Quantum electron dynamics in thin metal films

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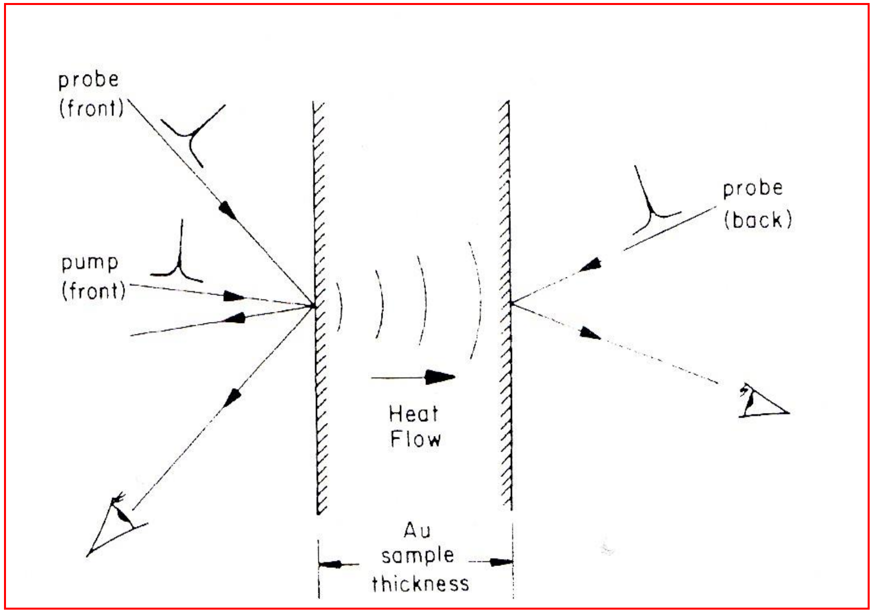
Outline

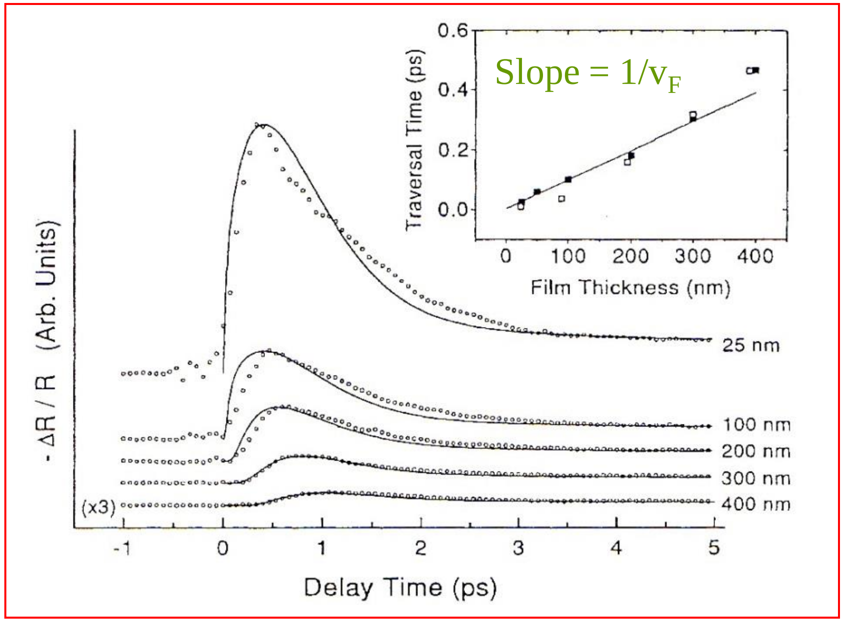
- **Motivations: quantum electron dynamics**
  - ***Electrons in solid-state nanometric devices***
- **Phase-space modeling: from classical to quantum**
  - ***Semi-classical: Vlasov equation***
  - ***Quantum: Wigner functions, hydrodynamics***
- **Results: Electron dynamics in thin metal films**
  - ***Quantum-classical transition***
  - ***Relaxation and decoherence***
- **Spin and relativistic effects**
  - ***Vlasov equation with spin***
  - ***Semi-relativistic approaches***

Quantum electron dynamics in thin metal films

- Experimental study of heat flow through Au films (“pump-probe”)
- Typical results:
  - **Heat transport is ballistic, not diffusive**
  - **It occurs at Fermi velocity of the metal**

> ➢ Ballistic: T = L / V
>
> ➢ Diffusive: $T = L^2 / D$

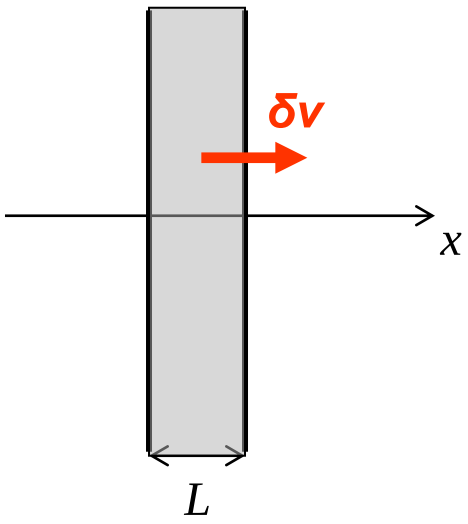




C. Suarez et al., Phys. Rev. Lett. **75**, 4536 (1995)

Classical model

- 1D slab geometry
  - Only fields normal to the film surface are considered
- Electron dynamics in the (x, v) phase space: classical Vlasov-Poisson system
- Ions: fixed ion density profile $n_i(x)$ (“jellium”)
- Initially, electrons are at equilibrium at finite temperature
- Electron are excited at t=0 by imparting a velocity ***δv*** to their equilibrium distribution



$$\frac{\partial f_e}{\partial t} + v\frac{\partial f_e}{\partial x} + \frac{e}{m_e}\frac{\partial \phi}{\partial x}\frac{\partial f_e}{\partial v} = 0$$

$$\frac{d^2\phi}{dx^2} = \frac{e}{\varepsilon_0}[n_e(x) - n_i(x)]$$

$$n_e(x,t) = \int f_e(x,v,t)dv$$

Physical parameters for sodium and gold films

	units	Au	Na
$\bar{n}_i$	m^{-3}	5.9×10^{28}	2.5×10^{28}
ω_p^{-1}	fs	0.07	0.11
$\hbar\omega_p$	eV	9.02	5.87
E_F	eV	5.53	3.12
T_F	K	6.41×10^4	3.62×10^4
L_F	nm	0.09	0.12
v_F	ms^{-1}	1.39×10^6	1.05×10^6
r_s	nm	0.16	0.21
r_s/a_0	—	3.01	4.0

$(L_F = v_F / \omega_p)$

NB: we will describe generic effects that do not depend on the specific properties of the metal

