Lecture 1

Nonadiabatic electron dynamics in TDDFT

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Benasque, September 2006

Overview

Lecture 1

Part I: Basics and formalism

- Beyond the adiabatic approximation: why and how?
- Current-TDDFT
- The VK functional: technical details

Part II: Applications in the linear response regime

- polarizabilities of polymers
- atomic excitation energies
- intersubband plasmons in quantum wells
- Spin-Current-TDDFT

Lecture 2

Part III: Nonlinear regime

- TDKS equation with memory
- Dissipation: where does the energy go?
- C-TDDFT versus L-TDDFT
- TDOEP

TDSE versus TDKS

$$\left[\sum_j\left(-\frac{\nabla_j^2}{2}+V\left(\vec{r}_j,t\right)\right)+\sum_{j\neq k}\frac{1}{\left|\vec{r}_j-\vec{r}_k\right|}-i\frac{\partial}{\partial t}\right]\Psi\left(\vec{r}_1,\ldots,\vec{r}_N,t\right)=0$$

Full many-body TDSE: linear equation, instantaneous interactions.

$$\left[-\frac{\nabla^2}{2}+V\left(\vec{r},t\right)+V_H\left(\vec{r},t\right)+V_{xc}\left(\vec{r},t\right)-i\frac{\partial}{\partial t}\right]\varphi_j\left(\vec{r},t\right)=0$$

TDKS equation: nonlinear (H+xc), memory-dependent (xc) Hamiltonian.

Via $\partial/\partial t$, both TDSE and TDKS carry the memory of the initial states from where the time propagation starts, $\Psi(0)$ and $\varphi_j(0)$.

Two kinds of xc memory in TDDFT

dependence on initial states, except when starting from the ground state (N. Maitra, TDDFT book, Ch.4)

$$V_{xc}\left[n, \overbrace{\Psi(0), \Phi_{KS}(0)}\right](\vec{r}, t)$$

dependence on densities:

$$n(\vec{r}', t'), \quad t' \leq t$$

(nonlocal in space and time)

The adiabatic approximation

$$V_{xc}^{A}[n](\vec{r},t) = V_{xc}^{static}[n(\vec{r},t)](\vec{r})$$

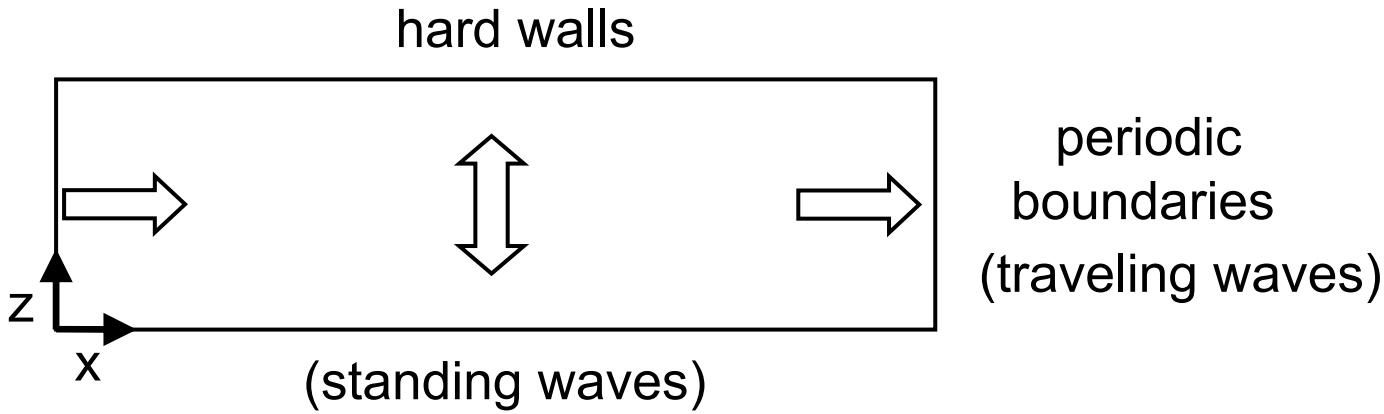
Take any approximate ground-state xc functional, and plug in a time-dependent density. Most widely used: ALDA

$$V_{xc}^{ALDA}(\vec{r},t) = \left.\frac{de_{xc}^{unif}(\bar{n})}{d\bar{n}}\right|_{\bar{n}=n(\vec{r},t)}$$

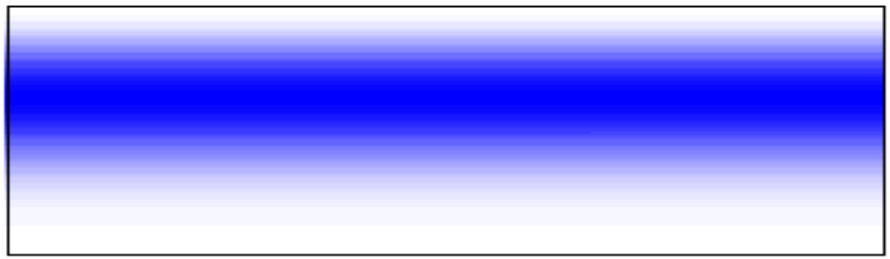
ALDA depends only on the density at the same space-time point: $n(\vec{r},t)$

"Adiabatic" means: no history dependence, no memory, no retardation.

Example: two electrons on a 2D quantum strip



Charge-density oscillations



Construction of the exact xc potential

Step 1: solve full 2-electron Schrödinger equation

$$\left[-\frac{\nabla_1^2}{2}-\frac{\nabla_2^2}{2}+V(z_1,t)+V(z_2,t)+\frac{1}{|\vec{r}_1-\vec{r}_2|}-i\frac{\partial}{\partial t}\right]\Psi(\vec{r}_1,\vec{r}_2,t)=0$$

Step 2: calculate the exact time-dependent density

$$\sum_{s_1,s_2}\int d\vec{r}_2\left|\Psi(\vec{r},\vec{r}_2,t)\right|^2=n(z,t)=2\left|\varphi(z,t)\right|^2$$

Step 3: find that TDKS system which reproduces the density

$$\left[-\frac{1}{2}\frac{d^2}{dz^2}+V(z,t)+V_H(z,t)+V_{xc}(z,t)-i\frac{\partial}{\partial t}\right]\varphi(z,t)=0$$

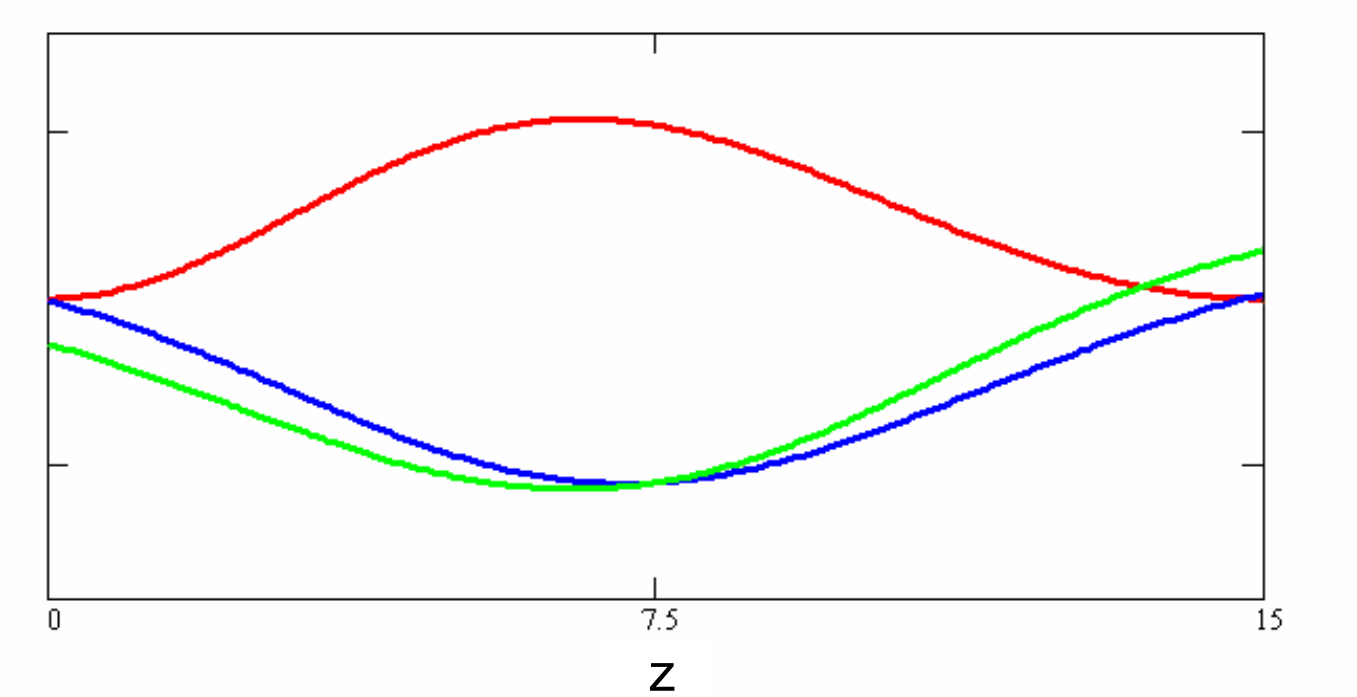
Construction of the exact xc potential

Ansatz: $$\varphi(\vec{r},t)=\sqrt{\frac{n(\vec{r},t)}{2}}\exp(i\alpha(\vec{r},t))$$

$$\Longrightarrow V_{xc}(\vec{r},t) = \underbrace{-V(\vec{r},t)-V_H(\vec{r},t) + \frac{1}{4}\nabla^2 \ln n(\vec{r},t)+\frac{1}{8}\left|\vec{\nabla} \ln n(\vec{r},t)\right|^2}_{V_{xc}^{A}} \underbrace{- \dot{\alpha}(\vec{r},t)-\frac{1}{2}\left|\vec{\nabla} \alpha(\vec{r},t)\right|^2}_{V_{xc}^{dyn}}$$

Exercise: verify this result!

2D quantum strip: charge-density oscillations



density

exact V_{xc}

adiabatic V_{xc}

The adiabatic approximation

- 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?

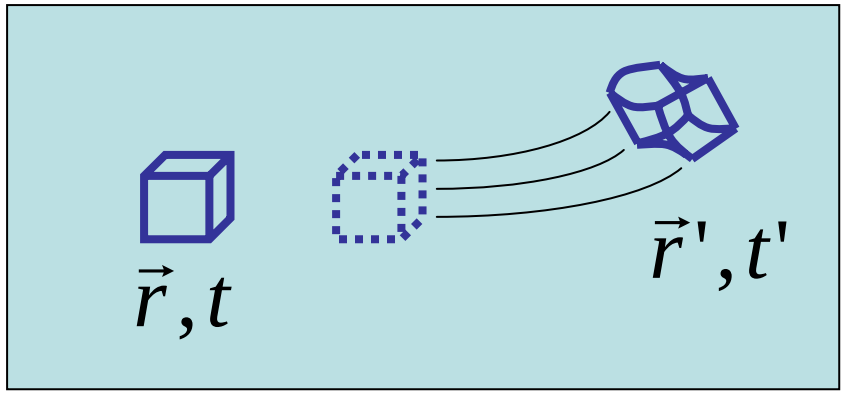
The frequency scale of f_{xc} is set by correlated multiple excitations, which are absent in the KS spectrum.

- Adiabatic approximation fails for more complicated excitations (multiple, charge-transfer)
- misses dissipation of long-wavelength plasmon excitations

Fundamental question: what is the proper extension of the LDA into the dynamical regime?

