Conductivity of dense plasmas

P. Renaudin

CEA, DAM, DIF
Bruyères-le-Châtel
91297 Arpajon Cedex, France

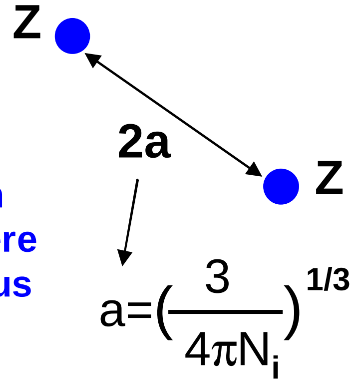
Acknowledgment
C. Blancard, J. Clérouin, G. Faussurier, S. Mazevet,
R. M. More, V. Recoules, H. Yoneda

High Energy Density Science using XFEL
Oxford, March 30-April 1, 2009

Outline

- **Theoretical point of view:**
  Dense plasmas may have very interesting transport regimes
  - **Transport *vs* Radiation theory**
  - **Classical *vs* Quantum theory**
- **Experimental point of view:**
  - **Electrically driven warm dense matter experiments**
  - **Ultrashort-pulse laser pump-probe experiments in the UV to IR range**
- **Quantum conduction phenomena and low-frequency opacity in dense plasmas**
- **Impact on the design of sub-ps XFEL experiments**

In dense plasmas, the ions are strongly correlated and coupled with complex atomic physics

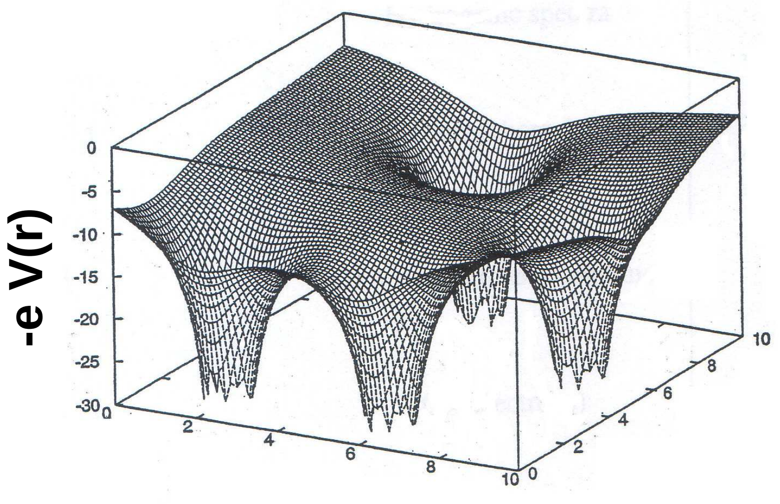


Ion sphere radius

$$a=\left(\frac{3}{4\pi N_i}\right)^{1/3}$$

Coulomb coupling parameter

$$\Gamma = \frac{(Ze)^2}{a\,k_B\,T}$$



Strongly coupled $\Gamma > 1$

- High ρ and low Te
- Non-perturbative methods
- No small parameters
- No clear distinction between atomic and plasma physics

Atoms interact in dense plasmas

- Disorder and fluctuations
- Spherical symmetry is broken
- Potentials change when ions move

Theoretically the difficulty is there are no small parameters

In the majority of the (ρ,kT,ħω) experimentally explored, the Boltzmann equation gives the correct answer

Assume free electrons, ions and coulomb collisions

- **Instantaneous collisions**
- **No interaction between the collisions**
- **Weak mean-free-path (LTE)**

Dielectric function $\epsilon(\omega) = 1 + 4\pi n_a \alpha(\omega) + \frac{4\pi i \sigma(\omega)}{\omega}$ **$\alpha(\omega)$ = atomic polarization**

DC conductivity $\sigma_{DC} = \frac{ne^2\tau}{m}$

AC conductivity (*Drude Formula*) $\sigma(\omega) = \frac{n_e\, \tau\, e^2}{m} \left(\frac{1}{1 - i\omega\tau}\right)$

Collision time τ (electron-phonon in solids, electron-ion in liquids & plasmas)

Many theories give similar results using different collision times (*Ziman*, *Kubo*, *Fokker Planck* etc)

Ziman formula $\frac{1}{\sigma_0} = -\frac{\eta}{3\pi Z^{*2} e^2 \rho_i} \int_0^\infty d\varepsilon\, f'(\varepsilon) \int_0^{2k} dq\, q^3 S(q) \Sigma_{sc}(q)$

