

# Time-dependent current-density-functional theory for extended systems

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Theory Days, November 29, 2012

## Outline

- ▶ Motivation: Polarization in extended systems
- ▶ Why do we need TD-current-DFT?
- ▶ The Vignale-Kohn functional
- ▶ Results
- ▶ Magnetization
- ▶ Conclusions

## Motivation

Description of the **electric response of extended systems** to a perturbation

### Macroscopic electric field

$$\mathbf{E}_{mac}(t) = \frac{1}{V} \int_V [\mathbf{E}_{ext}(\mathbf{r}, t) + \mathbf{E}_{ind}(\mathbf{r}, t)] d\mathbf{r}$$

### Macroscopic polarization

$$\mathbf{P}_{mac}(t) = \int dt' \chi_e(t - t') \cdot \mathbf{E}_{mac}(t')$$

$\chi_e$  is a **bulk property**

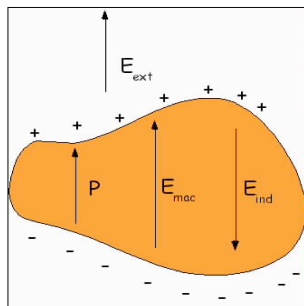
### Macroscopic dielectric function:

$$\epsilon_{M,ij} = 1 + 4\pi\chi_{e,ij}$$

Method:

A **density-functional theory**:

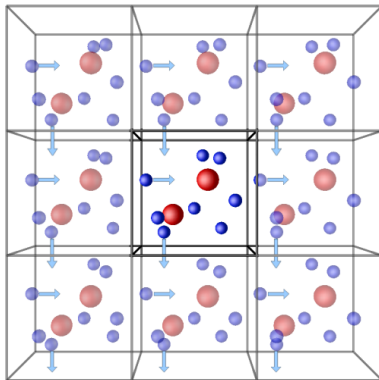
- ▶ All physical observables can be obtained exactly
- ▶ Allows for efficient calculations



## Periodic boundary conditions

Extended systems :

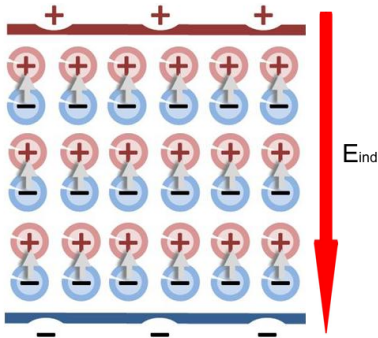
It is convenient to use **periodic boundary conditions** (PBC) within a Born-von Kármán cell approach to take advantage of **translational symmetry**



## Extended systems

PBC  $\rightarrow$  artificial removal of the surface density

Surface density is important to describe the macroscopic induced field



We need a DFT approach able to calculate polarization in systems described by PBC

## Polarization

We need a definition of **polarization density**  $\mathbf{P}(\mathbf{r}, t)$  **compatible with PBC**

A **natural definition** of  $\mathbf{P}(\mathbf{r}, t)$  should obey

$$\mathbf{p}(t) = \int_V \mathbf{P}(\mathbf{r}, t) d\mathbf{r} = \int_V \mathbf{r} \rho(\mathbf{r}, t) d\mathbf{r}$$

with  $\mathbf{p}(t)$  the total electric dipole moment

However,  $\mathbf{r}$  is **incompatible with PBC**

Using the **continuity equation**

$$\dot{\rho}(\mathbf{r}, t) + \nabla \cdot \mathbf{j}(\mathbf{r}, t) = 0$$

we can rewrite  $\mathbf{p}(t)$  as

$$\mathbf{p}(t) = \int_{t_0}^t dt' \int_V \mathbf{j}(\mathbf{r}, t') d\mathbf{r} - \int_{t_0}^t dt' \int_{\partial V} \mathbf{r} \hat{\mathbf{n}} \cdot \mathbf{j}(\mathbf{r}, t') d\mathbf{r}$$

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Therefore

$$\mathbf{P}(\mathbf{r}, t) = \int_{t_0}^t dt' \mathbf{j}(\mathbf{r}, t')$$



## Maxwell's equations

The polarization density

$$\mathbf{P}(\mathbf{r}, t) = \int_{t_0}^t dt' \mathbf{j}(\mathbf{r}, t')$$

is also **compatible** with **Maxwell's equations**:

"microscopic"

$$\nabla \cdot \mathbf{E} = 4\pi\rho_{tot}$$

$$c\nabla \times \mathbf{B} - \dot{\mathbf{E}} = 4\pi\mathbf{j}_{tot}$$

$$\nabla \cdot \mathbf{B} = 0$$

$$c\nabla \times \mathbf{E} + \dot{\mathbf{B}} = 0$$

"macroscopic"

$$\nabla \cdot \mathbf{D} = 4\pi\rho_{ext}$$

$$c\nabla \times \mathbf{H} - \dot{\mathbf{D}} = 4\pi\mathbf{j}_{ext}$$

$$\mathbf{D} = \mathbf{E} + 4\pi\mathbf{P}$$

$$\mathbf{H} = \mathbf{B} - 4\pi\mathbf{M}$$

From the Maxwell equations it follows that

$$\mathbf{j}(\mathbf{r}, t) = \dot{\mathbf{P}}(\mathbf{r}, t) + c\nabla \times \mathbf{M}(\mathbf{r}, t)$$