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)

Ultranonlocality in TDDFT

Zero-force theorem: $$\int d^3r\, n(\vec{r},t)\vec{\nabla}V_{xc}(\vec{r},t)=0$$

Linearized form: $$\int d^3r'\vec{\nabla}n_0(\vec{r}')\, f_{xc}(\vec{r},\vec{r}',\omega)=\vec{\nabla}V_{xc,0}(\vec{r})$$

If the xc kernel has a **finite range**, we can write for slowly varying systems:

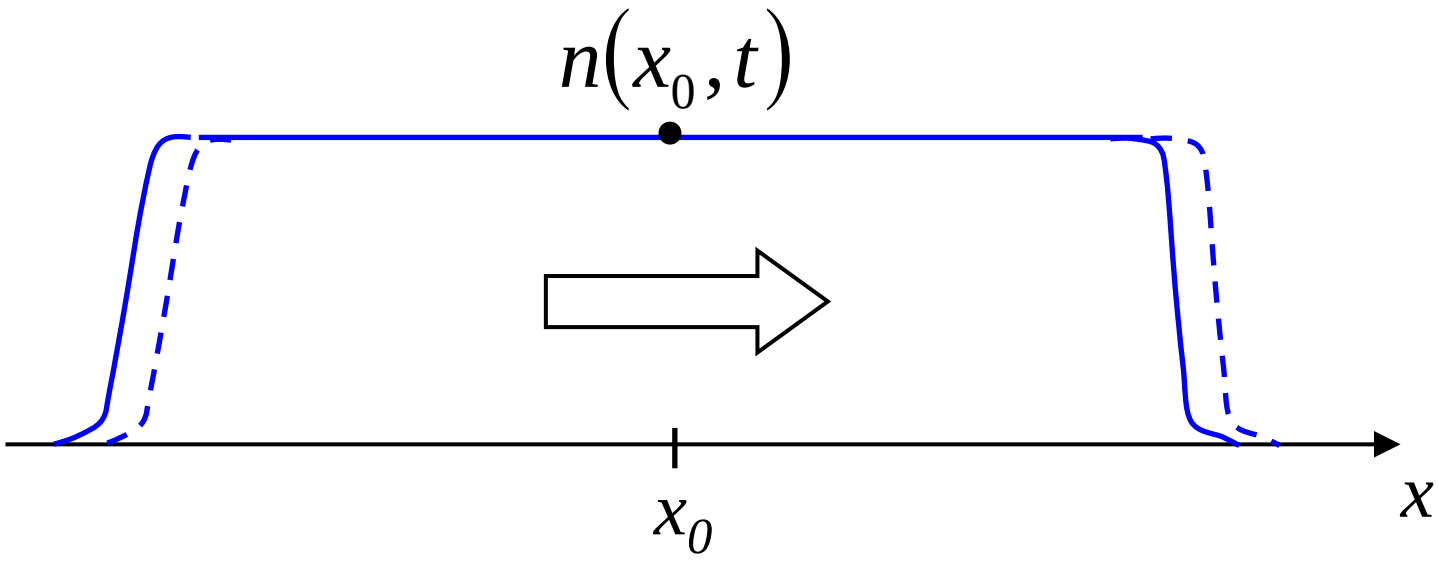
$$\vec{\nabla}n_0(\vec{r})\underbrace{\int d^3r'\, f_{xc}(\vec{r},\vec{r}',\omega)}=\vec{\nabla}V_{xc,0}(\vec{r})$$

$$\Rightarrow f_{xc}^{\text{hom}}(\vec{k}=0,\omega)$$

l.h.s. is frequency-dependent, r.h.s is not: **contradiction!**

$\Longrightarrow$ $f_{xc}(\vec{r},\vec{r}',\omega)$ has infinitely long spatial range!

Ultranonlocality and the density

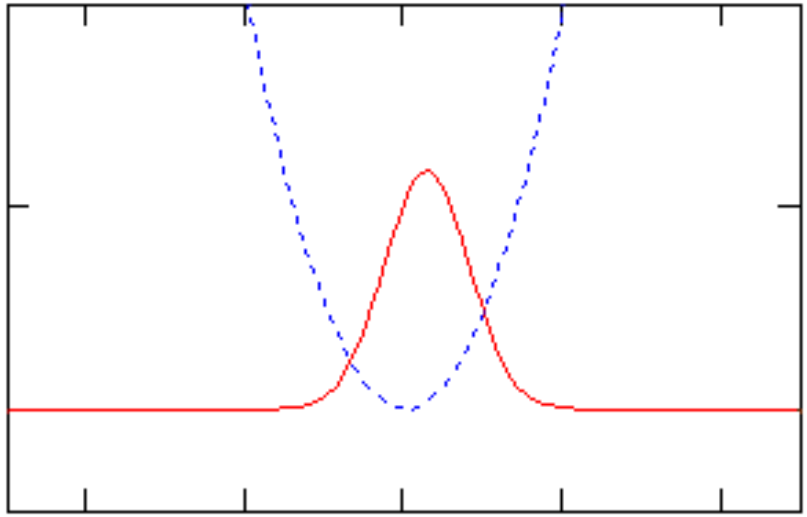


An xc functional that depends only on the local density (or its gradients) cannot see the motion of the entire slab.

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

Harmonic Potential Theorem – Kohn's mode

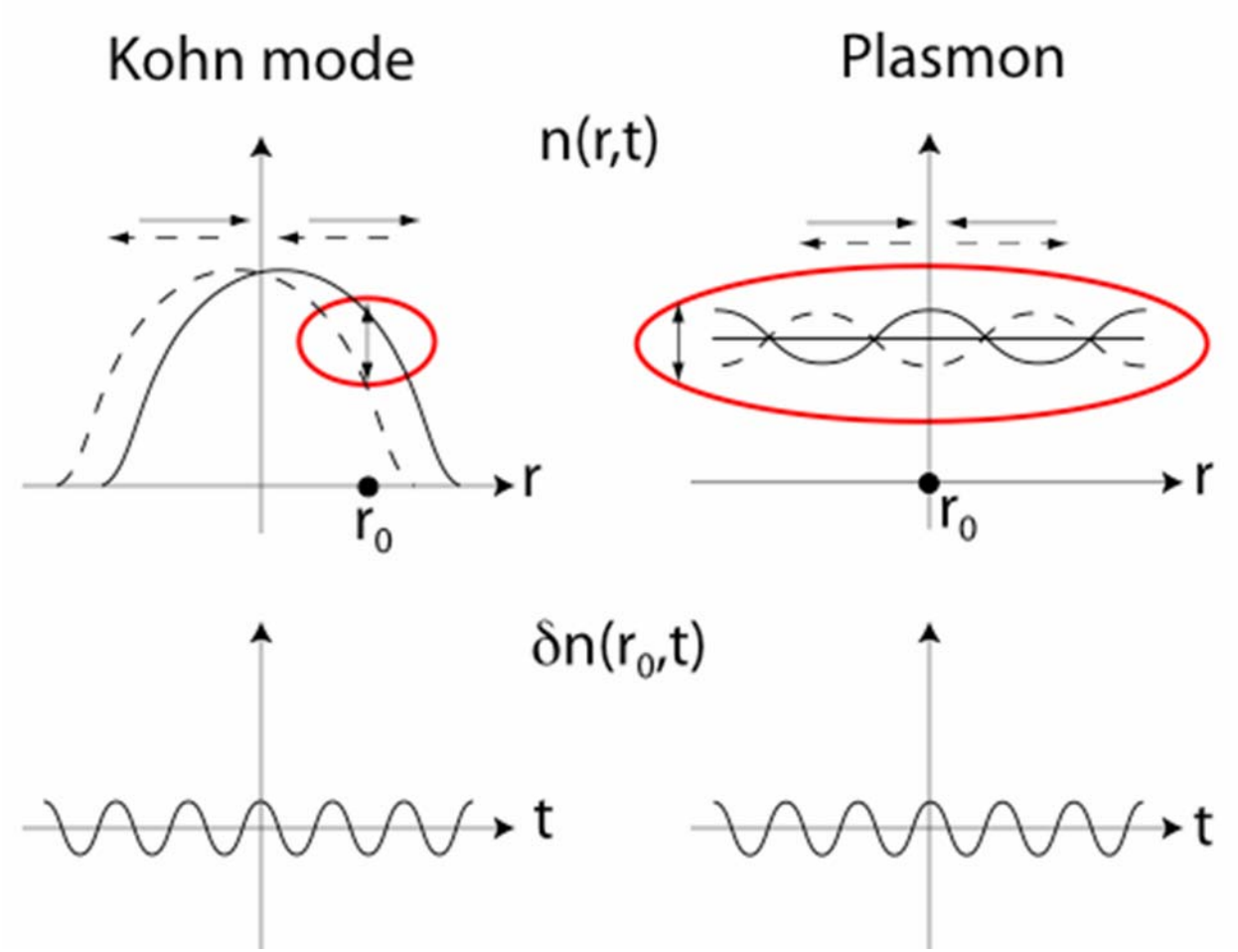
J.F. Dobson, PRL **73**, 2244 (1994)



A parabolically confined, interacting N-electron system can carry out an undistorted, undamped, collective "sloshing" mode, where $n(\vec{r},t) = n_0\left(\vec{r} - \vec{R}(t)\right)$, with the CM position $\vec{R}(t)$.

Exercise: Verify that the ALDA satisfies the HPT!

Point of view of the local density



global translation

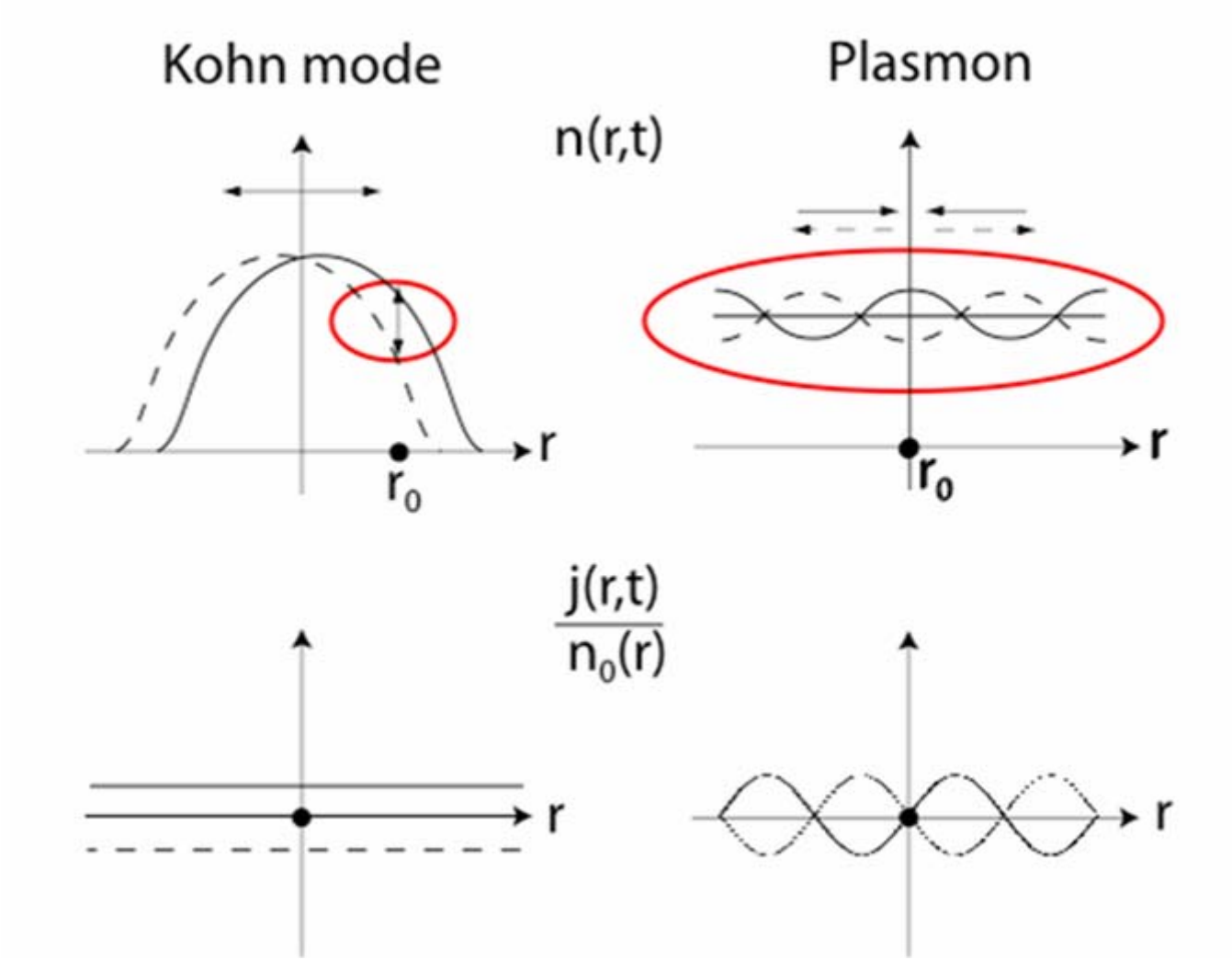
local compression and rarefaction

V_{xc} "rides along": undamped motion

V_{xc} is retarded: damped motion

➡ xc functionals based on local density can't distinguish the two cases!

