Time-dependent density functional theory

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OUTLINE

- **Basics of TDDFT**

- **Linear response regime:**
  -- Calculation of photo-absorption spectra

- **Beyond the linear regime:**
  -- TD transport
  -- TD Electron Localisation Function

What do we want to describe?

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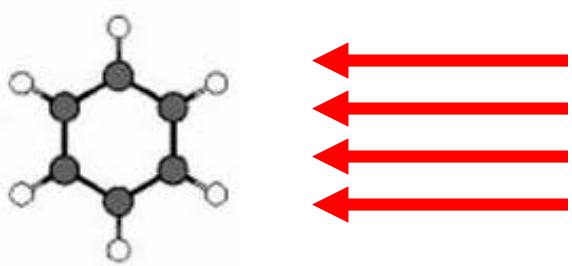
System in laser field:
Generic situation



$$\hat{H}(t) = \hat{T}_e + \hat{W}_{ee} + \sum_{j,\alpha} - \frac{Z_\alpha e^2}{|r_j - R_\alpha|} + \vec{r}_j \cdot \vec{E}(t) \cdot \sin \omega t$$

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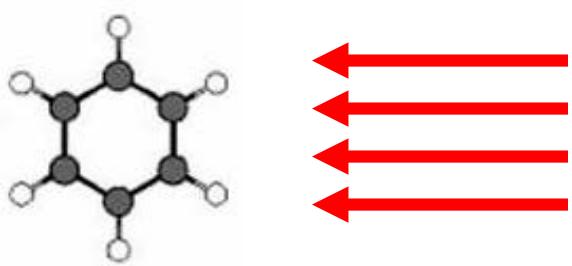

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Calculate electron spectra, ion spectra, photon spectra (e.g. HHG)

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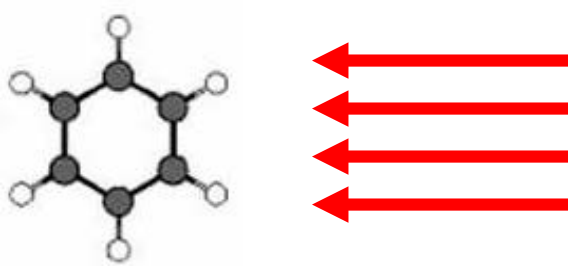

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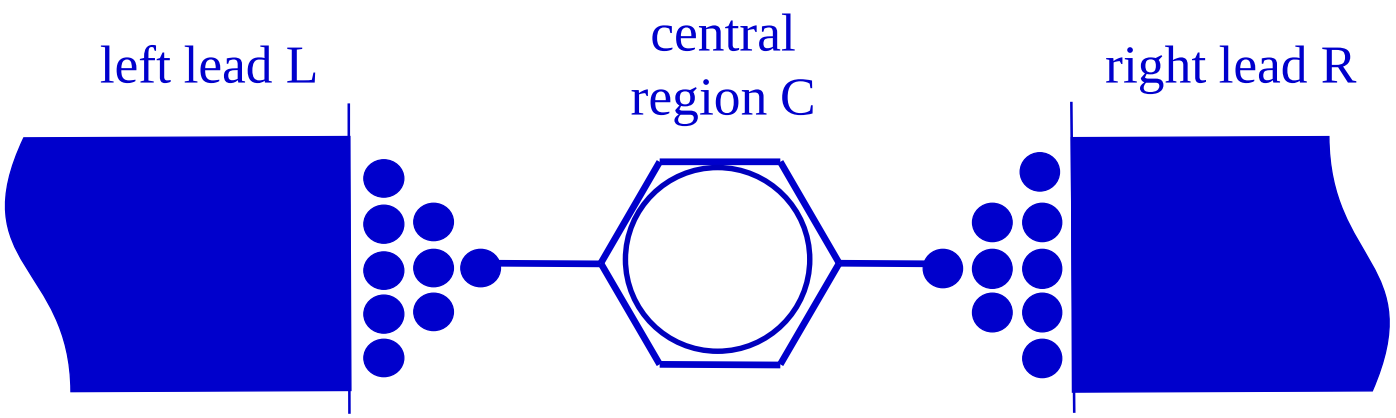
Electronic transport: Generic situation



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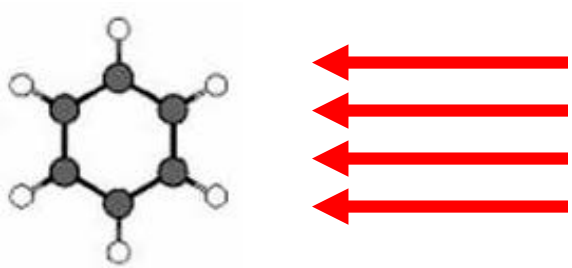

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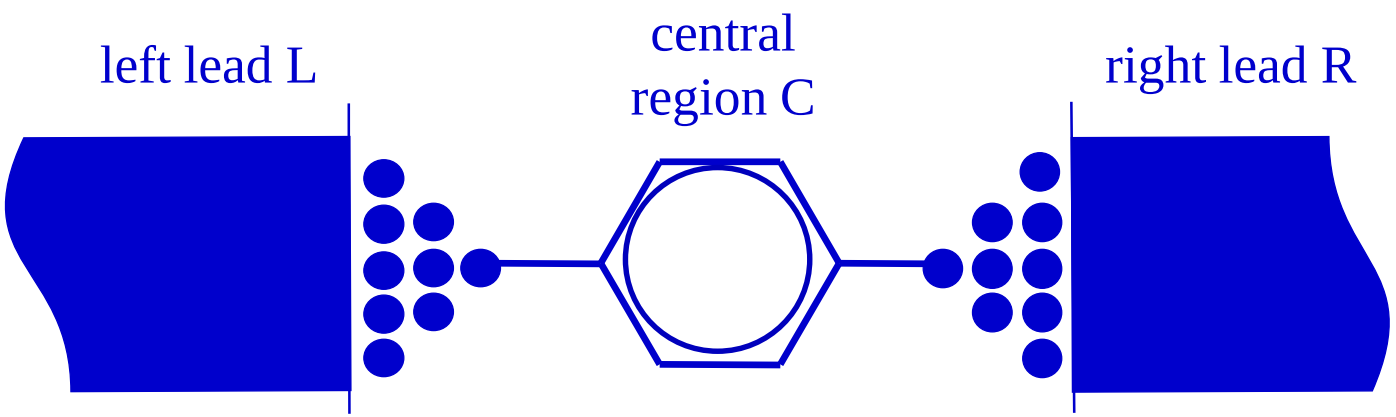


Bias U(t) between L and R is turned on: Calculate U-I characteristics

$$\hat{H}(t) = \hat{T}_e + \hat{W}_{ee} + \sum_{j,\alpha} - \frac{Z_\alpha e^2}{|r_j - R_\alpha|} + \vec{r}_j \cdot \vec{E}(t) \cdot \sin \omega t$$

Strong laser ($v_{laser}(t) \geq v_{en}$) :

Non-perturbative solution of full TDSE required

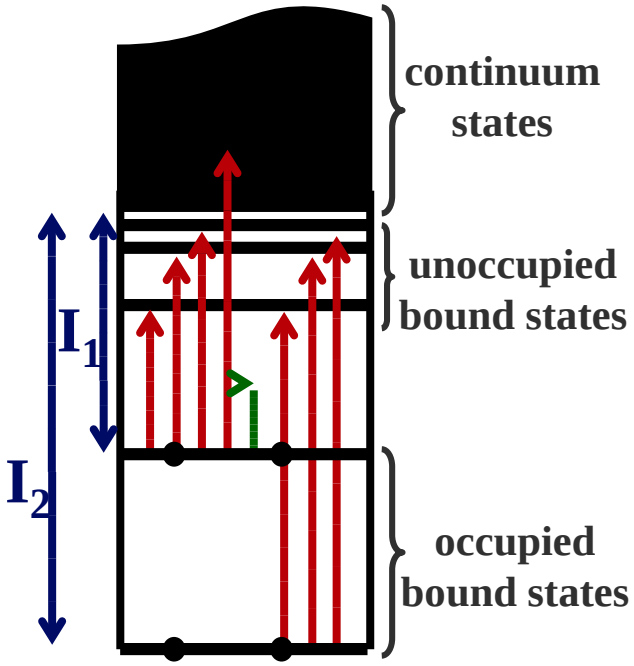
Weak laser ($v_{laser}(t) << v_{en}$) :

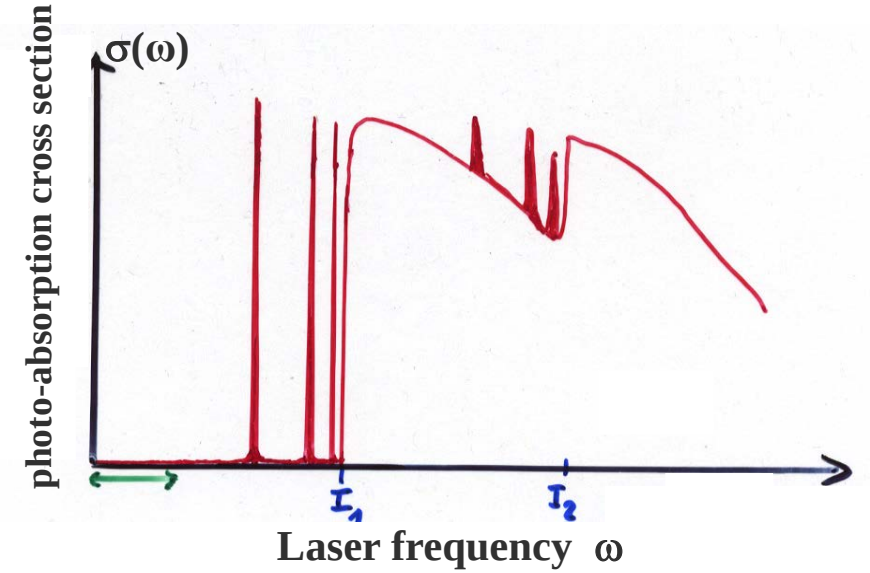
Calculate 1. Linear density response $\rho_1(\vec{r}\, t)$

2. Dynamical polarizability $\alpha(\omega) = -\frac{e}{E}\int z\, \rho_1(\vec{r}, \omega) d^3r$

3. Photo-absorption cross section $\sigma(\omega) = -\frac{4\pi\omega}{c} \mathrm{Im}\, \alpha$

Photo-absorption in weak lasers





Time-dependent density-functional formalism

(E. Runge, E.K.U.G., PRL 52, 997 (1984))

Basic 1-1 correspondence:

$$v(rt) \xleftrightarrow{1\text{-}1} \rho(rt)$$

The time-dependent density determines uniquely the time-dependent external potential and hence all physical observables for fixed initial state.

KS theorem:

The time-dependent density of the interacting system of interest can be calculated as density

$$\rho(rt) = \sum_{j=1}^{N} \left|\varphi_j(rt)\right|^2$$

of an auxiliary non-interacting (KS) system

$$i\hbar \frac{\partial}{\partial t}\varphi_j(rt) = \left(-\frac{\hbar^2\nabla^2}{2m} + v_S[\rho](rt)\right)\varphi_j(rt)$$

with the local potential

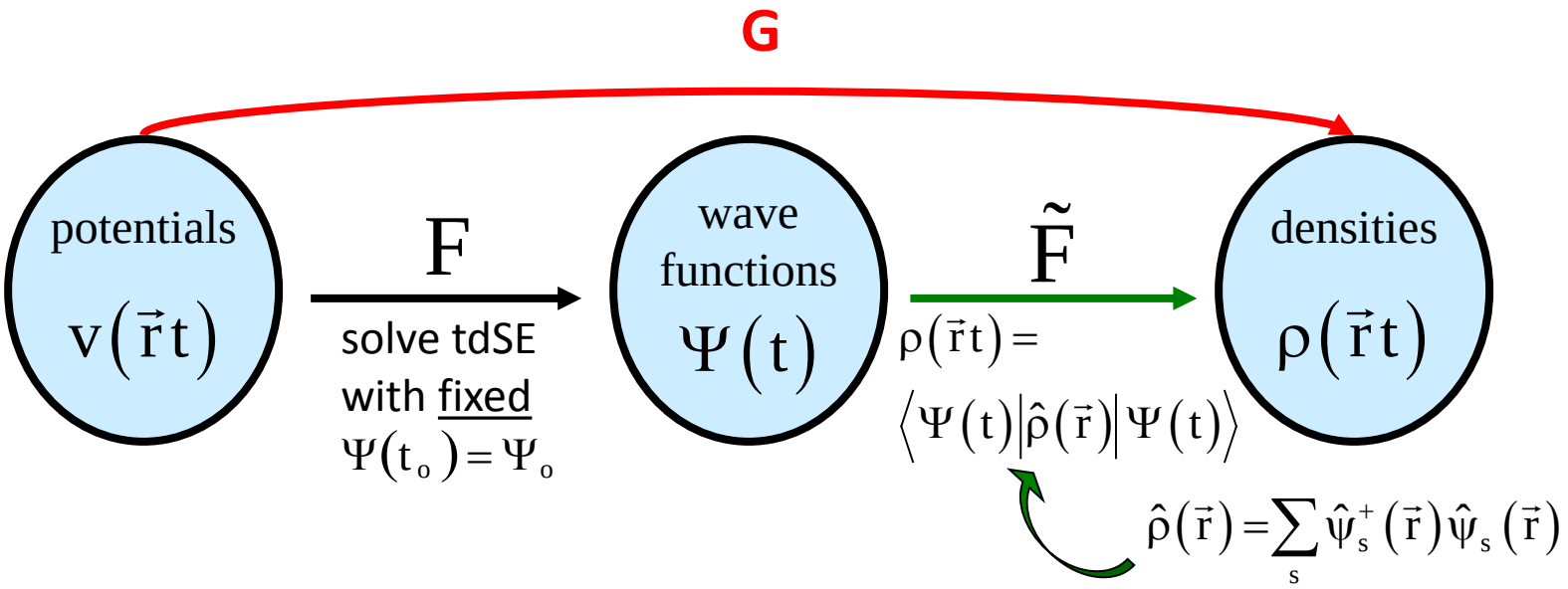
$$v_S\left[\rho(r't')\right](rt) = v(rt) + \int d^3r' \frac{\rho(r't)}{|r-r'|} + v_{xc}\left[\rho(r't')\right](rt)$$

The TDKS equations follow (like in the static case) from:

i. **the basic 1-1 mapping and**
ii. **the TD V-representability theorem (R. van Leeuwen, PRL 82, 3863 (1999)).**

Proof of basic 1-1 correspondence between $v(\vec{r}\,t)$ and $\rho(\vec{r}\,t)$

define maps $F:\ v(\vec{r}\,t) \mapsto \Psi(t)$ $\tilde{F}:\ \Psi(t) \mapsto \rho(\vec{r}\,t)$



$G:\ v(\vec{r}\,t) \mapsto \rho(\vec{r}\,t)$

complete 1 - 1 correspondence not to be expected!

$$i\frac{\partial}{\partial t}\Psi(t)=\left(\hat{T}+\underline{\underline{\hat{V}(t)}}+\hat{W}\right)\Psi(t) \qquad \Psi(t_o)=\Psi_o$$

$$i\frac{\partial}{\partial t}\Psi'(t)=\left(\hat{T}+\underline{\underline{\hat{V}'(t)}}+\hat{W}\right)\Psi'(t) \qquad \Psi'(t_o)=\Psi_o$$

$$\hat{V}'(t)=\hat{V}(t)+C(t) \;\Leftrightarrow\; \Psi'(t)=e^{-i\alpha(t)}\Psi(t)$$

"no operator"

with $\boxed{\dot{\alpha}(t)=C(t)}$

$$\Rightarrow \underline{\underline{\rho'(\vec{r}t)=\rho(\vec{r}t)}}$$

i.e. $\left\{\hat{V}(t)+C(t)\right\} \;\rightarrow\; \rho(\vec{r}t)$

If G is invertible up to within time-dependent function C(t)

$\Rightarrow \Psi = FG^{-1}\rho$ fixed up to within time-dependent phase

i.e. $\Psi = e^{-i\alpha(t)}\Psi[\rho]$

For any observable $\hat{O}$

$$\langle \Psi | \hat{O} | \Psi \rangle = \langle \Psi[\rho] | \hat{O} | \Psi[\rho] \rangle = O[\rho]$$

is functional of the density

THEOREM (time-dependent analogue of Hohenberg-Kohn theorem)

The map

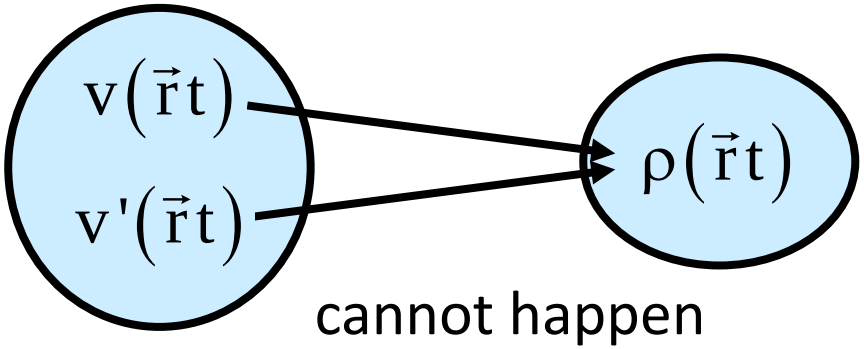
$$G: \; v(\vec{r}t) \mapsto \rho(\vec{r}t)$$

defined for all single-particle potentials $v(\vec{r}t)$ which can be expanded into a Taylor series with respect to the time coordinate around t_o

is invertible up to within an additive merely time-dependent function in the potential.