Relevant energy quantities

$$\text{Total energy}: \quad E_{tot} = E_{kin} + E_H$$

$$\text{Kinetic energy}: \quad E_{kin} = E_{cm} + E_{TF} + E_{th}$$

$$\text{Center-of-mass energy}: \quad E_{cm} = \frac{m}{2}\int u^2 n dx$$

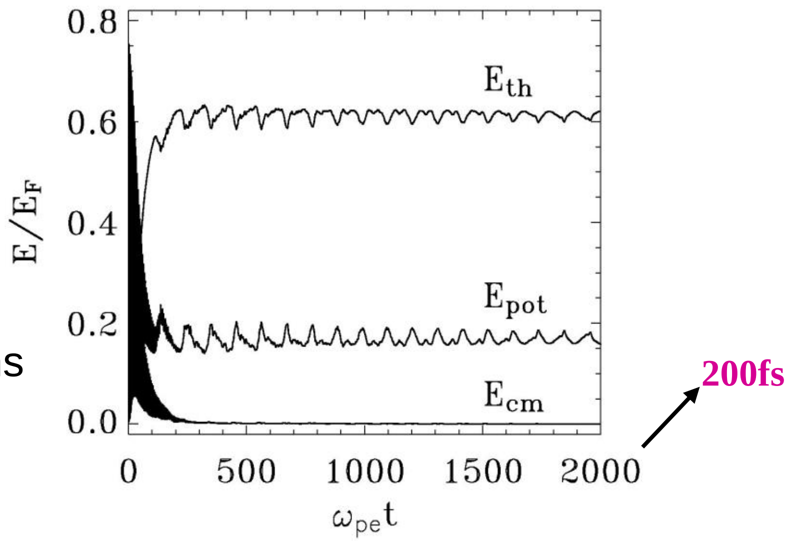
$$\text{Thomas-Fermi energy}: \quad E_{TF} = \frac{1}{5}\frac{\hbar^2}{2m}\left(3\pi^2\right)^{2/3} n^{5/3}$$

$$\text{Thermal energy}: \quad E_{th} = E_{kin} - E_{cm} - E_{TF}$$

Main results for sodium films

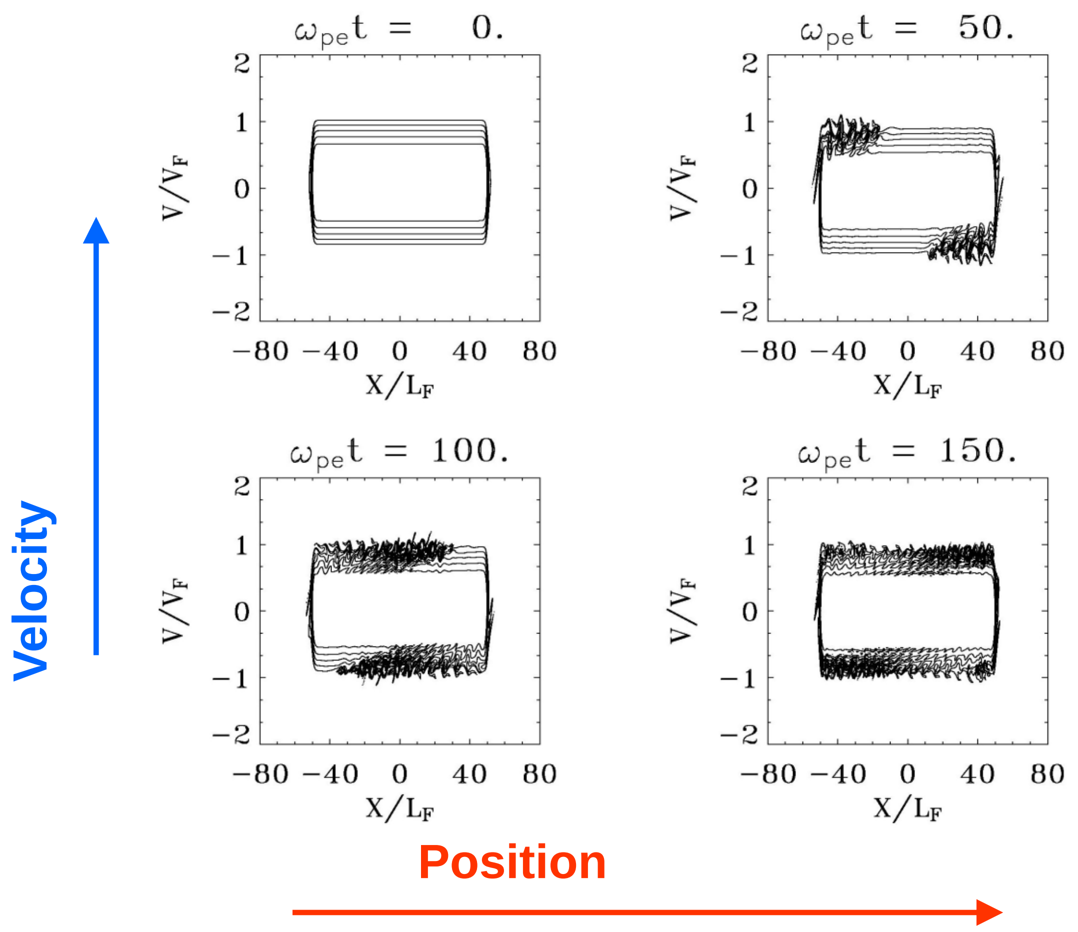
- **Initial phase** ($\omega_{pe}t < 200 \approx 20$ fs)
  - Damped oscillations at the plasma frequency.
  - Center-of-mass energy (E_{cm}) is converted into thermal energy (E_{th}).

- **Saturation phase** ($\omega_{pe}t > 200 \approx 20$ fs)
  - Low frequency oscillations (period $\approx 100\ \omega_{pe}^{-1} \approx 10$ fs).
  - Period is equal to the time-of-flight of electrons traveling at the Fermi velocity: $T = L / V_F$
  - Electrons bounce back and forth on the film surfaces.



G. Manfredi, P.-A. Hervieux, Phys. Rev. B **70**, 201402R (2004); Phys. Rev. B **72**, 155421 (2005).

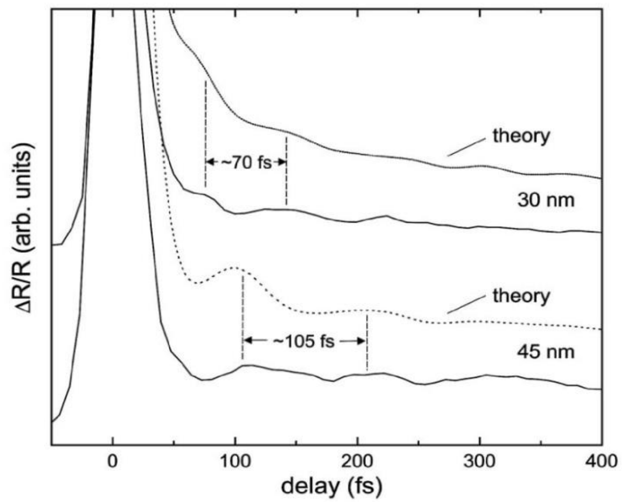
Phase-space dynamics



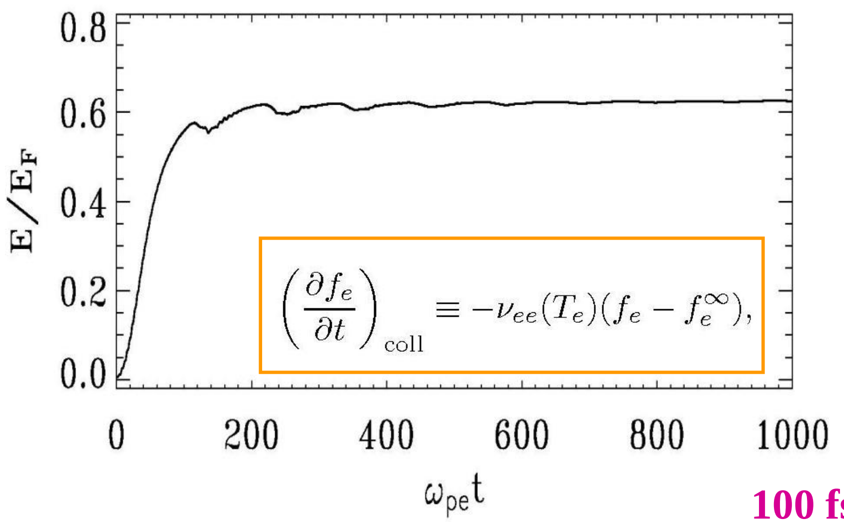
Experimental confirmation

- **X. Liu, R. Stock, W. Rudolph, Phys. Rev. B 72, 195431 (2005).**
- Measurements obtained with thin gold films.
- Ballistic electron oscillations with a period proportional to the film thickness.