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From the Maxwell equations it follows that

$$\mathbf{j}(\mathbf{r}, t) = \dot{\mathbf{P}}(\mathbf{r}, t) + \cancel{c\nabla \times \mathbf{M}(\mathbf{r}, t)}$$

Gauge freedom:  $\mathbf{M}(\mathbf{r}, t) = \mathbf{0}$

## Response of extended systems

### Macroscopic electric field

$$\mathbf{E}_{mac}(t) = \frac{1}{V} \int_V [\mathbf{E}_{ext}(\mathbf{r}, t) + \mathbf{E}_{ind}(\mathbf{r}, t)] d\mathbf{r}'$$

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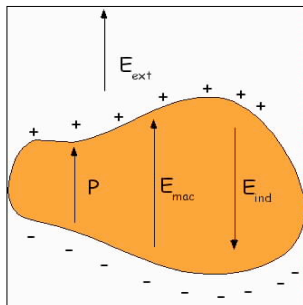
$\chi_e$  is a **bulk property**

$$\mathbf{P}_{mac}(t) = \int dt' \chi_e(t - t') \cdot \mathbf{E}_{mac}(t')$$

**Macroscopic dielectric function:**

$$\epsilon_{M,ij} = 1 + 4\pi\chi_{e,ij}$$

F. Kootstra, P. L. de Boeij, and J. G. Snijders, J. Chem. Phys. 112, 6517 (2000)



## Which DFT?

Semiconductor described by PBC in an electric field:

The **perturbing potential** is **not a functional** of the **bulk density** alone.  
It is a functional of  $\rho_{bulk}$  and  $\mathbf{P}_{mac}$

X. Gonze, Ph. Ghosez, and R.W. Godby, Phys. Rev. Lett. 74, 4035 (1995)

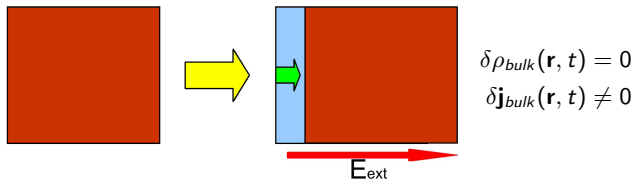
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Example: response of a homogeneous electron gas in a uniform field



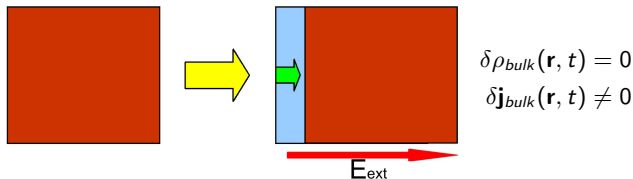
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The **perturbing potential** is a functional of the **bulk current**

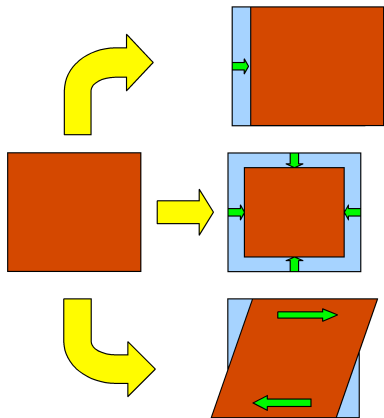
Current density is **local** indicator of **global** changes in the system

We need **TD Current-DFT!**

## TDCDFT vs TDDFT

TDCDFT allows description of

- Uniform macroscopic currents
- Transverse response.
- General electromagnetic fields (vector potentials)
- Nonlocal xc effects using local functional of the current.



Macroscopic response

$$\mathbf{j}(t) = \frac{1}{V} \int_V d\mathbf{r} \mathbf{j}(\mathbf{r}, t)$$

Longitudinal response

$$\nabla \cdot \mathbf{j}(\mathbf{r}, t) = -\frac{\partial}{\partial t} \rho(\mathbf{r}, t) \neq 0$$

$$\nabla \times \mathbf{j}(\mathbf{r}, t) = 0$$

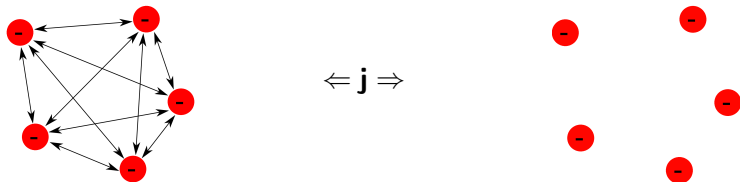
Transverse response

$$\nabla \cdot \mathbf{j}(\mathbf{r}, t) = -\frac{\partial}{\partial t} \rho(\mathbf{r}, t) = 0$$