Proof:

to be shown:



cannot happen

i.e. $\hat{v}(\vec{r}t) \neq \hat{v}'(\vec{r}t) + c(t) \overset{!}{\Rightarrow} \rho(\vec{r}t) \neq \rho'(\vec{r}t)$

potential expandable into Taylor series

$$\exists\, k \geq 0 \quad : \quad \frac{\partial^k}{\partial t^k}\left[v(\vec{r}t) - v'(\vec{r}t)\right]_{t=t_o} \neq \text{constant}$$

step 1

$$\vec{j}(\vec{r}t) \neq \vec{j}'(\vec{r}t)$$

step 2

$$\rho(\vec{r}t) \neq \rho'(\vec{r}t)$$

Step 1: Current densities

$$\vec{j}(\vec{r}t) = \left\langle \Psi(t) \middle| \hat{\vec{j}}(\vec{r}) \middle| \Psi(t) \right\rangle$$

with $$\hat{\vec{j}}(\vec{r}) = -\frac{1}{2i}\sum_s \left(\left[\vec{\nabla}\hat{\psi}_s^+(\vec{r}) \right] \hat{\psi}_s(\vec{r}) - \hat{\psi}_s^+(\vec{r}) \left[\vec{\nabla}\hat{\psi}_s(\vec{r}) \right] \right)$$

Use equation of motion:

$$i\frac{\partial}{\partial t}\left\langle \Psi(t) \middle| \hat{O}(t) \middle| \Psi(t) \right\rangle = \left\langle \Psi(t) \middle| i\frac{\partial}{\partial t}\hat{O}(t) + \left[\hat{O}(t), \hat{H}(t) \right] \middle| \Psi(t) \right\rangle$$

$$\Rightarrow \quad i\frac{\partial}{\partial t}\vec{j}(\vec{r}t) = \left\langle \Psi(t) \middle| \left[\hat{\vec{j}}(\vec{r}), \hat{H}(t) \right] \middle| \Psi(t) \right\rangle$$

$$i\frac{\partial}{\partial t}\vec{j}'(\vec{r}t) = \left\langle \Psi'(t) \middle| \left[\hat{\vec{j}}(\vec{r}), \hat{H}'(t) \right] \middle| \Psi'(t) \right\rangle$$

note: $$\vec{j}(\vec{r}t_o) = \vec{j}'(\vec{r}t_o) = \left\langle \Psi_o \middle| \hat{\vec{j}}(\vec{r}) \middle| \Psi_o \right\rangle \equiv \vec{j}_o(\vec{r})$$

$$\rho(\vec{r}t_o) = \rho'(\vec{r}t_o) = \left\langle \Psi_o \middle| \hat{\rho}(\vec{r}) \middle| \Psi_o \right\rangle \equiv \rho_o(\vec{r})$$

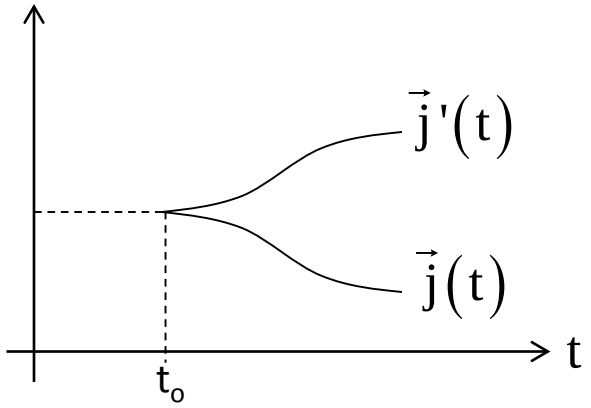
$$i\frac{\partial}{\partial t}\left[\vec{j}(\vec{r}t)-\vec{j}'(\vec{r}t)\right]_{t=t_o}=\left\langle\Psi_o\left|\left[\hat{\vec{j}}(\vec{r}),\hat{H}(t_o)-\hat{H}'(t_o)\right]\right|\Psi_o\right\rangle$$

$$=\left\langle\Psi_o\left|\left[\hat{\vec{j}}(\vec{r}),V(t_o)-V'(t_o)\right]\right|\Psi_o\right\rangle$$

$$=i\rho_o(\vec{r})\vec{\nabla}\left(v(\vec{r}t_o)-v'(\vec{r}t_o)\right)$$

if $\frac{\partial^k}{\partial t^k}\left[v(\vec{r}t)-v'(\vec{r}t)\right]_{t=t_o}\neq \text{constant}$ holds for k=0

then $i\frac{\partial}{\partial t}\left[\vec{j}(\vec{r}t)-\vec{j}'(\vec{r}t)\right]_{t=t_o}\neq 0$



$$\Rightarrow \quad \underline{\underline{\vec{j}(\vec{r}t)\neq\vec{j}'(\vec{r}t)}} \qquad \text{q.e.d.}$$

if $\frac{\partial^k}{\partial t^k}\left[v(\vec{r}t)-v'(\vec{r}t)\right]_{t=t_o} \neq \text{constant}$ holds for k>0

→ use equation of motion k+1 times:

$$\left(i\frac{\partial}{\partial t}\right)^2 \vec{j}(\vec{r}t) = i\frac{\partial}{\partial t}\left\langle \Psi(t)\left|\left[\hat{\vec{j}},\hat{H}(t)\right]\right|\Psi(t)\right\rangle$$

$$= \left\langle \Psi(t)\left| i\frac{\partial}{\partial t}\left[\hat{\vec{j}},\hat{H}(t)\right] + \left[\left[\vec{j},\hat{H}(t)\right],\hat{H}(t)\right]\right|\Psi(t)\right\rangle$$

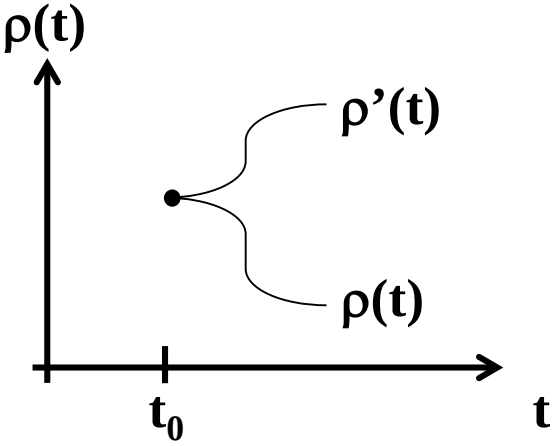
$$\left(i\frac{\partial}{\partial t}\right)^3 \vec{j}(\vec{r}t) = i\frac{\partial}{\partial t}\left\langle \Psi(t)\left| i\frac{\partial}{\partial t}\left[\vec{j},\hat{H}(t)\right] + \left[\left[\vec{j},H(t)\right],\hat{H}(t)\right]\right|\Psi(t)\right\rangle$$

$$\left(i\frac{\partial}{\partial t}\right)^{k+1}\left[\vec{j}(\vec{r}t)-\vec{j}'(\vec{r}t)\right]_{t=t_o} = i\rho_o(\vec{r})\vec{\nabla}\underbrace{\left(\left(i\frac{\partial}{\partial t}\right)^k\left[v(\vec{r}t)-v'(\vec{r}t)\right]_{t_o}\right)}_{\neq \text{constant}} \neq 0$$

$$\Rightarrow \quad \underline{\vec{j}(\vec{r}t) \neq \vec{j}'(\vec{r}t)} \qquad \text{q.e.d.}$$

Step 2: densities:

Show that ρ and ρ' will become different from each other infinitesimally later than t_0



Use continuity equation:

$$\frac{\partial}{\partial t}\left[\rho(\vec{r}\,t)-\rho'(\vec{r}\,t)\right]=-\mathrm{div}\left[\vec{j}(\vec{r}\,t)-\vec{j}\,'(\vec{r}\,t)\right]$$

$$\Rightarrow \quad \frac{\partial^{k+2}}{\partial t^{k+2}}\left[\rho(\vec{r}\,t)-\rho'(\vec{r}\,t)\right]_{t=t_o}=-\mathrm{div}\,\frac{\partial^{k+1}}{\partial t^{k+1}}\left[\vec{j}(\vec{r}\,t)-\vec{j}\,'(\vec{r}\,t)\right]_{t=t_o}$$

$$=-\mathrm{div}\;\rho_o(\vec{r})\vec{\nabla}\underbrace{\left(\frac{\partial^k}{\partial t^k}\left[v(\vec{r}\,t)-v'(\vec{r}\,t)\right]_{t=t_o}\right)}_{\neq\,\mathrm{constant}}$$

remains to be shown:

$$\mathrm{div}\left[\rho_o(\vec{r})\vec{\nabla}u(\vec{r})\right]\neq 0 \qquad \text{if} \qquad u(\vec{r})\neq \text{constant}$$

Proof: by reductio ad absurdum

Assume: $\operatorname{div}\left[\rho_o(\vec{r})\vec{\nabla}u(\vec{r})\right]=0$ with $u(\vec{r})\neq \text{constant}$

$$\int dr^3 \rho_o(\vec{r}) \left(\vec{\nabla}u(\vec{r})\right)^2$$

$$= -\int dr^3 u(\vec{r}) \underbrace{\operatorname{div}\left[\rho_o(\vec{r})\vec{\nabla}u(\vec{r})\right]}_{0} + \underbrace{\int \rho_o(\vec{r})u(\vec{r})\vec{\nabla}u(\vec{r})\cdot d\vec{S}}_{0} = 0$$

$$\Rightarrow \quad \rho_o(\vec{r})\left(\vec{\nabla}u(\vec{r})\right)^2 \equiv 0 \quad \longrightarrow \quad \text{contradiction to } u(\vec{r})\neq \text{constant}$$

Simplest possible approximation for $v_{xc}[\rho](\vec{r}\,t)$

Adiabatic Local Density Approximation (ALDA)

$$v_{xc}^{ALDA}(\vec{r}\,t) := v_{xc,stat}^{hom}(n)\Big|_{n=\rho(\vec{r}\,t)}$$

$v_{xc,stat}^{hom}$ **= xc potential of static homogeneous e-gas**

Approximation with correct asymptotic -1/r behavior: time-dependent optimized effective potential

(C. A. Ullrich, U. Gossmann, E.K.U.G., PRL 74, 872 (1995))

LINEAR RESPONSE THEORY

$t = t_0$: Interacting system in ground state of potential $v_0(r)$ with density $\rho_0(r)$

$t > t_0$: Switch on perturbation $v_1(r\,t)$ (with $v_1(r\,t_0)=0$).

Density: $\rho(r\,t) = \rho_0(r) + \delta\rho(r\,t)$

Consider functional $\rho[v](r\,t)$ defined by solution of interacting TDSE

Functional Taylor expansion of $\rho[v]$ around v_o:

$$\rho[v](r\,t) = \rho[v_0 + v_1](r\,t)$$

$$= \rho[v_0](r\,t) \longrightarrow \rho_o(r)$$

$$+ \int \left.\frac{\delta\rho[v](r\,t)}{\delta v(r'\,t')}\right|_{v_0} v_1(r'\,t')\,d^3r'\,dt' \longrightarrow \rho_1(r\,t)$$

$$+ \frac{1}{2}\int\int \left.\frac{\delta^2\rho[v](r\,t)}{\delta v(r'\,t')\delta v(r''\,t'')}\right|_{v_0} v_1(r',t')v_1(r'',t'')\,d^3r'\,d^3r''\,dt'\,dt'' \longrightarrow \rho_2(r\,t)$$

$$\vdots$$

$\rho_1(r,t)$ = **linear density response of interacting system**

$$\chi(r\,t, r'\,t') := \left.\frac{\delta\rho[v](r\,t)}{\delta v(r'\,t')}\right|_{v_0}$$ = **density-density response function of interacting system**

Lehmann representation of the full response function

$$\chi(r, r'; \omega) = \lim_{\eta\to 0^+} \sum_m \left(\frac{\langle 0|\hat{\rho}(r)|m\rangle\langle m|\hat{\rho}(r')|0\rangle}{\omega - (E_m - E_0) + i\eta} - \frac{\langle 0|\hat{\rho}(r')|m\rangle\langle m|\hat{\rho}(r)|0\rangle}{\omega + (E_m - E_0) + i\eta} \right)$$

with the exact many-body eigenfunctions and energies of the initial unperturbed interacting system Hamiltnian $H(t_0)|m\rangle = E_m|m\rangle$

$\Rightarrow$ **The exact linear density response**

$$\rho_1(\omega) = \chi(\omega)\, v_1$$

has poles at the exact excitation energies $\Omega = E_m - E_0$

Analogous function $\rho_s[v_s](r\,t)$ for <u>non</u>-interacting system

$$\rho_S[v_S](r\,t) = \rho_S[v_{S,0} + v_{S,1}](r\,t) = \rho_S[v_{S,0}](r\,t) + \int \left.\frac{\delta\rho_S[v_S](r\,t)}{\delta v_S(r'\,t')}\right|_{v_{S,0}} v_{S,1}(r'\,t')\, d^3r'\, dt' + \cdots$$

$$\chi_S(r\,t, r'\,t') := \left.\frac{\delta\rho_S[v_S](r\,t)}{\delta v_S(r'\,t')}\right|_{v_{S,0}}$$ = **density-density response function of <u>non</u>-interacting system**

GOAL: Find a way to calculate $\rho_1(r\,t)$ without explicitly evaluating $\chi(r\,t,r't')$ of the interacting system

starting point: Definition of xc potential

$$v_{xc}[\rho](r\,t) := v_S[\rho](r\,t) - v_{ext}[\rho](r\,t) - v_H[\rho](r\,t)$$

v_{xc} is well-defined through non-interacting / interacting 1-1 mapping.

$$\left.\frac{\delta v_{xc}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} = \left.\frac{\delta v_{S}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \left.\frac{\delta v_{ext}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \frac{\delta(t-t')}{|r-r'|}$$

$$\left.\frac{\delta v_{xc}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} = \left.\frac{\delta v_S[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \left.\frac{\delta v_{ext}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \frac{\delta(t-t')}{|r-r'|}$$

$$\uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow$$

$$f_{xc}(r\,t, r'\,t') \qquad \chi_S^{-1}(r\,t, r'\,t') \qquad \chi^{-1}(r\,t, r'\,t') \qquad W_C(r\,t, r'\,t')$$

$$\left.\frac{\delta v_{xc}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} = \left.\frac{\delta v_S[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \left.\frac{\delta v_{ext}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \frac{\delta(t-t')}{|r-r'|}$$

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$$f_{xc} + W_C = \chi_S^{-1} - \chi^{-1}$$

$$\left.\frac{\delta v_{xc}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} = \left.\frac{\delta v_S[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \left.\frac{\delta v_{ext}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \frac{\delta(t-t')}{|r-r'|}$$

$$\uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow$$

$$f_{xc}(r\,t, r'\,t') \qquad \chi_S^{-1}(r\,t, r'\,t') \qquad \chi^{-1}(r\,t, r'\,t') \qquad W_C(r\,t, r'\,t')$$

$$\chi_S \bullet \left| f_{xc} + W_C = \chi_S^{-1} - \chi^{-1} \right| \bullet \chi$$

$$\left.\frac{\delta v_{xc}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} = \left.\frac{\delta v_S[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \left.\frac{\delta v_{ext}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \frac{\delta(t-t')}{|r-r'|}$$

$$\uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow$$

$$f_{xc}(r\,t,r'\,t') \qquad \chi_S^{-1}(r\,t,r'\,t') \qquad \chi^{-1}(r\,t,r'\,t') \qquad W_C(r\,t,r'\,t')$$

$$\chi_S \bullet \left| f_{xc} + W_C = \chi_S^{-1} - \chi^{-1} \right| \bullet \chi$$

$$\chi_S\left(f_{xc} + W_C\right)\chi = \chi - \chi_S$$

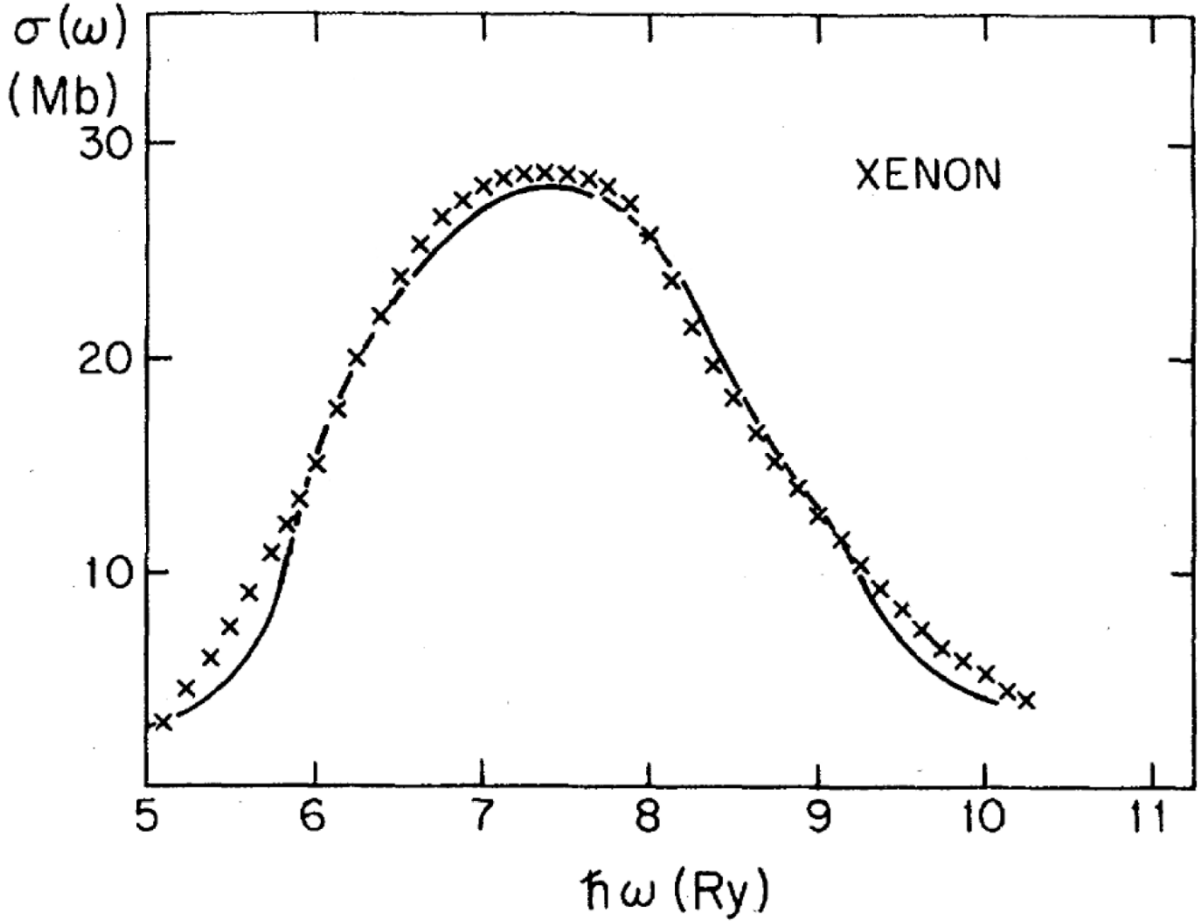
$$\chi = \chi_S + \chi_S \left(W_{ee} + f_{xc} \right) \chi$$

Act with this operator equation on arbitrary $v_1(r\,t)$ and use $\chi\, v_1 = \rho_1$:

$$\rho_1(r\,t) = \int d^3r'dt' \chi_S(r\,t, r't') \left[v_1(r\,t) + \int d^3r''dt'' \left\{ W_{ee}(r't', r''t'') + f_{xc}(r't', r''t'') \right\} \rho_1(r''t'') \right]$$

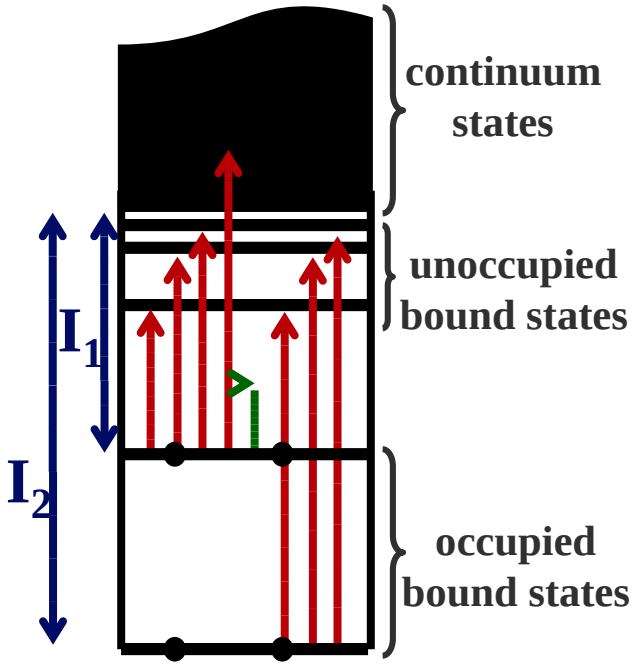
- Exact integral equation for $\rho_1(r\,t)$, to be solved iteratively
- Need approximation for $f_{xc}(r't', r''t'') = \left. \frac{\delta v_{xc}[\rho](r't')}{\delta \rho(r''t'')} \right|_{\rho_0}$
  (either for f_{xc} directly or for v_{xc})

Total photoabsorption cross section of the Xe atom versus photon energy in the vicinity of the 4d threshold.



Solid line: self-consistent time-dependent KS calculation [A. Zangwill and P. Soven, Phys. Rev. A 21, 1561 (1980)]; crosses: experimental data [R. Haensel, G. Keitel, P. Schreiber, and C. Kunz, Phys. Rev. 188, 1375 (1969)].

