

# **TDCDFT and plasmon damping: breakdown of the local approximation in quasi-2D systems**

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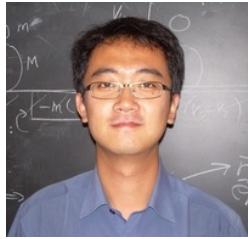


# Acknowledgments

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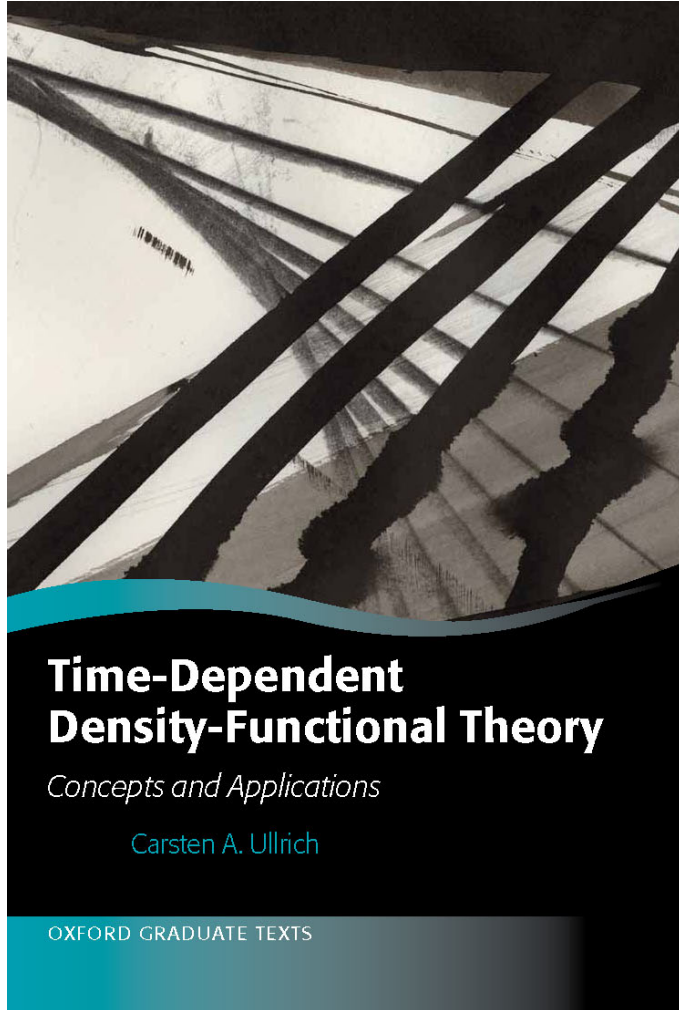


In collaboration with  
Irene D'Amico (York)



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**TDCDFT: basic formalism  
and ultranonlocality**

**Damping of charge and spin  
plasmons**

**Spin Coulomb drag: local  
approximation vs microscopic  
formulation**



# Why TDCDFT?

## **TDCDFT overcomes several formal limitations of TDDFT:**

allows treatment of **electromagnetic waves**, vector potentials, uniform applied electric fields.

works for all **extended systems**. One does not need the condition that the current density vanishes at infinity.

Vignale, PRB **70**, 201102 (2004), Maitra, Souza & Burke, PRB **68**, 045109 (2003)

## **But TDCDFT is also practically useful in situations that could, in principle, be fully described with TDDFT:**

Upgrading to the current density can be a more “natural” way to describe dynamical systems.

Helps to deal with the **ultranonlocality** problem of TDDFT

Provides ways to construct **nonadiabatic** approximations



# The adiabatic approximation

$$V_{xc}^A[n](\mathbf{r}, t) = V_{xc}^{static}[n(\mathbf{r}, t)](\mathbf{r}) \quad V_{xc}^{ALDA}(\mathbf{r}, t) = \left. \frac{de_{xc}^{unif}(\bar{n})}{d\bar{n}} \right|_{\bar{n}=n(\mathbf{r}, t)}$$

In general, the adiabatic approximation works well for excitations which have an analogue in the KS system (single excitations)

formally justified only for infinitely slow electron dynamics. But why is it that the frequency dependence seems less important?

The frequency scale of  $f_{xc}$  is set by correlated multiple excitations, which are absent in the KS spectrum.

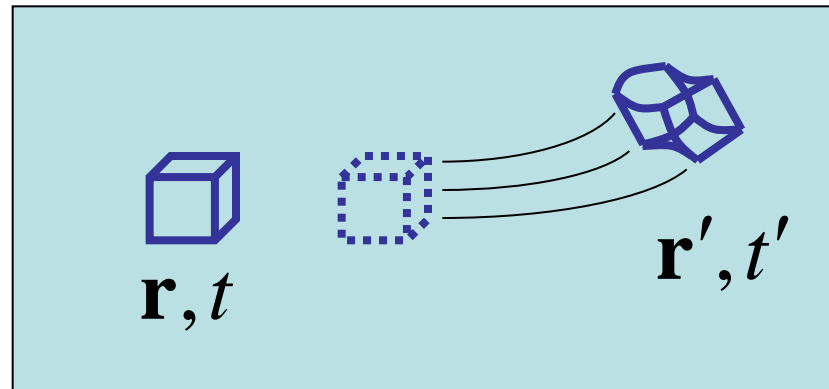
Adiabatic approximation fails for more complicated excitations (multiple, charge-transfer).

misses dissipation of long-wavelength plasmon excitations



# Nonlocality in space and time

Visualize electron dynamics as the motion (and deformation) of infinitesimal fluid elements:



**Nonlocality in time (memory) implies nonlocality in space!**

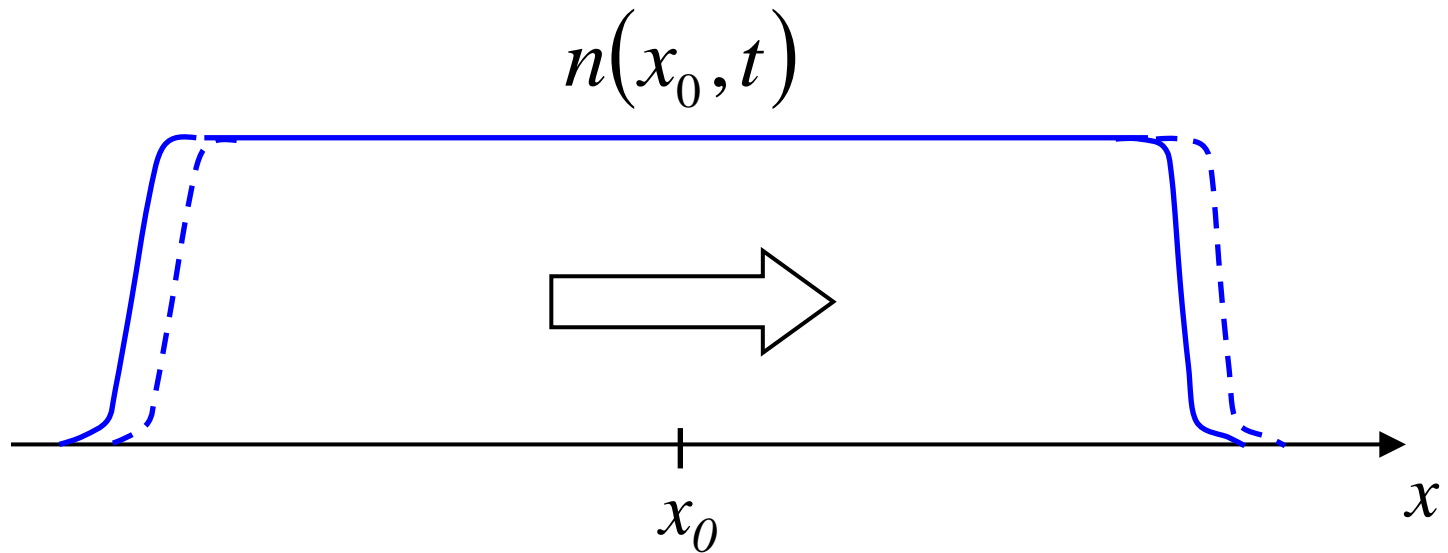
Dobson, Bünner, and Gross, PRL **79**, 1905 (1997)

I.V. Tokatly, PRB **71**, 165104 and 165105 (2005), PRB **75**, 125105 (2007)

C.A. Ullrich and I.V. Tokatly, PRB **73**, 235102 (2006)



# Ultranonlocality and the density

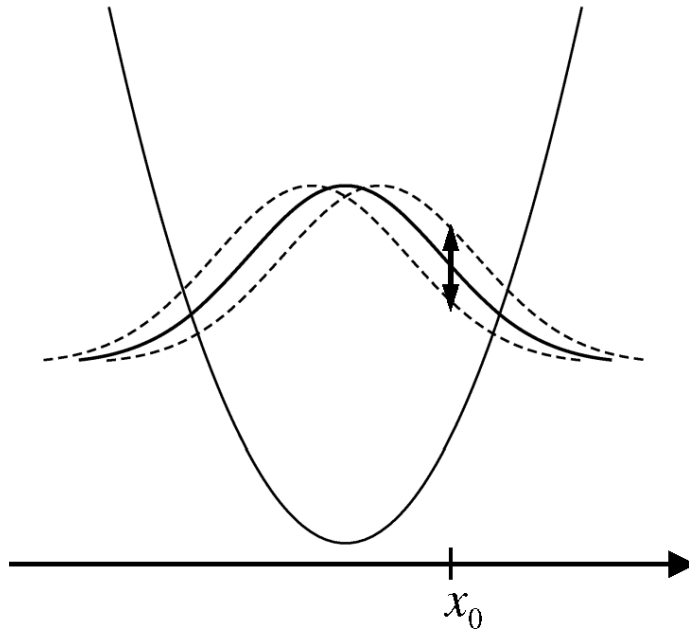


An xc functional that depends only on the local density (or its gradients) cannot see the motion of the entire slab.