Point of view of the local current

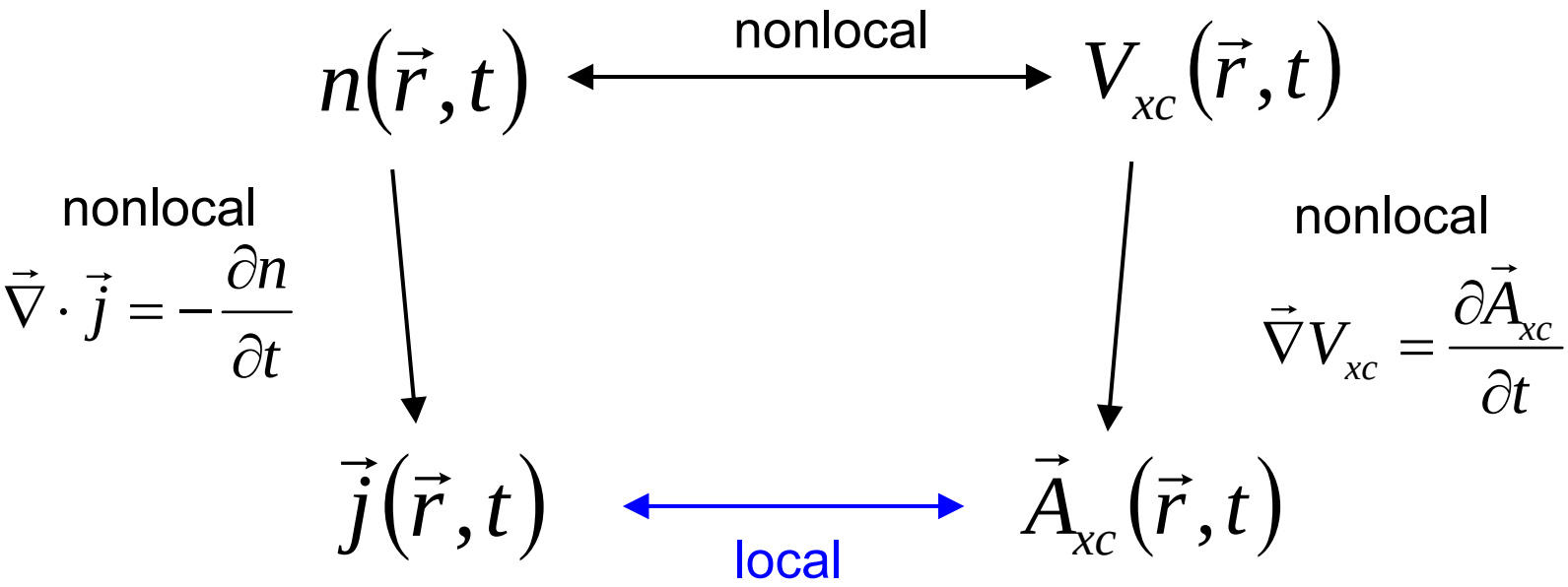


uniform velocity

oscillating velocity

much better chance to capture the physics correctly!

"Upgrading" TDDFT: Current-TDDFT



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

- Continuity equation only gives the longitudinal current
- C-TDDFT gives also the transverse current
- We can find a short-range current-dependent xc vector potential

C-TDDFT

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

$$\hat{H}_{\text{int}}(t) = \sum_i \left\{ \frac{1}{2}\left[\vec{p}_i + \tfrac{1}{c}\vec{A}_{ext}(\vec{r}_i,t)\right]^2 + V_{ext}(\vec{r}_i,t) \right\} + \sum_{i>j} U(\vec{r}_i - \vec{r}_j)$$

$\Downarrow$

$$\vec{j}(\vec{r},t) = \vec{j}_L(\vec{r},t) + \vec{j}_T(\vec{r},t)$$

full current can be represented by a KS system

$\Uparrow$

$$\hat{H}_{KS}(t) = \sum_i \left\{ \frac{1}{2}\left[\vec{p}_i + \tfrac{1}{c}\vec{A}_{KS}(\vec{r}_i,t)\right]^2 + V_{KS}(\vec{r}_i,t) \right\}$$

uniquely determined up to gauge transformation

C-TDDFT in the linear response regime

$$\vec{j}_1(\vec{r},\omega) = \int d^3r' \; \vec{\vec{\chi}}_{KS}(\vec{r},\vec{r}',\omega)\left\{\vec{A}_{ext,1}(\vec{r},\omega) + \vec{A}_{H,1}(\vec{r},\omega) + \vec{A}_{xc,1}(\vec{r},\omega)\right\}$$

KS current-current response tensor: diamagnetic + paramagnetic part

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

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

Exercise: show that $$\chi(\vec{r},\vec{r}',\omega) = \frac{1}{\omega^2}\sum_{\mu\nu}\nabla_\mu\nabla_\nu'\chi_{\mu\nu}(\vec{r},\vec{r}',\omega)$$

C-TDDFT: effective vector potential

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

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

gauge transformed Hartree potential

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

the xc kernel is now a tensor!

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

C-TDDFT beyond the ALDA: the VK functional

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

xc viscoelastic stress tensor:

$$\sigma_{xc,jk} = \tilde{\eta}_{xc}\left(\nabla_j v_{1,k} + \nabla_k v_{1,j} - \frac{2}{3}\vec{\nabla}\cdot\vec{v}_1\delta_{jk}\right) + \tilde{\zeta}_{xc}\vec{\nabla}\cdot\vec{v}_1\delta_{jk}$$

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

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

...but how on earth did they come up with this expression??

C-TDDFT beyond the ALDA

1 Derivation by "brute force" [G. Vignale and W. Kohn, PRL **77**, 2037 (1996)]

- Consider weakly inhomogeneous electron liquid, modulated by a charge-density wave of small amplitude and wavevector.
- Calculate xc kernel $f_{xc}(k+q,k,\omega)$ where $k,q << k_F, \omega / v_F$
- It turned out later that the resulting expression can be cast into hydrodynamic form

2 Physical insight [G. Vignale, C.A.U., and S. Conti, PRL **79**, 4878 (1997), G. Vignale, TDDFT-book, chapter 5]

- Classical theories of the dynamics of continuous media (elasticity and hydrodynamics) express many-body forces as divergences of stress tensors
- local functionals of the displacement or velocity field
- use general symmetries and sum rules

xc viscosity coefficients

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

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

In contrast with the classical case, the xc viscosities have both real and imaginary parts, describing dissipative and elastic behavior:

$$\tilde{\eta}(\omega) = \eta(\omega) - \frac{S_{xc}(\omega)}{i\omega}$$

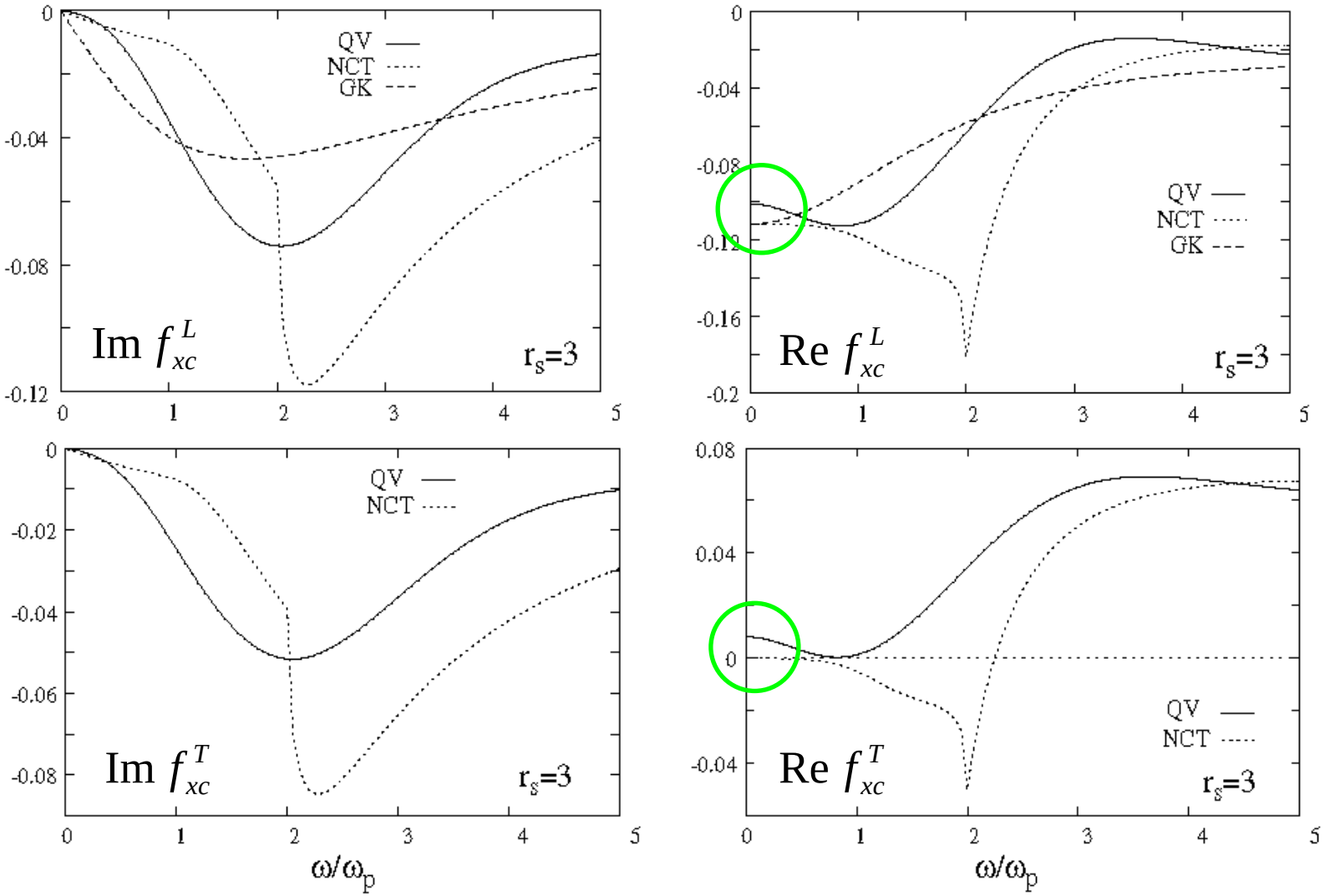
shear modulus

$$\tilde{\zeta}(\omega) = \zeta(\omega) - \frac{B_{xc}^{dyn}(\omega)}{i\omega}$$

dynamical bulk modulus

reflect the stiffness of Fermi surface against defor-mations

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)

static limits of the xc kernels

$$f_{xc}^{L}(0) = \frac{d^2 e_{xc}^{unif}(n)}{dn^2} + \frac{4}{3}\frac{S_{xc}(0)}{n^2}$$

$$f_{xc}^{T}(0) = \frac{S_{xc}(0)}{n^2}$$

The shear modulus of the electron liquid does **not** disappear for $\omega \to 0$. (as long as the limit q→0 is taken first). Physical reason:

- Even very small frequencies <<E_F are large compared to relaxation rates from electron-electron collisions.
- The zero-frequency limit is taken such that local equilibrium is not reached.
- The Fermi surface remains stiff against deformations.