Photo-absorption in weak lasers



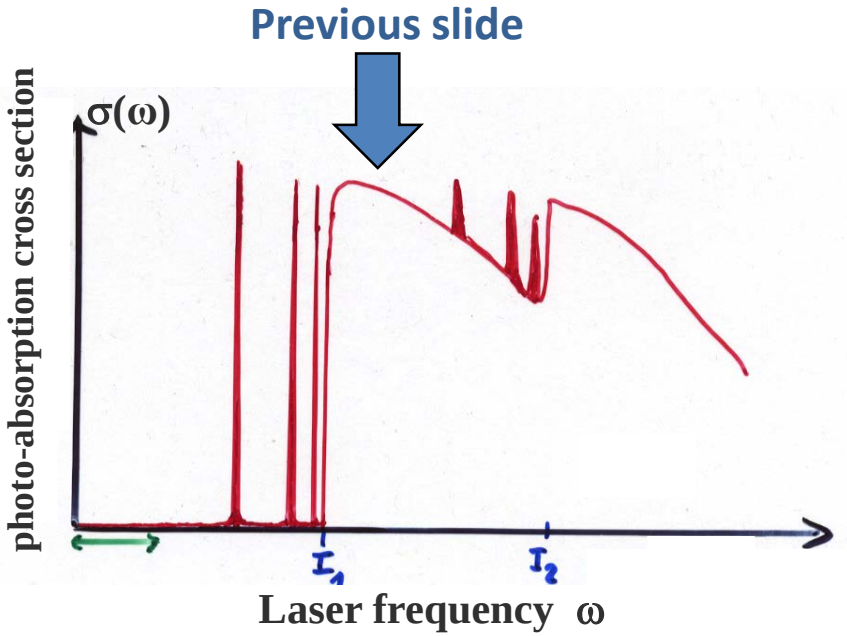
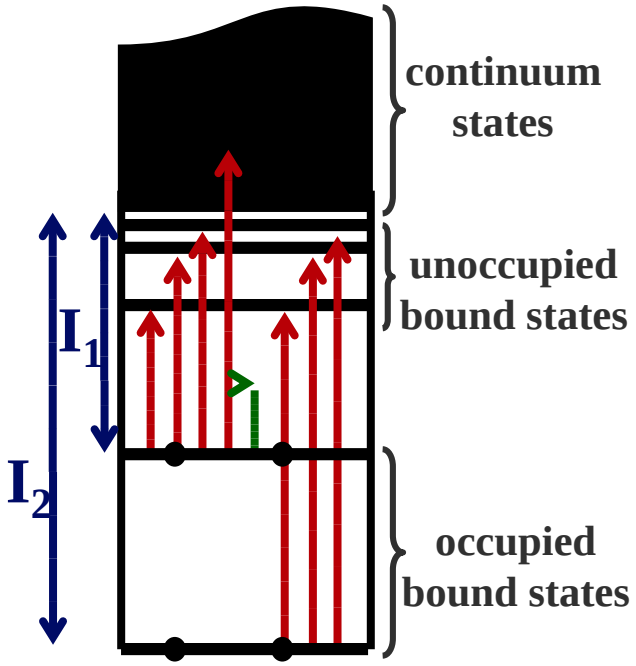
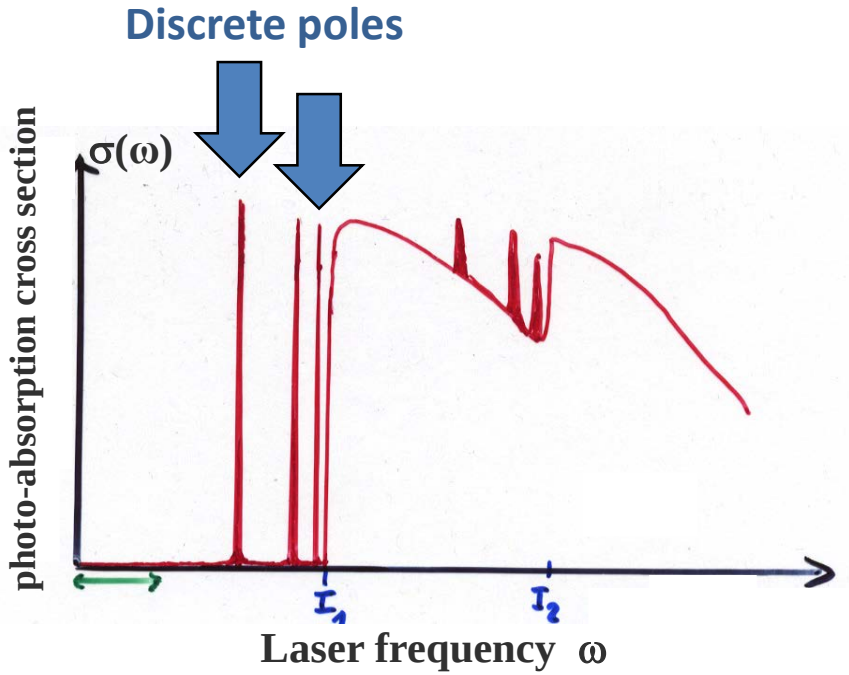


Photo-absorption in weak lasers





Looking at those frequencies, Ω, for which $\rho_1(\omega)$ has poles, leads to the (non-linear) eigenvalue equation

(T. Grabo, M. Petersilka, EKUG, J. Mol. Struc. (Theochem) 501, 353 (2000))

$$\sum_{q'}\left(A_{qq'}(\Omega)+\omega_q\delta_{qq'}\right)\beta_{q'}=\Omega\beta_q$$

where

$$A_{qq'}=\alpha_{q'}\int d^3r\int d^3r'\,\Phi_q(r)\left(\frac{1}{|r-r'|}+f_{xc}(r,r',\Omega)\right)\Phi_{q'}(r')$$

$q=(j,a)$ double index

$$\alpha_q=f_a-f_j$$

$$\Phi_q(r)=\varphi_a^*(r)\varphi_j(r)$$

$$\omega_q=\varepsilon_a-\varepsilon_j$$

Atom	Experimental Excitation Energies $^1S \rightarrow {}^1P$ (in Ry)	KS energy differences $\Delta\epsilon_{KS}$ (Ry)	TDDFT
Be	0.388	0.259	0.391
Mg	0.319	0.234	0.327
Ca	0.216	0.157	0.234
Zn	0.426	0.315	0.423
Sr	0.198	0.141	0.210
Cd	0.398	0.269	0.391

from: M. Petersilka, U. J. Gossmann, E.K.U.G., PRL 76, 1212 (1996)

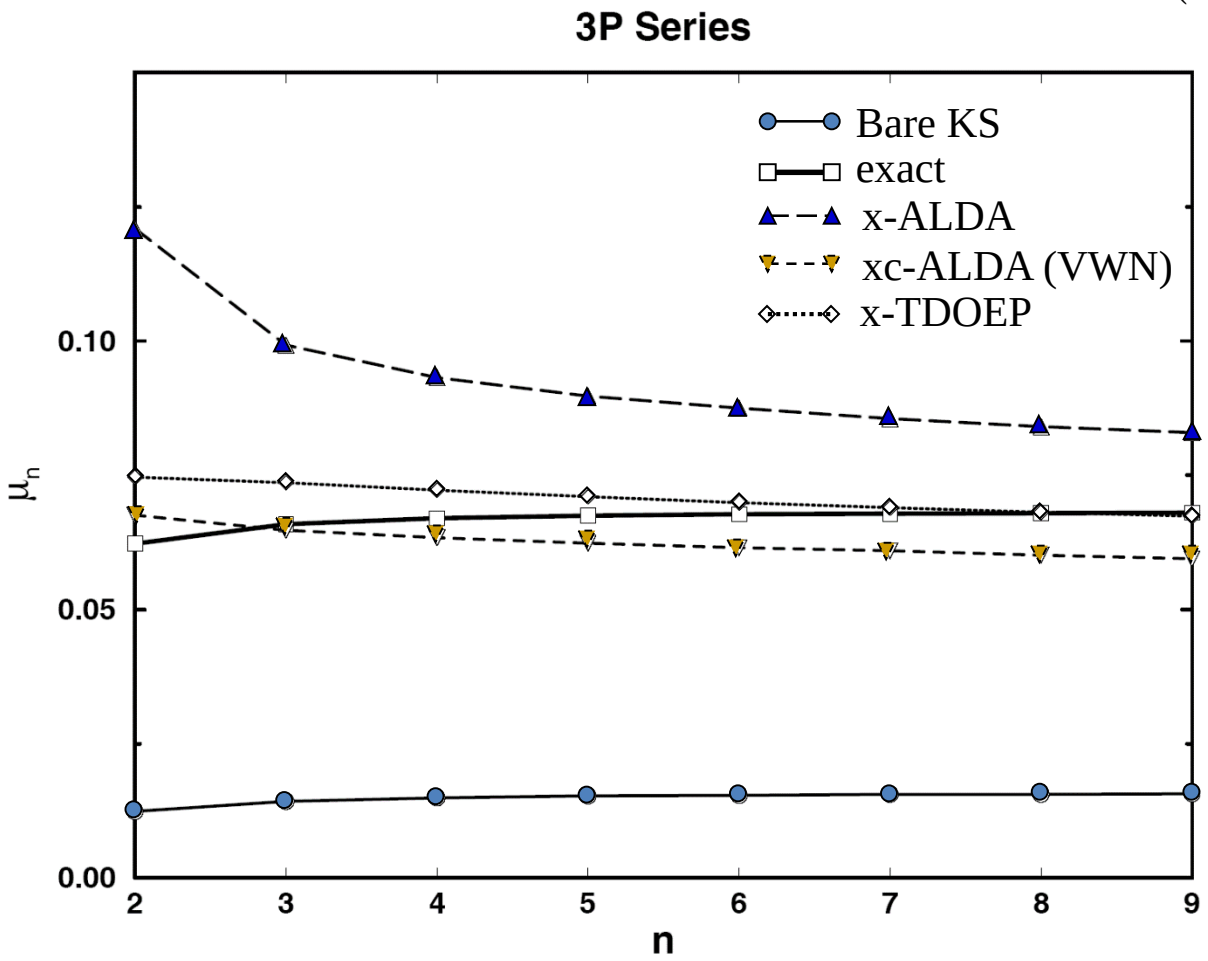
Excitation energies of CO molecule

State		Ω_{expt}	KS-transition	$\Delta\epsilon_{KS}$	TDDFT
A	$^1\Pi$	0.3127	$5\Sigma \rightarrow 2\Pi$	0.2523	0.3267
a	$^3\Pi$	0.2323			0.2238
I	$^1\Sigma^-$	0.3631	$1\Pi \rightarrow 2\Pi$	0.3626	0.3626
D	$^1\Delta$	0.3759			0.3812
a'	$^3\Sigma^+$	0.3127			0.3181
e	$^3\Sigma^-$	0.3631			0.3626
d	$^3\Delta$	0.3440			0.3404

T. Grabo, M. Petersilka and E.K.U. Gross, J. Mol. Struct. (Theochem) 501, 353 (2000)

approximations made: v_{xc}^{LDA} and f_{xc}^{ALDA}

Quantum defects in Helium $E_n = -\frac{1}{2(n-\mu_n)^2}$ [a.u.]



M. Petersilka, U.J. Gossmann and E.K.U.G., in: Electronic Density Functional Theory: Recent Progress and New Directions, J.F. Dobson, G. Vignale, M.P. Das, ed(s), (Plenum, New York, 1998), p 177 - 197.

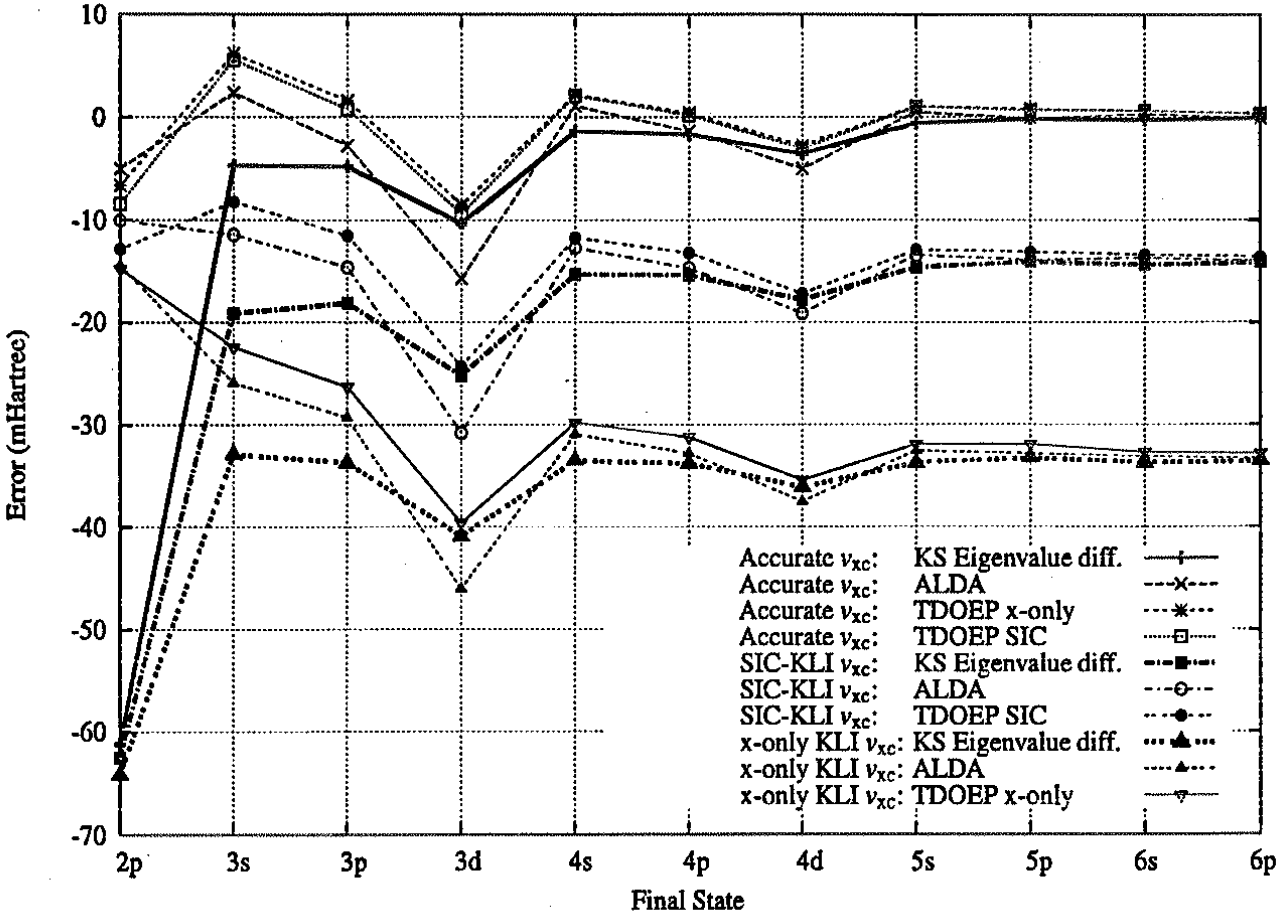
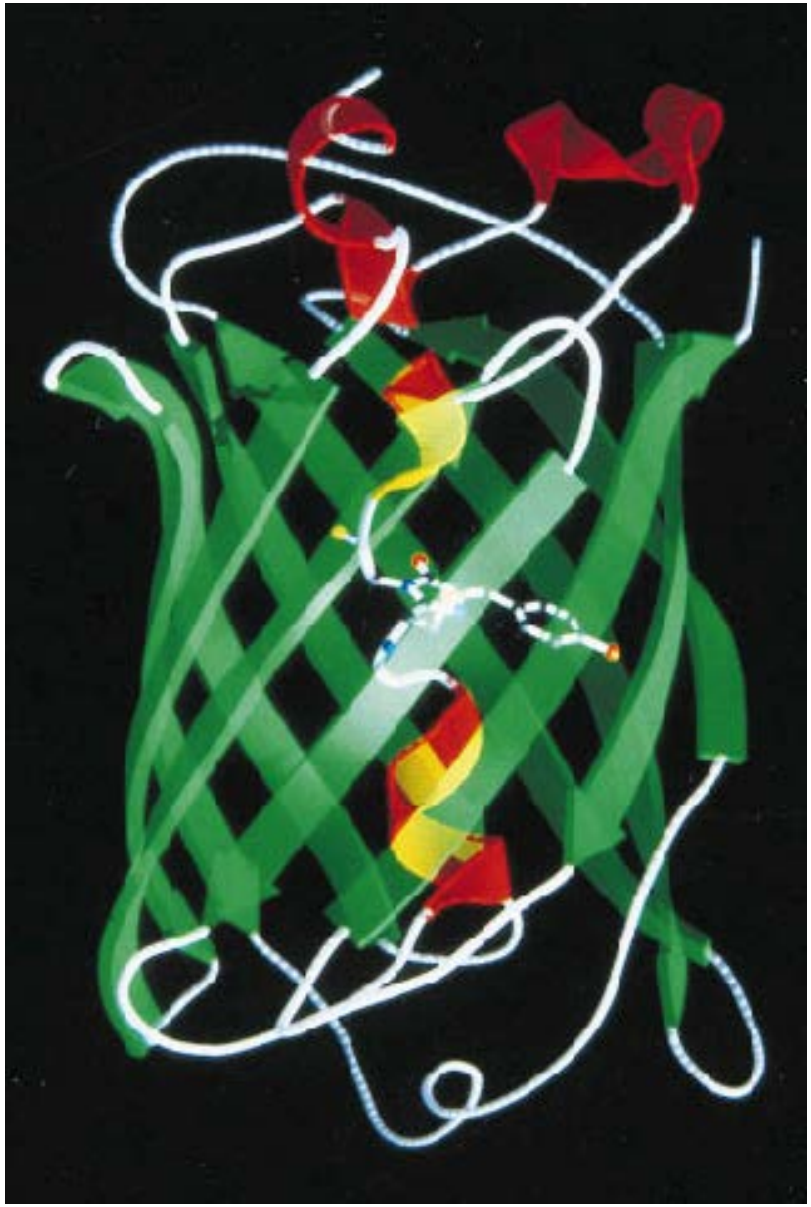


Figure 3.3: Errors of singlet excitation energies from the ground state of Be, calculated from the accurate, the OEP-SIC and x-only KLI exchange correlation potential and with different approximations for the exchange-correlation kernel (see text). The errors are given in mHartrees. To guide the eye, the errors of the discrete excitation energies were connected with lines.

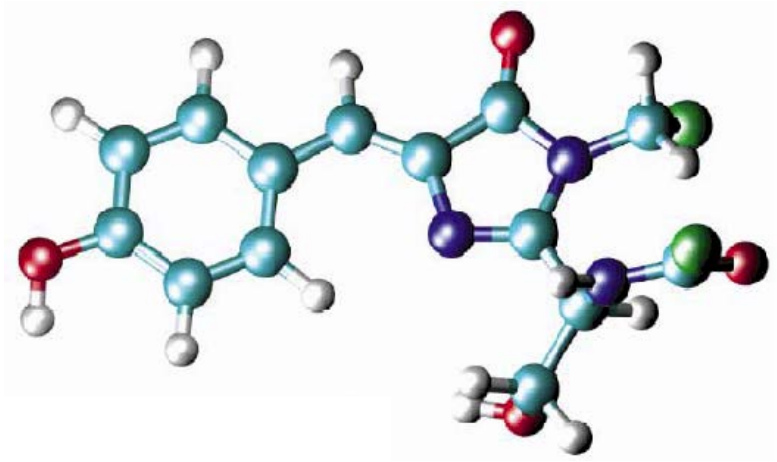
(M. Petersilka, E.K.U.G., K. Burke, Int. J. Quantum Chem. 80, 534 (2000))



Ribbon diagram of the wt-GFP structure. The α-helices are shown in red, the β-strands are shown in green, and the chromophore is shown as a ball-and-stick model.

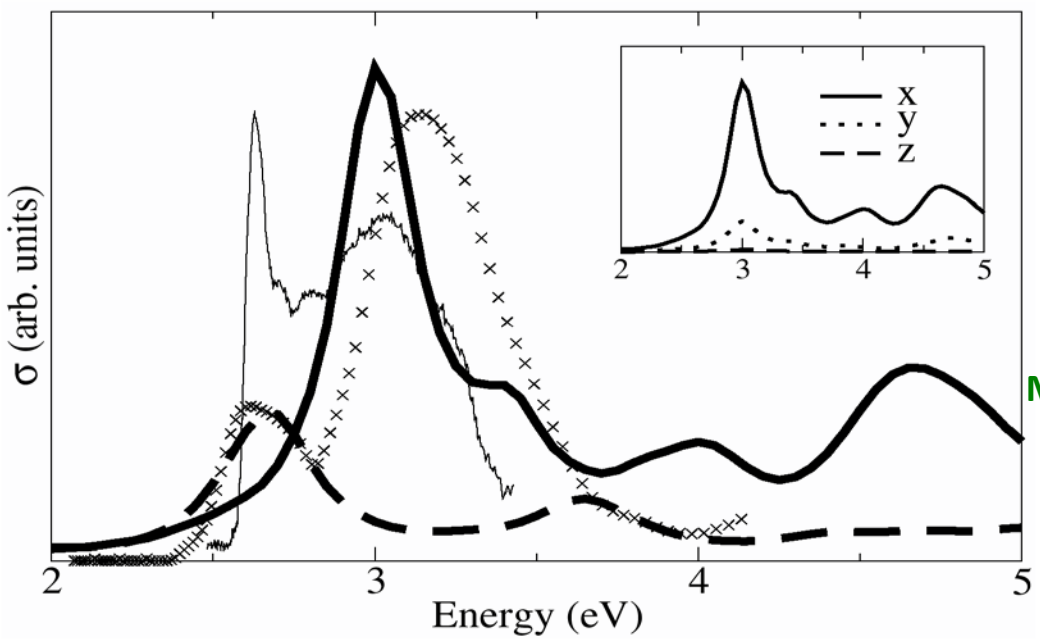
Brejc et al., Proc. Natl. Acad. Sci. 94, 2306 (1997)

GFP-chromophore



Computed photoabsorption cross section of the neutral (thick solid line) and anionic (thick dashed line) GFP chromophores, along with experimental results (thin solid line and crosses, respectively).