Experiment



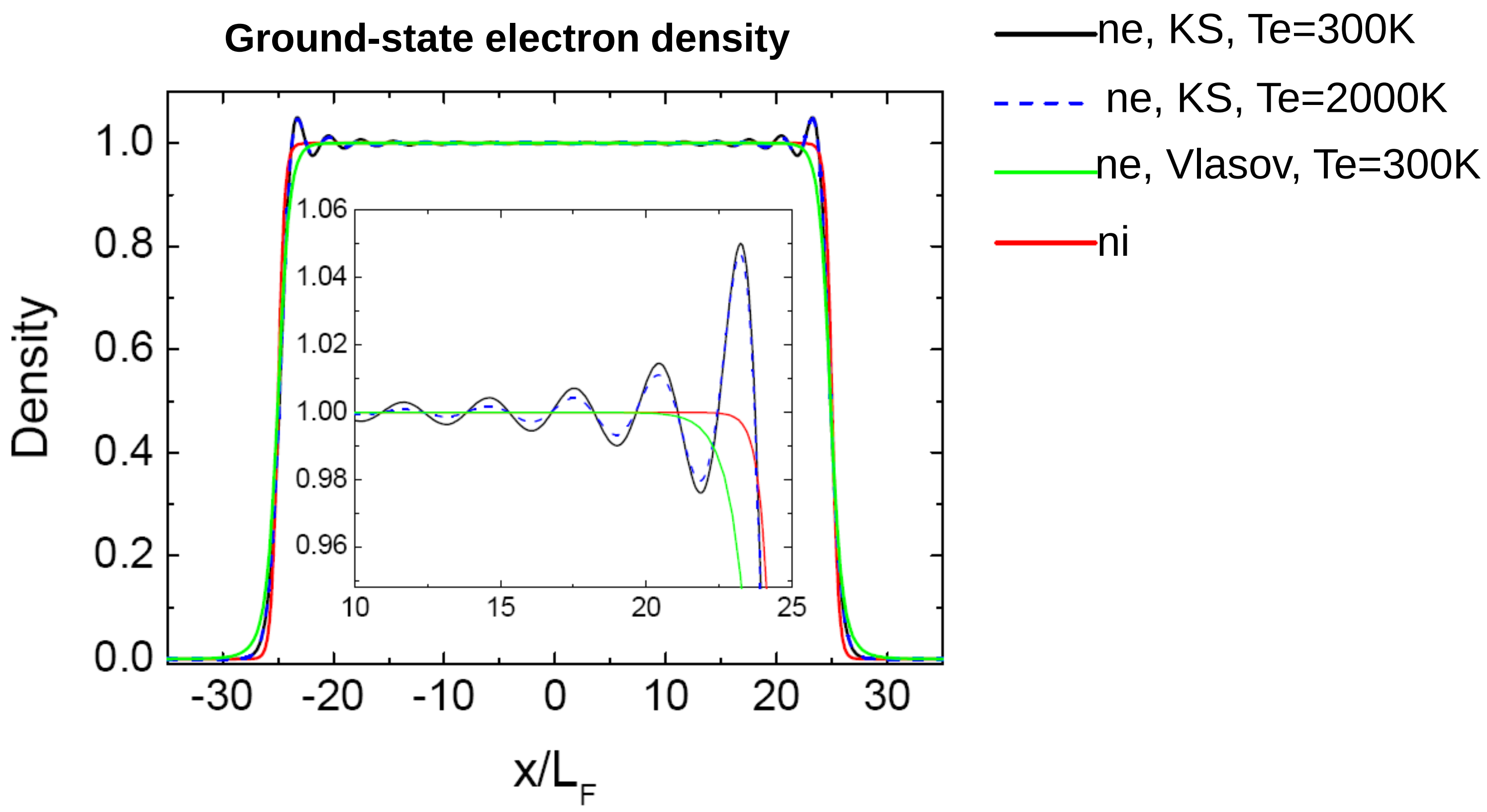
Simulation (with e-e collisions)



100 fs

Quantum effects: ground-state (Kohn-Sham)

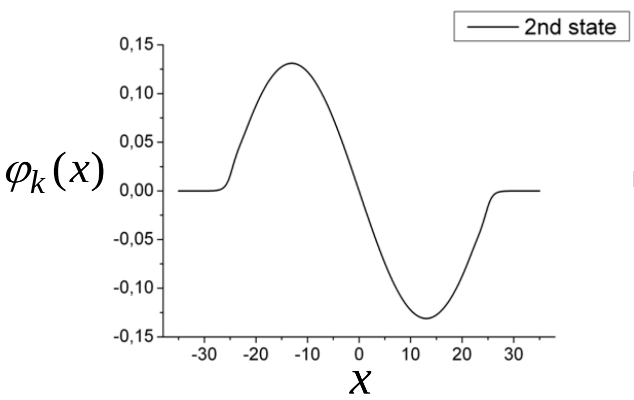
- Solution of the stationary Kohn-Sham equations (DFT) in 1D
- Provide the ground-state density



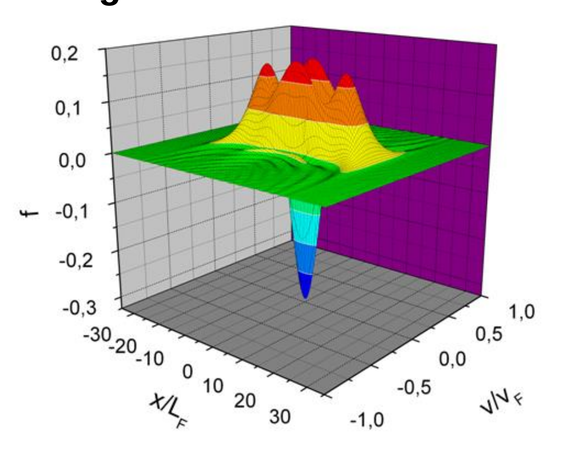
Quantum effects: ground-state Wigner functions