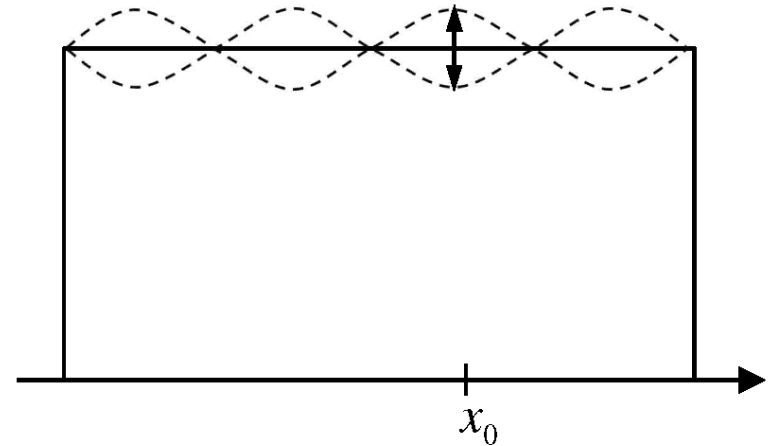
A density functional needs to have a long range to see the motion through the changes at the edges.



# Failure of nonadiabatic local density functionals



- ▶ undamped density oscillations
- ▶ xc potential rides along with density
- ▶ constant velocity field



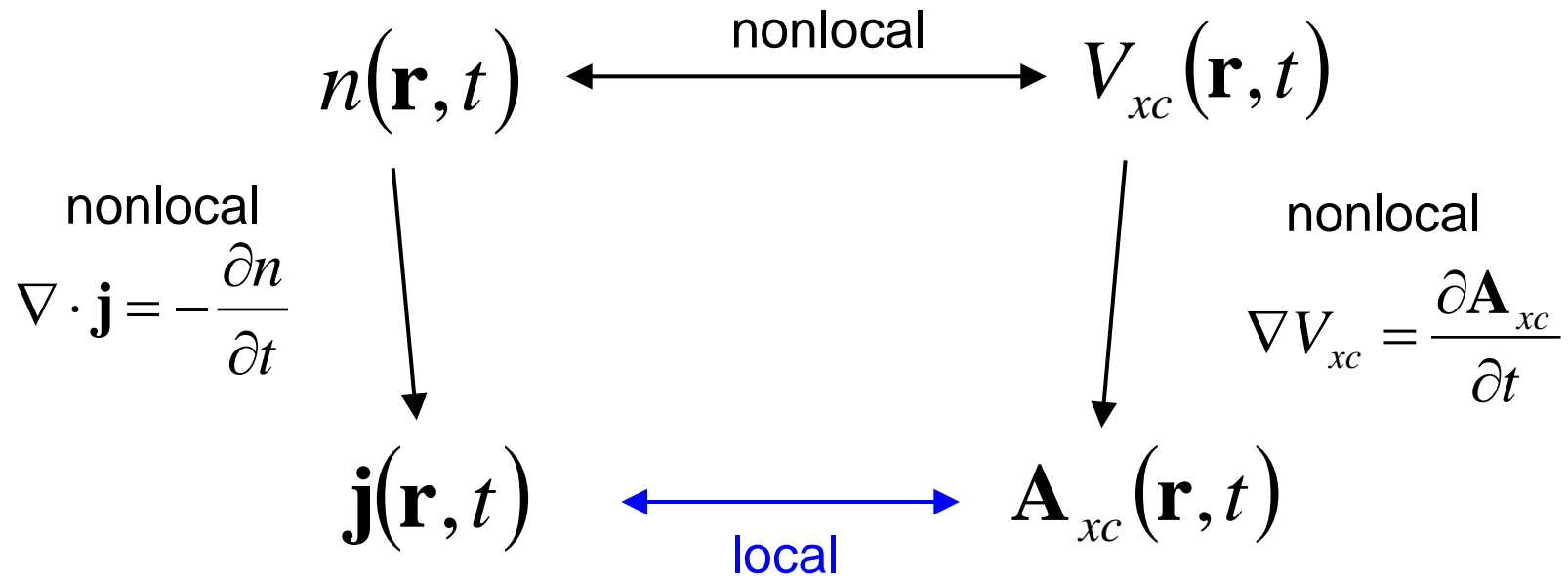
- ▶ bulk plasmon: periodic compression and rarefaction of the density
- ▶ intrinsic damping due to decay of collective mode into single-particle excitations
- ▶ oscillating velocity field

xc functionals based on local **density** can't distinguish the two cases!  
But one can capture the correct physics with **current** functionals.





# “Upgrading” TDDFT: Current-TDDFT



$$\mathbf{j}(\mathbf{r}, t) = \mathbf{j}_L(\mathbf{r}, t) + \mathbf{j}_T(\mathbf{r}, t), \quad \mathbf{j}_L(\vec{r}, t) = \frac{\nabla}{4\pi} \int \frac{\dot{n}(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|}$$

Continuity equation only gives the longitudinal current

TDCDFT gives also the transverse current

We can find a short-range current-dependent xc vector potential



# TDKS equation in TDCDFT

generalization of RG/vL theorems: Ghosh and Dhara, PRA **38**, 1149 (1988)  
G. Vignale, PRB **70**, 201102 (2004)

$$i \frac{\partial}{\partial t} \varphi_j(\mathbf{r}, t) = \left( \frac{1}{2} \left[ \frac{\nabla}{i} + \mathbf{A}_s(\mathbf{r}, t) \right]^2 + V_s(\mathbf{r}, t) \right) \varphi_j(\mathbf{r}, t)$$

Gauge-invariant physical current density:

$$\begin{aligned} \mathbf{j}(\mathbf{r}, t) &= n(\mathbf{r}, t) \mathbf{A}_s(\mathbf{r}, t) + \frac{1}{2i} \sum_{j=1}^N \left[ \varphi_j^*(\mathbf{r}, t) \nabla \varphi_j(\mathbf{r}, t) - \varphi_j(\mathbf{r}, t) \nabla \varphi_j^*(\mathbf{r}, t) \right] \\ &= \mathbf{j}_{\text{dia}}(\mathbf{r}, t) + \mathbf{j}_{\text{para}}(\mathbf{r}, t) \end{aligned}$$

Scalar and vector potentials:

$$V_s[\mathbf{j}](\mathbf{r}, t) = V(\mathbf{r}, t) + V_H(\mathbf{r}, t) + V_{xc}(\mathbf{r}, t)$$

$$\mathbf{A}_s[\mathbf{j}](\mathbf{r}, t) = \mathbf{A}(\mathbf{r}, t) + \mathbf{A}_{xc}(\mathbf{r}, t) \quad (\text{ignore Hartree vector potential from induced currents})$$



## TDCDFT in the linear response regime

$$\mathbf{j}_1(\mathbf{r}, \omega) = \int d^3 r' \tilde{\chi}_s(\mathbf{r}, \mathbf{r}', \omega) \{ \mathbf{A}_{ext,1}(\mathbf{r}, \omega) + \mathbf{A}_{H,1}(\mathbf{r}, \omega) + \mathbf{A}_{xc,1}(\mathbf{r}, \omega) \}$$

KS current-current response tensor: diamagnetic + paramagnetic part

$$\chi_{s,\mu\nu}(\mathbf{r}, \mathbf{r}', \omega) = n_0(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}') \delta_{\mu\nu} + \frac{1}{2} \sum_{j,k}^{\infty} \frac{f_k - f_j}{\varepsilon_k - \varepsilon_j + \omega + i\eta} P_{\mu}^{kj}(\mathbf{r}) P_{\nu}^{jk}(\mathbf{r}')$$

where 
$$P_{\mu}^{kj} = \varphi_k^*(\mathbf{r}) \nabla_{\mu} \varphi_j(\mathbf{r}) - \varphi_j(\mathbf{r}) \nabla_{\mu} \varphi_k^*(\mathbf{r})$$

Note:

$$\chi_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{\omega^2} \sum_{\mu\nu} \nabla_{\mu} \nabla'_{\nu} \chi_{s,\mu\nu}(\mathbf{r}, \mathbf{r}', \omega)$$



# TDCDFT: effective vector potential

$\mathbf{A}_{ext,1}(\mathbf{r}, \omega)$ : external perturbation. Can be a true vector potential, or a gauge transformed scalar perturbation:  $\mathbf{A}_{ext,1} = \frac{1}{i\omega} \nabla V_{ext,1}$

$$\mathbf{A}_{H,1}(\mathbf{r}, \omega) = \frac{\nabla}{(i\omega)^2} \int d^3 r' \frac{\nabla' \cdot \mathbf{j}_1(\mathbf{r}', \omega)}{|\mathbf{r} - \mathbf{r}'|}$$

gauge transformed Hartree potential

$$\mathbf{A}_{xc,1}(\mathbf{r}, \omega) = \int d^3 r' \vec{f}_{xc}(\mathbf{r}, \mathbf{r}', \omega) \mathbf{j}_1(\mathbf{r}', \omega)$$

the xc kernel is now a tensor!