For comparative purposes, the anionic results is divided by 4 with respect to the neutral results.



Inset: decomposition of the computed spectra of the neutral chromophore in the three directions, showing the inherent anisotropy of the GFP molecule.

Marques et al., PRL 90, 258101 (2003)

Failures of ALDA in the linear response regime

- **H_2 dissociation is incorrect:**

$$E\left(^1\Sigma_u^+\right) - E\left(^1\Sigma_g^+\right) \xrightarrow[R\to\infty]{} 0 \quad \textbf{(in ALDA)}$$

(see: Gritsenko, van Gisbergen, Görling, Baerends, J. Chem. Phys. 113, 8478 (2000))

- **response of long chains strongly overestimated**

(see: Champagne et al., J. Chem. Phys. 109, 10489 (1998) and 110, 11664 (1999))

- **in periodic solids,** $f_{xc}^{ALDA}(q,\omega,\rho) = c(\rho)$ **whereas,**

for insulators, $f_{xc}^{exact} \xrightarrow[q\to 0]{} 1/q^2$ **divergent.**

- **charge-transfer excitations not properly described**

(see: Dreuw et al., J. Chem. Phys. 119, 2943 (2003))

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These difficulties have largely been solved by xc functionals more advanced than ALDA

How good is ALDA for periodic solids?

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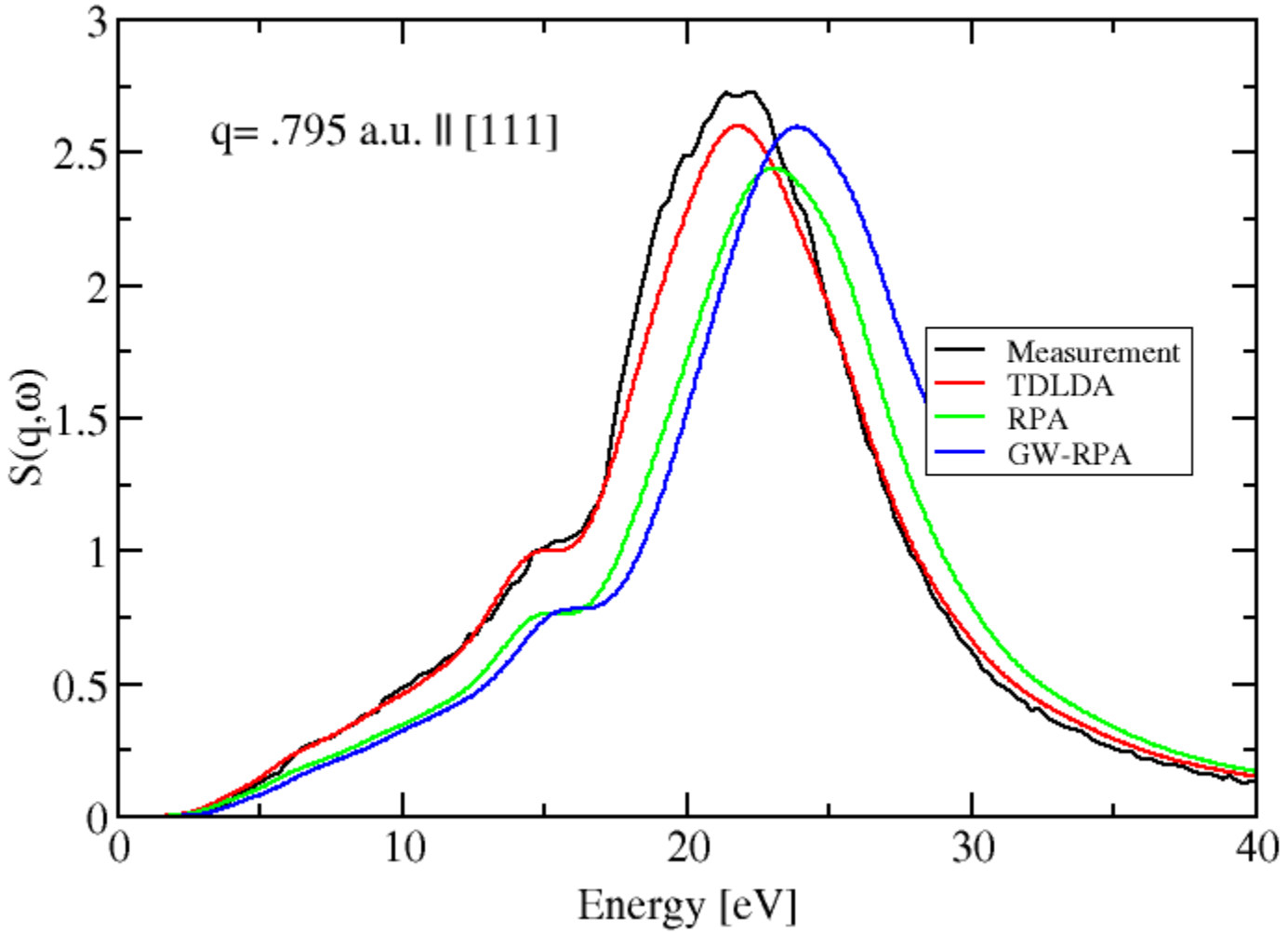
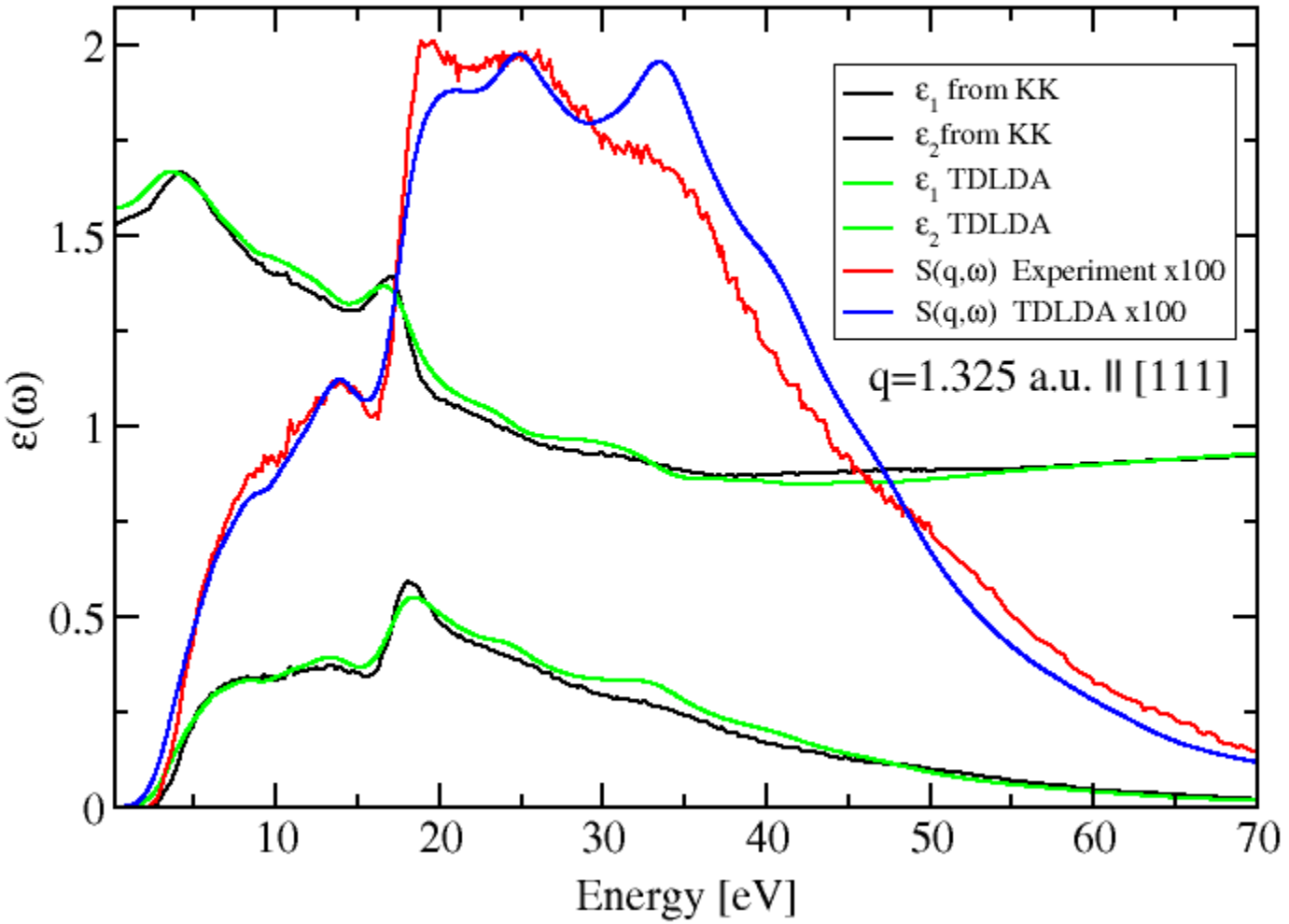
Look at "EELS": Im $\chi(q,\omega)$ at finite q

H.C. Weissker, J. Serrano, S. Huotari,
F. Bruneval, F. Sottile, G. Monaco, M. Krisch,
V. Olevano, L. Reining,
Phys. Rev. Lett. 97, 237602 (2006)

Silicon: Loss function Im $\chi(q,\omega)$




ε(ω)
Energy [eV]
ε1 from KK
ε2 from KK
ε1 TDLDA
ε2 TDLDA
S(q,ω) Experiment x100
S(q,ω) TDLDA x100
q=1.325 a.u. ∥ [111]
0
0.5
1
1.5
2
10
20
30
40
50
60
70

Excitons in TDDFT

TDDFT response equation (formally exact)

$$\varepsilon^{-1}(\mathbf{q},\omega) = 1 + \chi_S(\mathbf{q},\omega) v(\mathbf{q}) \left[1 - \left(v(\mathbf{q}) + f_{xc}(\mathbf{q},\omega) \right) \chi_S(\mathbf{q},\omega) \right]^{-1}$$

all quantities are matrices w.r.t. G G'

$f_{xc}(\mathbf{r},\mathbf{r}',t-t') \equiv \delta v_{xc}(\mathbf{r},t) / \delta\rho(\mathbf{r}',t')$ xc kernel

$\chi_S(\mathbf{q},\omega)$ Kohn-Sham response function

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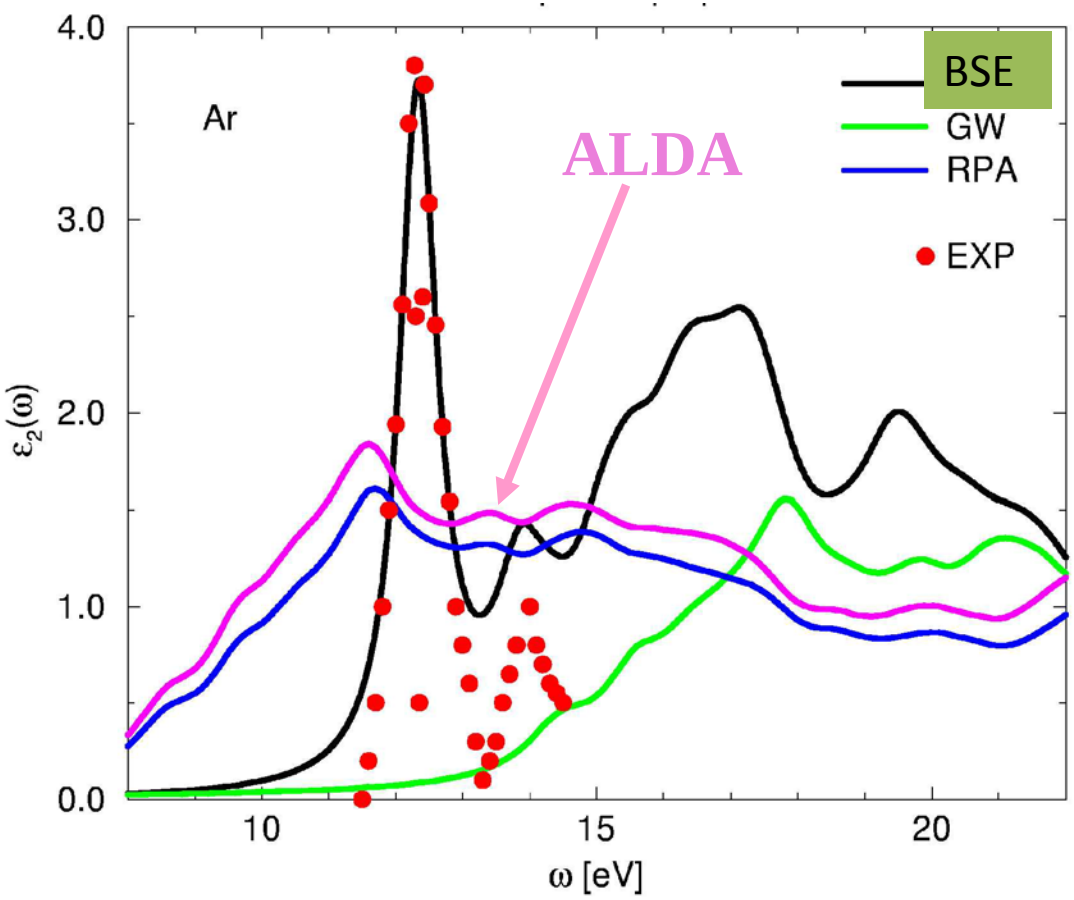
$\chi_S(\mathbf{q},\omega)$ Kohn-Sham response function

Need approximation for f_{xc}

RPA: $f_{xc} \equiv 0$

ALDA: $f_{xc}^{ALDA}(\mathbf{r},\mathbf{r}',t-t') = \delta(\mathbf{r}-\mathbf{r}')\delta(t-t')\, \partial v_{xc}^{LDA}(\rho(\mathbf{r})) / \partial\rho(\mathbf{r})$

Solid Argon



L. Reining, V. Olevano, A. Rubio, G. Onida, PRL 88, 066404 (2002)

Excitons are completely missing for simple xc functionals like ALDA!

EXPLANATION:

TDDFT response equation: bare Coulomb interaction and xc kernel only appear as a sum $(v + f_{xc})$.

$v(q)$ diverges like $1/q^2$ for $q \longrightarrow 0$

$f_{xc} \longrightarrow$ const (in ALDA)

Hence results are close to $f_{xc} = 0$ (RPA) in the $q \longrightarrow 0$ limit.

CONSEQUENCE:

Approximations for f_{xc} are needed which, for $q \longrightarrow 0$, correctly diverge like $1/q^2$. Such approximations can be derived from many-body perturbation theory (see, e.g., L. Reining, V. Olevano, A. Rubio, G. Onida, PRL 88, 066404 (2002)).

$$\varepsilon^{-1}(\mathbf{q},\omega) = 1 + \chi_S(\mathbf{q},\omega) v(\mathbf{q}) \left[1 - \left(v(\mathbf{q}) + f_{xc}^{approx}(\mathbf{q},\omega) \right) \chi_S(\mathbf{q},\omega) \right]^{-1}$$

Two problems of ALDA need to be fixed:

- **Onset of absorption is dictated by χ_S , i.e. is identical to the LDA gap for ω-independent kernel (such as ALDA)**

- **Description of excitons requires $1/q^2$ behavior**

Bootstrap kernel (Sharma, Dewhurst, Sanna, EKUG, PRL **107**, 186401 (2011))

$$f_{xc}^{boot}(\mathbf{q},\omega) = \frac{\varepsilon^{-1}(\mathbf{q},\omega=0)}{\tilde{\chi}^{00}(\mathbf{q},\omega=0)}$$

$$f_{xc}^{appr}(\mathbf{q},\omega) = \frac{1}{\chi_S(\mathbf{q},\omega)} - \frac{1}{\tilde{\chi}(\mathbf{q},\omega)} + f_{xc}^{boot}(\mathbf{q})$$

where $\tilde{\chi}$ is a single-particle response function that has the right gap, e.g. from G_0W_0 , or LDA/GGA+Scissors.

$$\varepsilon^{-1}(\mathbf{q},\omega) = 1 + \chi_0(\mathbf{q},\omega)v(\mathbf{q})\left[1 - \left(v(\mathbf{q}) + f_{xc}^{appr}(\mathbf{q},\omega)\right)\chi_0(\mathbf{q},\omega)\right]^{-1}$$

Bootstrap kernel (Sharma, Dewhurst, Sanna, EKUG, PRL **107**, 186401 (2011))