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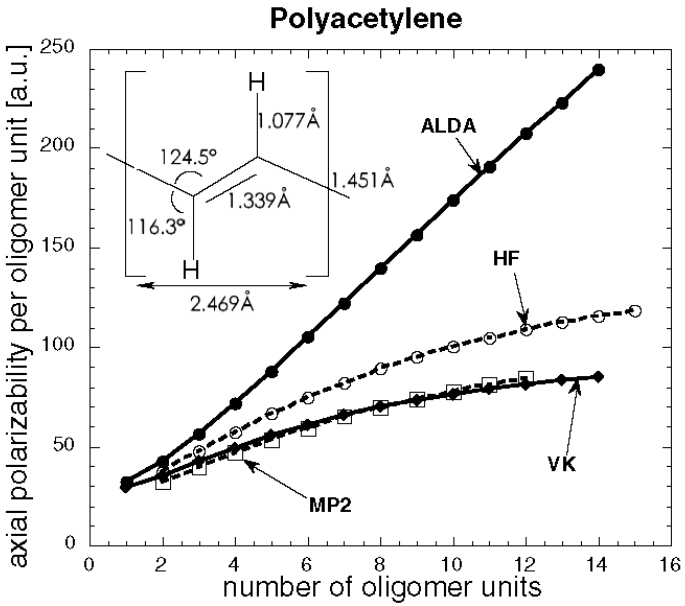
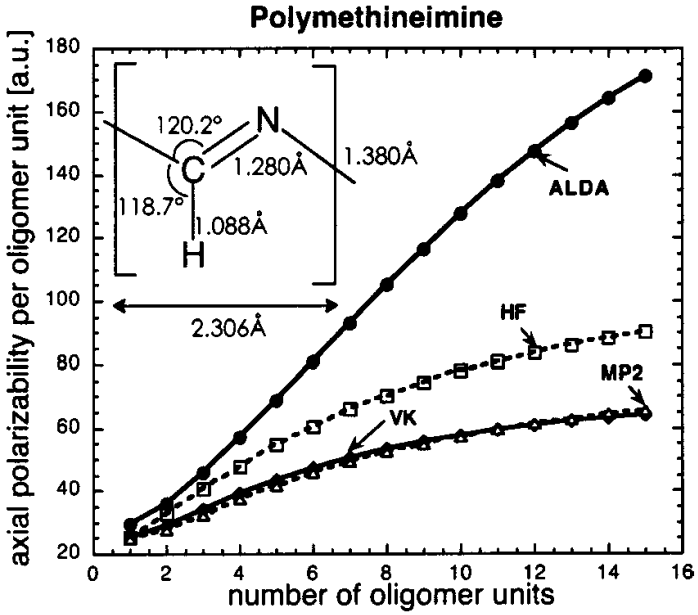
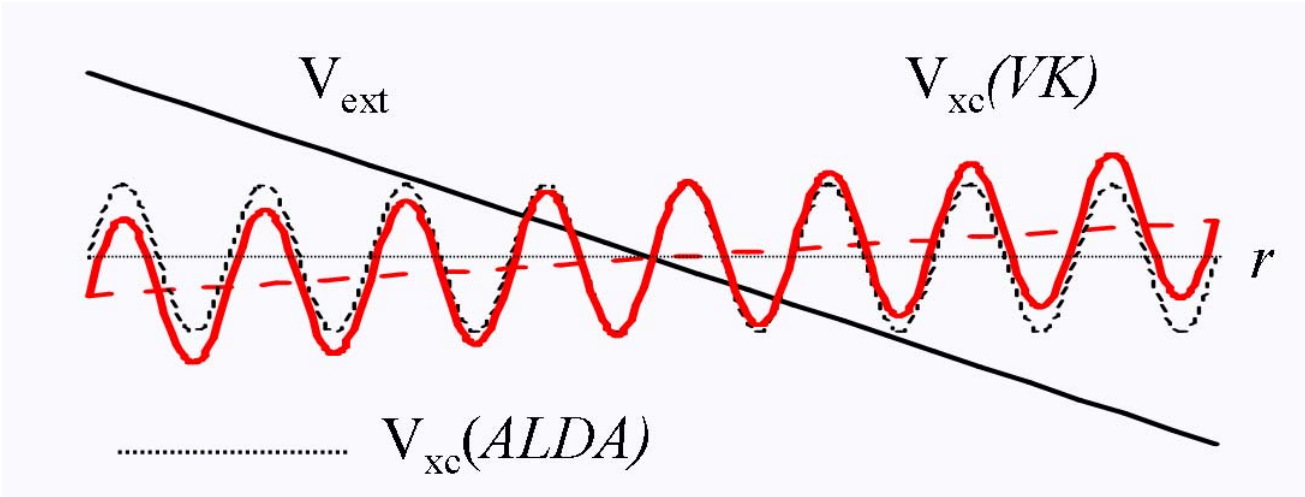
C-TDDFT for conjugated polymers





ALDA overestimates polarizabilities of long molecular chains. The long-range VK functional produces a counteracting field, due to the finite shear modulus at $\omega \to 0$.



M. van Faassen et al., PRL **88**, 186401 (2002) and JCP **118**, 1044 (2003)

Atomic excitation energies

lowest $^1S \rightarrow {}^1S$ (in eV)

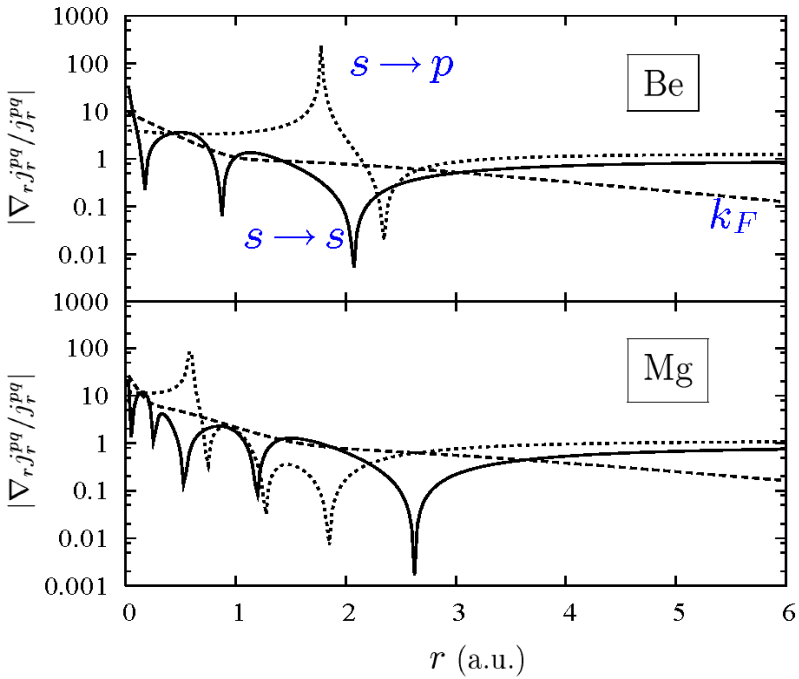
	Exp	bare KS	ALDA	VK
Be $2s \rightarrow 3s$	6.78	5.56	5.62	$5.67 - 0.04\,i$
Mg $3s \rightarrow 4s$	5.39	4.72	4.78	$4.83 - 0.05\,i$
Ca $4s \rightarrow 5s$	4.13	3.77	3.81	$3.87 - 0.06\,i$
Sr $5s \rightarrow 6s$	3.79	3.50	3.54	$3.59 - 0.06\,i$

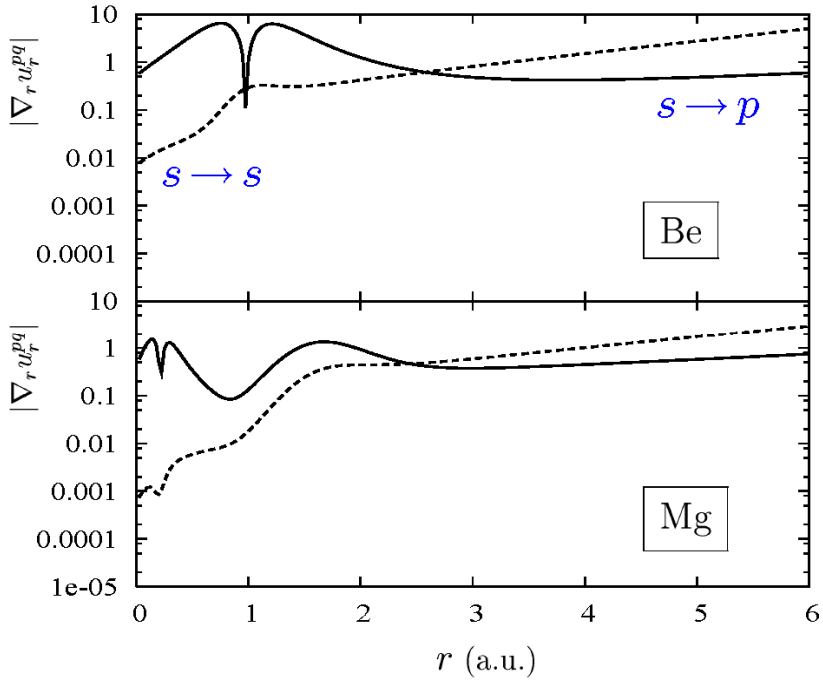
lowest $^1S \rightarrow {}^1P$ (in eV)

	Exp	bare KS	ALDA	VK
Be $2s \rightarrow 2p$	5.28	3.50	5.08	$6.24 - 0.69\,i$
Mg $3s \rightarrow 3p$	4.35	3.39	4.57	$4.86 - 0.09\,i$
Ca $4s \rightarrow 4p$	2.93	2.39	3.38	$3.22 - 0.06\,i$
Sr $5s \rightarrow 5p$	2.69	2.22	3.11	$-1.63 - 0.06\,i$

C.A.U. and K. Burke, JCP **121**, 28 (2004)

Analysis of the VK functional for atoms





conditions for validity of the VK functional:

$$\left|\frac{\nabla n_0}{n_0}\right|, \left|\frac{\nabla j_\nu}{j_\nu}\right|, \left|\frac{\nabla v_\nu}{v_\nu}\right| << k_F, \frac{\omega}{v_F}$$

OK for s→s, but violated for s→p!

Analysis of the VK functional for atoms

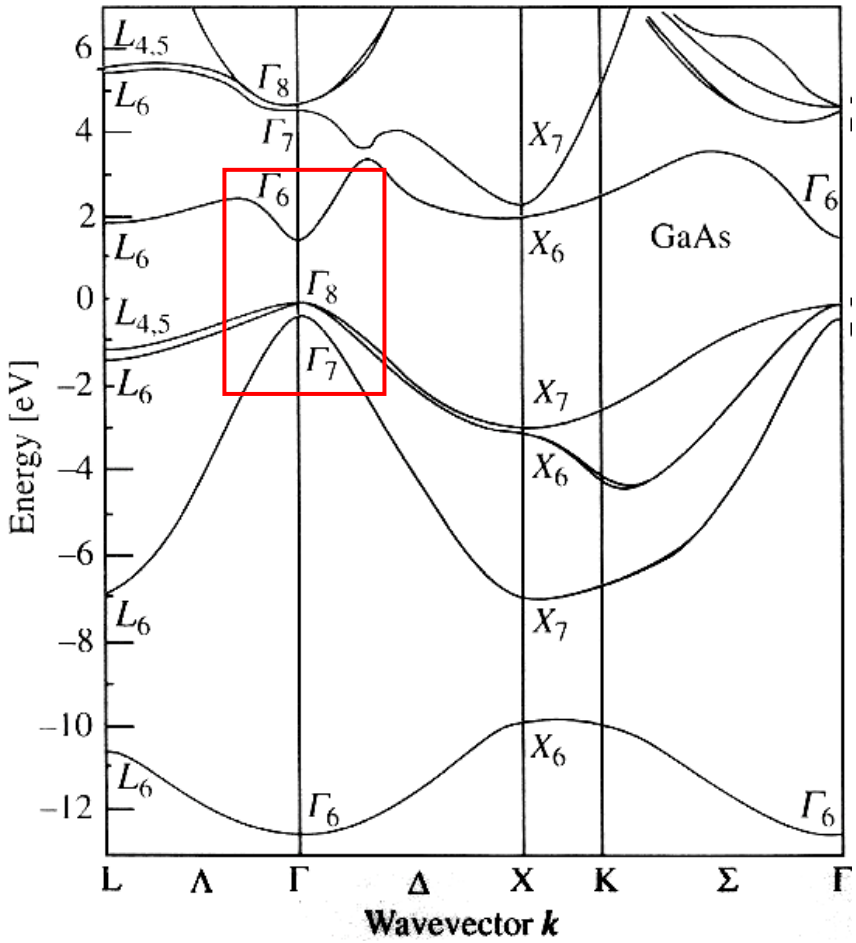
► Frequency shifts:

- small for s→s, but tend to overshoot for s→p excitations
- need more accurate $f_{xc}^{L,T}(\omega)$, especially around nucleus (r_s<<1)
- excitations with large ∇j are problematic. Need higher gradients.
  [partial cure: Tao and Vignale, PRL (2006)]