$$\nabla \times \mathbf{j}(\mathbf{r}, t) \neq 0$$

## TDCDFT: Kohn-Sham equations

The full **interacting** system can be **mapped** onto a **Kohn-Sham** system



$$H_{KS}(\mathbf{r}_i, t) = \sum_{i=1}^N h_{KS}(\mathbf{r}_i, t) \quad h_{KS}(\mathbf{r}, t) = \frac{1}{2} [-i\nabla + \mathbf{A}_{KS}(\mathbf{r}, t)]^2 + v_{KS}(\mathbf{r}, t)$$
$$i \frac{\partial}{\partial t} \phi_i(\mathbf{r}, t) = h_{KS}(\mathbf{r}, t) \phi_i(\mathbf{r}, t)$$

$$\mathbf{j}(\mathbf{r}, t) = \sum_{i=1}^N \left[ \frac{1}{2i} \phi_i^*(\mathbf{r}, t) \nabla \phi_i(\mathbf{r}, t) + \text{c.c.} \right] + \rho(\mathbf{r}, t) \mathbf{A}_{KS}(\mathbf{r}, t)$$

$$\rho(\mathbf{r}, t) = \sum_{i=1}^N |\phi_i(\mathbf{r}, t)|^2,$$

$$v_{KS}[\mathbf{j}] = v_{\text{ext}} + v_H + v_{xc}$$

$$\mathbf{A}_{KS}[\mathbf{j}] = \mathbf{A}_{\text{ext}} + \mathbf{A}_{xc}$$

Equations need to be solved **self-consistently**



## Microscopic Coulomb gauge

Choice of **gauge**:

$V_{KS}$  is lattice periodic and can only contain **microscopic** contributions

$$V_{KS} = V_{H,mic} + V_{xc,mic}$$

$\mathbf{A}_{KS}$  contains all **macroscopic** contributions

$$\mathbf{A}_{KS} = \mathbf{A}_{mac} + \mathbf{A}_{xc}$$

where

$$\mathbf{A}_{mac} = \mathbf{A}_{ext} + \mathbf{A}_{ind,mac}$$

We fix  $\mathbf{E}_{mac} (= -\dot{\mathbf{A}}_{mac})$  and calculate **self-consistently** the corresponding  $\mathbf{j}$

F. Kootstra, P. L. de Boeij, and J. G. Snijders, J. Chem. Phys. 112, 6517 (2000)  
AB, P. L. de Boeij, and R. van Leeuwen, Phys. Rev. B 71, 155104 (2005)

## TDCDFT: linear response

Small perturbations  $\rightarrow$  **linear response** regime.

$$\delta\rho(\mathbf{r}, \omega) = \int d\mathbf{r}' \chi_{\rho j_p}^{KS}(\mathbf{r}, \mathbf{r}', \omega) \cdot \delta\mathbf{A}_{KS}(\mathbf{r}', \omega) + \int d\mathbf{r}' \chi_{\rho\rho}^{KS}(\mathbf{r}, \mathbf{r}', \omega) \delta v_{KS}(\mathbf{r}', \omega)$$

$$\delta\mathbf{j}(\mathbf{r}, \omega) = \int d\mathbf{r}' \chi_{j j}^{KS}(\mathbf{r}, \mathbf{r}', \omega) \cdot \delta\mathbf{A}_{KS}(\mathbf{r}', \omega) + \int d\mathbf{r}' \chi_{j\rho}^{KS}(\mathbf{r}, \mathbf{r}', \omega) \delta v_{KS}(\mathbf{r}', \omega)$$

with

$$\chi_{j j}^{KS}(\mathbf{r}, \mathbf{r}', \omega) = \chi_{j\rho j_p}^{KS}(\mathbf{r}, \mathbf{r}', \omega) + \rho_0(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')$$

The KS response functions depend on **ground-state properties** only.

$$\chi_{ab}^{KS}(\mathbf{r}, \mathbf{r}', \omega) = \sum_{i,j} (f_i - f_j) \frac{[\phi_i^*(\mathbf{r})\hat{a}(\mathbf{r})\phi_j(\mathbf{r})][\phi_j^*(\mathbf{r}')\hat{b}(\mathbf{r}')\phi_i(\mathbf{r}')] }{\omega - (\epsilon_j - \epsilon_i) + i\eta}$$

where  $\hat{a}(\mathbf{r})$  and  $\hat{b}(\mathbf{r})$  are either  $\hat{\rho} = 1$  or  $\hat{\mathbf{j}}_p = 1/(2i)(\nabla - \nabla^\dagger)$

## The adiabatic local-density approximation

The **adiabatic local-density approximation** (ALDA) is given by

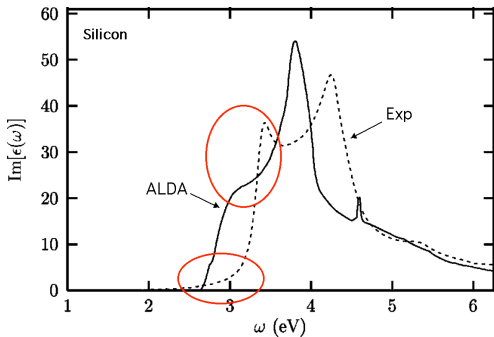
$$\begin{aligned}\delta v_{xc,mic}(\mathbf{r}, \omega) &= \int d\mathbf{r}' f_{xc}^{ALDA}(\mathbf{r}, \mathbf{r}', \omega) \delta\rho(\mathbf{r}', \omega) \\ \delta \mathbf{A}_{xc}(\mathbf{r}, \omega) &= 0\end{aligned}$$

where the **exchange-correlation kernel**  $f_{xc}^{ALDA}$  is given by

$$f_{xc}^{ALDA}(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \left. \frac{d^2 \epsilon_{xc}^{hom}}{d\rho^2} \right|_{\rho=\rho_0(\mathbf{r})}$$