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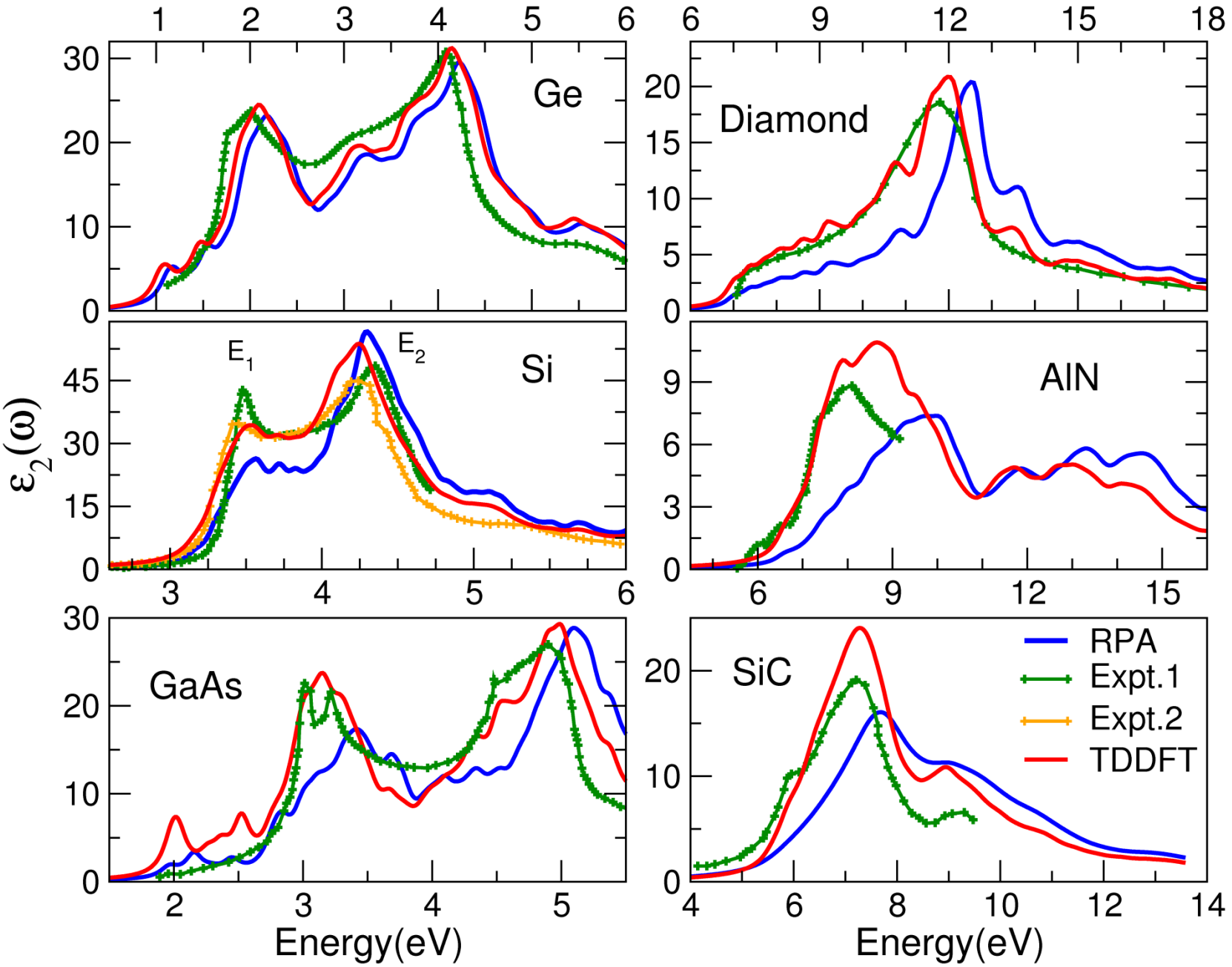
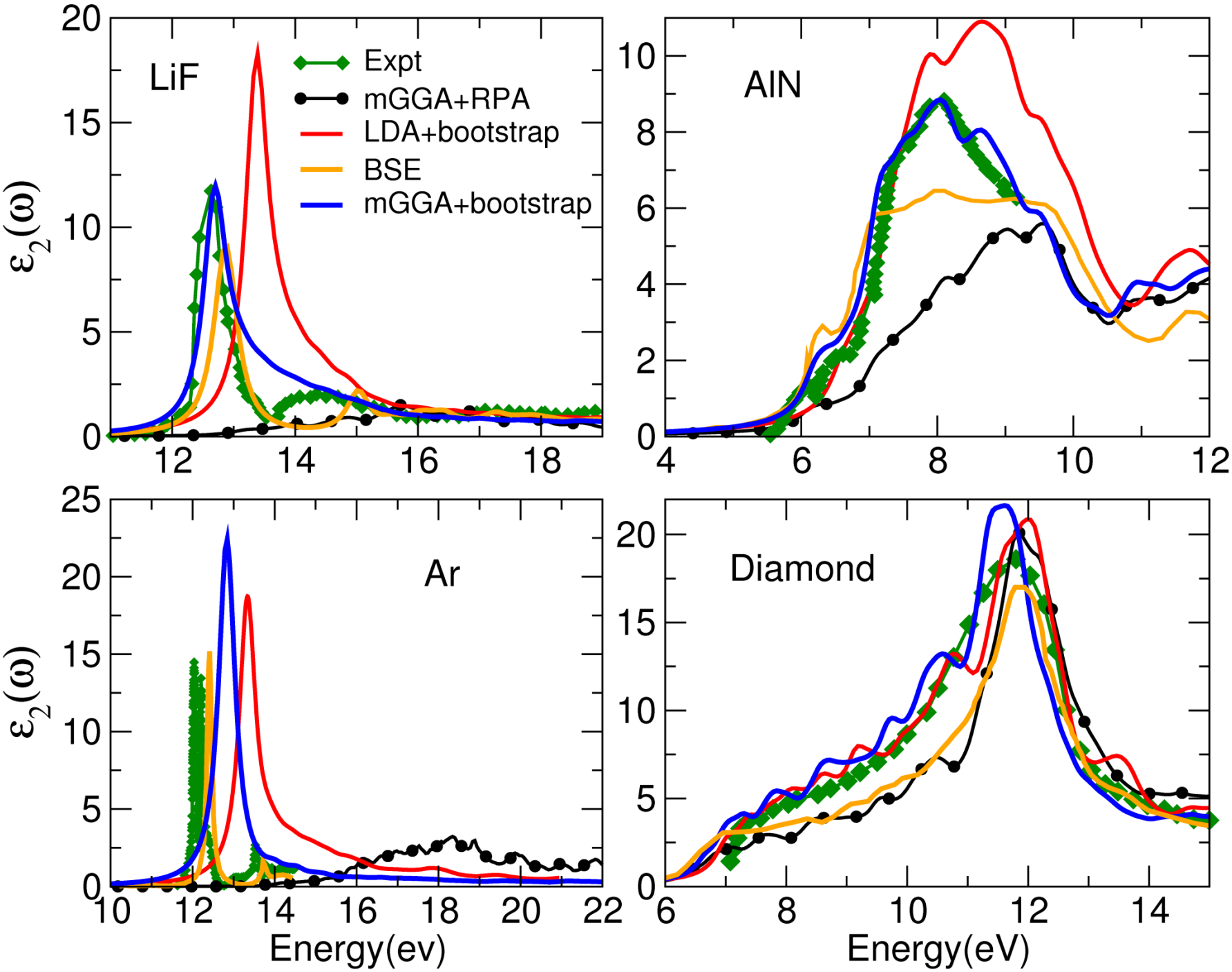
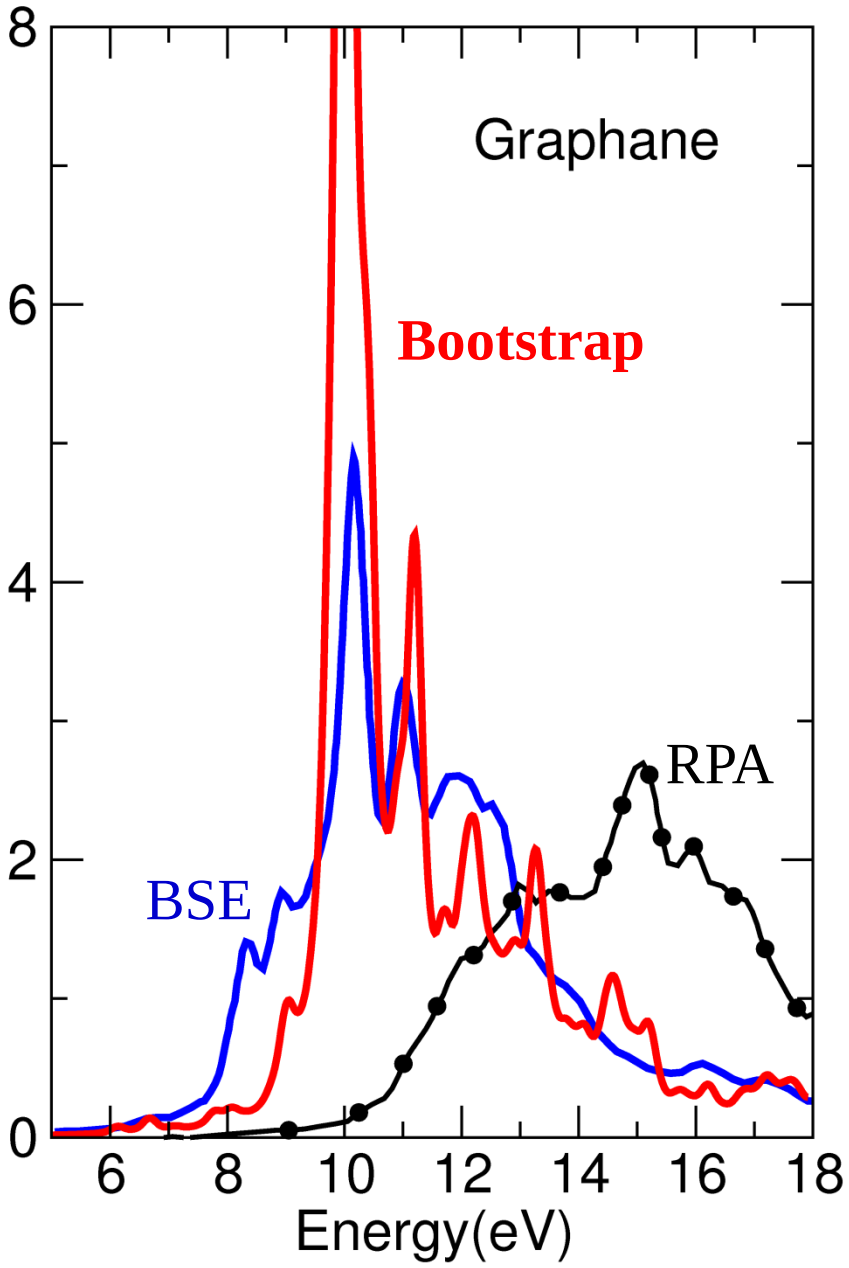
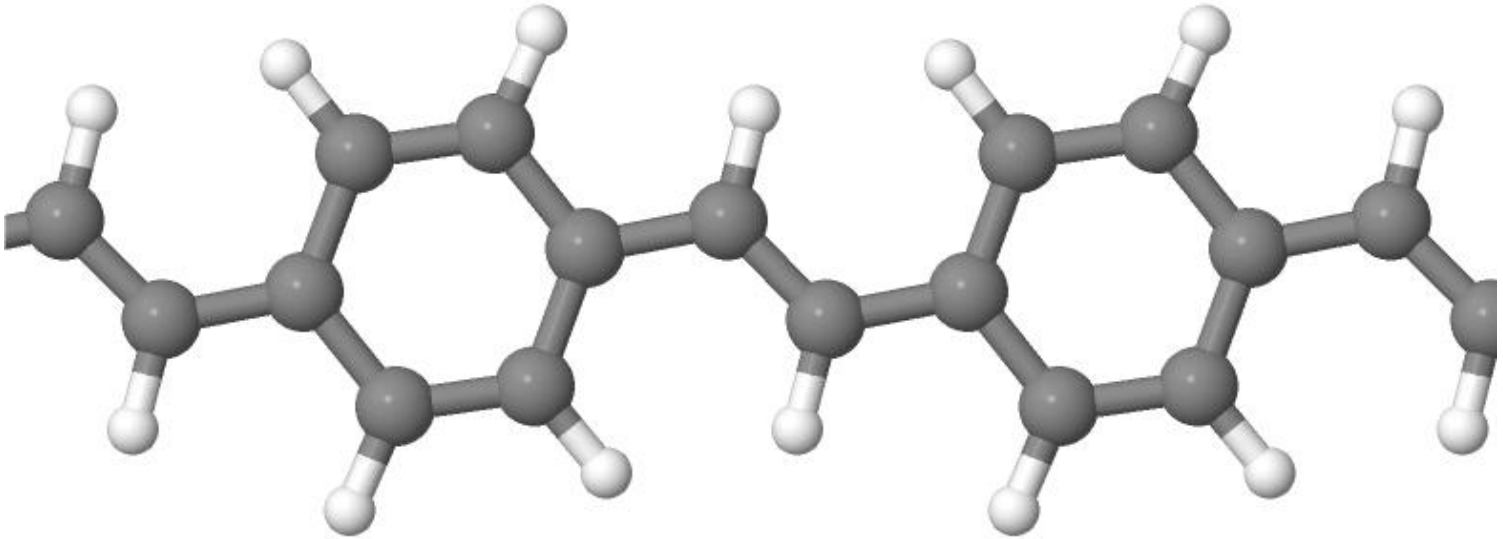
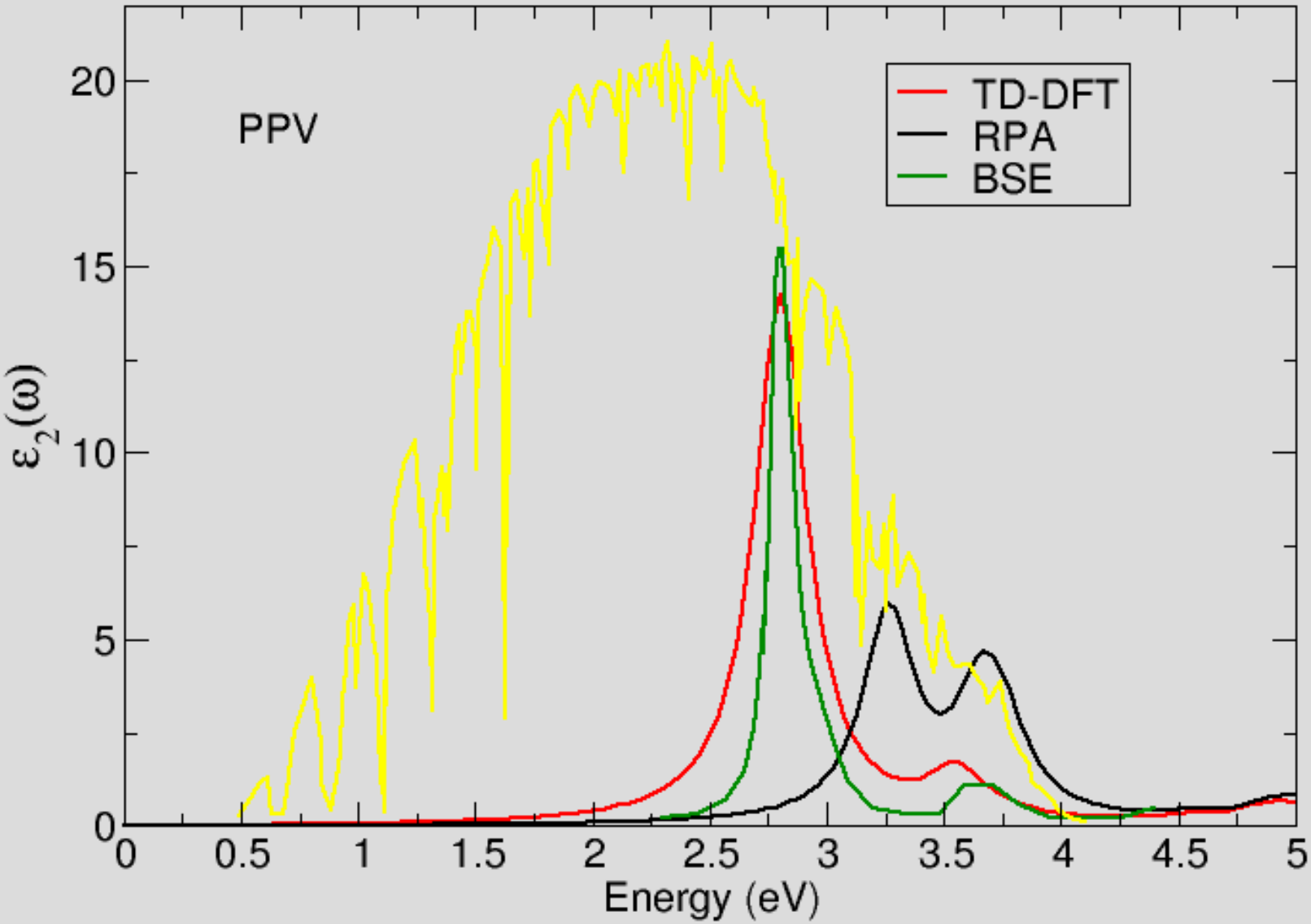

Ge
Diamond
Si
E1
E2
AlN
GaAs
SiC
RPA
Expt.1
Expt.2
TDDFT
ε2(ω)
Energy(eV)
Energy(eV)


LiF
Expt
mGGA+RPA
LDA+bootstrap
BSE
mGGA+bootstrap
AlN
Ar
Diamond
$\varepsilon_2(\omega)$
Energy(ev)
Energy(eV)

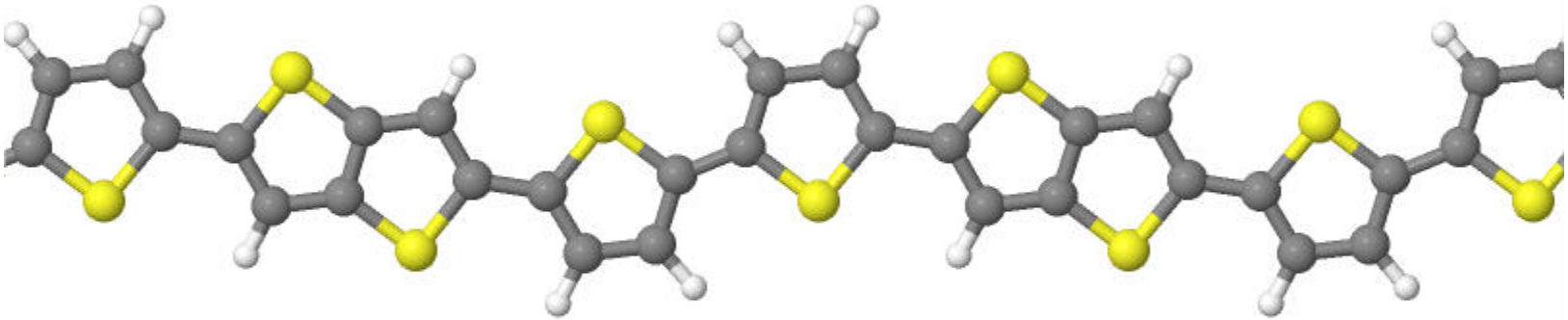

Graphane
Bootstrap
BSE
RPA
0
2
4
6
8
6
8
10
12
14
16
18
Energy(eV)

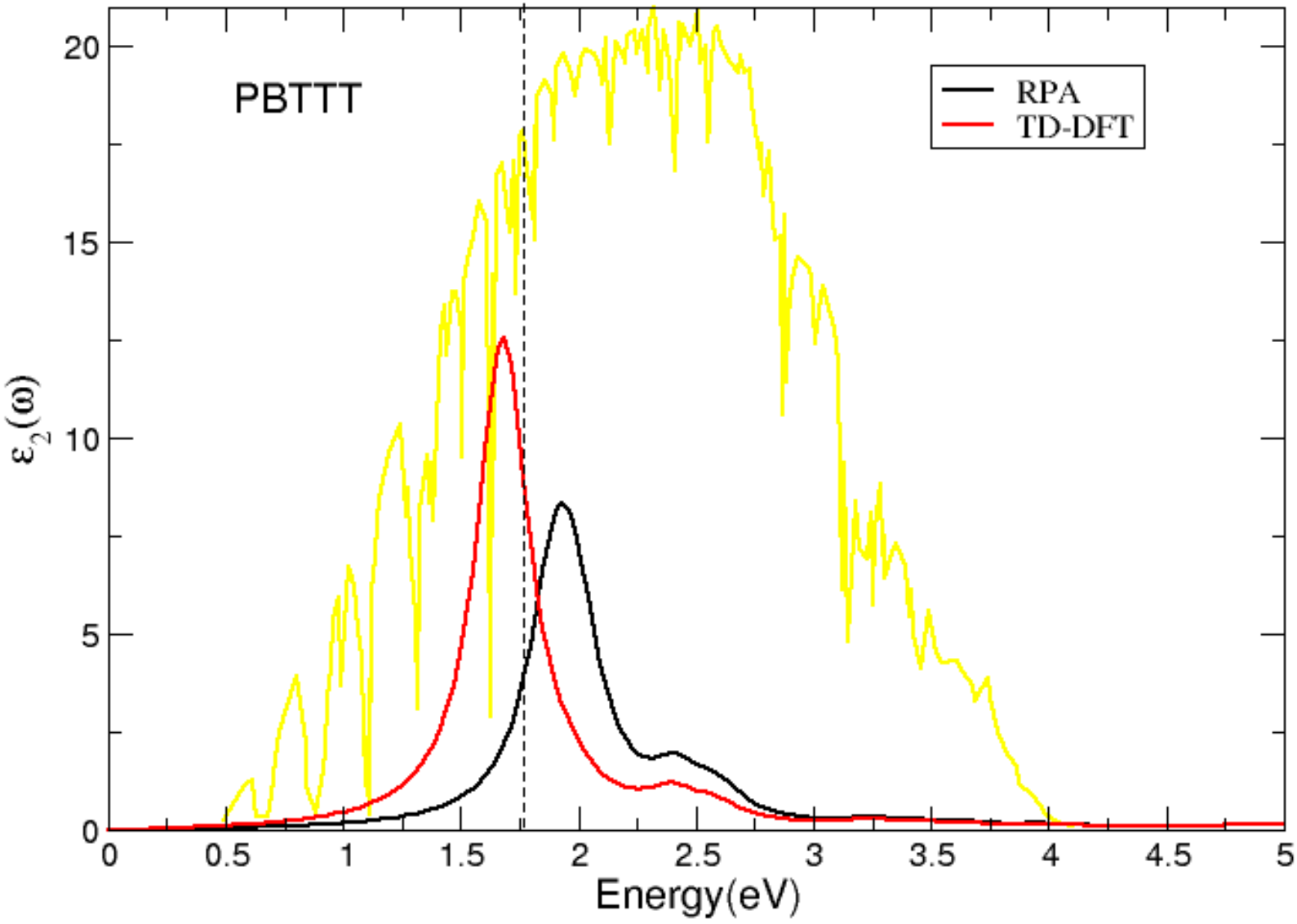
PPV



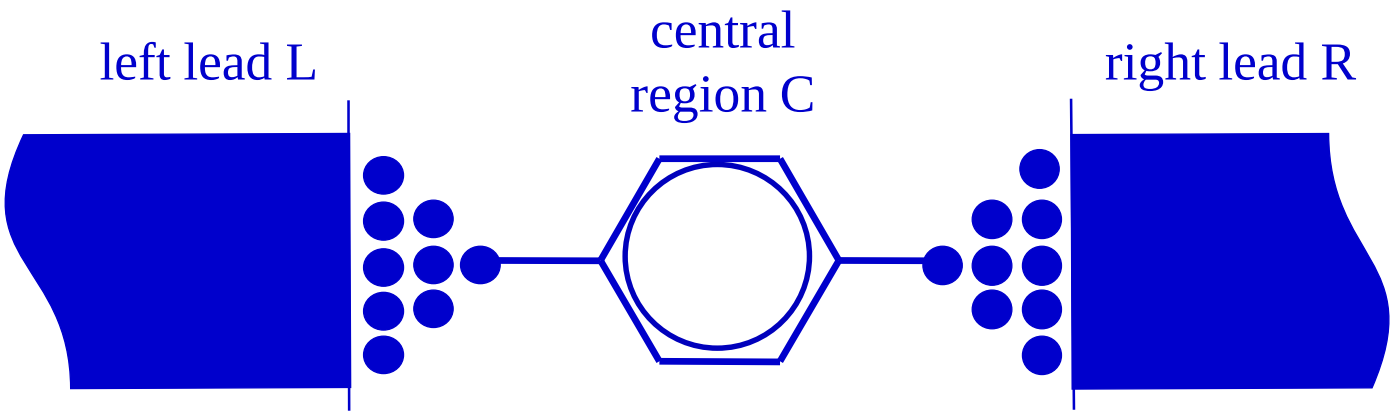

PPV
TD-DFT
RPA
BSE
$\varepsilon_2(\omega)$
Energy (eV)
0
5
10
15
20
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5