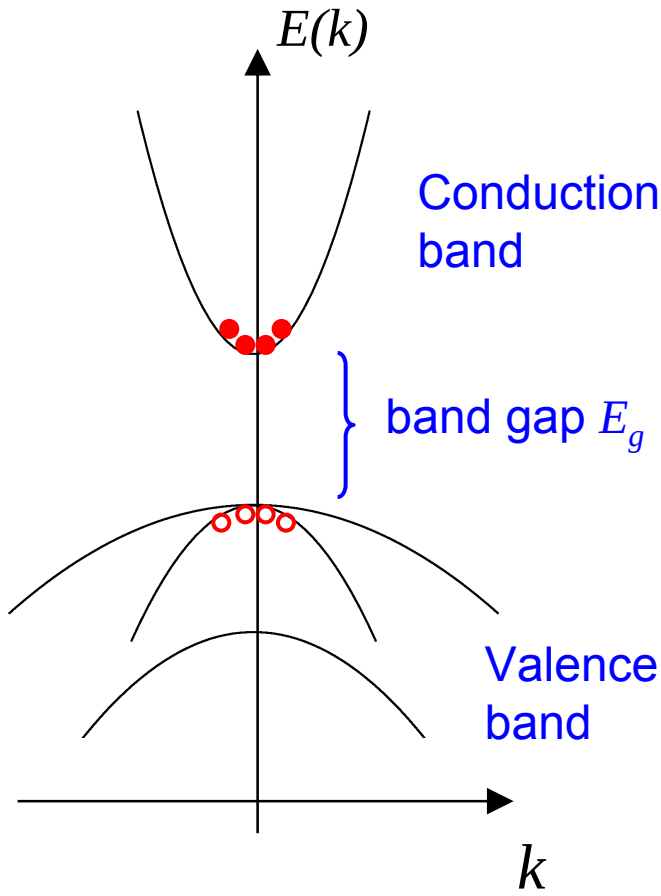
► Imaginary parts:

- small but finite, often of the same order as frequency shifts.
- unphysical: a finite system ought to have zero linewidth.
  Difficult to achieve for a functional with the homogeneous electron gas as reference system!

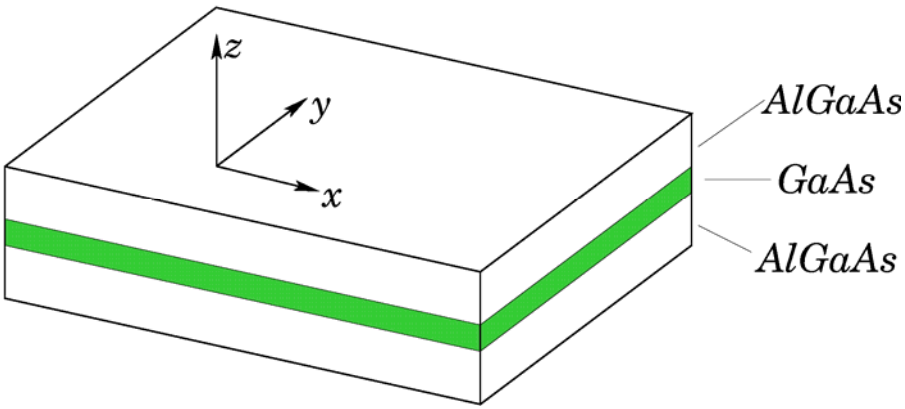
GaAs band structure



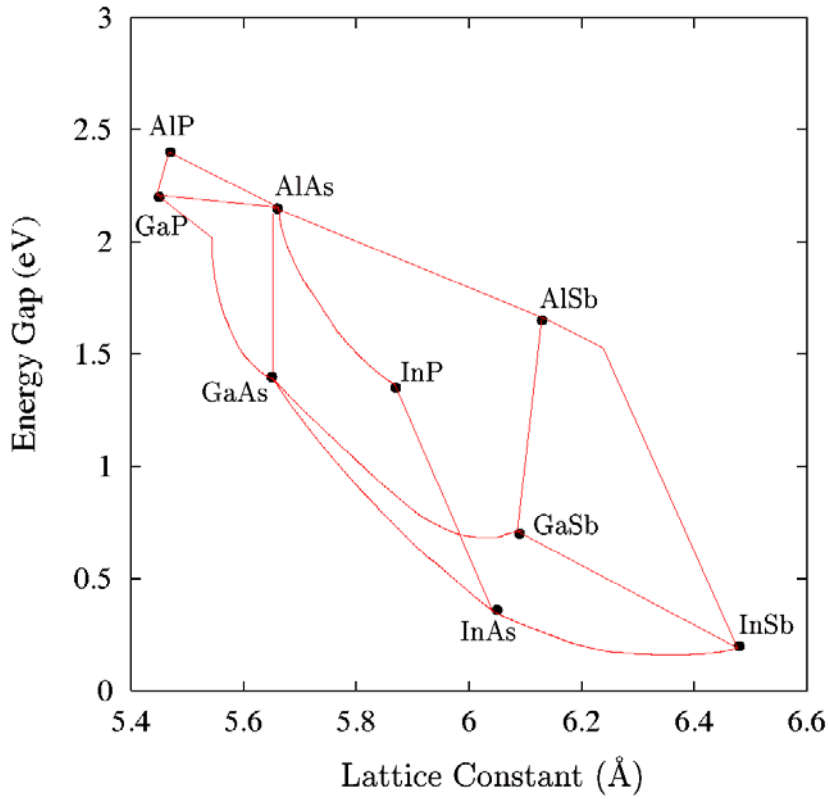
Around the zone center:



Quantum well basics

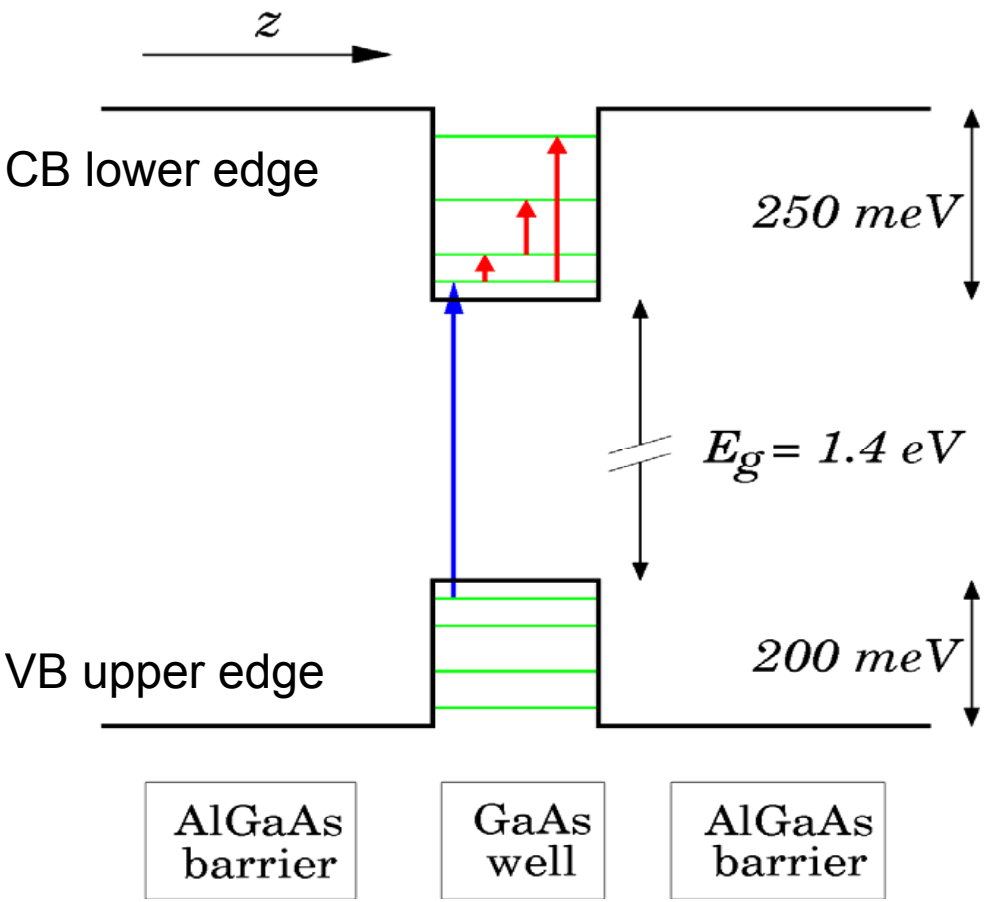


"Map of the World"



- semiconductor heterostructures are grown with MBE or MOCVD
- control and design through layer-by-layer variation of material composition
- widely used class or materials: III-V compounds

Electronic transitions in quantum wells



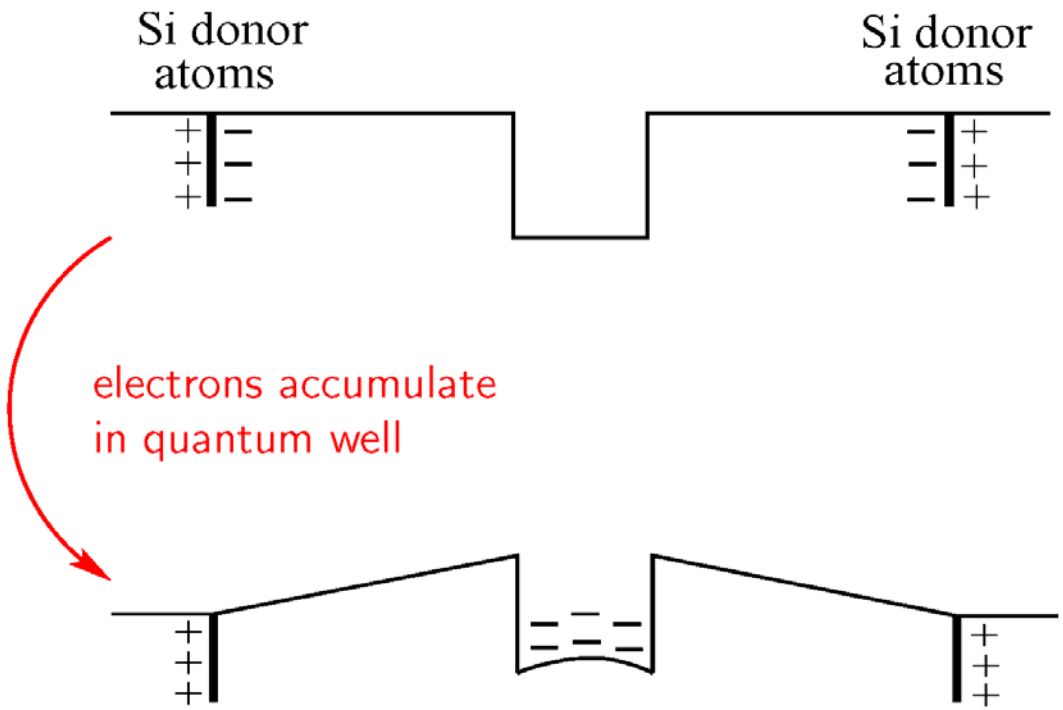
Interband transitions:
of order eV
(visible to near-IR)

Intersubband transitions:
of order meV
(mid- to far-IR)

10 meV = 2.4 THz

n-doped quantum wells

- Donor atoms separated from quantum well: **modulation delta doping**
- Total sheet density N_s typically ~10^{11} cm^{-2}