**ALDA:**  $\mathbf{A}_{xc,1}^{ALDA}(\mathbf{r}, \omega) = \frac{\nabla}{(i\omega)^2} \int d^3 r' f_{xc}^{ALDA}(\mathbf{r}, \mathbf{r}') \nabla \cdot \mathbf{j}_1(\mathbf{r}', \omega)$



# TDCDFT beyond the ALDA: the VK functional

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)

$$\mathbf{A}_{xc,1}^{VK}(\mathbf{r}, \omega) = \mathbf{A}_{xc,1}^{ALDA}(\mathbf{r}, \omega) - \frac{1}{i\omega n_0(\mathbf{r})} \nabla \cdot \vec{\sigma}_{xc}(\mathbf{r}, \omega)$$

xc viscoelastic stress tensor:

$$\sigma_{xc,\mu\nu}(\omega) = \eta_{xc} \left( \nabla_\nu u_{1,\mu} + \nabla_\mu u_{1,\nu} - \frac{2}{3} \nabla \cdot \mathbf{u}_1 \delta_{\mu\nu} \right) + \zeta_{xc} \nabla \cdot \mathbf{u}_1 \delta_{\mu\nu}$$

$$\mathbf{u}(\mathbf{r}, \omega) = \mathbf{j}(\mathbf{r}, \omega) / n_0(\mathbf{r}) \quad \text{velocity field}$$

automatically satisfies zero-force theorem/Newton's 3<sup>rd</sup> law  
automatically satisfies the Harmonic Potential theorem  
is local in the current, but nonlocal in the density  
introduces dissipation/retardation effects



## xc viscosity coefficients

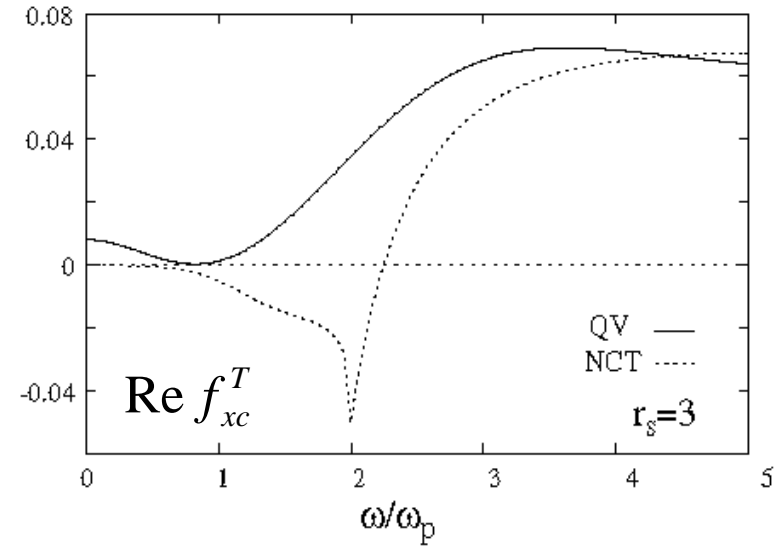
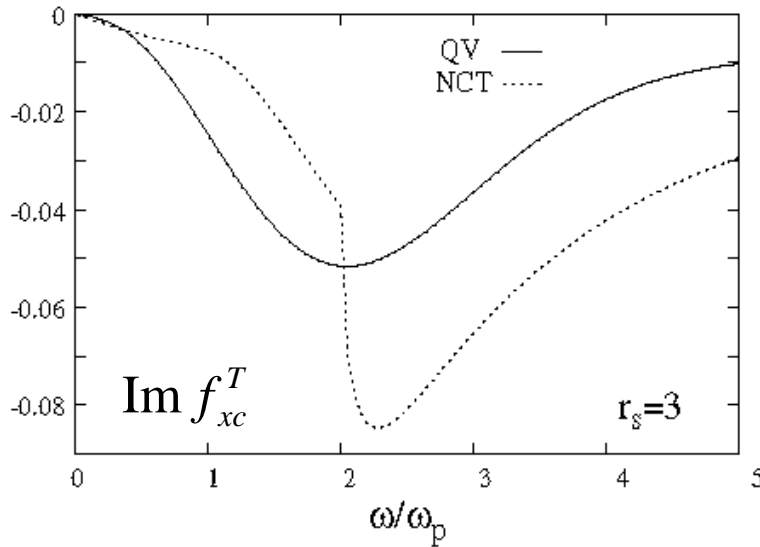
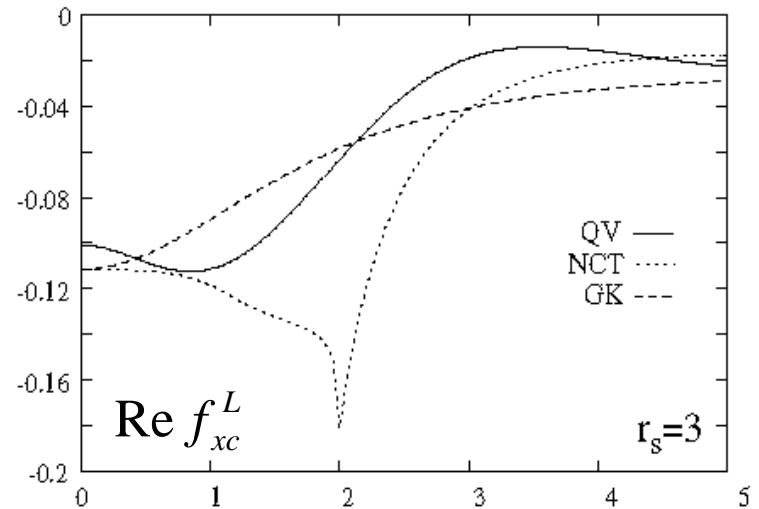
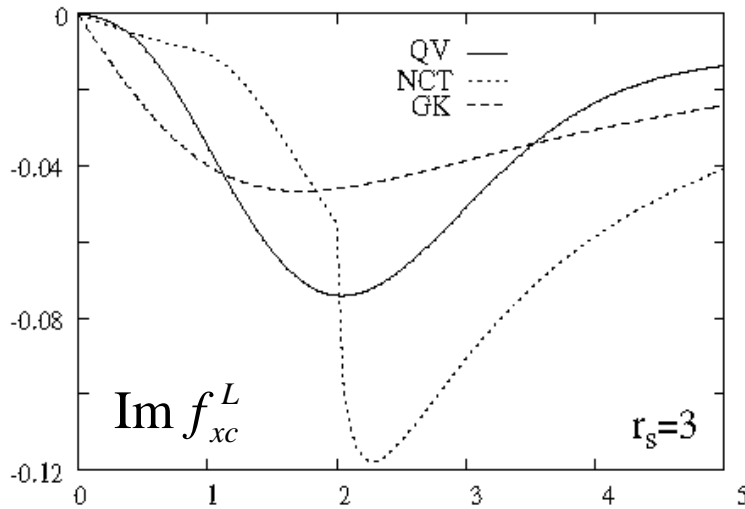
$$\eta_{xc}(n, \omega) = -\frac{n^2}{i\omega} f_{xc}^T(n, \omega)$$

$$\zeta_{xc}(n, \omega) = -\frac{n^2}{i\omega} \left( f_{xc}^L(n, \omega) - \frac{4}{3} f_{xc}^T(n, \omega) - \frac{d^2 e_{xc}^{unif}}{dn^2} \right)$$

The xc viscosities have both real and imaginary parts, describing **dissipative** and **elastic** behavior.



# xc kernels of the homogeneous electron gas



**GK:** E.K.U. Gross and W. Kohn, PRL **55**, 2850 (1985)

**NCT:** R. Nifosi, S. Conti, and M.P. Tosi, PRB **58**, 12758 (1998)

**QV:** X. Qian and G. Vignale, PRB **65**, 235121 (2002)



# Applications of the VK functional

## (A) In the (quasi)-static $\omega \rightarrow 0$ limit:

Polarizabilities of  $\pi$ -conjugated polymers (van Faassen et al.)

Nanoscale transport (Vignale, Di Ventura et al.)

Stopping power of slow ions in metals (Nazarov et al.)

These applications profit from the fact that VK does not reduce to the ALDA in the static limit.