The ALDA kernel is **frequency independent** and **local**

## The adiabatic local-density approximation

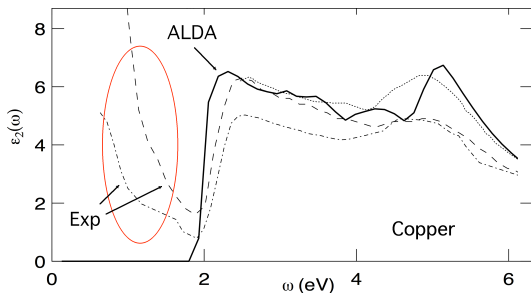


- Too low onset of the absorption
- No excitonic effects

F. Kootstra, P. L. de Boeij, and J. G. Snijders, J. Chem. Phys. 112, 6517 (2000)

## The adiabatic local-density approximation

ALDA results are not always satisfactory, especially for extended systems.



- No Drude-tails in absorption spectra of metals

P. Romaniello and P. L. de Boeij, Phys. Rev. B 71, 155108 (2005)

- Overestimation of static polarizabilities of conjugated polymers

## Beyond the adiabatic local-density approximation

How to find better xc functionals?

For any DFT: No simple systematic nor intuitive way to improve on xc functionals.

Impose exact constraints

Example: **zero-force theorem**: xc potentials should not exert a net force on the system.

## Beyond the adiabatic local-density approximation

How to find better xc functionals?

For any DFT: No simple systematic nor intuitive way to improve on xc functionals.

Impose exact constraints

Example: **zero-force theorem**: xc potentials should not exert a net force on the system.

TDDFT:

$$\int d\mathbf{r}' \nabla' \rho_0(\mathbf{r}') f_{xc}(\mathbf{r}, \mathbf{r}', \omega) = \nabla v_{xc,0}(\mathbf{r})$$

**$\omega$ -dependent LDA is not compatible with basic conservation laws**

**TDCDFT**:  $\omega$ -dependent **local-current-density** approximation is compatible with basic conservation laws

G. Vignale, Phys. Lett. A 209, 206 (1995)

G. Vignale and W. Kohn, Phys. Rev. Lett. 77, 2037 (1996)

## An explicit current functional: The Vignale-Kohn functional

Vignale and Kohn considered a **slightly inhomogeneous** electron gas

$$\rho_0(\mathbf{r}) = \rho(1 + 2\gamma \cos(\mathbf{q} \cdot \mathbf{r})) \quad |\mathbf{q}| \ll k_F, \omega/v_F, \gamma \ll 1$$

that is **weakly perturbed**, i.e.,  $|\mathbf{k}| \ll k_F, \omega/v_F$



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that is **weakly perturbed**, i.e.,  $|\mathbf{k}| \ll k_F, \omega/v_F$

$f_{xc,ij}$  is completely **fixed** by imposing several exact **constraints**:

- zero-force theorem
- zero-torque theorem
- Onsager symmetry relation ( $f_{xc,ij}(\mathbf{r}, \mathbf{r}', \omega) = f_{xc,ji}(\mathbf{r}', \mathbf{r}, \omega)$ )
- a Ward identity

G. Vignale and W. Kohn, Phys. Rev. Lett. 77, 2037 (1996)

## An explicit current functional: The Vignale-Kohn functional

The **Vignale-Kohn functional** is a **nonadiabatic local** current-density approximation

$$i\omega A_{xc,i}^{VK}(\mathbf{r}, \omega) = \partial_i v_{xc}^{ALDA}(\mathbf{r}, \omega) - \frac{1}{\rho_0(\mathbf{r})} \sum_j \partial_j \sigma_{xc,ij}(\mathbf{r}, \omega)$$
$$\sigma_{xc,ij} = \tilde{\eta}_{xc} \left[ \partial_i u_j + \partial_j u_i - \frac{2}{3} \delta_{ij} (\nabla \cdot \mathbf{u}) \right] + \tilde{\zeta}_{xc} \delta_{ij} (\nabla \cdot \mathbf{u}),$$

where  $\mathbf{u}(\mathbf{r}, \omega) = \delta \mathbf{j}(\mathbf{r}, \omega) / \rho_0(\mathbf{r})$ .

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where  $\mathbf{u}(\mathbf{r}, \omega) = \delta \mathbf{j}(\mathbf{r}, \omega) / \rho_0(\mathbf{r})$ .