Electronic ground state: subband levels

Effective-mass approximation: $m^* = \mu m \qquad e^* = e/\sqrt{\kappa}$

$(\text{for GaAs}: \mu = 0.067,\ \kappa = 13)$

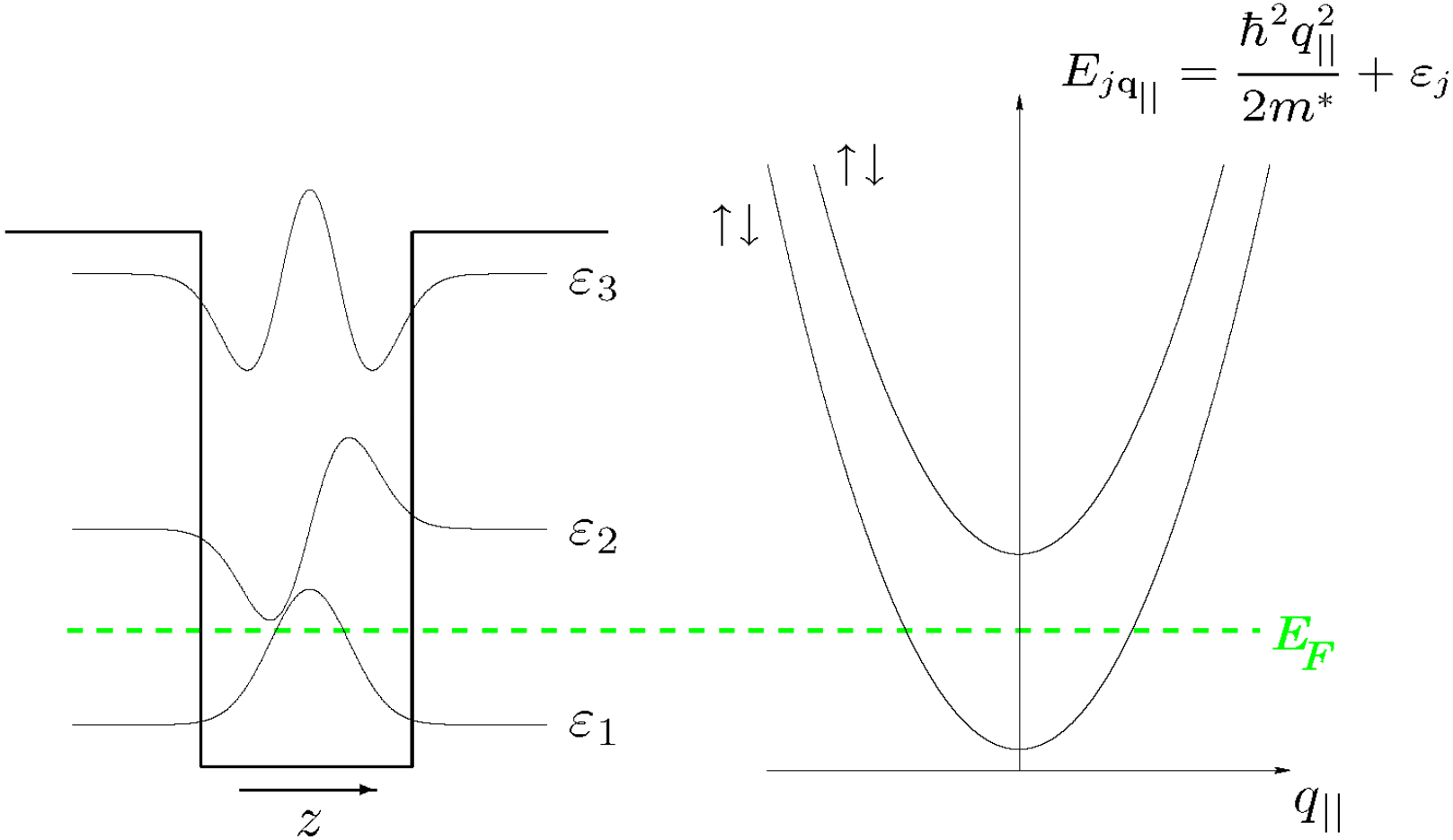
Electrons in a quantum well: plane waves in *x-y* plane, confined along *z*

$$\psi_{jq_\parallel}(\vec{r}) = \frac{1}{\sqrt{A}} e^{iq_\parallel r_\parallel} \varphi_j(z) \quad \text{with energies} \quad E_{jq_\parallel} = \frac{\hbar^2 q_\parallel^2}{2m^*} + \varepsilon_j$$

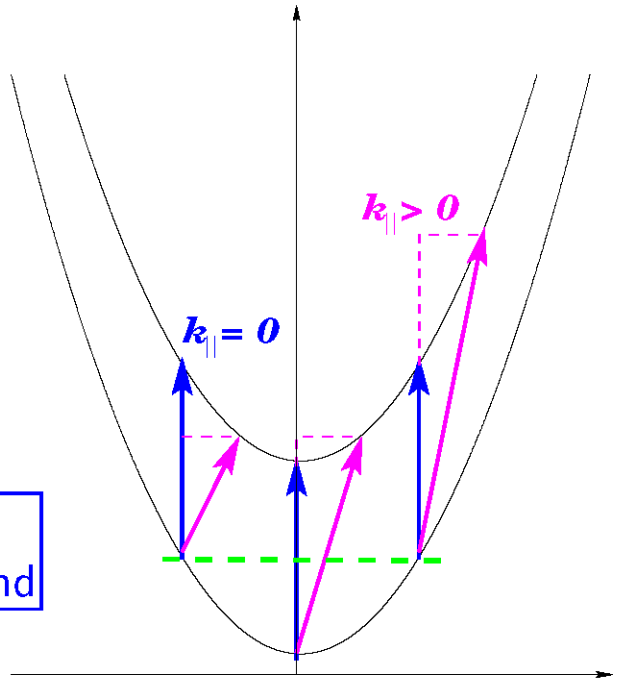
quantum well confining potential

$$\left[-\frac{\hbar^2}{2m^*}\frac{d^2}{dz^2} + V_{conf}(z) + V_H(z) + V_{xc}^{LDA}(z) \right] \varphi_j(z) = \varepsilon_j \varphi_j(z)$$

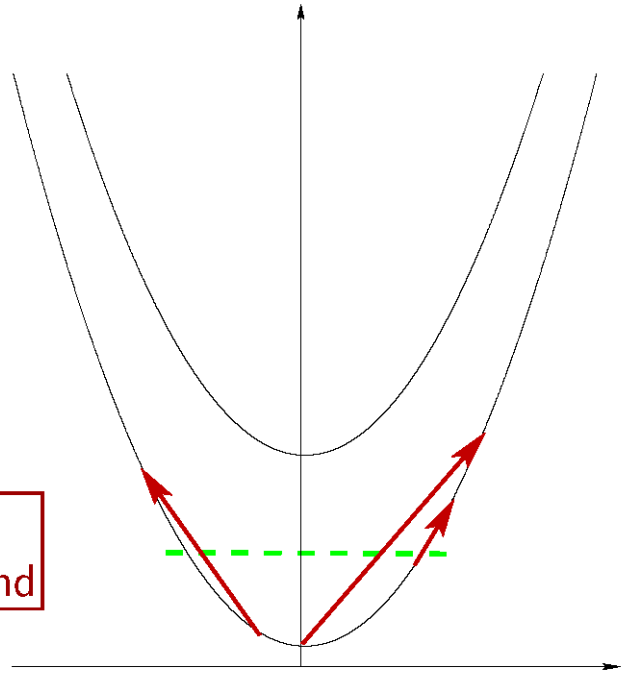
Quantum well subbands



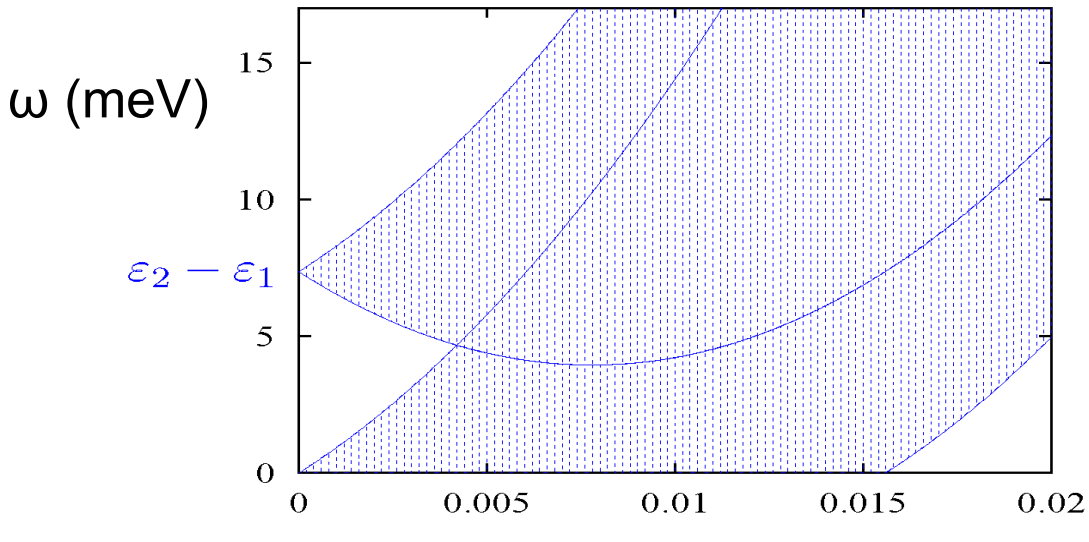
Single-particle excitations



Inter-subband

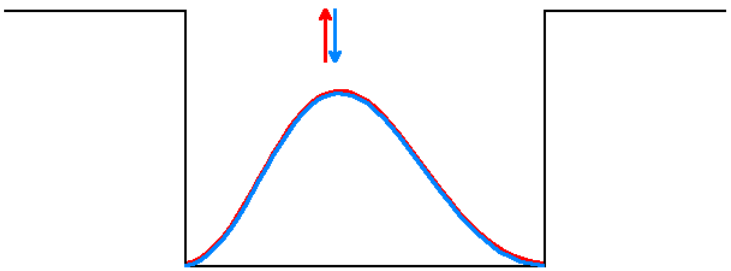


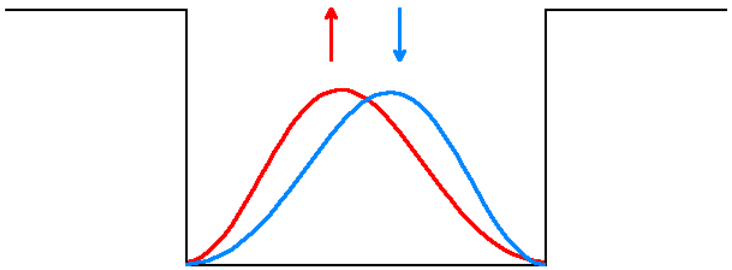
Intra-subband



40 nm GaAs/AlGaAs quantum well

Collective excitations

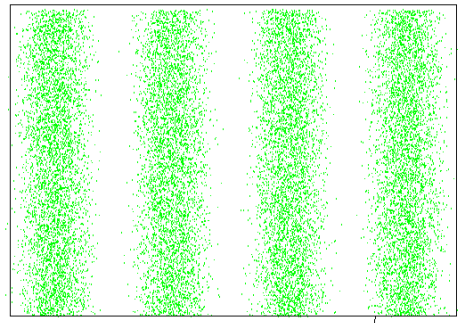


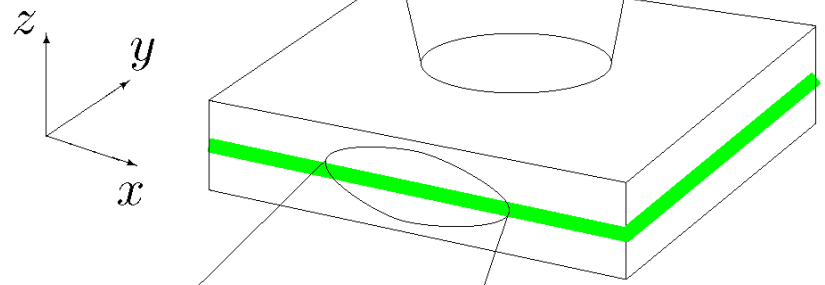


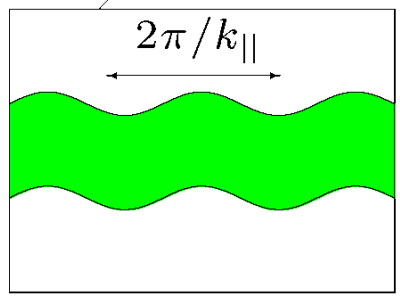
Intersubband charge and spin plasmons: ↑ and ↓ densities in and out of phase

2D Plasmon

(in-plane motion)







Intersubband Plasmon

(out-of-plane motion)