PBTTT



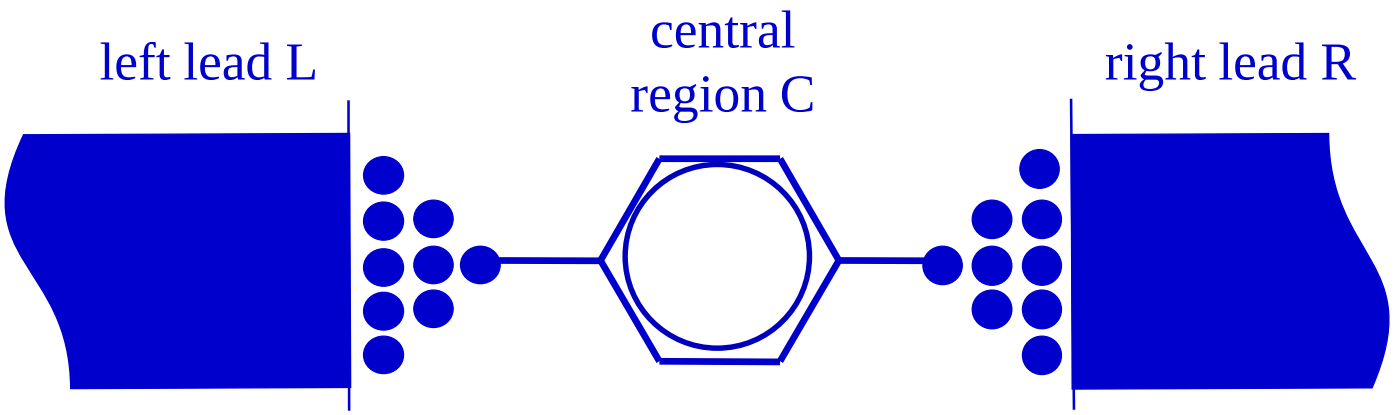

PBTTT
RPA
TD-DFT
$\varepsilon_2(\omega)$
Energy(eV)
0
5
10
15
20
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5

Electronic transport with TDDFT



Bias between L and R is turned on: U(t) $\longrightarrow$ V for large t

Electronic transport with TDDFT

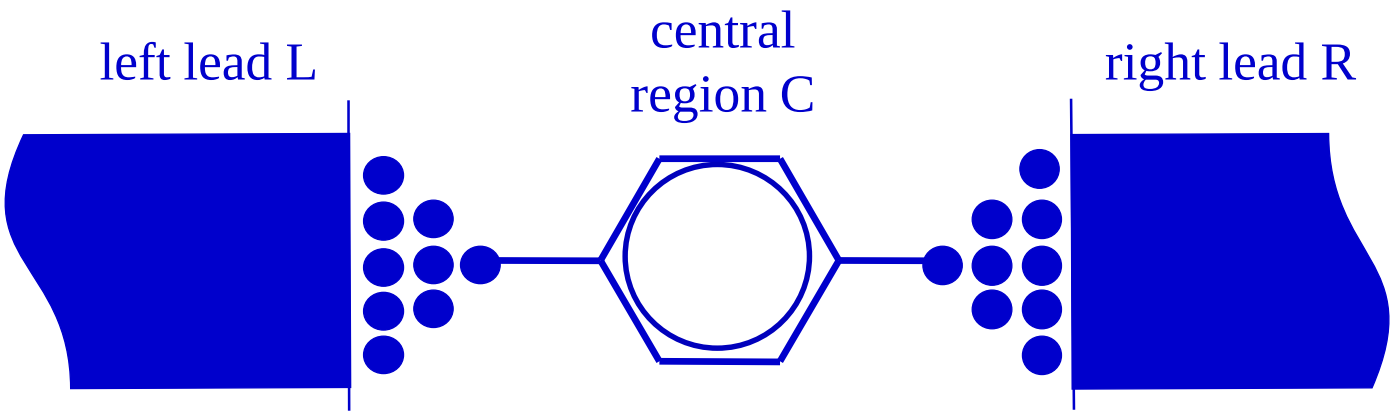


Bias between L and R is turned on: U(t) $\longrightarrow$ V for large t

Questions:

- After switching-on, does one always reach a steady state?
- Is the steady state unique?
- How to deal with time-dependent external fields?

Electronic transport with TDDFT

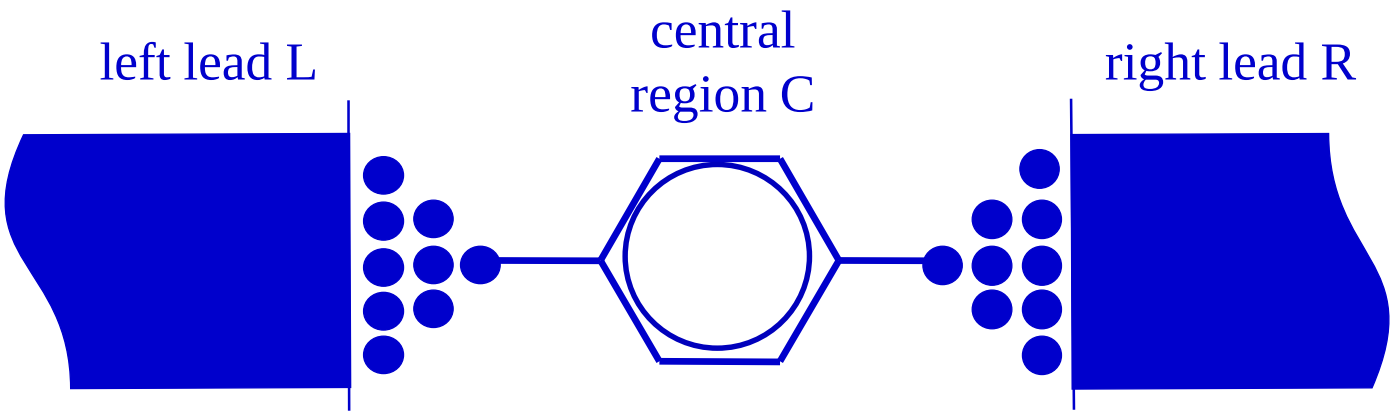


TDKS equation (E. Runge, EKUG, PRL **52**, 997 (1984))

$$i\hbar\frac{\partial}{\partial t}\varphi_j(rt)=\left(-\frac{\hbar^2\nabla^2}{2m}+v_{KS}[\rho](rt)\right)\varphi_j(rt)$$

$$v_{KS}[\rho(r't')](rt)=v(rt)+\int d^3r'\frac{\rho(r't)}{|r-r'|}+v_{xc}[\rho\,(r't')](r\,t)$$

Electronic transport with TDDFT



TDKS equation

$$i\frac{\partial}{\partial t}\begin{pmatrix}\varphi_L(t)\\ \varphi_C(t)\\ \varphi_R(t)\end{pmatrix}=\begin{pmatrix}H_{LL}(t) & H_{LC}(t) & H_{LR}(t)\\ H_{CL}(t) & H_{CC}(t) & H_{CR}(t)\\ H_{RL}(t) & H_{RC}(t) & H_{RR}(t)\end{pmatrix}\begin{pmatrix}\varphi_L(t)\\ \varphi_C(t)\\ \varphi_R(t)\end{pmatrix}$$

Effective TDKS Equation for the central (molecular) region only

S. Kurth, G. Stefanucci, C.O. Almbladh, A. Rubio, E.K.U.G.,
Phys. Rev. B 72, 035308 (2005)

$$i\frac{\partial}{\partial t}\varphi_C(t) = H_{CC}(t)\varphi_C(t) + \int_0^t dt' \left[H_{CL}G_L(t,t')H_{LC} + H_{CR}G_R(t,t')H_{RC}\right]\varphi_C(t') + iH_{CL}G_L(t,0)\varphi_L(0) + iH_{CR}G_R(t,0)\varphi_R(0)$$

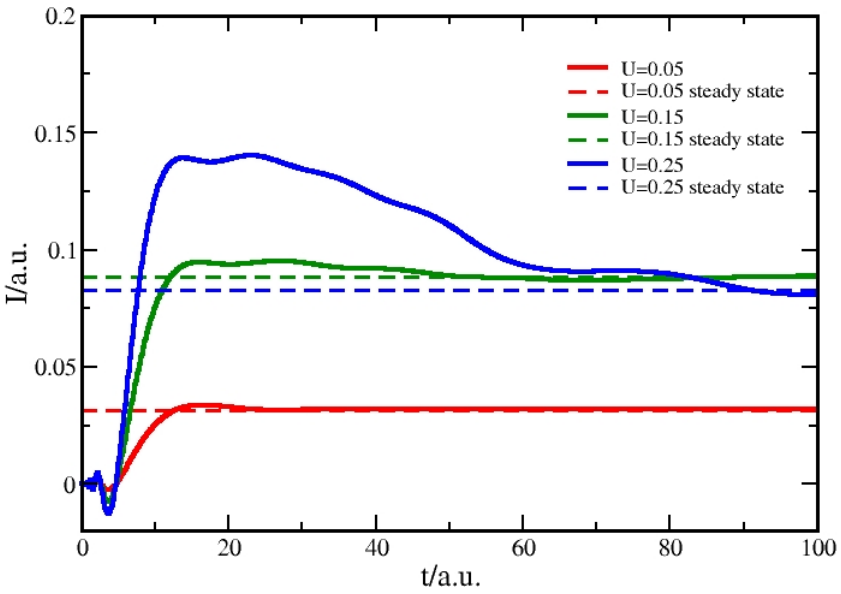
source term: $L \rightarrow C$ and $R \rightarrow C$ charge injection

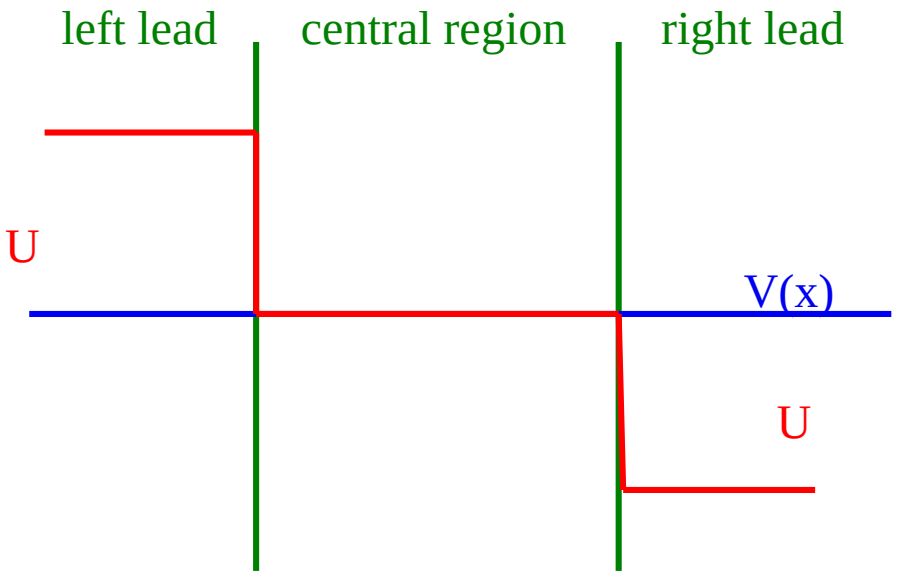
memory term: $C \rightarrow L \rightarrow C$ and $C \rightarrow R \rightarrow C$ hopping

Note: So far, no approximation has been made.

Numerical examples for non-interacting electrons

Recovering the Landauer steady state





Time evolution of current in response to bias switched on at time t = 0,

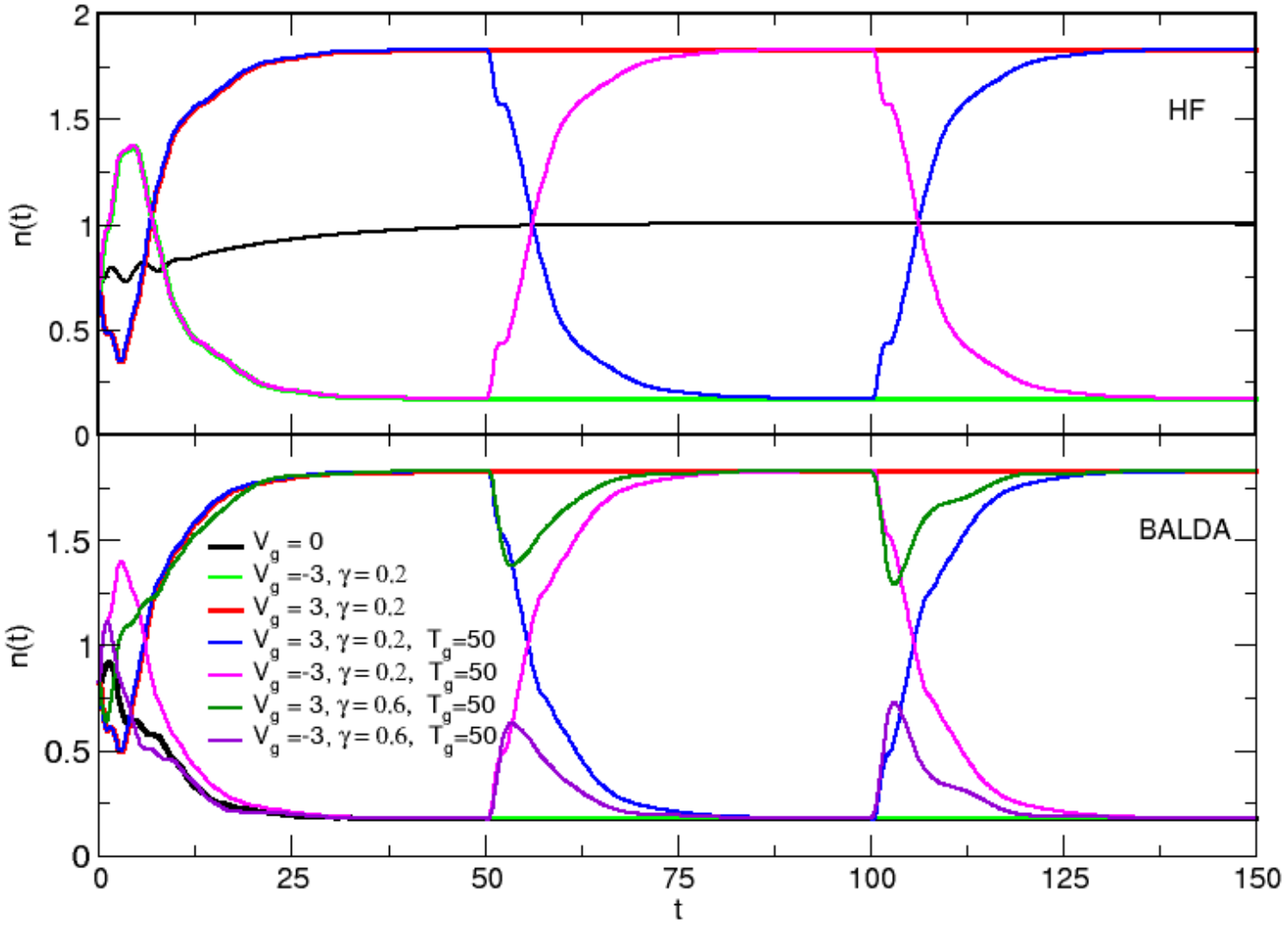
Fermi energy $\varepsilon_F = 0.3$ a.u.

Steady state coincides with Landauer formula

and is reached after a few femtoseconds

Can there be more than one steady state?

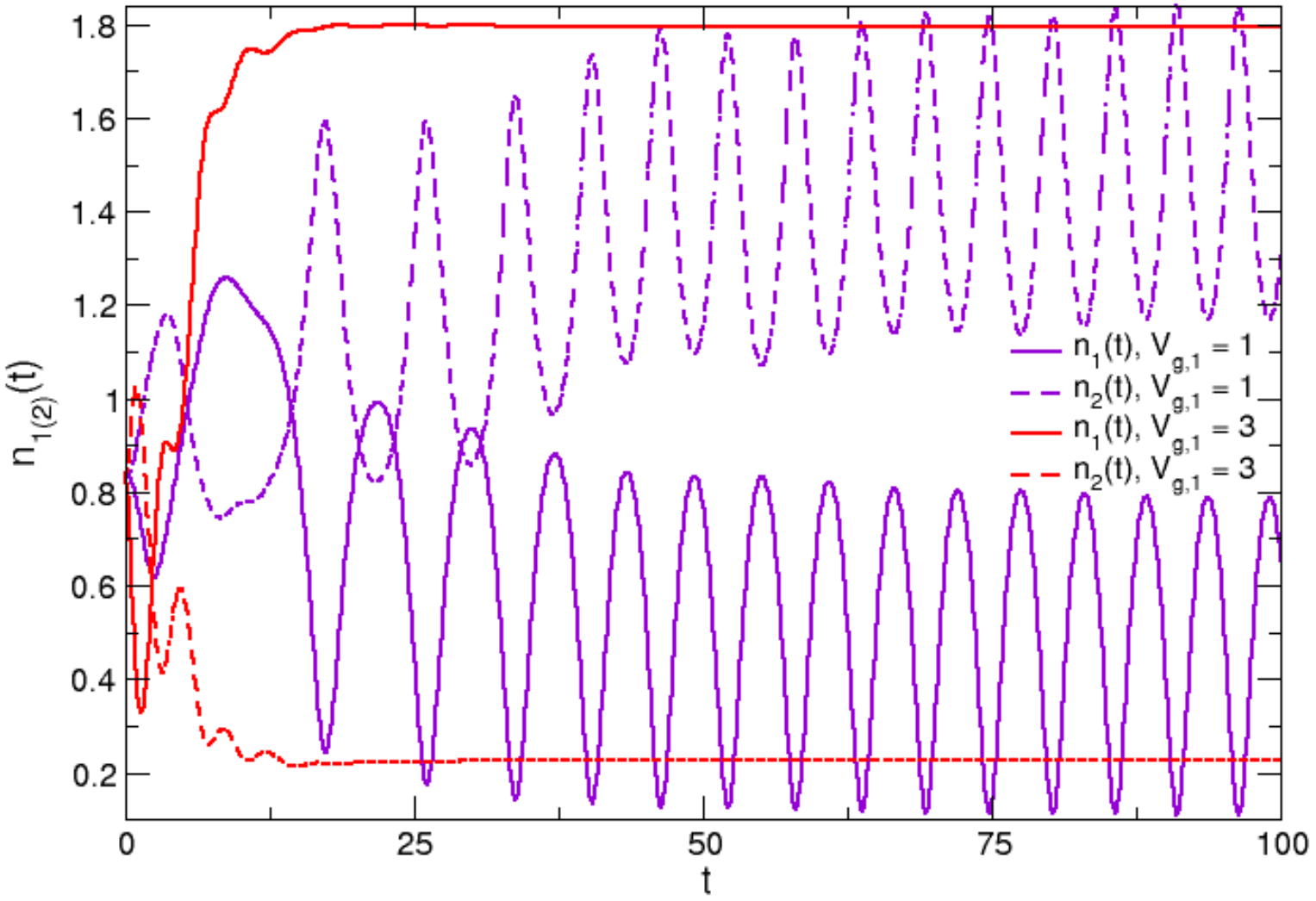
Multi-stability in TDHF and TDDFT for one-site Anderson model



E. Khosravi, A.M. Uimonen, A. Stan, G. Stefanucci, S. Kurth, R. van Leeuwen, E.K.U.G. Phys. Rev. B 85, 075103 (2012)

Is there always a steady state?