## (B) To describe excitations at finite frequencies:

atomic and molecular excitation energies

plasmon excitations in doped semiconductor structures

optical properties of bulk metals and insulators

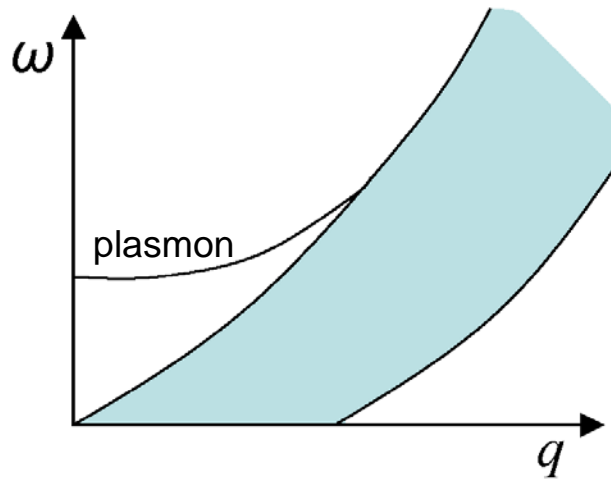
(De Boeij, Berger, Romaniello, van Leeuwen)

Here the picture is less clear. Some situations are well described, others fail.





# Metallic systems vs. insulators

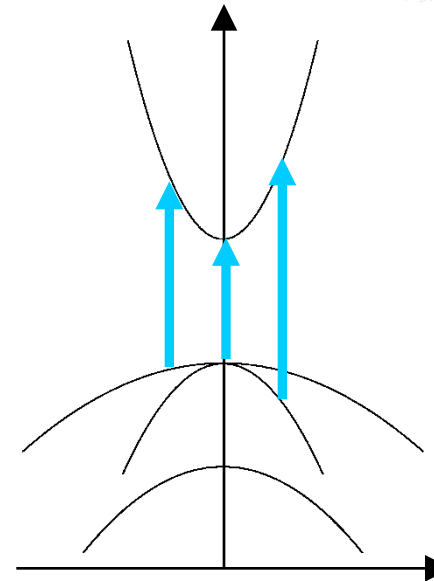
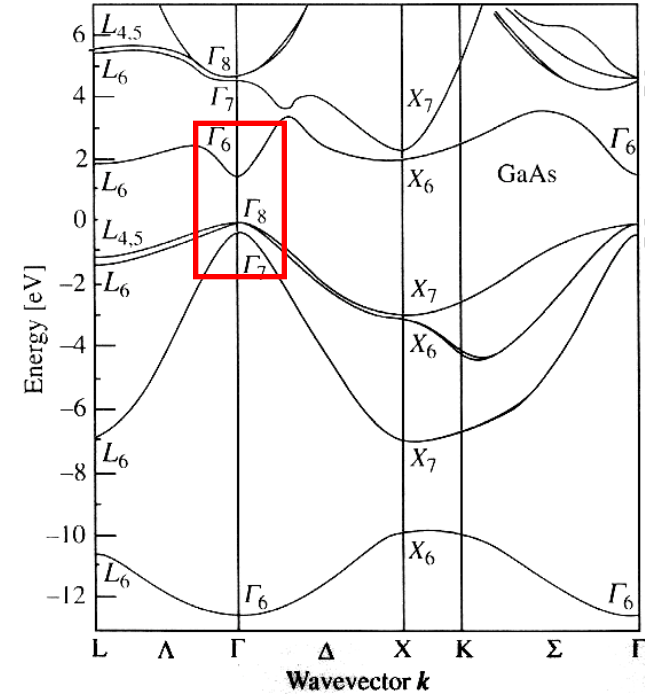


Excitation spectrum of simple metals:

single particle-hole continuum  
(incoherent)

collective plasmon mode

RPA/ALDA misses plasmon  
damping (multiple e-h excitations)

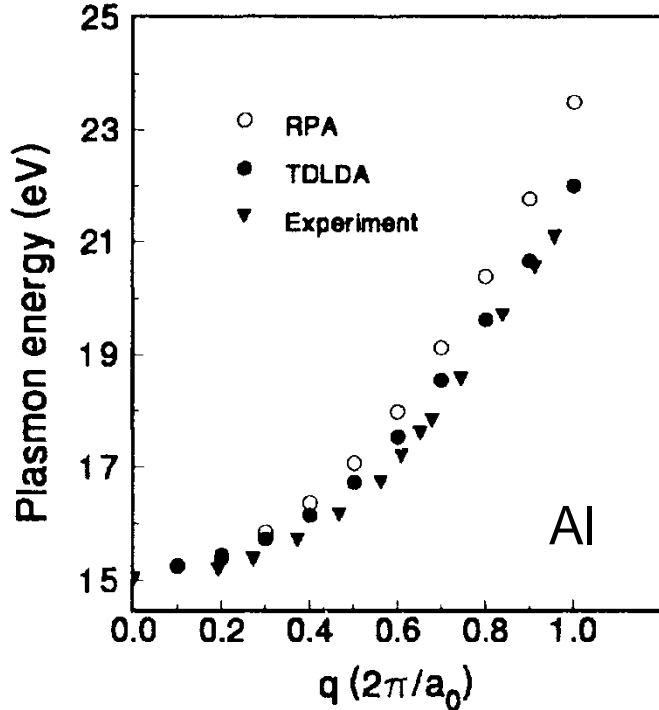


Optical excitations  
of insulators:

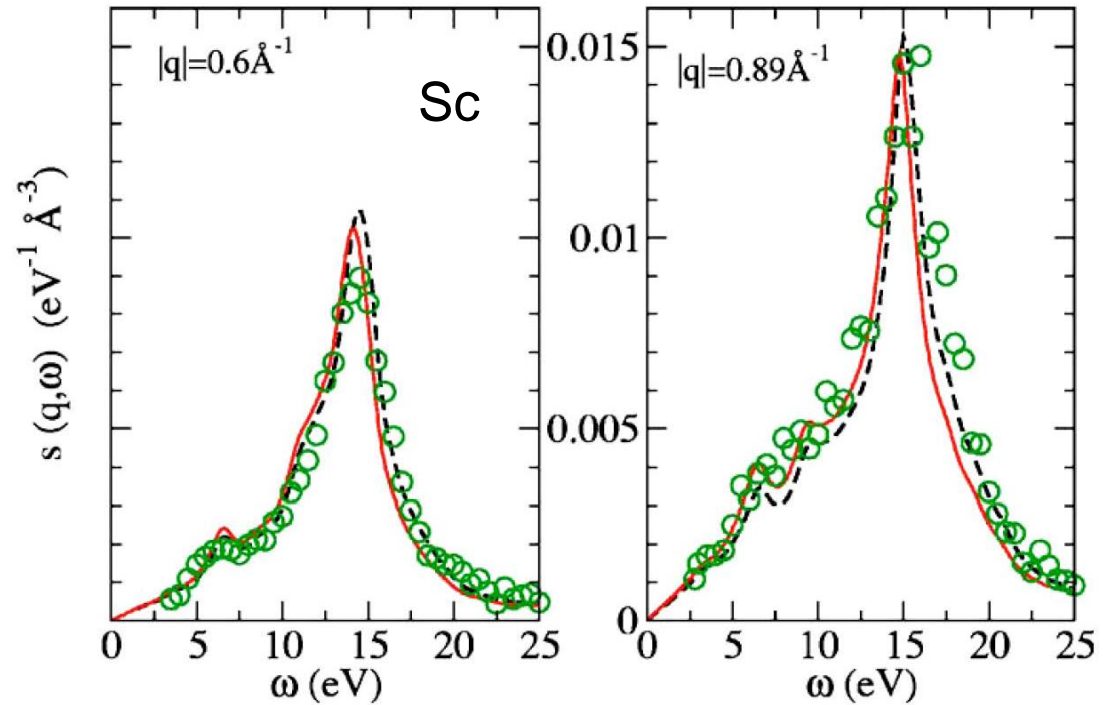
interband transitions  
excitons (bound  
electron-hole pairs)



# Plasmon excitations in bulk metals



Quong and Eguiluz,  
PRL **70**, 3955 (1993)