TDDFT linear response theory for quantum wells

C.A.U. and G. Vignale, PRB **58**, 15756 (1998)

$$n_{1\sigma}(k_{\|},z,\omega)=\sum_{\sigma'}\int dz'\,\chi^{KS}_{\sigma\sigma'}(k_{\|},z,z'\omega)V^{ext+Hxc}_{1\sigma'}(k_{\|},z',\omega)$$

$$\chi^{KS}_{\sigma\sigma'}(k_{\|},z,z',\omega)=\delta_{\sigma\sigma'}\sum_{\mu=1}^{N_{occ}}\sum_{\nu=1}^{\infty}F_{\mu\nu}(k_{\|},\omega)\varphi_{\mu}(z)\varphi_{\nu}(z)\varphi_{\mu}(z')\varphi_{\nu}(z')$$

$$F_{\mu\nu}(k_{\|},\omega)=\int\frac{d^2q_{\|}}{(2\pi)^2}\left[\frac{\theta(E_F-E_{\mu q_{\|}})}{\omega-\omega_{\nu\mu}-k_{\|}q_{\|}-\frac{k_{\|}^2}{2}+i\eta}-\frac{\theta(E_F-E_{\mu q_{\|}})}{\omega+\omega_{\nu\mu}+k_{\|}q_{\|}+\frac{k_{\|}^2}{2}+i\eta}\right]$$

$$V^{Hxc}_{1\sigma}(k_{\|},z,\omega)=\sum_{\sigma'}\int dz'\left[2\pi\frac{e^{-k_{\|}|z-z'|}}{k_{\|}}+f_{xc,\sigma\sigma'}(k_{\|},z,z',\omega)\right]n_{1\sigma'}(k_{\|},z',\omega)$$

Collective excitations in ALDA ($k_{||}$=0)

$$n_{1\sigma}(z,\Omega)=\sum_{\sigma'\sigma''}\int dz'\,\chi^{KS}_{\sigma\sigma'}(z,z',\Omega)\left[-2\pi\,|z-z'|+f^{ALDA}_{xc,\sigma'\sigma''}(z,z',\Omega)\right]n_{1\sigma''}(z',\Omega)$$

Find those frequencies Ω where there is a finite solution of the response equation without any external perturbation (eigenmodes of the system).

Consider the case N_{occ}=1 (only the lowest subband is occupied:

$$\chi^{KS}_{\sigma\sigma'}(z,z',\omega)=N_s\delta_{\sigma\sigma'}\sum_{\nu=1}^{\infty}\left[\frac{1}{\omega-\omega_{\nu 1}}-\frac{1}{\omega+\omega_{\nu 1}}\right]\varphi_1(z)\varphi_\nu(z)\varphi_1(z')\varphi_\nu(z')$$

ALDA xc kernel: $$f^{ALDA}_{xc,\sigma\sigma'}(z,z')=\delta(z-z')\left.\frac{d^2e^{unif}_{xc}(n_\uparrow,n_\downarrow)}{dn_\sigma dn_{\sigma'}}\right|_{n=n_0(z)}$$

Since the system is not spin polarized: $$f^{ALDA}_{xc,\uparrow\uparrow}=f^{ALDA}_{xc,\downarrow\downarrow},\quad f^{ALDA}_{xc,\uparrow\downarrow}=f^{ALDA}_{xc,\downarrow\uparrow}$$

The small-matrix approximation

Approximate as a two-level system:

$$\chi^{KS}_{\sigma\sigma'}(z,z',\omega) \approx N_s \delta_{\sigma\sigma'} \left[\frac{1}{\omega - \omega_{21}} - \frac{1}{\omega + \omega_{21}} \right] \varphi_1(z)\varphi_2(z)\varphi_1(z')\varphi_2(z')$$

Define double-matrix element:

$$S_{\sigma\sigma'} = \frac{N_s}{2} \int dz \int dz' \left[-2\pi \,|\, z - z' \,| + f^{ALDA}_{xc,\sigma\sigma'}(z,z') \right] \varphi_1(z)\varphi_2(z)\varphi_1(z')\varphi_2(z')$$

$$\Omega^2_{\pm} = \omega^2_{21} + 2\omega_{21}\left(S_{\uparrow\uparrow} \pm S_{\uparrow\downarrow} \right)$$

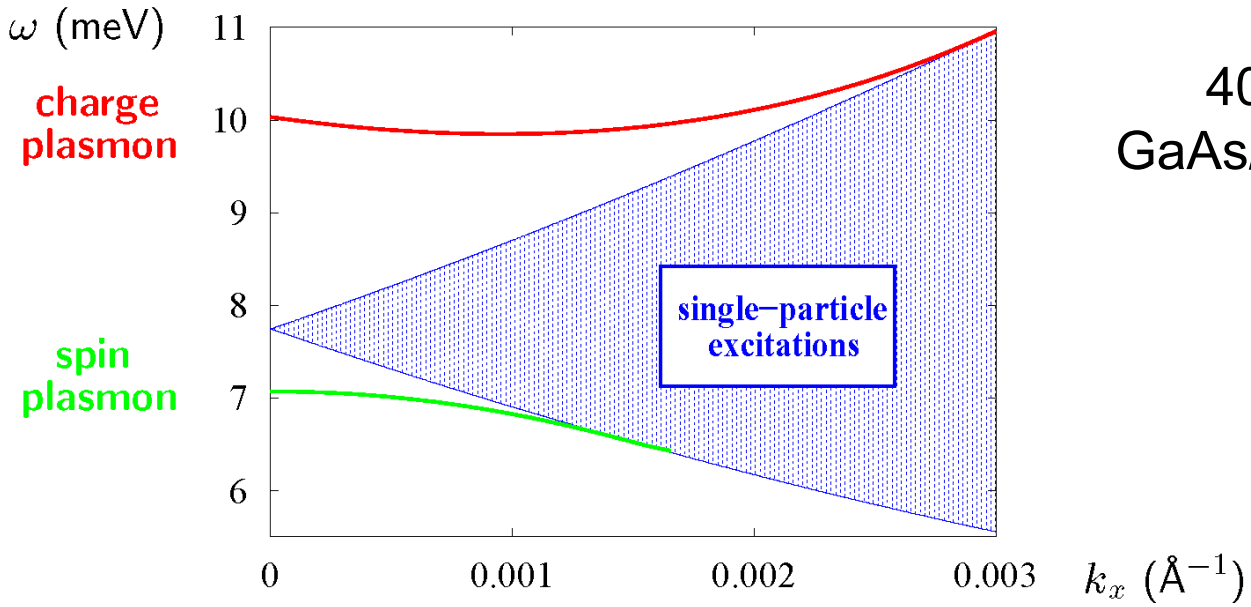
+ Charge Plasmon: large Hartree up-shift and smaller xc down-shift

– Spin Plasmon: Hartree cancels out, only small xc down-shift

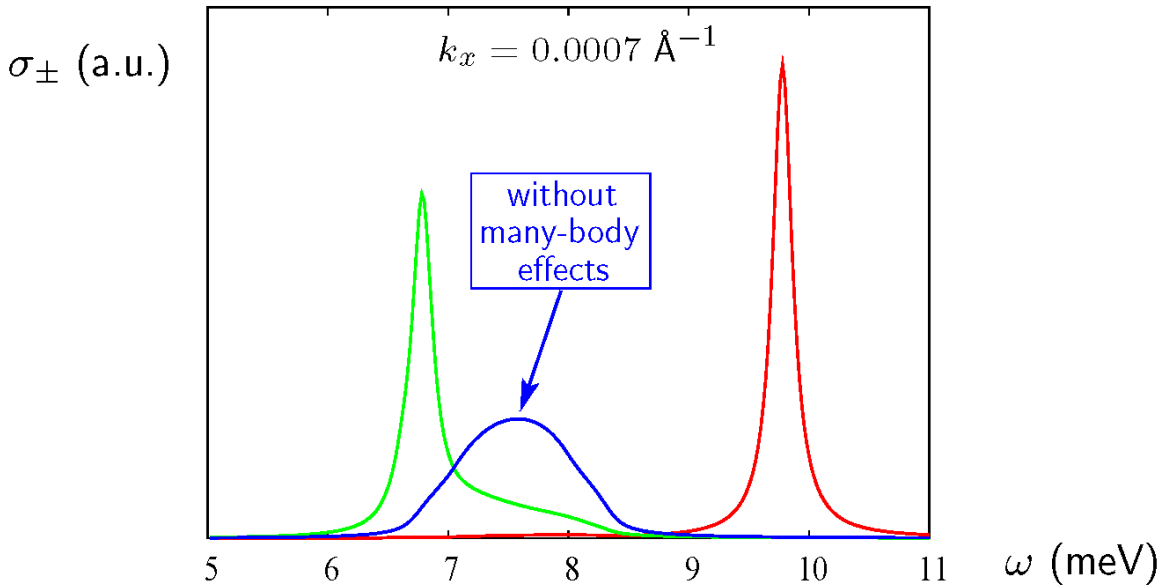
The Hartree contribution is also known as “depolarization shift”

[S.J. Allen, D.C. Tsui, and B. Vinter, Solid State Commun. **20**, 425 (1976)

Intersubband plasmon dispersions



40 nm
GaAs/AlGaAs

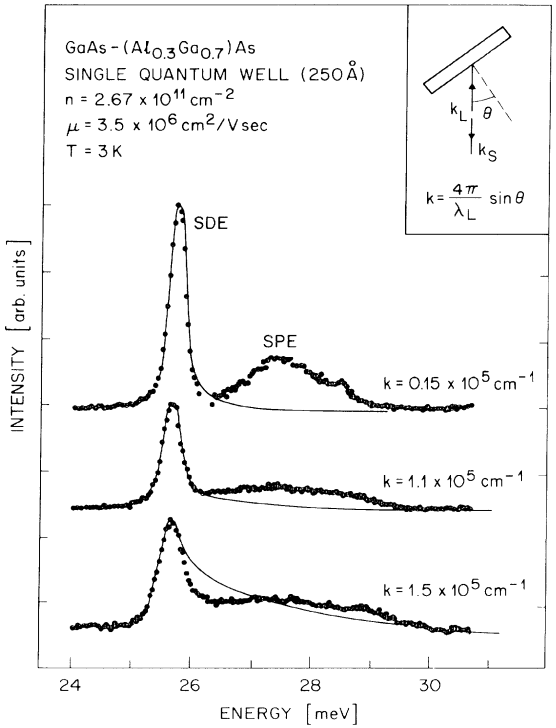


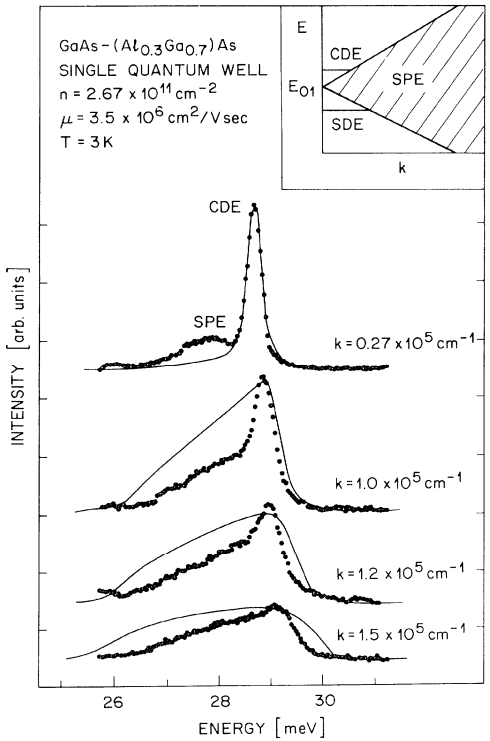
Inelastic light scattering

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





"depolarized"

spin plasmons

"polarized"

charge plasmons

C-TDDFT for intersubband plasmons

Since this is a "1D" system, we can integrate the continuity equation:

$$\frac{\partial j_1}{\partial z} = i\omega n_1 \quad \Rightarrow \quad j_1(z,\omega) = i\omega \int_{-\infty}^{z} n_1(z',\omega)dz'$$

and we can construct the xc scalar potential from the xc vector potential:

$$\frac{\partial V_{xc,1}}{\partial z} = i\omega A_{xc,1} \quad \Rightarrow \quad V_{xc,1}(z,\omega) = i\omega \int_{-\infty}^{z} A_{xc,1}(z',\omega)dz'$$