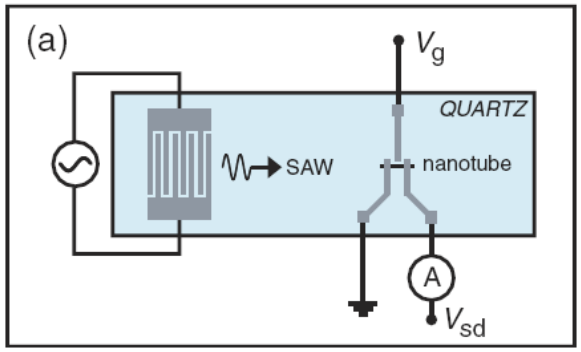
No steady state in two-site Anderson model

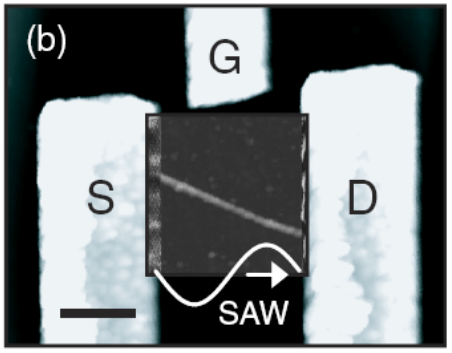


ELECTRON PUMP

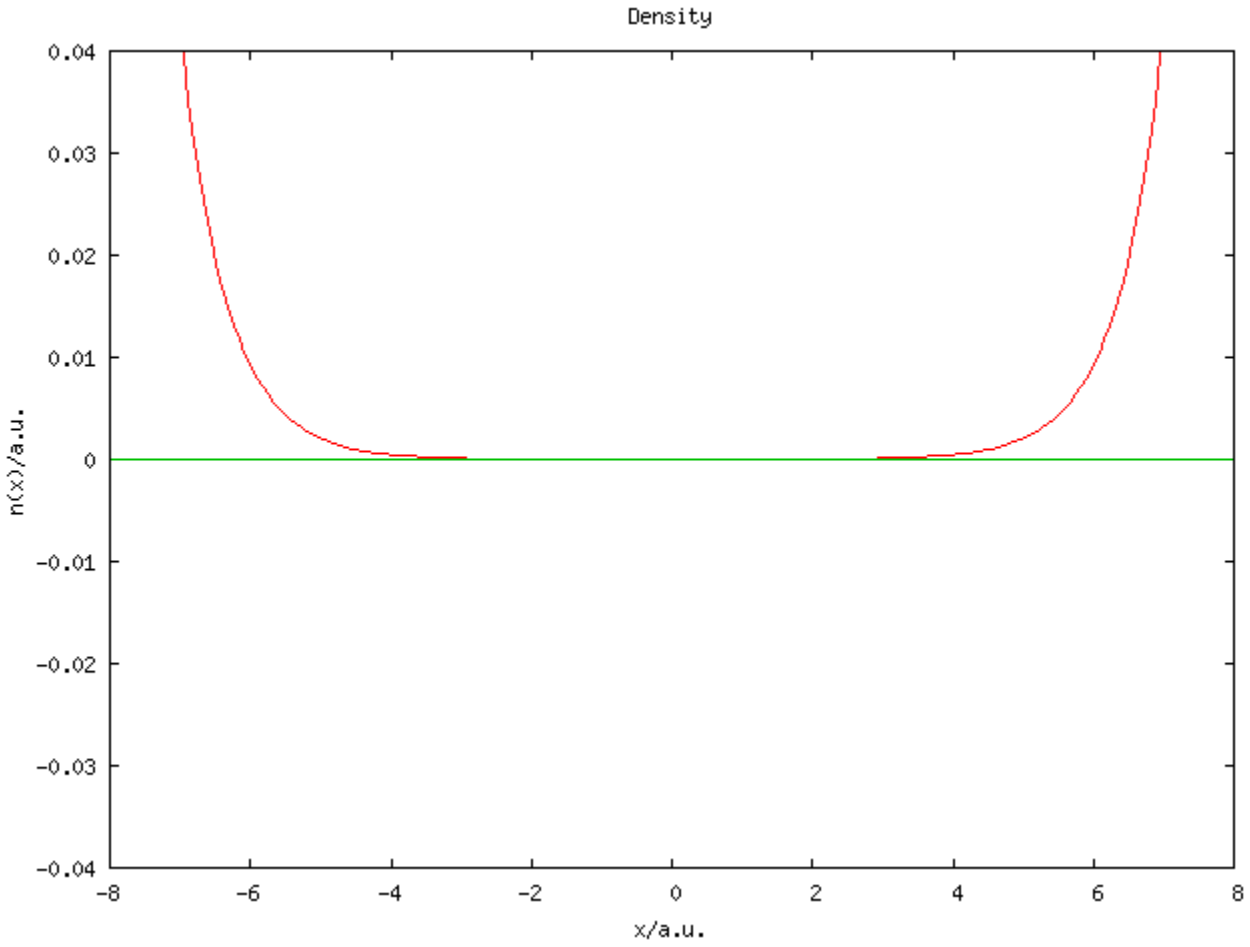
Device which generates a net current between two electrodes (with no static bias) by applying a time-dependent potential in the device region

Experimental realization : Pumping through carbon nanotube by surface acoustic waves on piezoelectric surface (Leek et al, PRL 95, 256802 (2005))



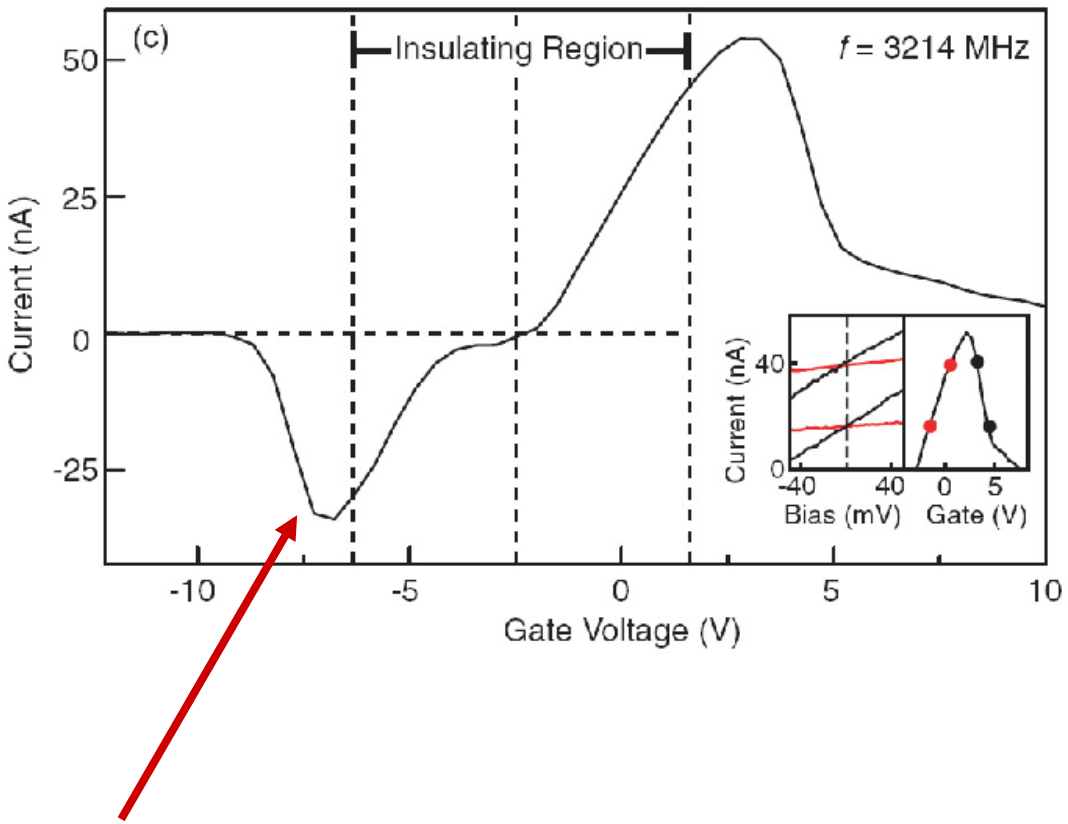


Pumping through a square barrier (of height 0.5 a.u.) using a travelling wave in device region
$U(x,t) = U_o \sin(kx-\omega t)$ ($k = 1.6$ a.u., $\omega = 0.2$ a.u. Fermi energy = 0.3 a.u.)

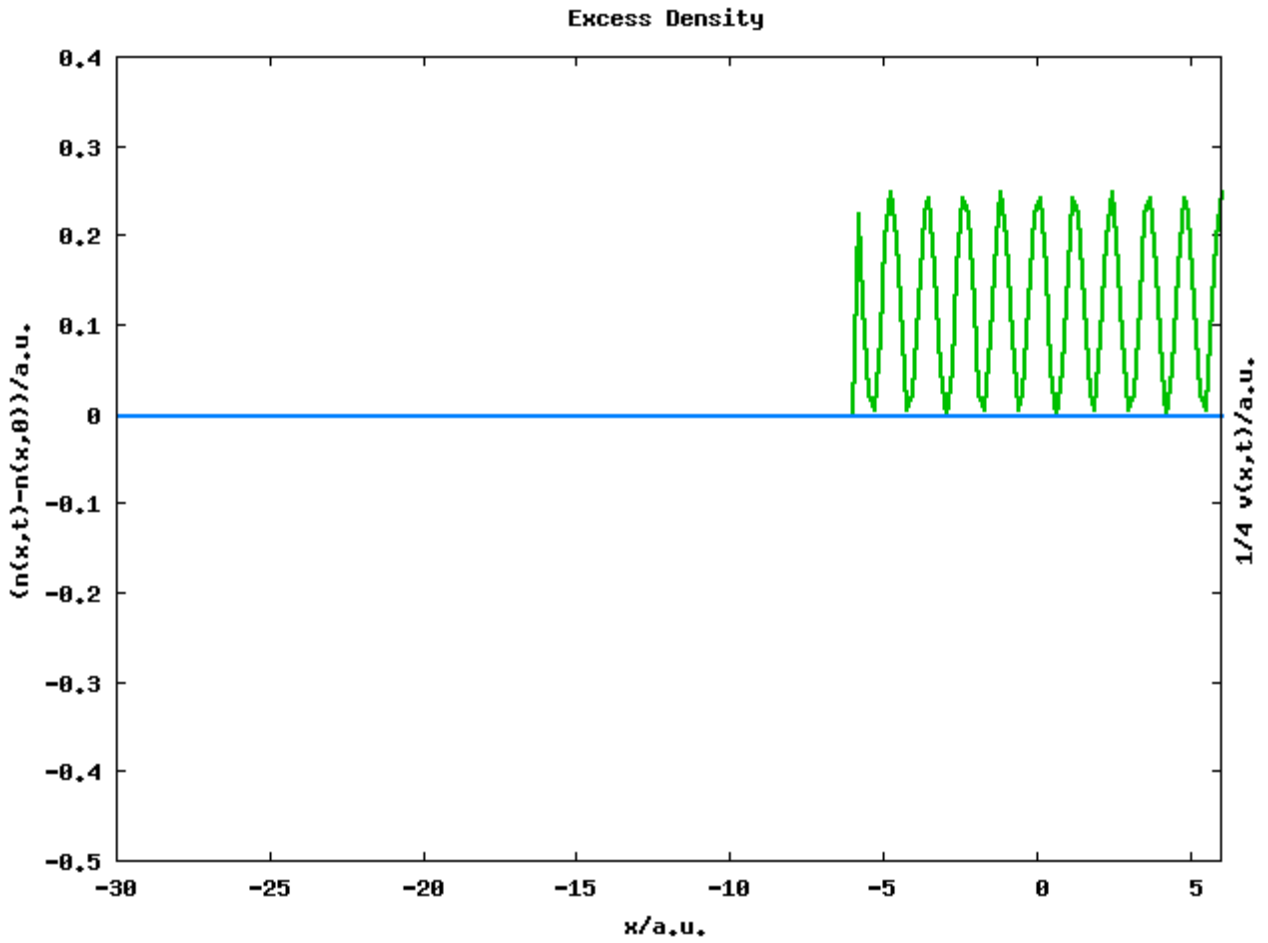


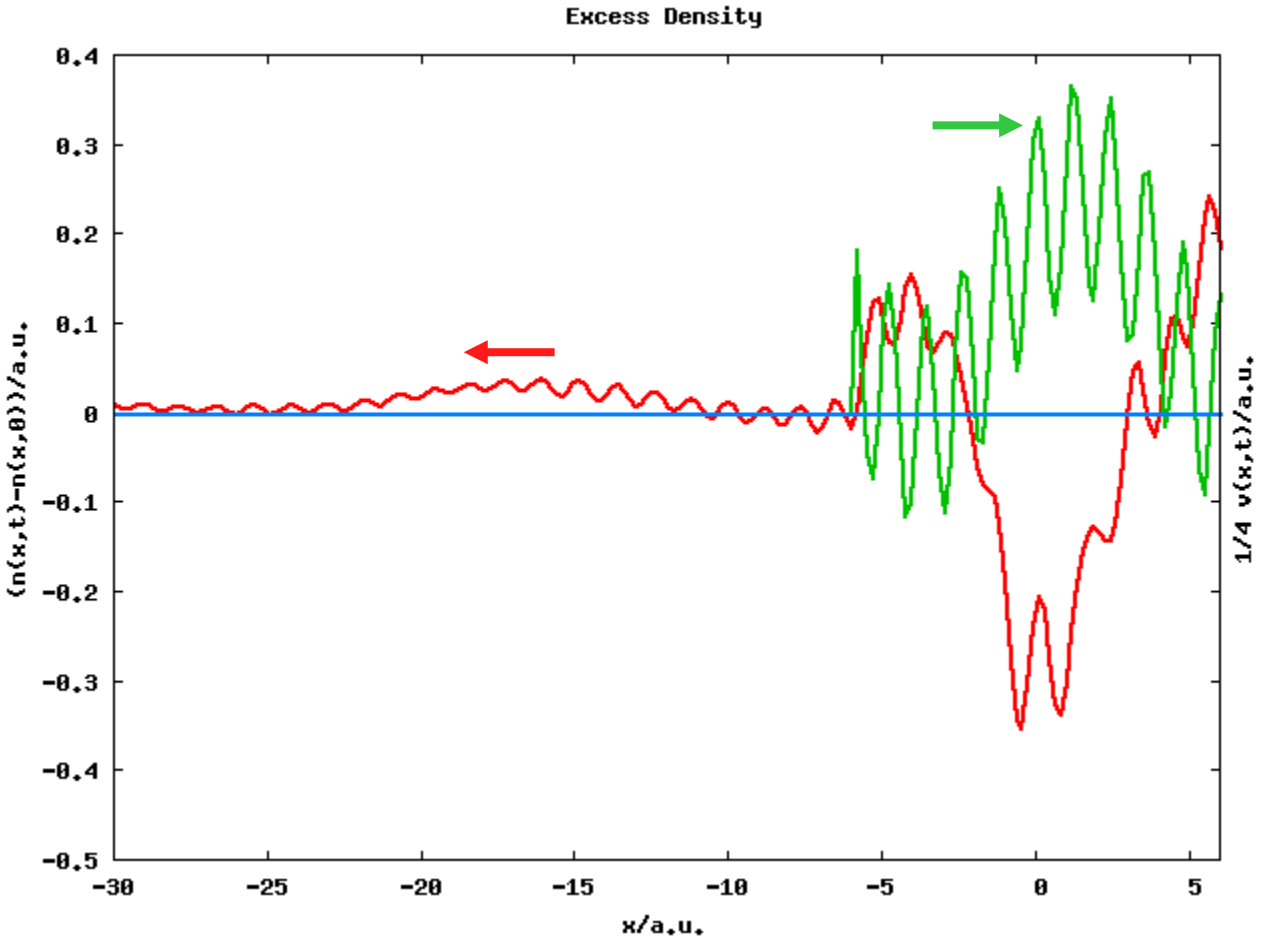
Archimedes' screw: patent 200 b.c.

Experimental result:



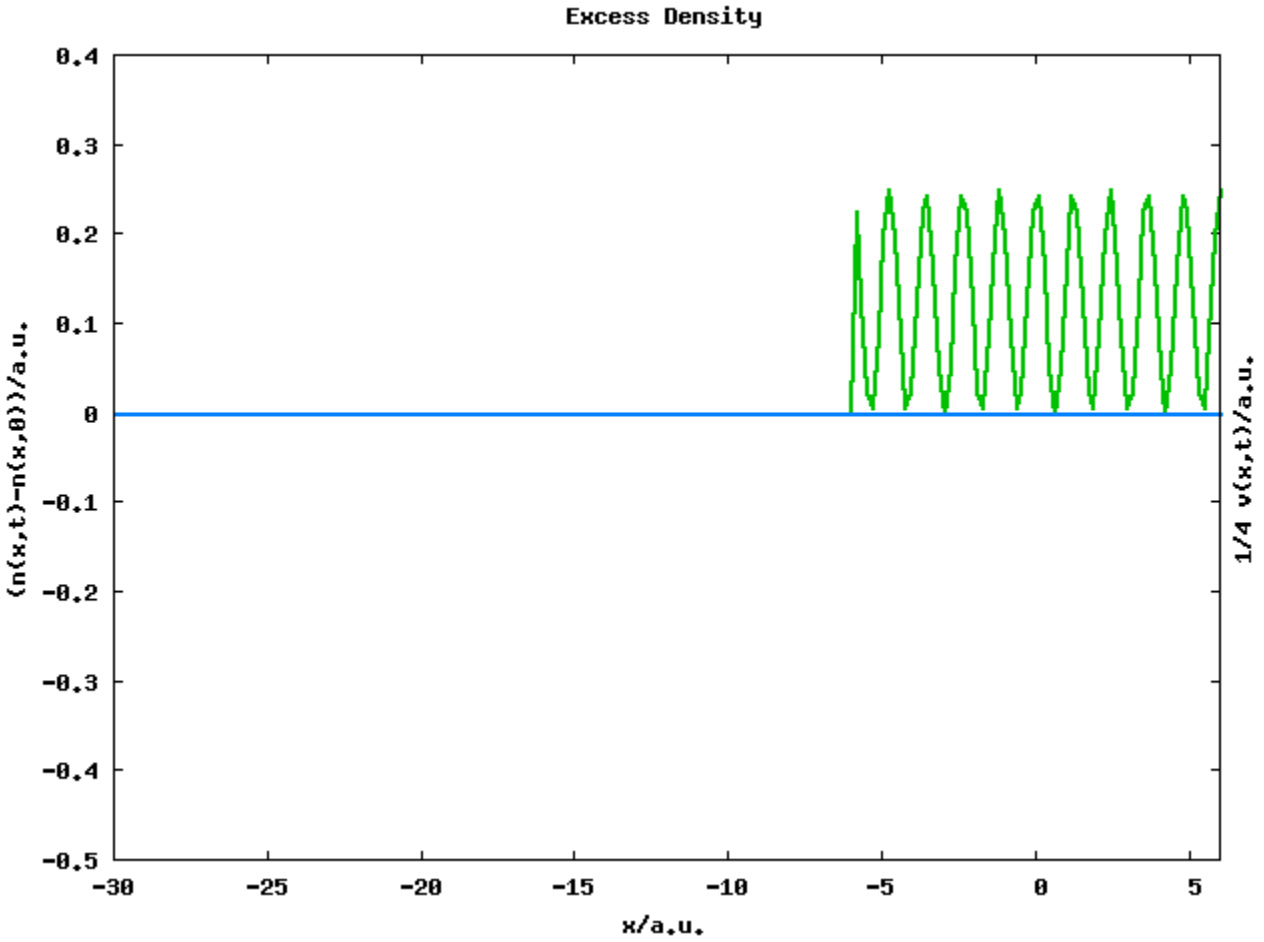
Current flows in direction opposite to sound wave


Excess Density
(n(x,t)-n(x,0))/a.u.
1/4 v(x,t)/a.u.
x/a.u.
0.4
0.3
0.2
0.1
0
-0.1
-0.2
-0.3
-0.4
-0.5
-30
-25
-20
-15
-10
-5
0
5



Current goes in direction opposite to the external field !!