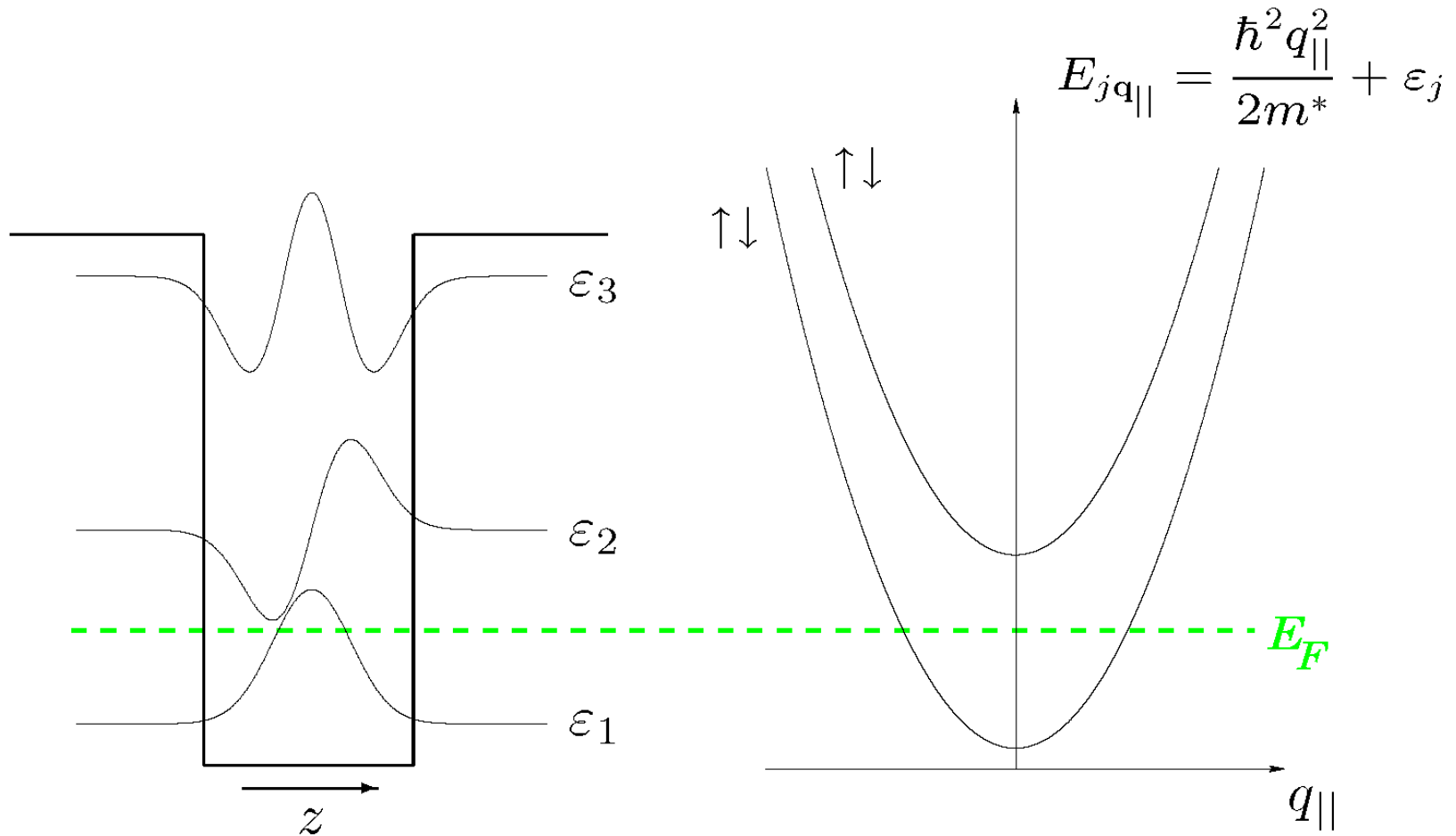


Gurtubay et al., PRB **72**, 125114 (2005)

- In general, excitations in (simple) metals very well described by ALDA.
- Time-dependent Hartree already gives the dominant contribution
- $f_{xc}$  typically gives some (minor) corrections (damping!)
- This is also the case for 2DEGs in doped semiconductor heterostructures



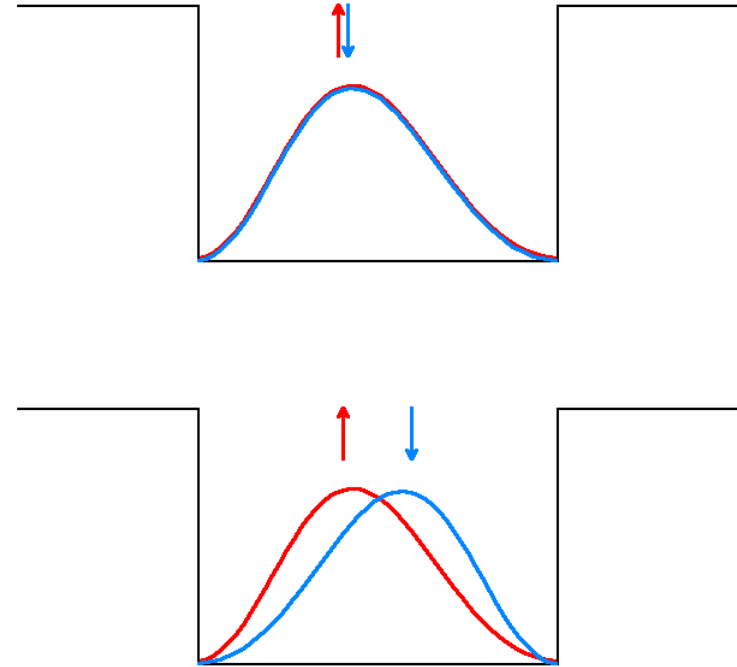
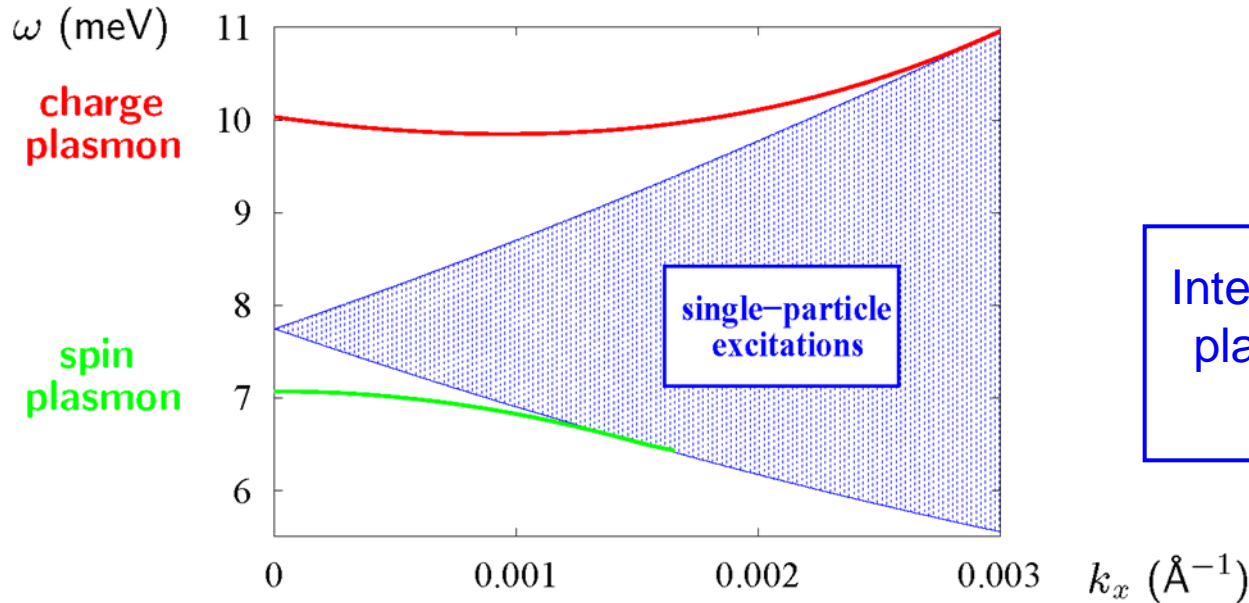
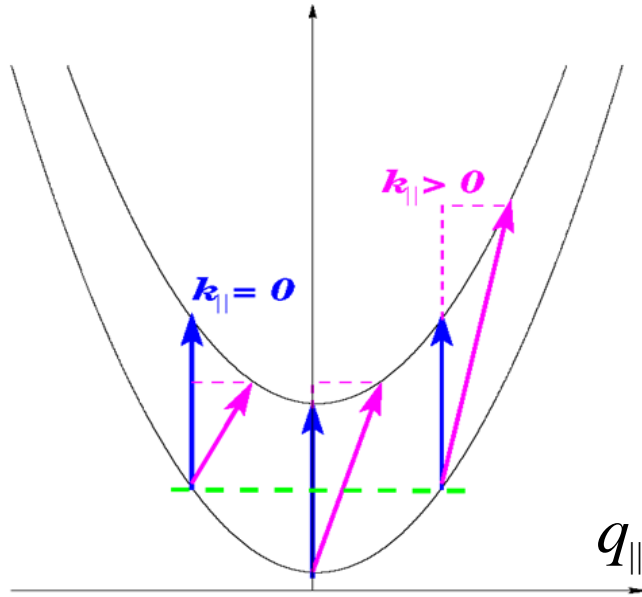
# Quantum well subbands



- Electrons in a quantum well:
- quantized in  $z$ -direction (discrete subbands)
  - free in the  $x$ - $y$  plane (each subband is parabolic)