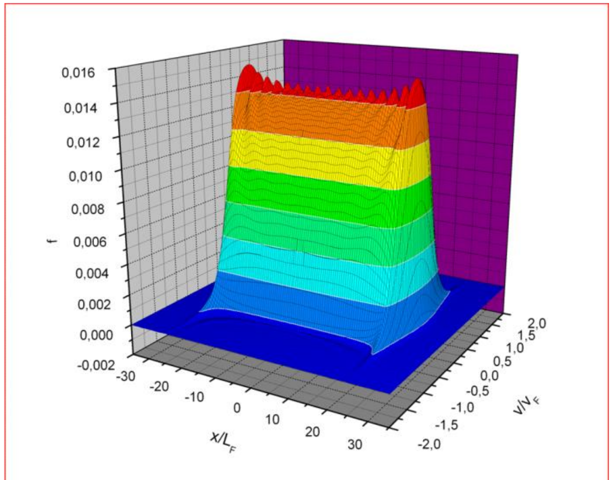
$$f_k(x,v) = \frac{m}{2\pi\hbar}\int_{-\infty}^{+\infty} \varphi_k^*\left(x+\frac{\lambda}{2},t\right)\varphi_k\left(x-\frac{\lambda}{2},t\right) e^{imv\lambda/\hbar}\, d\lambda.$$

Kohn-Sham wavefunction



Wigner function





Total ground-state Wigner function

$$f(x,v) = \sum_{k=1}^{N} p_k f_k(x,v).$$

p_k = Fermi-Dirac weights at finite Te

Quantum effects: dynamics

- **Dynamical model: Wigner evolution equation**

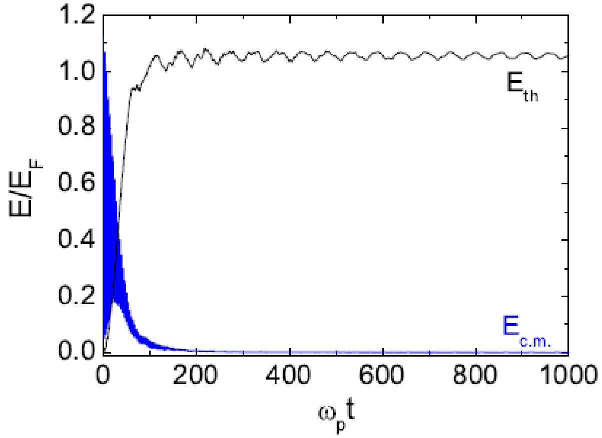
$$\frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x} + i\frac{em}{2\pi\hbar^2} \times$$
$$\int d\lambda dv' e^{im(v-v')\lambda/\hbar} \left[V_{\text{eff}}\left(x+\frac{\lambda}{2},t\right) - V_{\text{eff}}\left(x-\frac{\lambda}{2},t\right)\right] f(x,v',t) = 0$$

$$V_{\text{eff}} = V_H + V_{xc}$$
$$V_H = \text{Hartree potential (from Poisson's eq.)}$$
$$V_{xc} = \text{exchange - correlations}$$

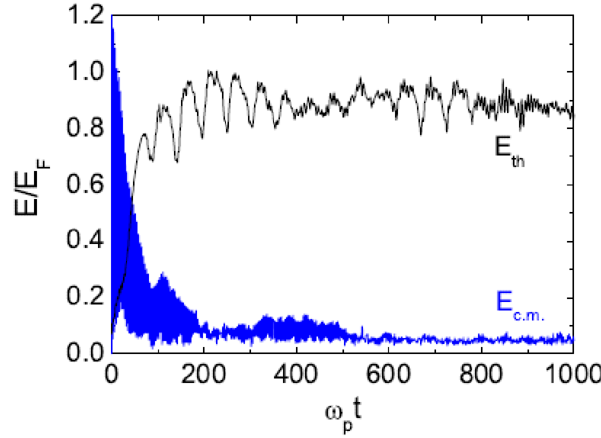
- Reduces to the Vlasov equation in the classical limit.
- Electron are excited at t=0 by imparting a **velocity "kick" *δv*** to their velocity distribution