The coefficients  $\tilde{\eta}_{xc}$  and  $\tilde{\zeta}_{xc}$  are determined by kernels of the **homogeneous electron gas**

$$\tilde{\eta}_{xc}(\mathbf{r}, \omega) = -\frac{\rho_0^2(\mathbf{r})}{i\omega} f_{xcT}^{hom}(\rho_0, \omega)$$
$$\tilde{\zeta}_{xc}(\mathbf{r}, \omega) = -\frac{\rho_0^2(\mathbf{r})}{i\omega} \left[ f_{xcl}^{hom}(\rho_0, \omega) - \frac{4}{3} f_{xcT}^{hom}(\rho_0, \omega) - \frac{d^2 \epsilon_{xc}^{hom}}{d\rho}(\rho_0) \right].$$

G. Vignale and W. Kohn, Phys. Rev. Lett. 77, 2037 (1996)

G. Vignale, C. A. Ullrich, and S. Conti, Phys. Rev. Lett. 79, 4878 (1997)

## Parametrizations of the xc-kernels and static limit

**Parametrizations** for  $f_{xcL,T}^{hom}$  are given by:

- Conti, Nifosi, and Tosi (CNT), J. Phys. Condens. Matter 9, L475 (1997).
- Qian and Vignale (QV), Phys. Rev. B 65, 235121 (2002).

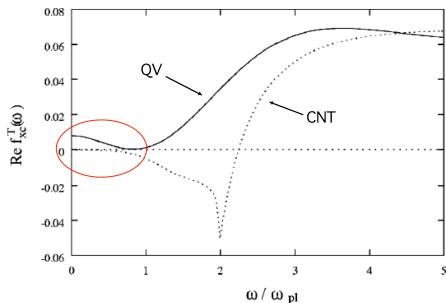
Important **difference**: treatment of the **static limit**

$$\lim_{\omega \rightarrow 0} \omega \tilde{\zeta}_{xc} = 0$$

$$\lim_{\omega \rightarrow 0} -i\omega \tilde{\eta}_{xc} = \rho_0^2 f_{xcT}^h(\omega = 0) = \mu_{xc}$$

CNT:  $\mu_{xc} = 0$  (ALDA)

QV:  $\mu_{xc} \neq 0$



## Simplified VK: A polarization functional

Neglect microscopic contributions to  $\delta \mathbf{A}_{xc}^{VK}$  and  $\delta \mathbf{j} \rightarrow$  polarization functional

$$\delta A_{xc,mac,i}(\omega) = \frac{i}{\omega} \sum_j Y_{ij}[\rho_0](\omega) P_{mac,j}(\omega)$$

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Effect of macroscopic vector potential can be applied **post-SCF**.

$$\chi_e(\omega) = (1 - \chi_e^{ALDA}(\omega) Y(\omega))^{-1} \chi_e^{ALDA}(\omega)$$

Absolute value of  $Y$  too big. Scale with empirical prefactor: 0.4.

P. L. de Boeij, F. Kootstra, AB, R. van Leeuwen, J.G. Snijders, JCP 115, 1995 (2001)

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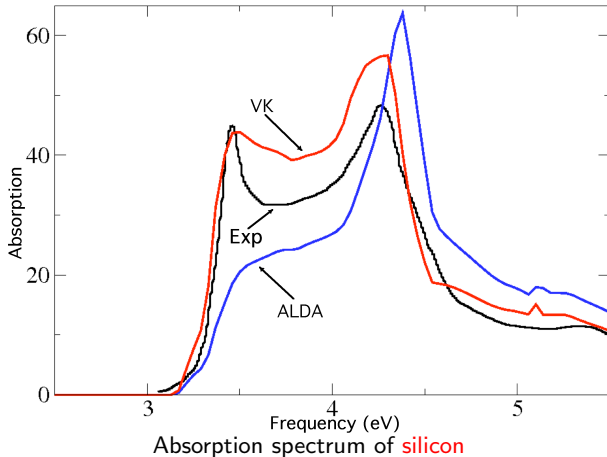
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Note: **Long-range** scalar  $f_{xc}$  equivalent to **local** tensor  $f_{xc,ij}$

$$f_{xc} = \frac{\alpha}{q^2} \Rightarrow f_{xc}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\alpha}{4\pi} \frac{1}{|\mathbf{r} - \mathbf{r}'|} \sim f_{xc,ij} = -\frac{\alpha}{4\pi} \frac{1}{\omega^2} \delta(\mathbf{r} - \mathbf{r}')$$

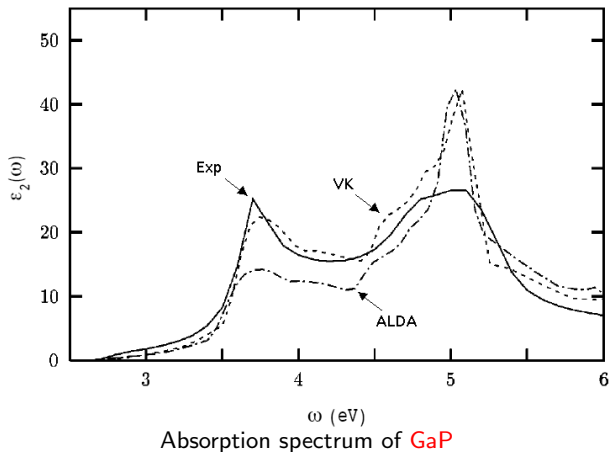
## Polarization functional: Excitonic effects