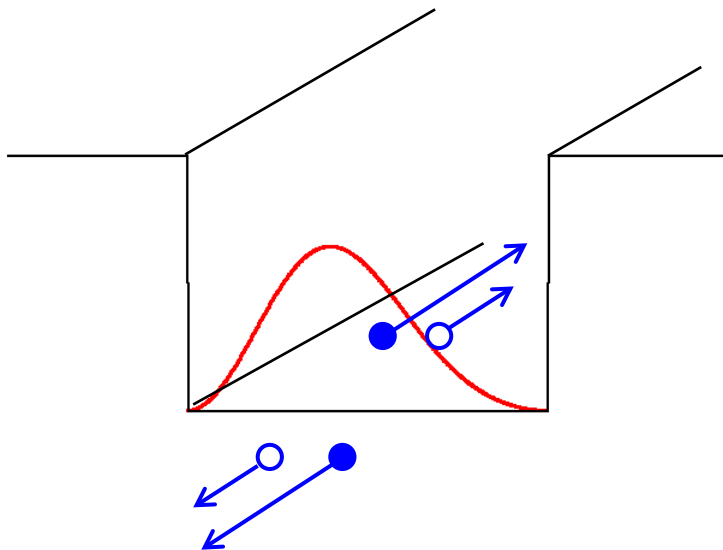
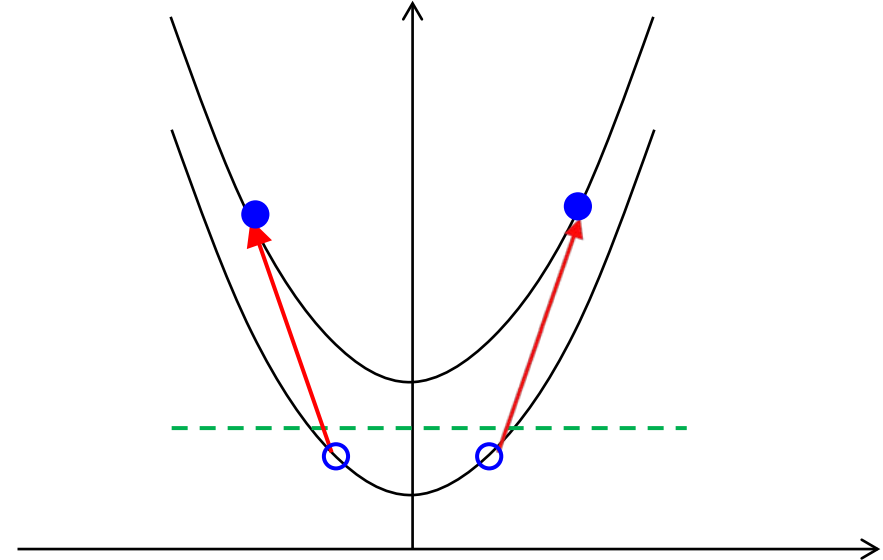
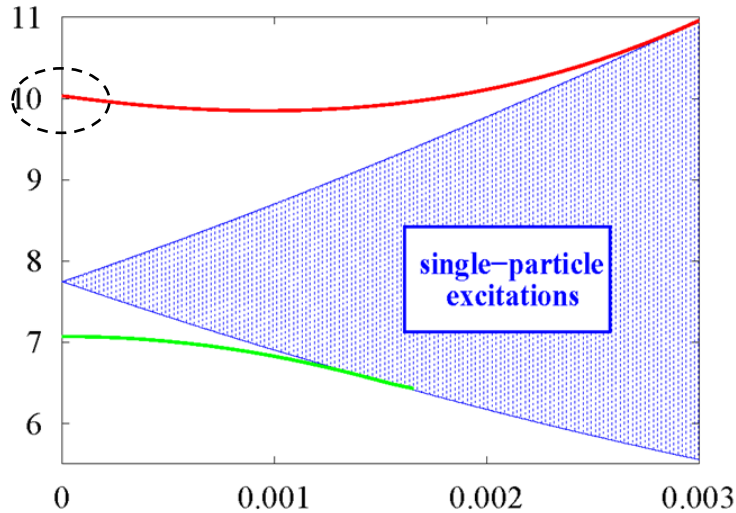
# Single-particle and collective excitations



Intersubband charge and spin plasmons:  $\uparrow$  and  $\downarrow$  densities in and out of phase



# Intrinsic plasmon damping mechanism



- ▶ Plasmon has energy and momentum different from any single p-h pair  
→ **plasmon is robust**
- ▶ But, can find two p-h pairs at right energy, and combined right total momentum  
→ **(weak) decay channel, requires Coulomb correlation beyond ALDA**



# ISB plasmon frequency $\Omega$ and linewidth $\Gamma$



	Experiment	ALDA	GK <sub>GK</sub>	GK <sub>NCT</sub>	VK <sub>GK</sub>	VK <sub>NCT</sub>
$\Omega$ (single)	10.7	10.25	10.63	10.23	10.31	10.24
$\Gamma$ (single)	0.53	—	0.683	0.655	0.128	0.104
$\Omega$ (double)	14.6	13.85	14.24	13.88	20.64	12.55
$\Gamma$ (double)	1.17	—	1.00	0.403	8.55	4.15

Single well: VK gives good results for electronic linewidth

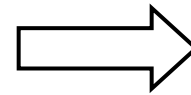
Double well: VK overestimates linewidth. The tunneling barrier makes the electronic flow very “non-hydrodynamic”.



# Ultranonlocality in DFT: “upgrades”

Band insulators:

$$f_{xc}(\mathbf{k}, \mathbf{k}, 0) \xrightarrow{k \rightarrow 0} \frac{\alpha}{k^2}$$

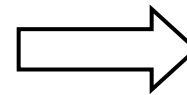


**Polarization DFT**

Gonze, Ghosez, and Godby,  
PRL **74**, 4035 (1995)

TDDFT:

$$f_{xc}(\mathbf{k}, \mathbf{k}, \omega) \xrightarrow{k \rightarrow 0} \frac{\alpha(\omega)}{k^2}$$

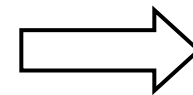


**TDCDFT**

Vignale and Kohn,  
PRL **77**, 2037 (1996),  
Nazarov, Vignale & Chang,  
PRL **102**, 113001 (2009)

Spin-TDDFT:

$$f_{xc, \sigma\sigma'}^{unif}(\mathbf{k}, \omega) \xrightarrow{k \rightarrow 0} \frac{A(\omega)}{k^2} \frac{\sigma\sigma' n^2}{4n_\sigma n_{\sigma'}} + B_{\sigma\sigma'}(\omega)$$



**TDSCDFT**

Situation even worse: ultranonlocality  
appears in the homogeneous case!

Qian, Constantinescu, Vignale,  
PRL **90**, 066402 (2003)



# Spin-dependent generalization of VK functional

Qian, Constantinescu, Vignale,  
PRL **90**, 066402 (2003)

spin-dependent generalization of  
the xc viscoelastic stress tensor  
depends on velocity gradients

$$\mathbf{A}_{xc,1\sigma}(\mathbf{r}, \omega) = \mathbf{A}_{xc,1\sigma}^{ALDA}(\mathbf{r}, \omega) - \frac{1}{i\omega n_{\sigma}(\mathbf{r})} \sum_{\sigma'} \nabla \cdot \vec{\sigma}_{xc,\sigma\sigma'}(\mathbf{r}, \omega) \\ + \frac{n_{\uparrow}(\mathbf{r})n_{\downarrow}(\mathbf{r})\rho_{\uparrow\downarrow}(\mathbf{r}, \omega)}{i\omega} \sum_{\sigma'} \frac{\sigma\sigma'}{n_{\sigma}(\mathbf{r})n_{\sigma'}(\mathbf{r})} \mathbf{j}_{\sigma'}(\mathbf{r}, \omega)$$

$\rho_{\uparrow\downarrow}(\omega)$   
spin-transresistivity

new term, depends on the velocities themselves  
disappears in the static limit ( $\rho_{\uparrow\downarrow} \sim \omega$ )  
real part: spin mass  
imaginary part: spin Coulomb drag





# Microscopic theory of SCD (hom. systems)

Kubo formula: 
$$\sigma_{\alpha\beta}(\omega) = -\frac{1}{i\omega} \frac{e^2}{m} \left( n_{\alpha} \delta_{\alpha\beta} + \frac{1}{m} \langle\langle \mathbf{P}_{\alpha}; \mathbf{P}_{\beta} \rangle\rangle_{\omega} \right)$$

$$\Rightarrow \rho_{\uparrow\downarrow}(\omega) = \frac{i\omega}{e^2 n_{\uparrow} n_{\downarrow}} \langle\langle \mathbf{P}_{\uparrow}; \mathbf{P}_{\downarrow} \rangle\rangle_{\omega}$$

can re-write this in terms of a force-force correlation function:

$$\text{Re } \rho_{\uparrow\downarrow}(\omega) = -\frac{1}{e^2 n_{\uparrow} n_{\downarrow} \omega} \text{Im} \langle\langle \mathbf{F}_{\uparrow}; \mathbf{F}_{\downarrow} \rangle\rangle_{\omega}$$

The diagram illustrates the decomposition of a four-point correlation function into two-point functions. On the left, a light blue rectangular box contains the correlation function  $\langle\langle \hat{n}_{-q\uparrow} \hat{n}_{q\downarrow}; \hat{n}_{q'\uparrow} \hat{n}_{-q'\downarrow} \rangle\rangle$ . The four corners of the box are labeled with the corresponding operators:  $\hat{n}_{-q\uparrow}$  (top-left),  $\hat{n}_{q'\uparrow}$  (top-right),  $\hat{n}_{q\downarrow}$  (bottom-left), and  $\hat{n}_{-q'\downarrow}$  (bottom-right). To the right of the box is a tilde symbol ( $\approx$ ) followed by the text "mode coupling approx.". This is followed by a plus sign between two groups of two-point functions, each enclosed in a light blue oval and grouped by a large curly bracket. The first group contains  $\chi_{\uparrow\uparrow}(q, \omega')$  and  $\chi_{\downarrow\downarrow}(q, \omega')$ . The second group contains  $\chi_{\downarrow\uparrow}(q, \omega - \omega')$  and  $\chi_{\uparrow\downarrow}(q, \omega - \omega')$ .