G. Stefanucci, S. Kurth, A. Rubio, E.K.U. Gross, Phys. Rev. B 77, 075339 (2008)



G. Stefanucci, S. Kurth, A. Rubio, E.K.U. Gross, Phys. Rev. B 77, 075339 (2008)

Time-Dependent Electron Localization Function (TD-ELF)

Time-Dependent Electron Localization Function (TD-ELF)

GOAL

Time-resolved visualization of the breaking and formation of chemical bonds.

Time-Dependent Electron Localization Function (TD-ELF)

How can one give a rigorous mathematical meaning to chemical concepts such as

- **Single, double, triple bonds**
- **Lone pairs**

Note:

- **Density $\rho_\sigma(r)$ is not useful!**
- **Orbitals are ambiguous (w.r.t. unitary transformations)**

$$D_\sigma(\vec{r},\vec{r}') = \sum_{\sigma_3\sigma_4\ldots\sigma_N} \int d^3r_3 \ldots \int d^3r_N \left|\Psi(\vec{r}\sigma, \vec{r}'\sigma, \vec{r}_3\sigma_3 \ldots, \vec{r}_N\sigma_N)\right|^2$$

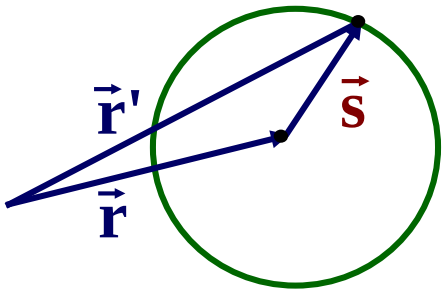
= diagonal of two-body density matrix

= probability of finding an electron with spin σ at $\vec{r}$ and another electron with the same spin at $\vec{r}'$.

$$P_\sigma(\vec{r},\vec{r}') := \frac{D_{\sigma\sigma}(\vec{r},\vec{r}')}{\rho_\sigma(\vec{r})}$$

= conditional probability of finding an electron with spin σ at $\vec{r}'$ if we know with certainty that there is an electron with the <u>same</u> spin at $\vec{r}$.

Coordinate transformation



If we know there is an electron with spin σ at $\vec{\mathbf{r}}$, then $P_\sigma\left(\vec{\mathbf{r}},\vec{\mathbf{r}}+\vec{\mathbf{s}}\right)$ is the (conditional) probability of finding another electron at $\vec{\mathbf{r}}+\vec{\mathbf{s}}$

Spherical average $$p_\sigma\left(\vec{r},|\vec{s}|\right)=\frac{1}{4\pi}\int_0^{\pi}\sin\theta d\theta\int_0^{2\pi}d\varphi P_\sigma\left(\vec{r},|\vec{s}|,\theta,\varphi\right)$$

If we know there is an electron with spin σ at $\vec{\mathbf{r}}$, then $p_\sigma\left(\vec{\mathbf{r}},\mathbf{s}\right)$ is the conditional probability of finding another electron at the distance **s** from .

Expand in a Taylor series:

$$p_\sigma\left(\vec{r},s\right)=\underbrace{p_\sigma\left(\vec{r},0\right)}_{0}+\underbrace{\left.\frac{dp_\sigma\left(\vec{r},s\right)}{ds}\right|_{s=0}}_{0}\cdot s+\frac{1}{3}C_\sigma\left(\vec{r}\right)s^2$$

The first two terms vanish.

$C_\sigma(\vec{r})$ is a measure of electron localization.

Why? $C_\sigma(\vec{r})$, being the s^2-coefficient, gives the probability of finding a second <u>like-spin</u> electron <u>very near</u> the reference electron. If this probability <u>very near</u> the reference electron is low then this reference electron must be very localized.

$C_\sigma(\vec{r})$ small means strong localization at $\vec{r}$

C_σ is always ≥ 0 (because p_σ is a probability) and $C_\sigma(\vec{r})$ is not bounded from above.

Define as a useful visualization of localization
(A.D. Becke, K.E. Edgecombe, JCP 92, 5397 (1990))

$$\mathrm{ELF} = \frac{1}{1+\left(\frac{C_\sigma(\vec{r})}{C_\sigma^{\mathrm{uni}}(\vec{r})}\right)^2}$$

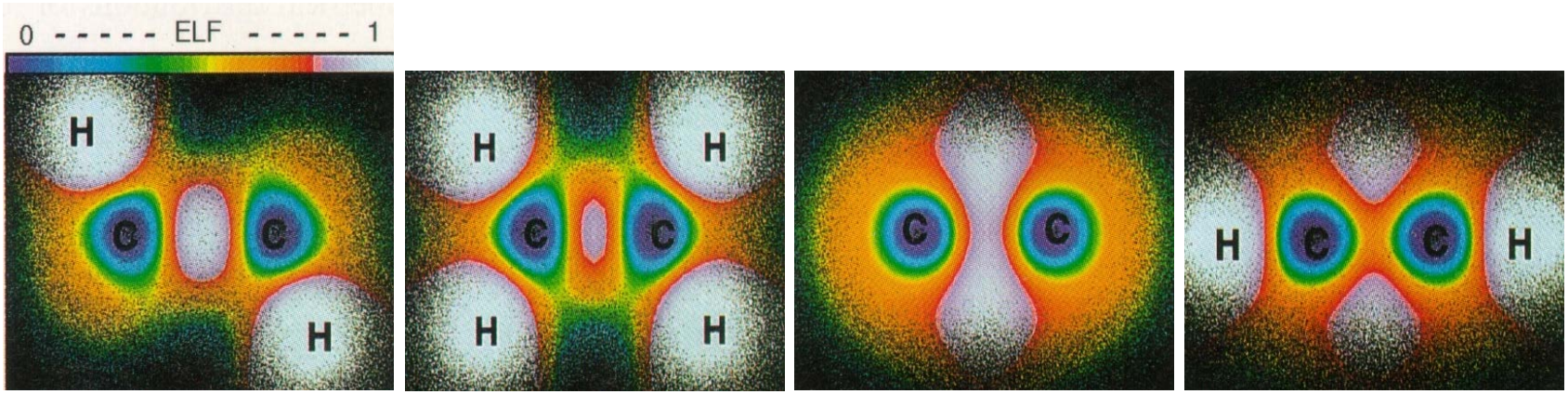
where

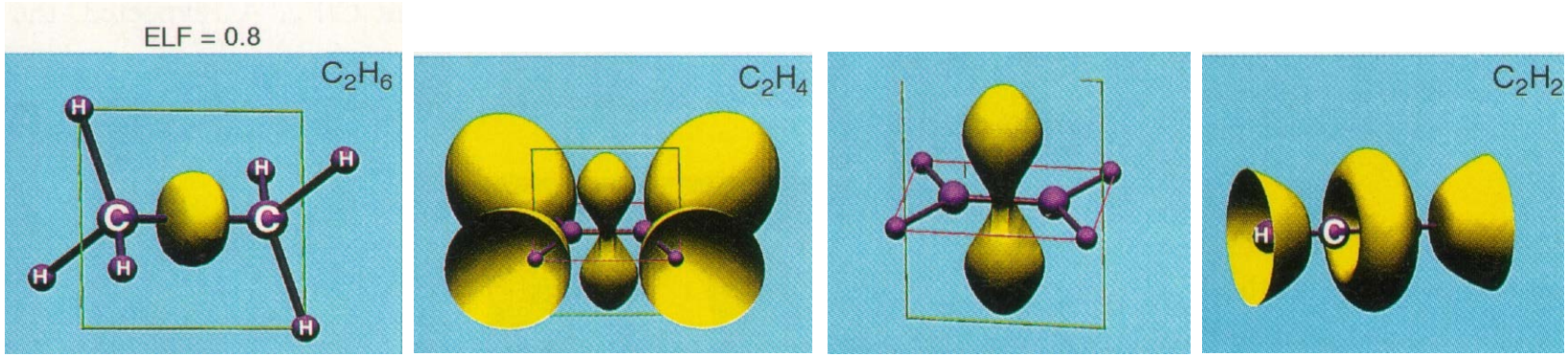
$$C_\sigma^{\mathrm{uni}}(\vec{r}) = \frac{3}{5}(6\pi^2)^{2/3}\rho_\sigma^{5/3}(\vec{r}) = \tau_\sigma^{\mathrm{uni}}(\vec{r})$$

is the kinetic energy density of the uniform gas.

Advantage: ELF is dimensionless and $0 \leq \mathrm{ELF} \leq 1$

ELF

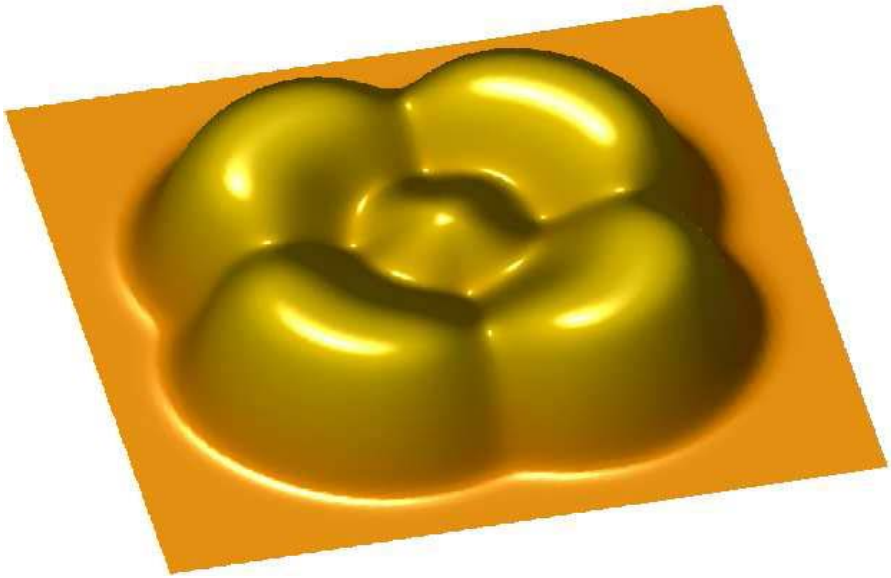




A. Savin, R. Nesper, S. Wengert, and T. F. Fässler, Angew. Chem. Int. Ed. 36, 1808 (1997)

12-electron 2D quantum dot with four minima

Density



ELF

E. Räsänen, A. Castro and E.K.U. Gross, Phys. Rev. B 77, 115108 (2008).

For a determinantal wave function one obtains

in the static case:

$$C_\sigma^{det}\left(\vec{r}\right) = \sum_{i=1}^{N_\sigma} \left|\nabla\varphi_{i\sigma}\left(\vec{r}\right)\right|^2 - \frac{1}{4}\frac{\left(\nabla\rho_\sigma\left(\vec{r}\right)\right)^2}{\rho_\sigma\left(\vec{r}\right)}$$

(A.D. Becke, K.E. Edgecombe, JCP 92, 5397 (1990))

in the time-dependent case:

$$C_\sigma^{det}\left(\vec{r},t\right) = \sum_{i=1}^{N_\sigma} \left|\nabla\varphi_{i\sigma}\left(\vec{r},t\right)\right|^2 - \frac{1}{4}\frac{\left(\nabla\rho_\sigma\left(\vec{r},t\right)\right)^2}{\rho_\sigma\left(\vec{r},t\right)} - j_\sigma\left(\vec{r},t\right)^2 \Big/ \rho_\sigma\left(\vec{r},t\right)$$

(T. Burnus, M. Marques, E.K.U.G., PRA (Rapid Comm) 71, 010501 (2005))

TDELF movies produced from TD Kohn-Sham equations

$$i\hbar\frac{\partial}{\partial t}\varphi_j(rt) = \left(-\frac{\hbar^2\nabla^2}{2m} + v_{KS}[\rho](rt)\right)\varphi_j(rt)$$

$$v_{KS}[\rho(r't')](rt) = v(rt) + \int d^3r' \frac{\rho(r't)}{|r-r'|} + v_{xc}[\rho\,(r't')](r\,t)$$

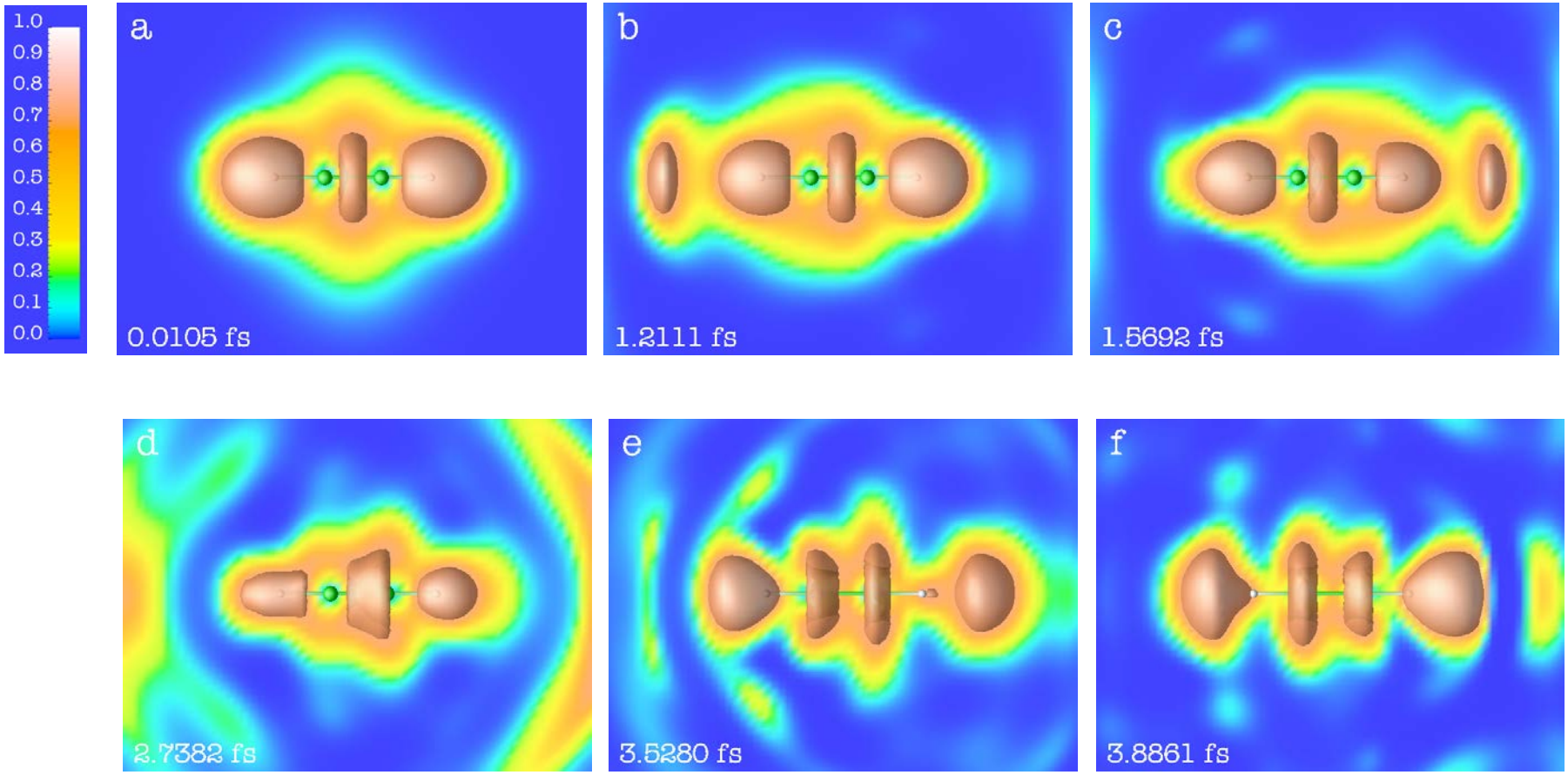
propagated numerically on real-space grid using **octopus** code

***octopus: a tool for the application of time-dependent density functional theory*, A. Castro, M.A.L. Marques, H. Appel, M. Oliveira, C.A. Rozzi, X. Andrade, F. Lorenzen, E.K.U.G., A. Rubio, Physica Status Solidi 243, 2465 (2006).**

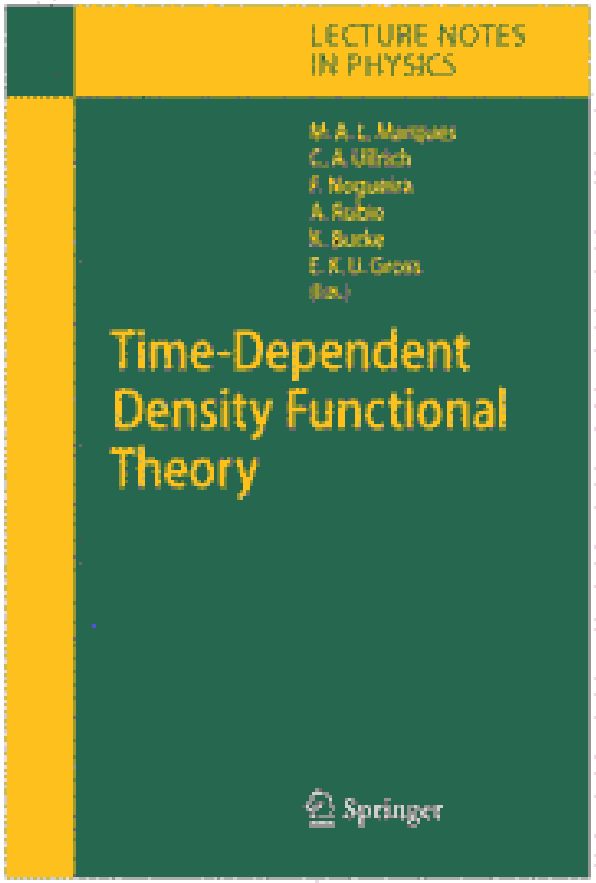
T. Burnus, M. Marques, E.K.U.G, PRA (Rapid Comm) 71, 010501 (2005)

TDELF for acetylene in strong laser field

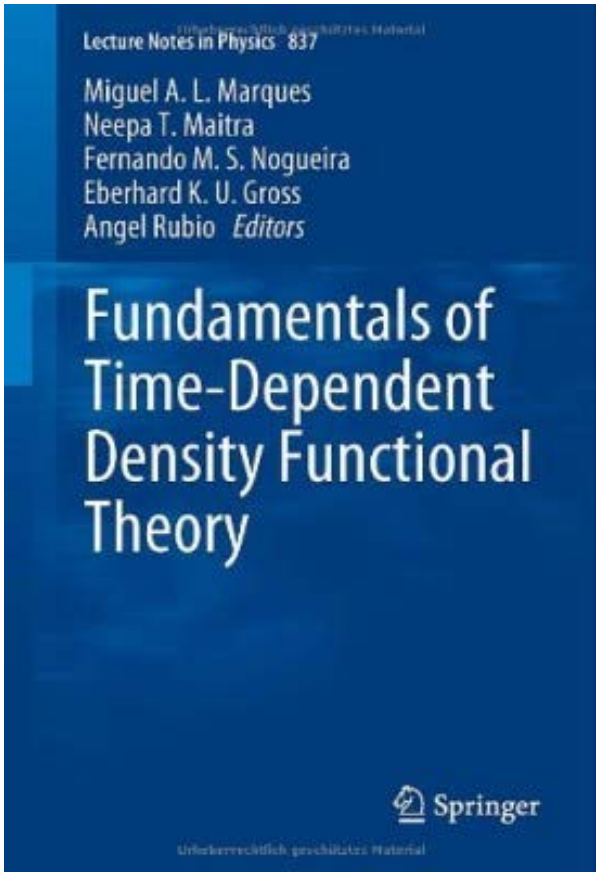
($\hbar\omega$ = 17.15 eV, I = 1.2×10^{14} W/cm^2)



More on TDDFT…..



Lecture Notes in Physics 706 (Springer, 2006)



Lecture Notes in Physics 837 (Springer, 2012)