Long-range electron-hole effects can be described with a local functional of the current density



## Polarization functional: Excitonic effects



P. L. de Boeij, F. Kootstra, AB, R. van Leeuwen, J.G. Snijders, JCP 115, 1995 (2001)

## Implementation of the full VK functional

$\delta \mathbf{A}_{xc}^{VK}$ : **third-order derivatives** of the KS wave functions

$\delta \mathbf{A}_{xc}^{VK}$  can be written in a more convenient manner:

$$\delta \mathbf{A}_{xc}^{VK} = -\frac{i}{\omega} \nabla [\delta v_{xc}^{ALDA} + \delta u_{xc}] + \delta \mathbf{a}_{xc} + \nabla \times \delta \mathbf{b}_{xc}$$

where

$$\begin{pmatrix} \delta u_{xc} \\ i\omega \delta \mathbf{a}_{xc} \\ i\omega \delta \mathbf{b}_{xc} \end{pmatrix} = \begin{pmatrix} y_{\rho\rho} & y_{\rho j} & 0 \\ y_{j\rho} & y_{jj} & y_{jm} \\ y_{m\rho} & y_{mm} & 0 \end{pmatrix} \begin{pmatrix} \delta\rho \\ i\delta\mathbf{j}/\omega \\ i\delta\mathbf{m}/\omega \end{pmatrix}$$

with

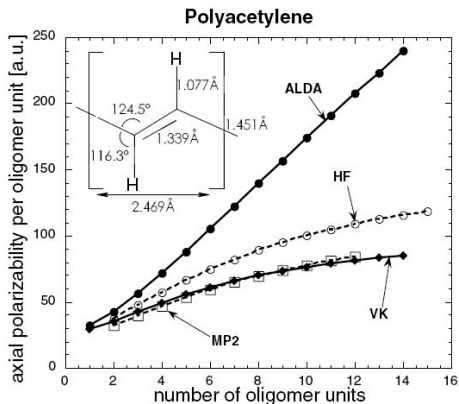
$$\delta \mathbf{m} = \nabla \times \delta \mathbf{j}$$

-**First-order derivatives** of the KS wave functions only

-The various **y-components** are **local**  $\omega$ -dependent functions of the ground-state density  $\rho_0$

AB, P. L. de Boeij, and R. van Leeuwen, Phys. Rev. B 71, 155104 (2005)

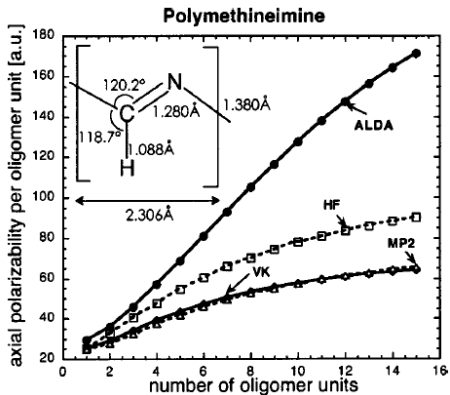
## Full VK: Static polarizabilities of conjugated polymers



- The **current density** can account for **counteracting xc-potential** induced by **charge buildup at end points**
- It is essential that  $\mu_{xc} \neq 0$ , otherwise ALDA results

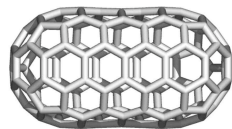
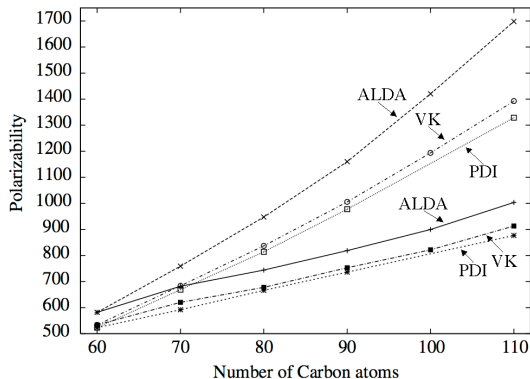
M. van Faassen, P. L. de Boeij, R. van Leeuwen, AB, and J. G. Snijders, PRL 88, 186401 (2002)

## Full VK: Static polarizabilities of conjugated polymers



M. van Faassen, P. L. de Boeij, R. van Leeuwen, AB, and J. G. Snijders, JCP 118, 1044 (2003)

## Full VK: Static polarizabilities of fullerenes



$C_{110}$  fullerene

$C_{60}$ :

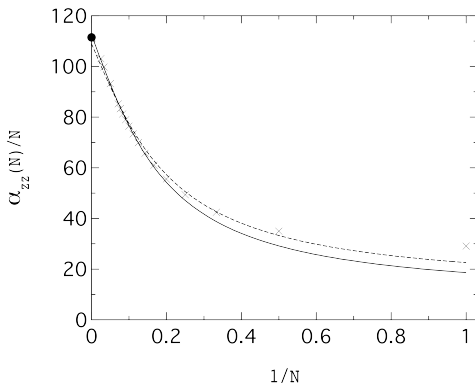
Exp:  $516 \pm 54$  a.u.