## Microscopic theory of the SCD

$$\text{Re } \rho_{\uparrow\downarrow}(\omega, T) = \frac{\hbar^2}{n_{\uparrow}n_{\downarrow}Ve^2} \sum_q \frac{q^2 v_q^2}{d} \frac{(e^{-\hbar\omega/k_B T} - 1)}{\omega} \\ \times \int \frac{d\omega'}{\pi} \frac{[\chi''_{\uparrow\uparrow}(q, \omega')\chi''_{\downarrow\downarrow}(q, \omega - \omega') - \chi''_{\uparrow\downarrow}(q, \omega')\chi''_{\downarrow\uparrow}(q, \omega - \omega')]}{(e^{-\hbar\omega'/k_B T} - 1)(e^{-\hbar(\omega - \omega')/k_B T} - 1)}$$

In the DC limit, one obtains

$$\rho_{\uparrow\downarrow}(T) = \frac{\hbar^2}{4n_{\uparrow}n_{\downarrow}k_B T V e^2} \sum_q \frac{q^2 v_q^2}{d} \int_0^{\infty} \frac{d\omega'}{\pi} \frac{\text{Im } \chi_{0\uparrow}(q, \omega') \text{Im } \chi_{0\downarrow}(q, \omega')}{|\varepsilon(q, \omega')|^2 \sinh^2(\hbar\omega' / 2k_B T)}$$

RPA: D'Amico and Vignale, PRB **62**, 4853 (2000)

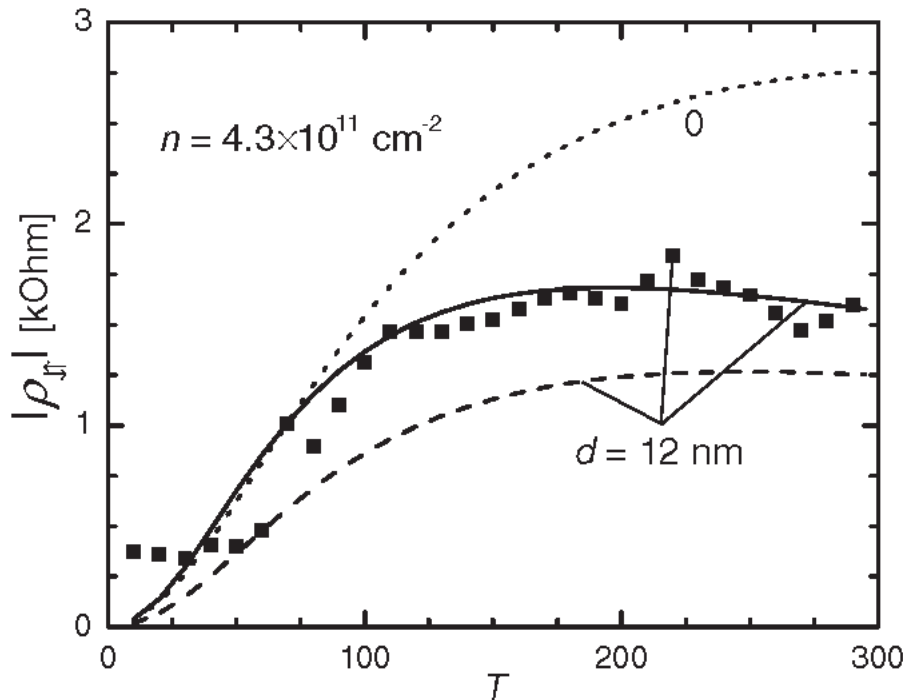
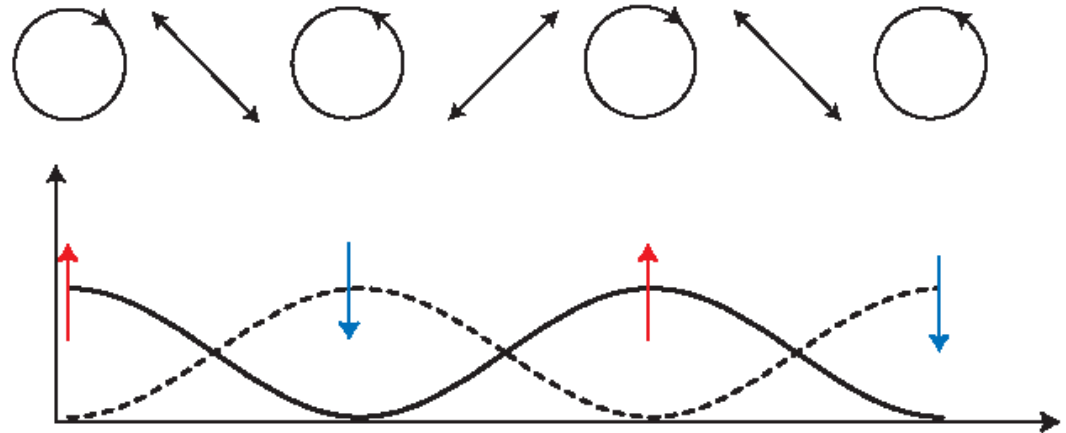
Beyond RPA: Badalyan, Kim, and Vignale, PRL **100**, 016603 (2008)



# Experimental evidence of SCD

Weber, Gedik, Moore, Orenstein, Stephens, and Awschalom, Nature **437**, 1330 (2005)

Transient spin grating plus time-resolved Kerr effect spectroscopy to measure spin diffusion constant



RPA, 2D

RPA+XC, finite width

RPA, finite width

Badalyan, Kim, and Vignale, PRL **100**, 016603 (2008)



# Spin-resolved excitation spectrum in TDDFT

(complex) excitation frequency

$$\Omega_{\pm\sigma}^2 \approx \omega_{pq\sigma}^2 + 2\omega_{pq\sigma} S_{\pm\sigma}$$

linewidth

$$\Gamma_{\pm} \approx \text{Im } S_{\pm\sigma}$$

$\omega_{pq\sigma}$  = bare single-particle excitation frequency between levels p and q

$$S_{\pm\sigma} = \left( S_{\sigma\bar{\sigma}}^{H+ALDA} \pm S_{\sigma\bar{\sigma}}^{H+ALDA} \right) + \left( S_{\sigma\bar{\sigma}}^{VE} \pm S_{\sigma\bar{\sigma}}^{VE} \right) + \left( S_{\sigma\bar{\sigma}}^{SCD} \pm S_{\sigma\bar{\sigma}}^{SCD} \right)$$

Hartree + ALDA: real  
(no dissipation)

Viscoelastic + SCD: complex  
(dissipative)

$$\text{Total linewidth: } \Gamma_{\pm} = \Gamma_{\pm}^{disorder} + \Gamma_{\pm}^{VE} + \Gamma_{\pm}^{SCD}$$



# SC-TDDFT energy shift for excitation energies

$$S_{\sigma\sigma'}^{H+ALDA} = \int d^3r \int d^3r' \left[ \frac{1}{|\vec{r} - \vec{r}'|} + f_{xc,\sigma\sigma'}^{ALDA}(\vec{r}, \vec{r}') \right] \varphi_{p\sigma}(\vec{r}) \varphi_{q\sigma}(\vec{r}) \varphi_{p\sigma'}(\vec{r}') \varphi_{q\sigma'}(\vec{r}')$$

$$S_{\sigma\sigma'}^{VE} = \frac{i\omega}{\omega_{pq\sigma}^2} \int d^3r \vec{\sigma}_{\sigma\sigma'}^{xc,pq}(\vec{r}, \omega) \vec{\nabla} \left[ \frac{\vec{j}_{pq\sigma}(\vec{r})}{n_{\sigma}(\vec{r})} \right]$$

$$\left( S_{\sigma\bar{\sigma}}^{SCD} \pm S_{\bar{\sigma}\sigma}^{SCD} \right) = \frac{ie^2\omega}{\omega_{pq\sigma}^2} \int d^3r \left[ \frac{n_{\bar{\sigma}}}{n_{\sigma}} |\vec{j}_{pq\sigma}|^2 \mp \vec{j}_{pq\bar{\sigma}} \cdot \vec{j}_{pq\sigma} \right] \rho_{\uparrow\downarrow}(\omega; n_{\uparrow}, n_{\downarrow})$$

structure of a (complex) power loss term in an AC spintronics circuit



# Comparison with experiment

VOLUME 63, NUMBER 15

PHYSICAL REVIEW LETTERS

9 OCTOBER 1989

## Large Exchange Interactions in the Electron Gas of GaAs Quantum Wells

A. Pinczuk, S. Schmitt-Rink, G. Danan, J. P. Valladares, L. N. Pfeiffer, and K. W. West

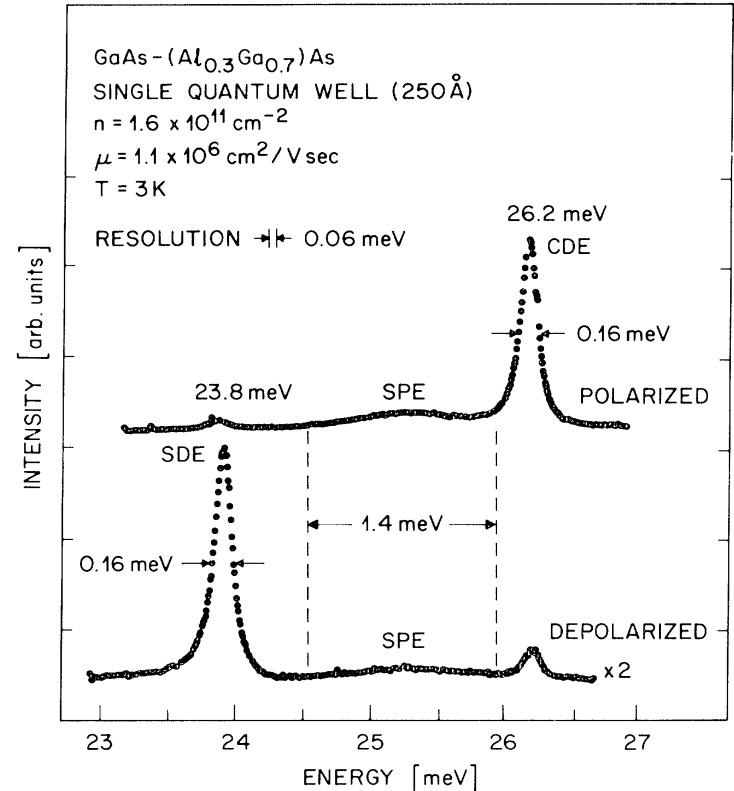
*AT&T Bell Laboratories, Murray Hill, New Jersey 07974*

