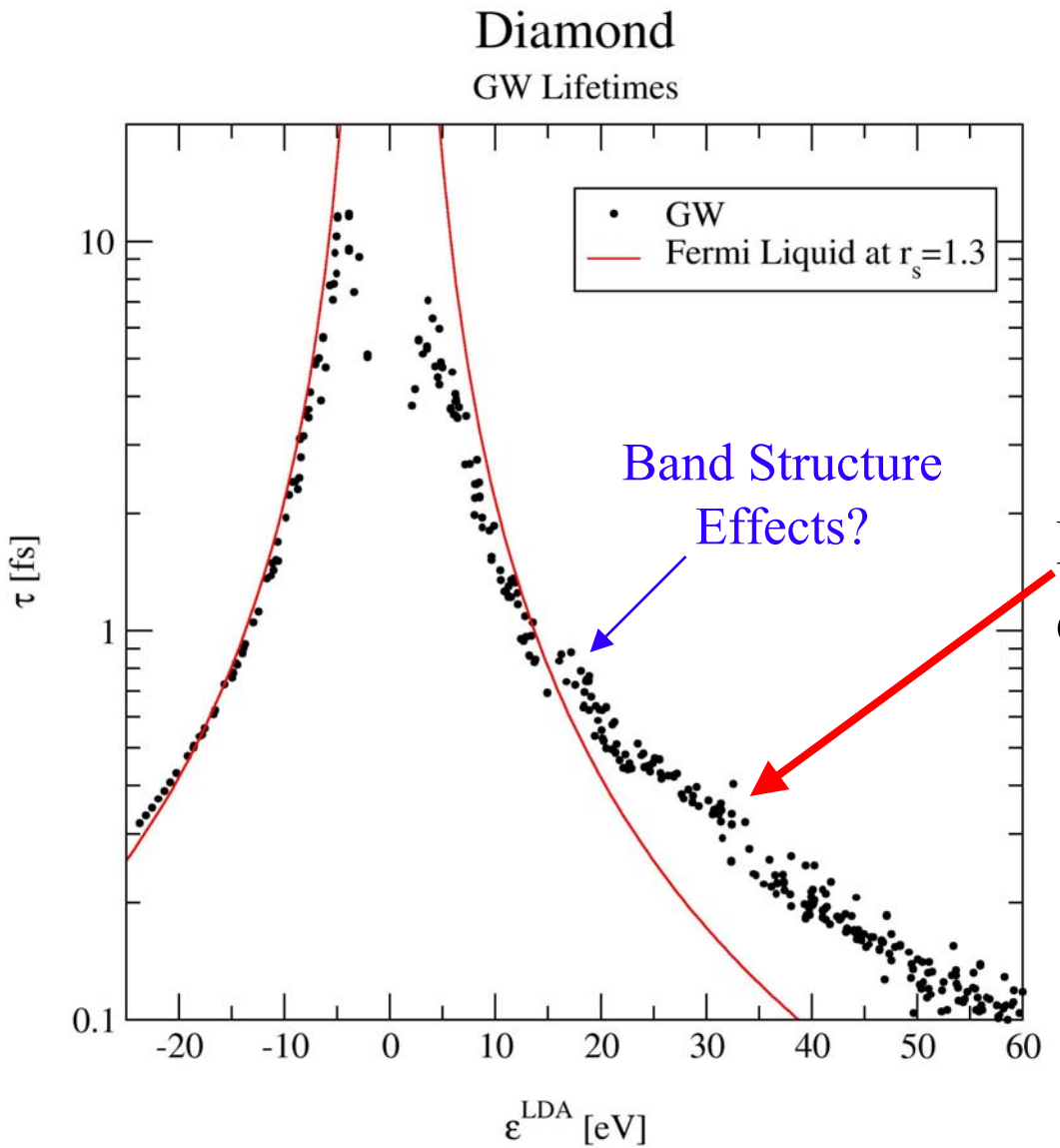
$$\tau_i \propto r_s^{-5/2} \left(\varepsilon_i - \varepsilon_F\right)^{-2}$$

Lifetimes in Silicon

Phonon Channels only region



Lifetimes in Diamond



Comparison with future Slicing sur Soleil Data and Conclusions

- With a 20fs resolution, the measure of the lifetime of states close to the Fermi level, becomes accessible. And a comparison with the Theory is possible. Especially in metals.
- The use of X-rays is preferable in order to remove unwished dependence of the results on the probe energy.
- Perspectives from the Theory point of view: accounting of the Phonon contributions.
- Perspective from the Experimental point of view: hope for further improvement of the Slicing sur Soleil's Laser time-resolution (10fs? 5fs?).

Numerical Evaluation of the Self-Energy

$$\Sigma^{GW}(\mathbf{r},\mathbf{r}',\omega) = \frac{i}{2\pi}\int d\omega' e^{-i\delta\omega'} G(\mathbf{r},\mathbf{r}',\omega-\omega')W(\mathbf{r},\mathbf{r}',\omega')$$

Fourier Transform of the GW Self-Energy

Three methods:

1. Brute force ω' **Real Axis Integration**
2. ω' Imag Axis Integration, **Analytical Continuation** from Imag ω to ε_i
3. **Contour Deformation** ω' Integration and direct calculation at $\omega = \varepsilon_i$