ALDA: 581.6 a.u.

VK: 533.6 a.u.

## Full VK: polarizabilities of infinite polymer chains

Polyacetylene:



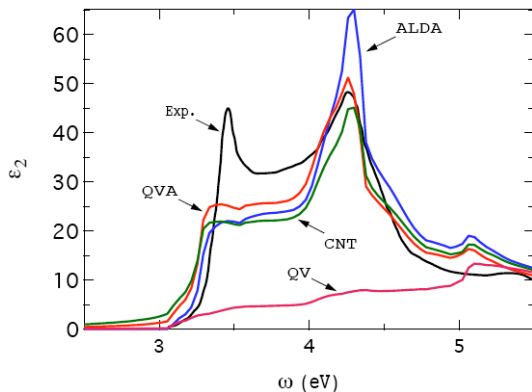
Extrapolation: **112.4** a.u.

PBC: **111.5** a.u.

Extrapolation based on a **physical model**:

AB, P. L. de Boeij, R. van Leeuwen, JCP 123, 174910 (2005)

## Full VK: Absorption spectrum of silicon



QVA = QV with  $\mu_{xc} = 0$

- Collapse of spectra using  $\mu_{xc} \neq 0$
- Slight improvement over ALDA using  $\mu_{xc} = 0$

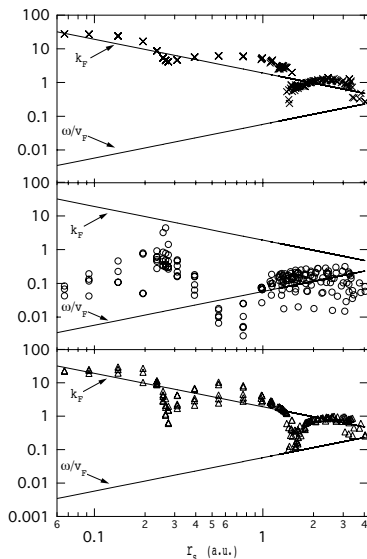
AB, P. L. de Boeij, R. van Leeuwen, PRB 75, 035116 (2007)

## Violation of constraints on VK functional in silicon

$$|\mathbf{q}| \ll k_F, \omega/v_F \sim \frac{|\nabla \rho_0|}{\rho_0} \ll k_F, \omega/v_F$$

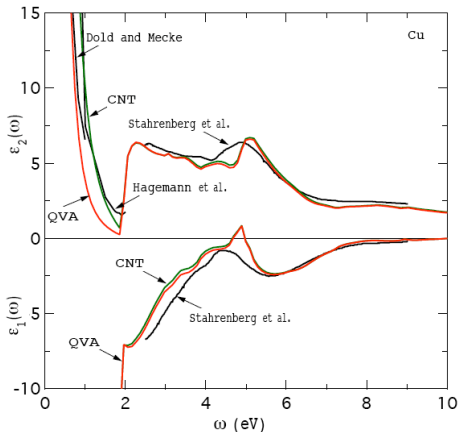
$$|\mathbf{k}| \ll k_F, \omega/v_F \sim \frac{\nabla \cdot \delta \mathbf{j}}{|\delta \mathbf{j}|} \ll k_F, \omega/v_F$$

$$|\mathbf{k}| \ll k_F, \omega/v_F \sim \frac{|\nabla \times \delta \mathbf{j}|}{|\delta \mathbf{j}|} \ll k_F, \omega/v_F$$





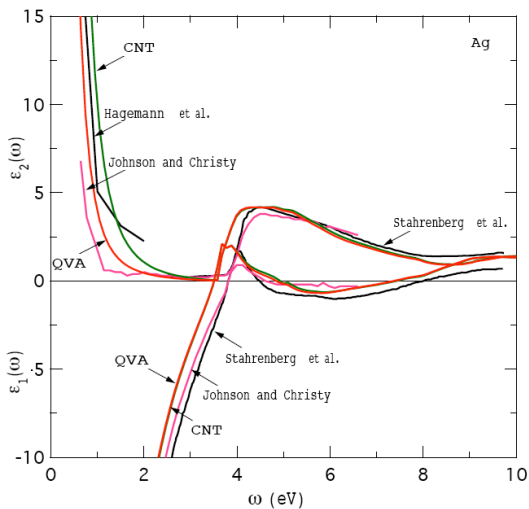
## Drude tail in absorption spectra of metals: Copper



Dielectric function of **copper**

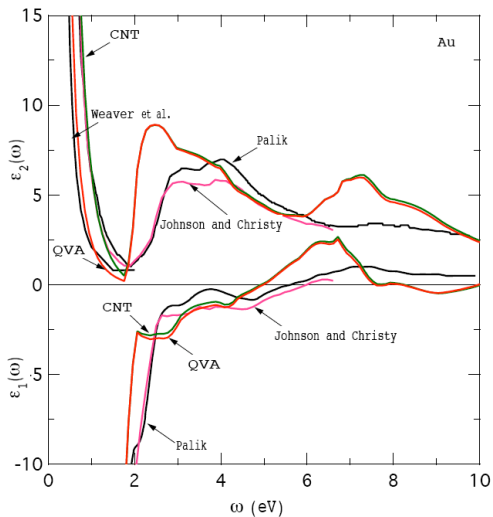
- The **frequency-dependent** VK functional can describe **relaxation effects** due to **scattering**
- $\mu_{xc}$  needs to be set to **zero** to avoid collapse.