Quantum dynamics — results

Vlasov



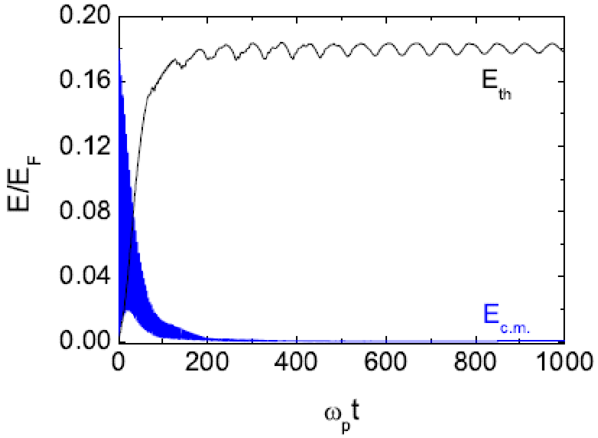
Wigner



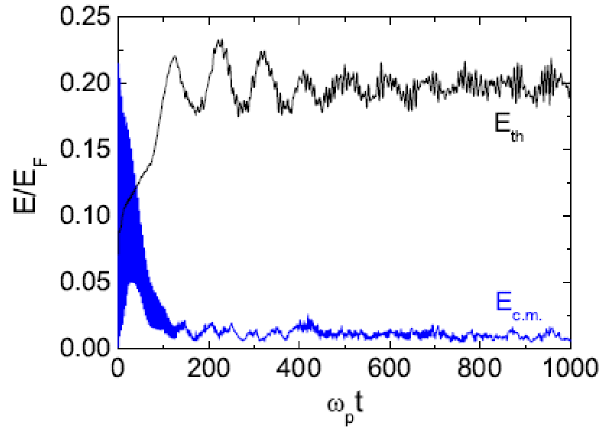
≈ 100 fs

Large excitation

Vlasov



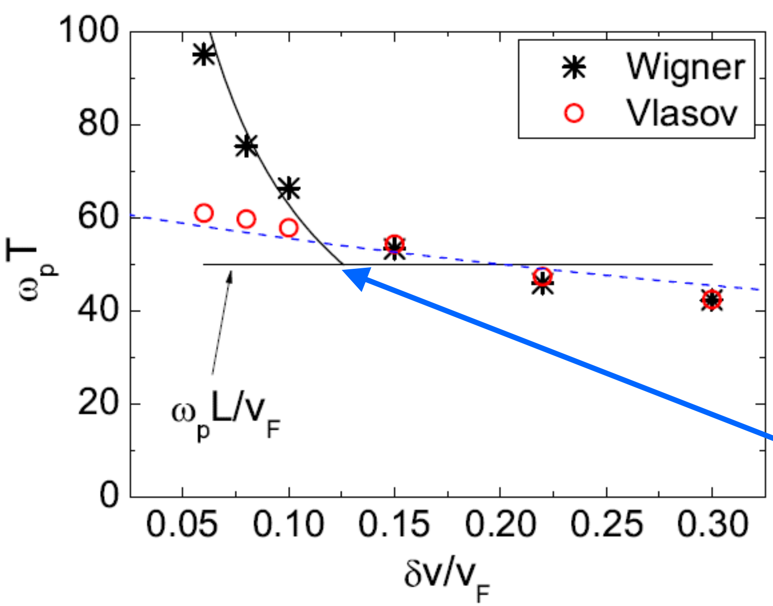
Wigner



Small excitation

Quantum dynamics — results

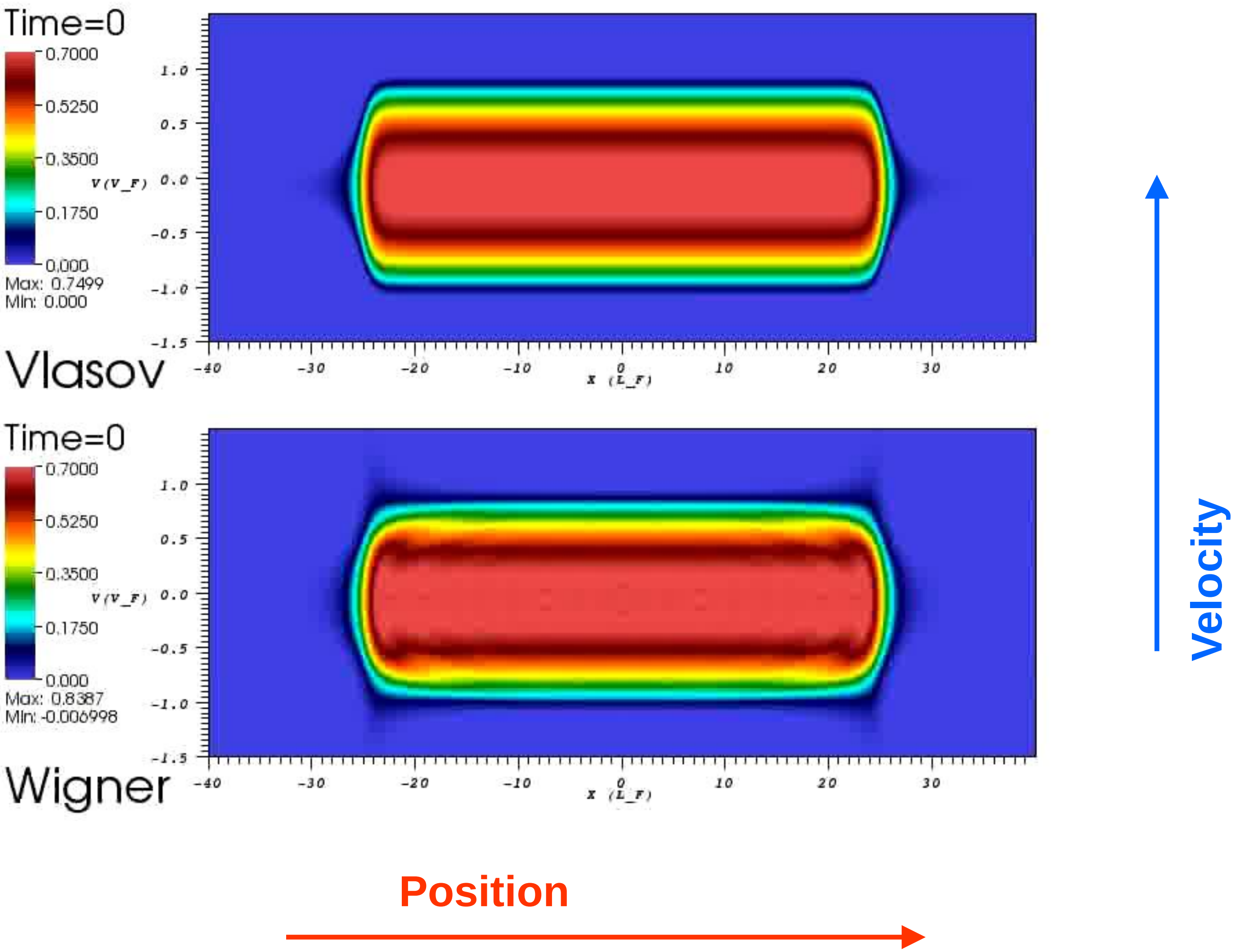
- Period of slow oscillations is identical for Wigner and Vlasov simulations for <u>large excitations</u>
- They diverge for <u>small excitations</u>: quantum effects



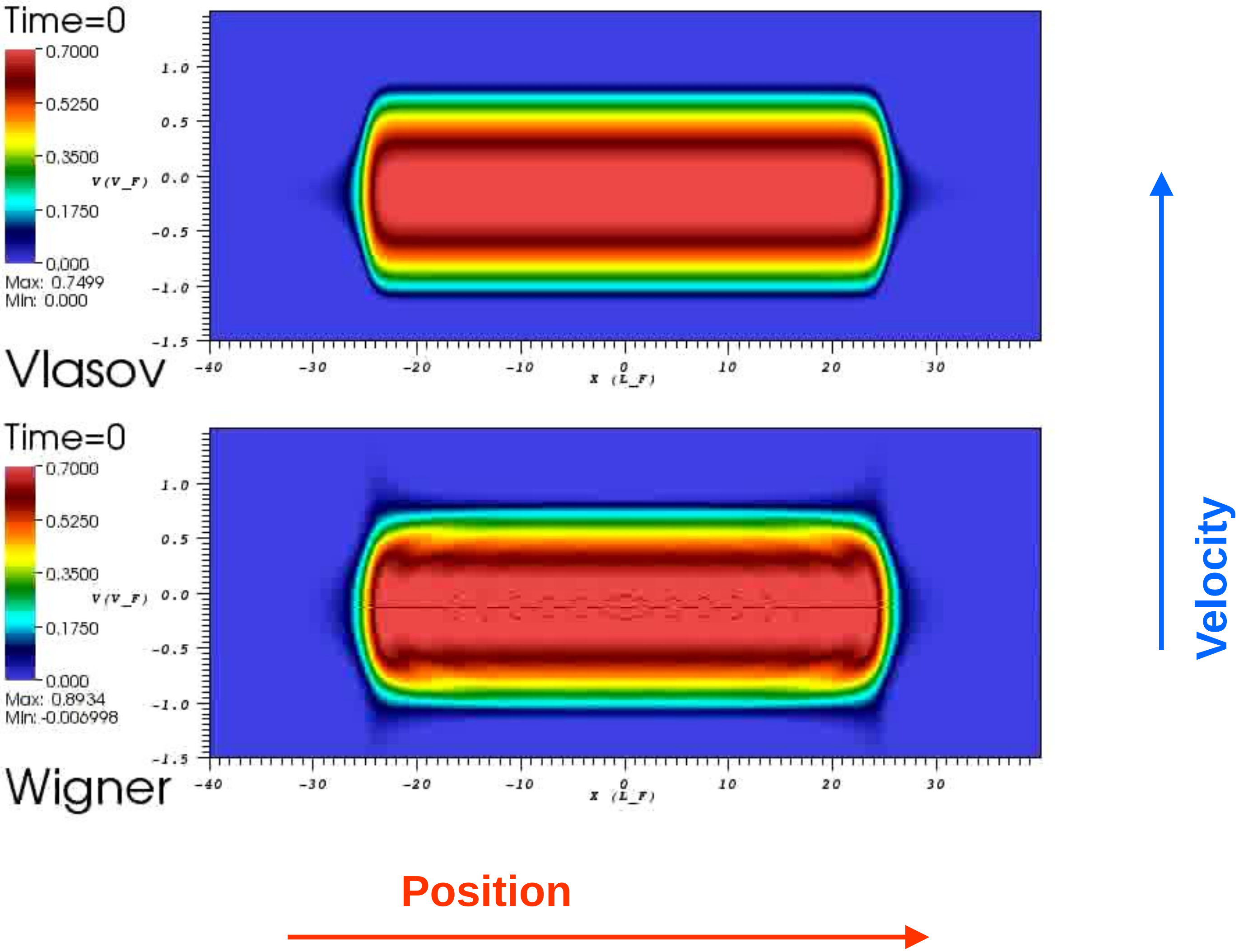
Threshold occurs when excitation energy is close to plasmon energy

$$\boxed{\delta E = m_e v_F \delta v \approx \hbar \omega_p}$$

Small excitation dynamics



Large excitation dynamics



Coupling to phonons: quantum Fokker-Planck equation

$$\frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x} + i\frac{em}{2\pi\hbar^2} \times$$
$$\int d\lambda dv' e^{im(v-v')\lambda/\hbar} \left[V_{\text{eff}}\left(x+\frac{\lambda}{2},t\right) - V_{\text{eff}}\left(x-\frac{\lambda}{2},t\right)\right] f(x,v',t) = \boxed{\left(\frac{\partial f}{\partial t}\right)_{\text{scatt}}}$$

$$\boxed{\left(\frac{\partial f}{\partial t}\right)_{\text{scatt}} = \gamma \frac{\partial}{\partial v}\left[v\,G(f)\right] + D\frac{\partial^2 f}{\partial v^2}}$$