Results with local approximation:

$$\text{SDE: } \begin{cases} \Gamma^{SCD} = 0.40 \text{ meV} \\ \Gamma^{VE} = 0.04 \text{ meV} \end{cases}$$

$$\text{CDE: } \Gamma^{VE} = 0.30 \text{ meV}$$

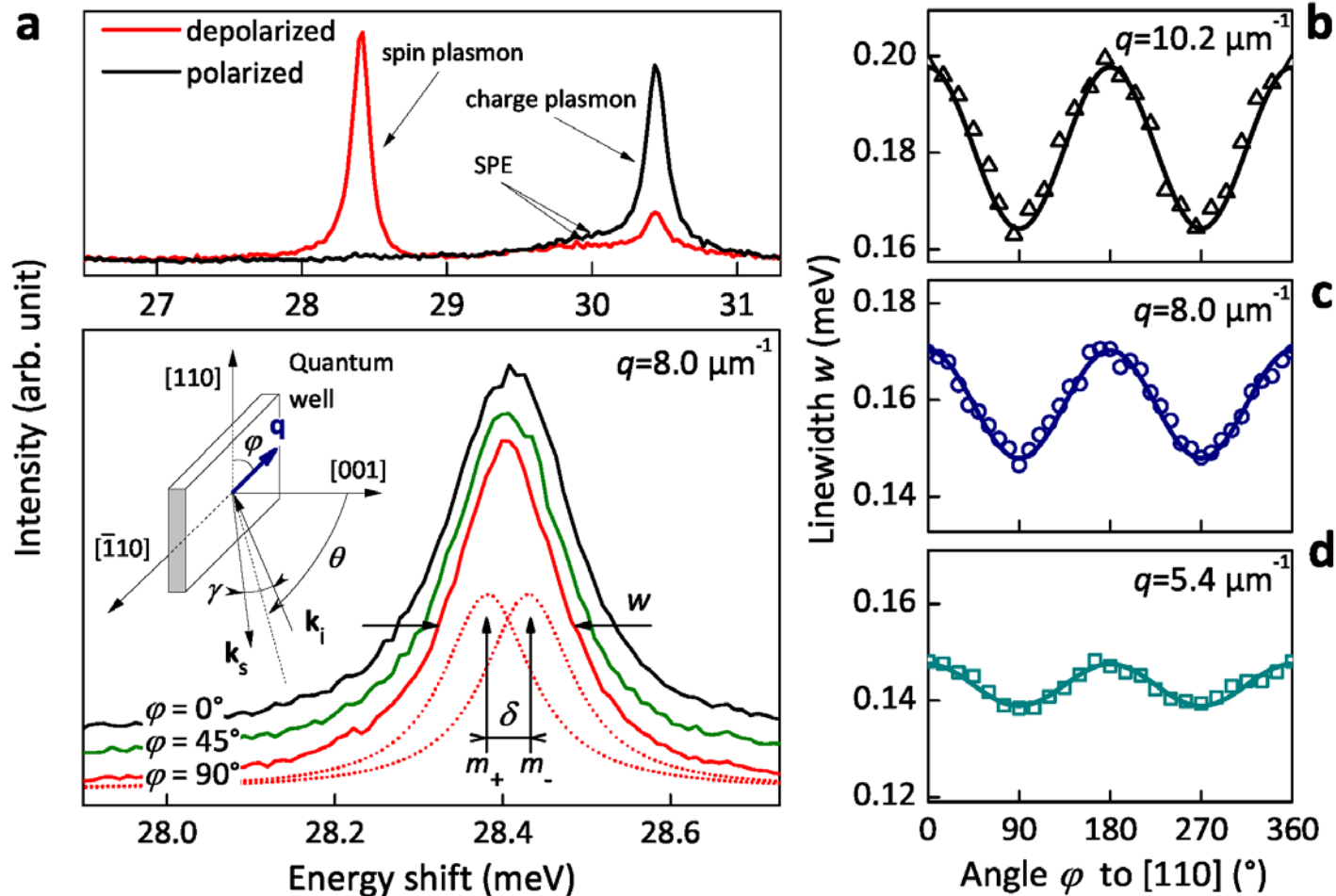
**overdamping!**





# Comparison with experiment

F. Baboux, F. Perez, C.A. Ullrich, I. D'Amico, J. Gomez, M. Bernard, PRL **109**, 166401 (2012)



Linewidth modulated by spin-orbit coupling

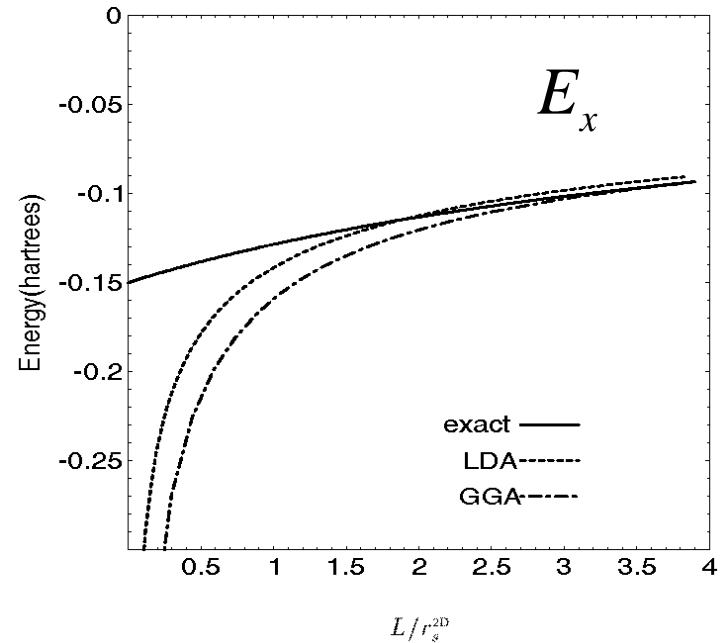
Again, SCD linewidth overestimated by factor 2-3



# The local approximations in quasi-2D

For structure, the 3D LDA is fine,  
as long as the quantum well is not  
too narrow:  $L \geq r_s^{2D}$

L. Pollack and J.P. Perdew,  
J. Phys.: Condens. Matter **12**, 1239 (2000)



Exchange and correlation energies in  
typical quantum wells are well reproduced.

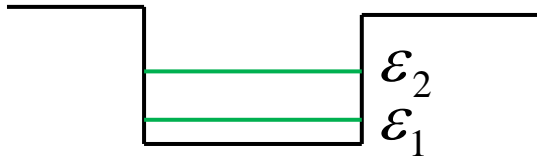




# The local approximations in quasi-2D

**For dynamics, it depends:**

**Plasmon dispersions are fine with 3D ALDA (about as accurate as singlet-triplet excitation energies in atoms).**



ISB plasmon is dominated by  $\Delta\epsilon = \epsilon_2 - \epsilon_1$  with relatively minor many-body corrections.

$$\Delta\epsilon \gg \Gamma$$

**Plasmon damping fails with 3D local approximation: size quantization creates a bottleneck for dissipation into lateral electronic degrees of freedom.**

Dissipation is purely nonclassical many-body physics:

The physics is not contained in the Kohn-Sham reference system

Need detailed information on the structure of the single-particle spectrum:

**Difficult to do in terms of the local current only!**

But the p-h spectrum can be accurately described in LDA (even RPA)



# Conclusion

- ▶ TDCDFT can be used for nonadiabatic dynamics, but choosing the right reference system is crucial!
- ▶ Local approximations (based on 3D HEG) work well in the thermodynamical/bulk limit, but fail for quantum confined systems
- ▶ Plasmon damping in quantum well as an example of the crossover 3D/2D: “tough” dynamics has no counterpart in the Kohn-Sham system
- ▶ In general, multiple/correlated excitations are better described using many-body techniques.
- ▶ A microscopic calculation of the spin plasmon linewidth was presented, which gives good agreement with experiment (the slides were removed, since they contain unpublished results).