## Drude tail in absorption spectra of metals: Silver



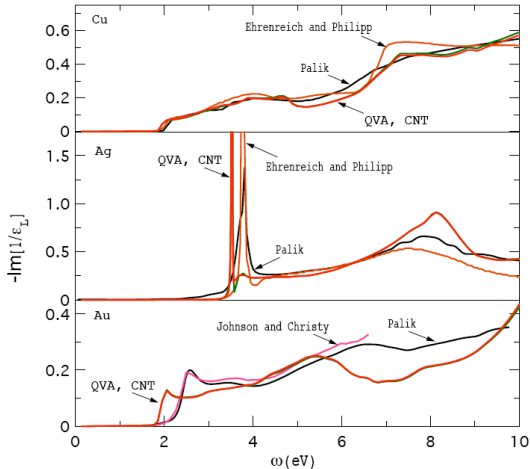
Dielectric function of silver

## Drude tail in absorption spectra of metals: Gold



Dielectric function of **gold**

## Loss functions of metals

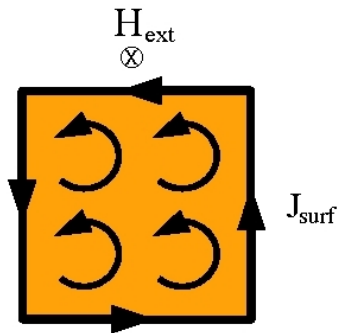


Loss functions of copper, silver and gold

The frequency-dependent VK functional can describe the finite width of the plasmon in silver.

## Magnetization

We want to describe **magnetization** within **TDCDFT** for systems described by **PBC**



## Magnetization

We need a definition of **magnetization density**  $\mathbf{M}(\mathbf{r}, t)$  that is compatible with **PBC**

A **natural definition** of  $\mathbf{M}(\mathbf{r}, t)$  should obey

$$\mathbf{m}(t) = \int_V \mathbf{M}(\mathbf{r}, t) d\mathbf{r} = \frac{1}{2} \int_V \mathbf{r} \times \mathbf{j}(\mathbf{r}, t) d\mathbf{r}$$

with  $\mathbf{m}(t)$  the total magnetic dipole moment.

However,  $\mathbf{r}$  is incompatible with PBC.

By analogy with polarization: Can we use the **continuity equation** of  $\mathbf{j}(\mathbf{r}, t)$

$$\dot{\mathbf{j}}(\mathbf{r}, t) + \nabla \cdot \mathbf{T} = 0 \quad ?$$

to find  $\mathbf{M}(\mathbf{r}, t)$ ?

# Magnetization

Relation between  $\mathbf{M}$  and  $\mathbf{j}$  from Maxwell's equations

$$\mathbf{j}(\mathbf{r}, t) = \dot{\mathbf{P}}(\mathbf{r}, t) + c\nabla \times \mathbf{M}(\mathbf{r}, t)$$

The gauge  $\mathbf{M}(\mathbf{r}, t) = \mathbf{0}$  is not convenient

Gauge:

$$\begin{aligned}\mathbf{j}_L(\mathbf{r}, t) &= \dot{\mathbf{P}}(\mathbf{r}, t) \\ \mathbf{j}_T(\mathbf{r}, t) &= c\nabla \times \mathbf{M}(\mathbf{r}, t)\end{aligned}$$

This gauge is compatible with a natural definition of magnetization

We can try to solve for  $\mathbf{M}(\mathbf{r}, t)$

$$\mathbf{M}(\mathbf{r}, t) = \frac{1}{4\pi} \int_V d\mathbf{r}' \frac{\nabla' \times \mathbf{j}(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} - \int_{\partial V} d\mathbf{r}' \left[ \frac{\hat{\mathbf{n}} \times \mathbf{M}(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} + \frac{\hat{\mathbf{n}} \cdot \mathbf{M}(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} \right]$$

## Conclusions

- ▶ The current density is the natural fundamental quantity in which to formulate a time-dependent Density Functional Theory for extended systems
- ▶ TD-CDFT provides an efficient way to include contributions of (ultra-)nonlocal exchange-correlation effects using a local functional of the current
- ▶ With the VK functional we can improve on ALDA results but it depends strongly on the static limit of the xc kernels of the electron gas
- ▶ We need more accurate values for the static limit of  $f_{xcT}$  (or  $\mu_{xc}$ )
- ▶ Outlook: Magnetization

Acknowledgments: Paul de Boeij, Robert van Leeuwen, Pina Romaniello, Meta van Faassen, Nathaniel Raimbault