$\gamma =$ e - ph relaxation rate

$D = \gamma(k_B T_{\text{latt}} / m_e) =$ decoherence

$$\gamma = g/c_e$$

$$c_e(T_e) = \pi^2 n_0 k_B (T_e/2T_F)$$

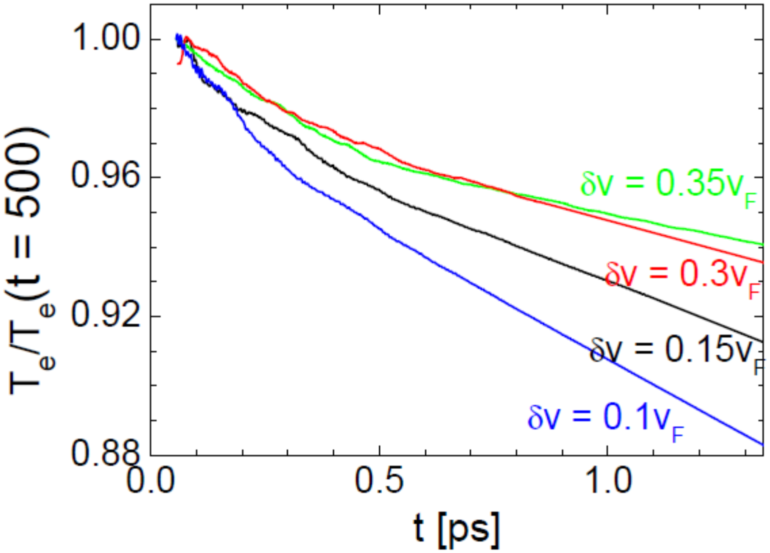
$g = 2 \times 10^{16} \mathrm{W/m^3K}$: e-ph coupling constant

c_e = electron heat capacity

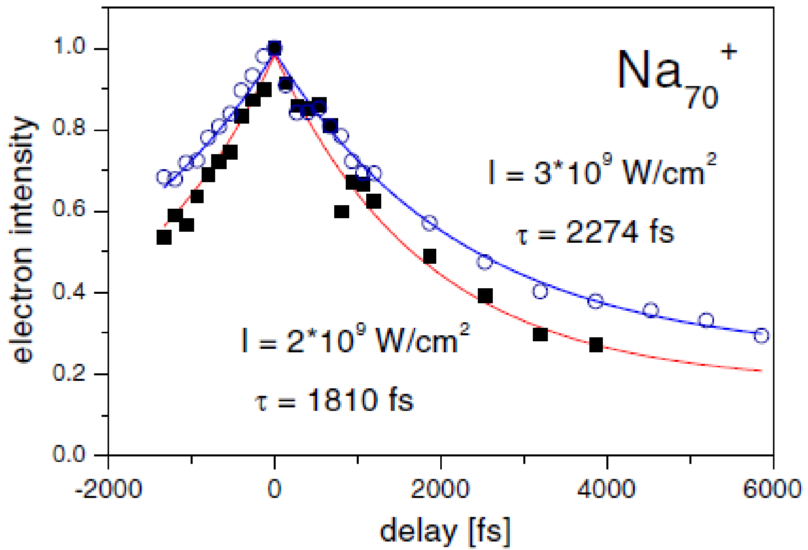
Equilibrium solution (Fermi-Dirac): $f_{eq}(v_x) = \frac{3}{4}\frac{n_0}{v_F}\frac{T_i}{T_F} \ln\left[1+\exp\left(-\frac{m_e v_x^2/2-\mu}{k_B T_i}\right)\right]$

Relaxation of the electron temperature

Simulations



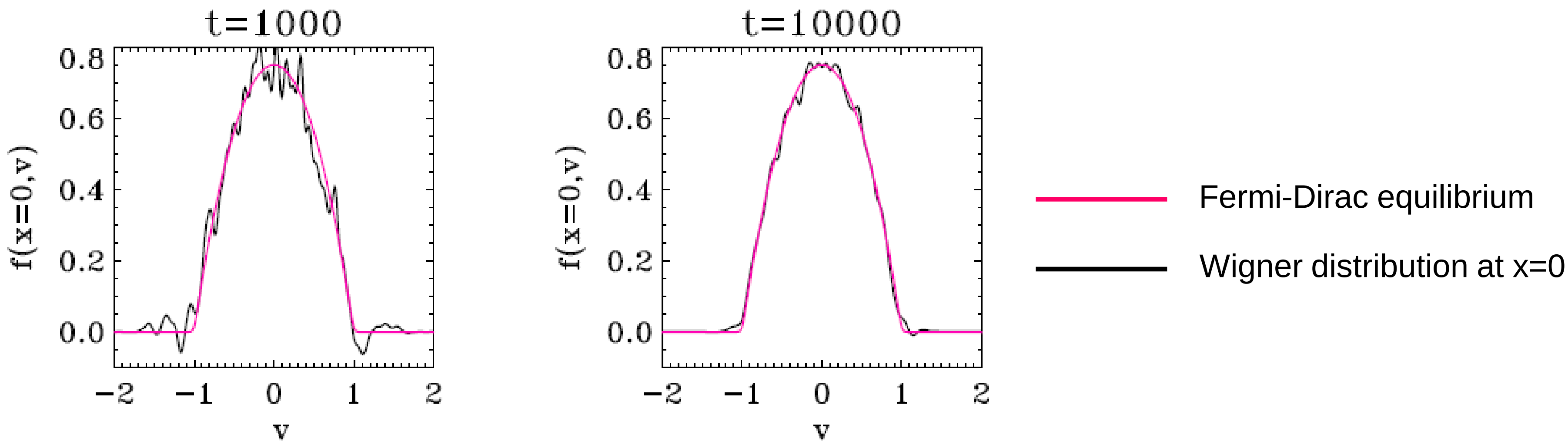
Experiments



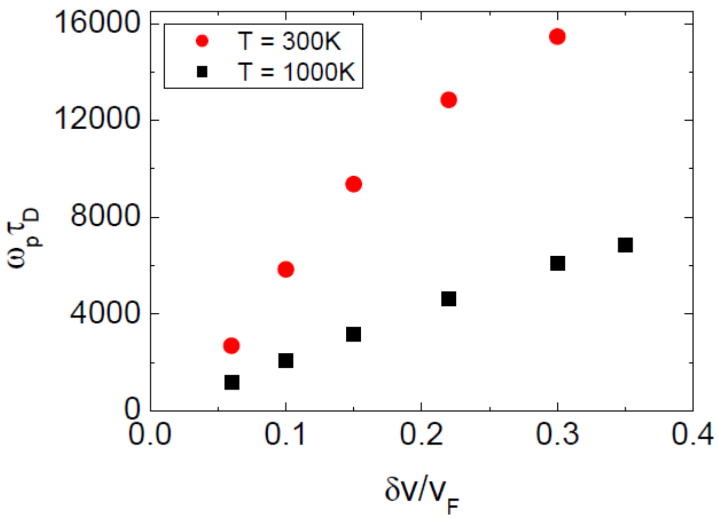
M. Maier et al., Phys. Rev. Lett. **96**, 117405 (2006)