We only need the zz component of the xc stress tensor:

$$\sigma_{xc,zz}(z,\omega) = \left(\zeta_{xc} + \frac{4}{3}\eta_{xc}\right)\frac{\partial v(z,\omega)}{\partial z}$$

where

$$\zeta_{xc} + \frac{4}{3}\eta_{xc} = -\frac{n_0^2(z)}{i\omega}\left(f_{xc}^{L}(n,\omega) - \frac{d^2 e_{xc}^{unif}}{dn^2}\right)_{n=n_0(z)}$$

f_{xc}^{dyn}

Explicit expression for the xc kernel in 1D

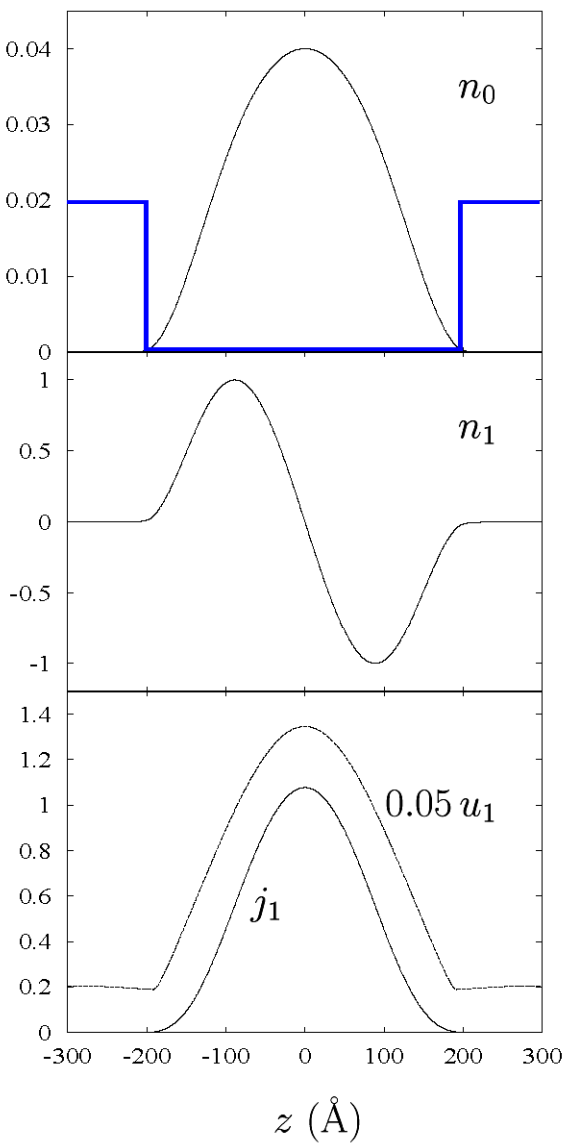
$$f_{xc}^{VK}(z,z',\omega) = f_{xc}^{L}(z,\omega)\delta(z-z')$$
$$- f_{xc}^{dyn}(z,\omega)\frac{n_0'(z)}{n_0(z)}\theta(z-z') - f_{xc}^{dyn}(z',\omega)\frac{n_0'(z')}{n_0(z')}\theta(z'-z)$$
$$+ \int dz''\theta(z''-z)\theta(z''-z')\, f_{xc}^{dyn}(z'',\omega)\left(\frac{n_0'(z'')}{n_0(z)}\right)^2$$

- long-range nature of the xc kernel explicitly visible
- satisfies Harmonic Potential Theorem and other symmetries

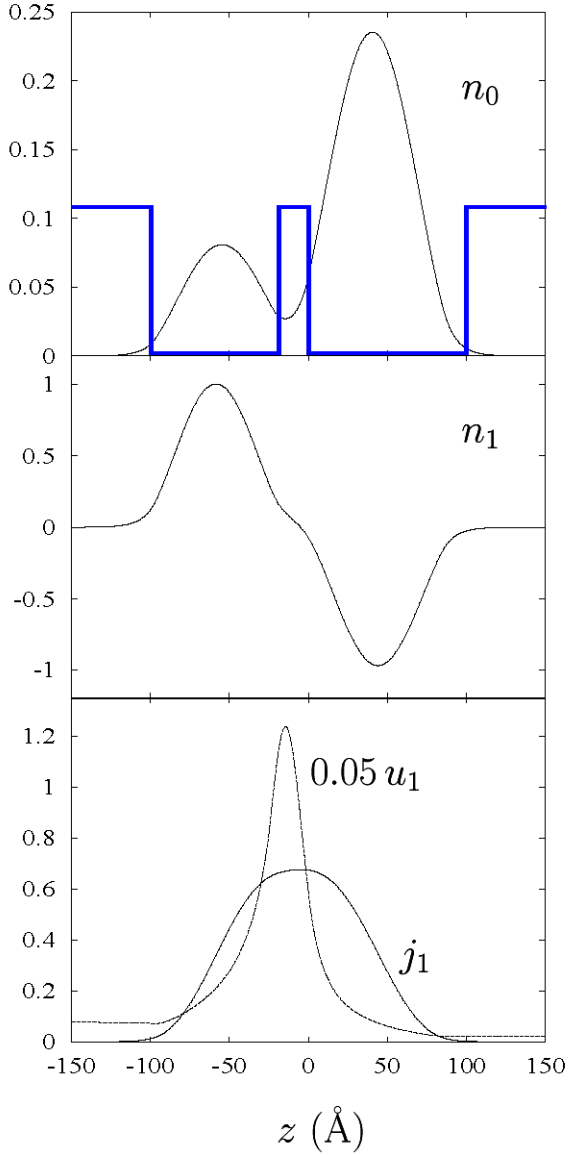
G. Vignale and W. Kohn, in “Electronic DFT” (Plenum, 1998)

Exercise: derive this expression!

Intersubband plasmons: mode profiles

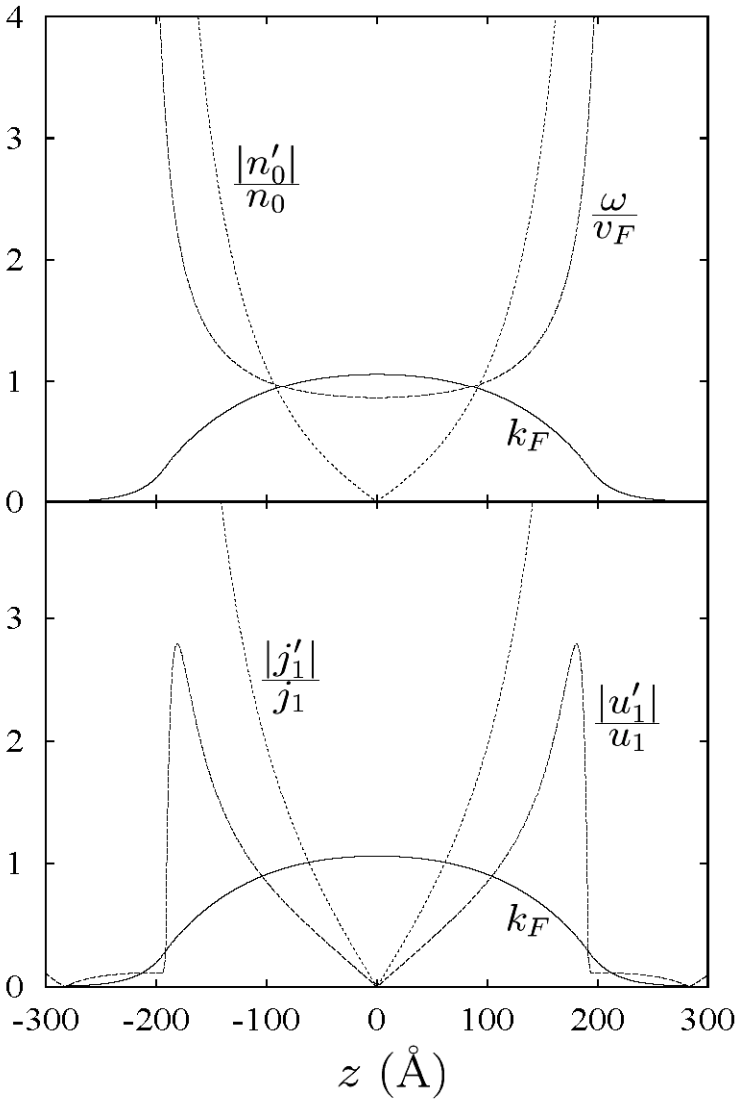


single well

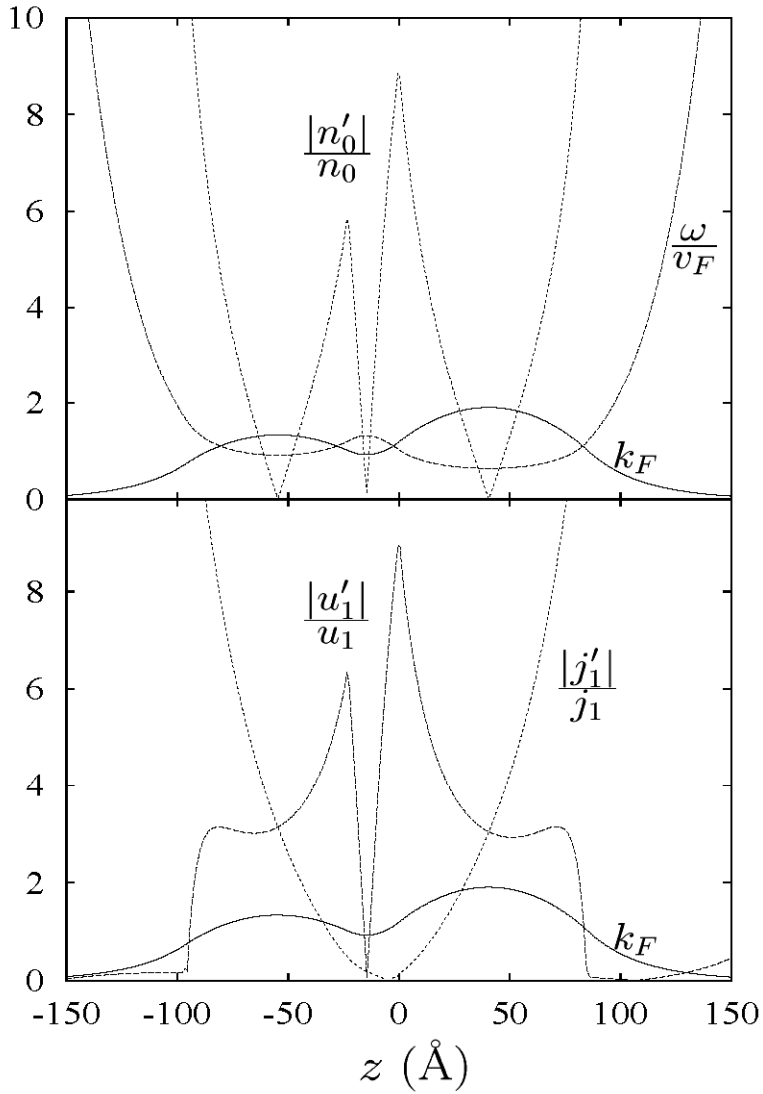


double well

Analysis of the VK functional for ISB plasmons



single well



double well

Analysis of the VK functional for ISB plasmons

Single well

	ALDA	VK [1]	VK [2]	Exp
Ω	10.25	10.31	10.24	10.7
Γ		0.128	0.104	0.53

Double well

	ALDA	VK [1]	VK [2]	Exp
Ω	13.85	20.64	12.55	14.6
Γ		8.55	4.15	1.17

VK [1]: f_{xc} Gross/Kohn 1985
VK [2]: f_{xc} Nifosi/Conti/Tosi 1998

Single well: VK functional works fine (linewidth Γ below exp. value)

Double well: VK functional breaks down (tunneling through barrier)

detailed comparison with experimental data, including disorder effects:
C.A.U. and G. Vignale, PRL **87**, 036402 (2001)
C.A.U. and G. Vignale, PRB **65**, 245102 (2002)

End of the first lecture

Today's summary:

- density-based nonadiabatic xc functionals in TDDFT are plagued by ultranonlocality
- upgrading to C-TDDFT makes a local approximation possible
- natural way of describing dynamical xc effects via viscoelastic stresses in the electron liquid
- works great for polarizabilities of polymers and plasmon linewidths, not so good for atoms.

Tomorrow:

- dynamics in the time domain: memory and dissipation in TDKS
- a rigorous extension of the LDA: L-TDDFT versus C-TDDFT
- more examples: spin Coulomb drag, and some toy models