- Using similar physical parameters, we obtain relaxation times compatible with experiments on sodium clusters
- **Relaxation time (2-3 ps) increases with excitation**

Quantum decoherence



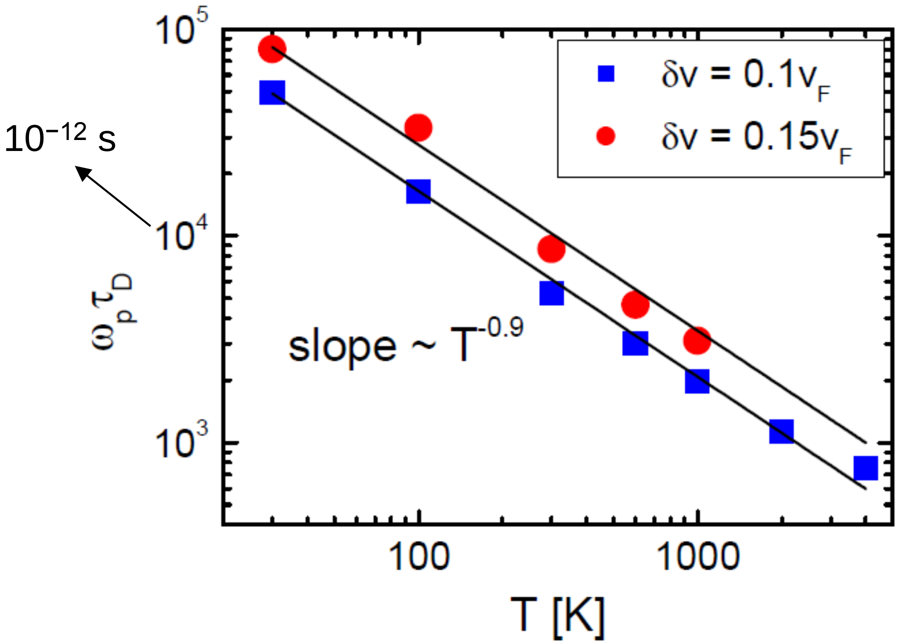
Decoherence time vs. excitation strength



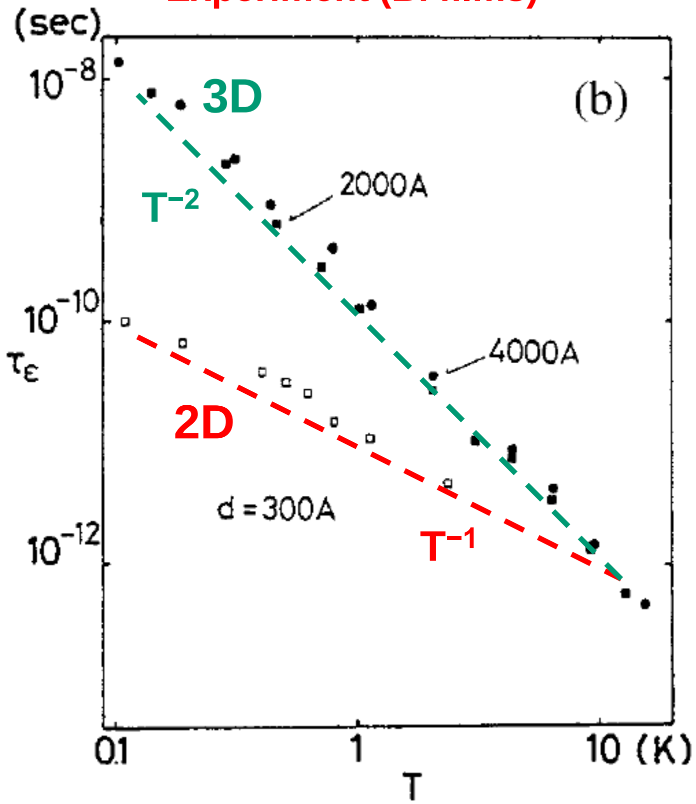
- **Decoherence time ≈ 0.4 ps**
- **Relaxation time ≈ 3 ps**

Decoherence vs. ion lattice temperature

Simulation



Experiment (Bi films)



Komori F, et al. 1987 *J. Phys. Soc. Japan* **56** 691

Quantum hydrodynamics for thin metal films

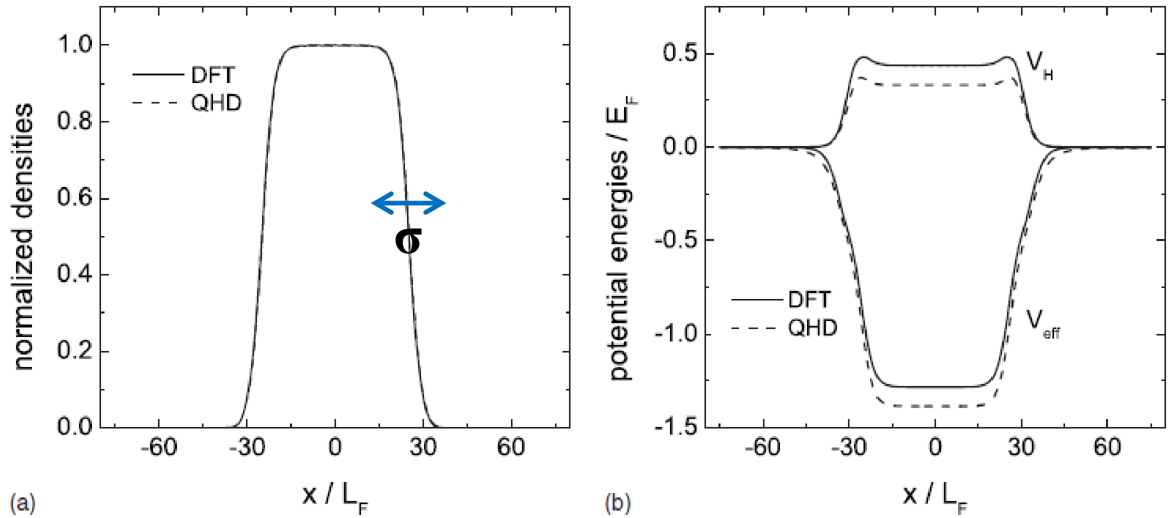
$$\begin{cases} \dfrac{\partial n}{\partial t} + \dfrac{\partial (nu)}{\partial x} = 0\,, \\ \dfrac{\partial u}{\partial t} + u\dfrac{\partial u}{\partial x} = \dfrac{e}{m}\dfrac{\partial \phi}{\partial x} - \dfrac{1}{mn}\dfrac{\partial P^C}{\partial x} - \dfrac{1}{mn}\dfrac{\partial P^Q}{\partial x}\,. \end{cases}$$

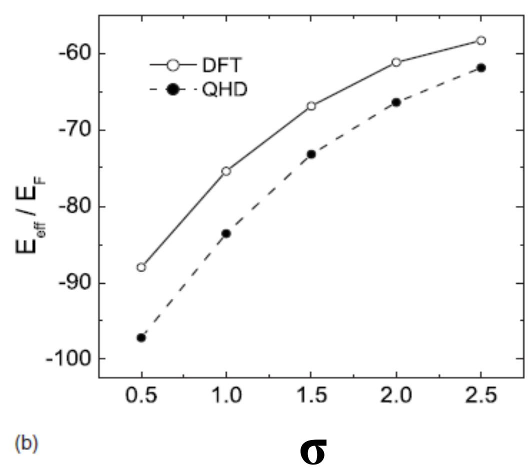
$P^C = P^C(n)$: "equation of state"

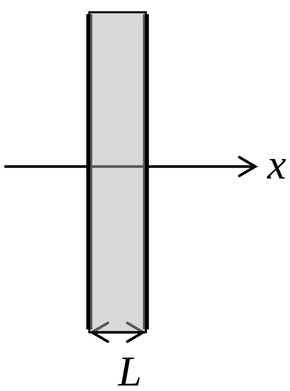
$$P^Q = \frac{\hbar^2}{2m}\left[\left(\frac{\partial}{\partial x}\sqrt{n}\right)^2 - \sqrt{n}\frac{\partial^2}{\partial x^2}\sqrt{n}\right]$$

+ Poisson equation

QHD for thin metal films: ground state

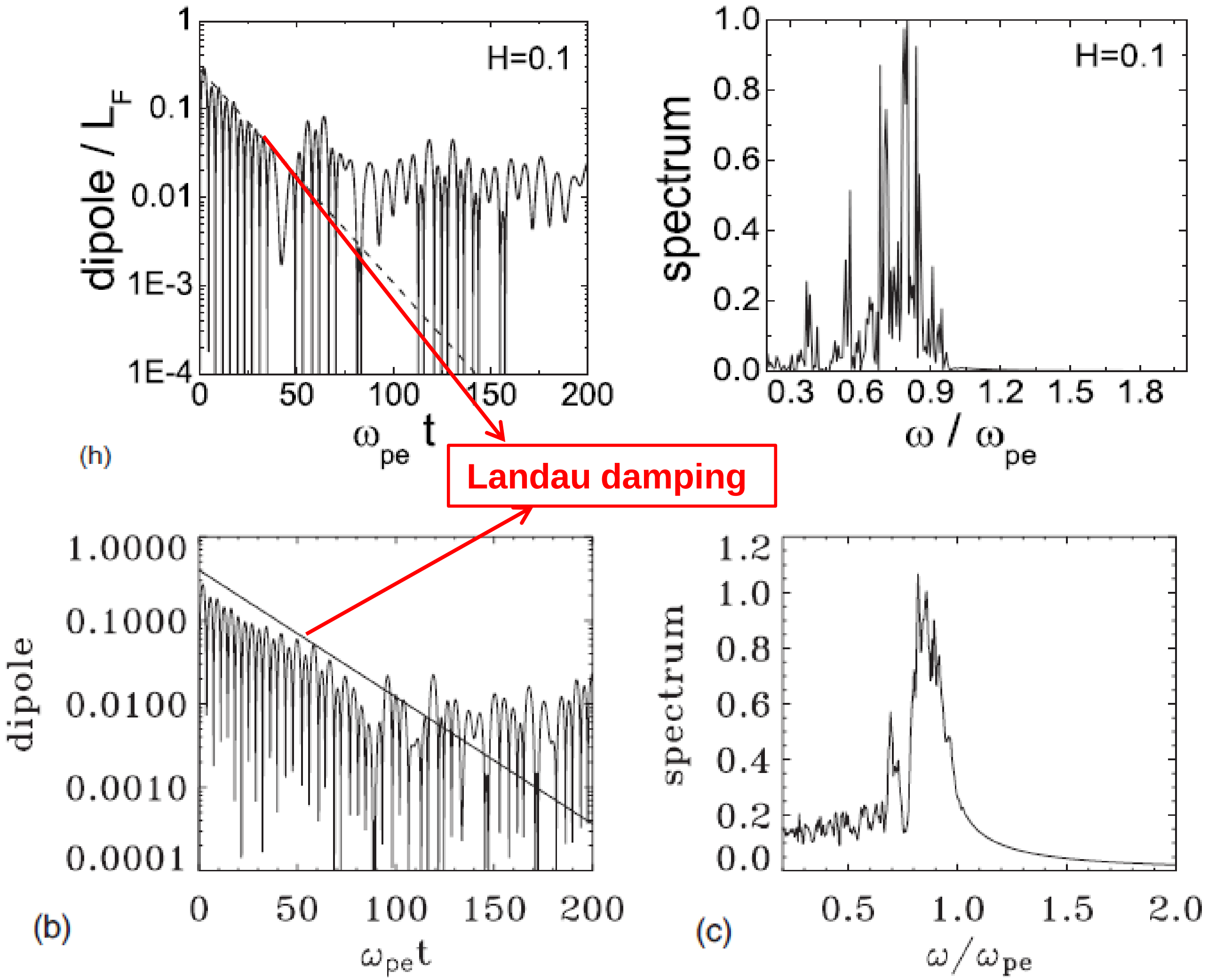






N. Crouseilles, P.-A. Hervieux, G. Manfredi, Phys. Rev. B **78**, 155412 (2008).

QHD for thin metal films: dipole motion



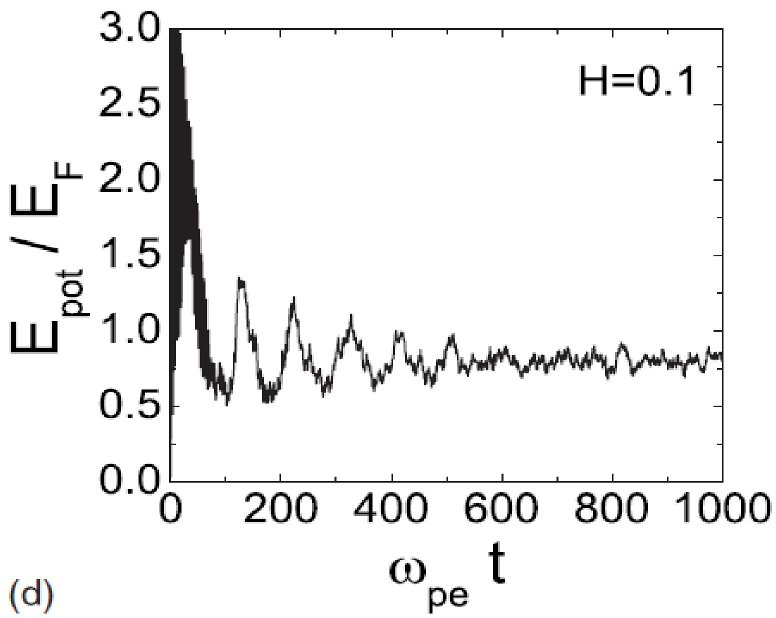
Quantum Hydro-dynamics

Semiclassical (Vlasov)

N. Crouseilles, P.-A. Hervieux, G. Manfredi, Phys. Rev. B **78**, 155412 (2008).

QHD for thin metal films: nonlinear oscillations

QHD



Vlasov (semiclassical)