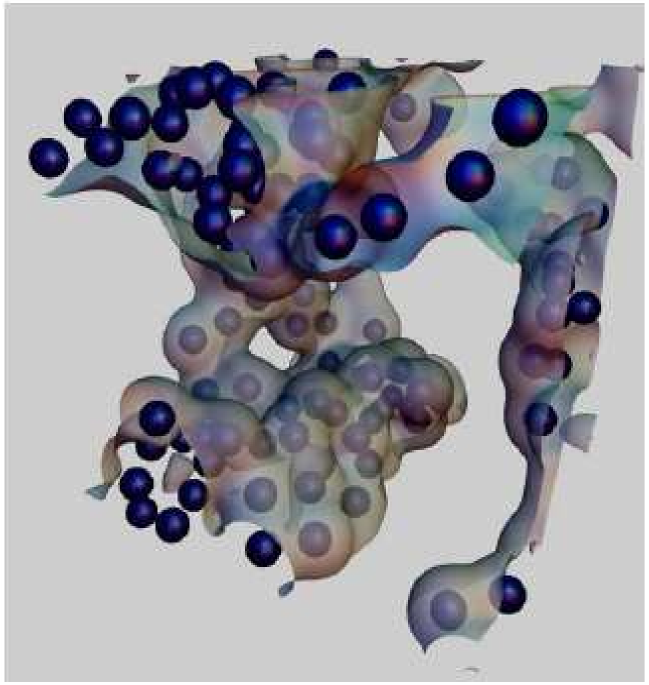
Opacity $K(\omega)$ and refraction index $n(\omega)$ are obtained from the dielectric functions $\varepsilon_r(\omega)$ and $\varepsilon_i(\omega)$ (*Kramers-Kronig relations*)

$$K(\omega) = \frac{4\,\pi\,\sigma_{DC}}{n(\omega)\, c\, \rho\, (1 + \omega^2\tau^2)}$$

Fluctuation-dissipation theorem coupled with QMD calculations provides a consistent approach of the structure and transport properties in dense plasmas

QMD code

- Pseudo potentials
- Small number of bands
- Small number of k-points
- Ionic trajectories



Post-Processor

Detailed electronic structure

- Ionic Configurations
- Large number of bands
- Several k-points
- Eigenvalues

Matrix elements

$$\sigma(\omega, R_I) = \frac{2\pi}{3\omega} \int_{BZ} dk \sum_{n,m} \left(f_n^k - f_m^k\right) \left|\left\langle \psi_n^k \middle| \hat{v} \middle| \psi_m^k \right\rangle\right|^2 \delta\left(E_i - E_j - \omega\right)$$

Optical conductivity σ(ω)

Kubo-Greenwood formula generalized to *k*-vector sampling

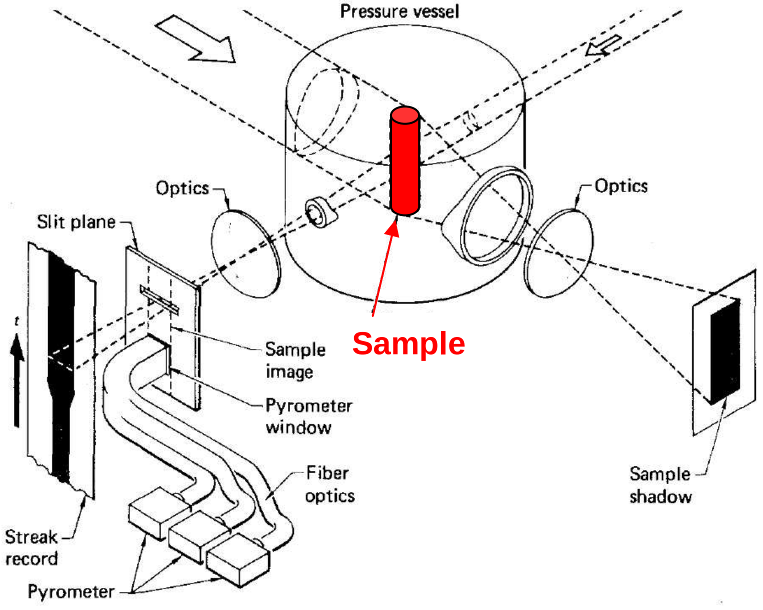
Optical properties

Desjarlais *et al*., Phys Rev E 66 (2002)
Recoules *et al*., Phys Rev E 66 (2002)

Using fast transient methods allowed to measure DC conductivity of hot expanded liquids

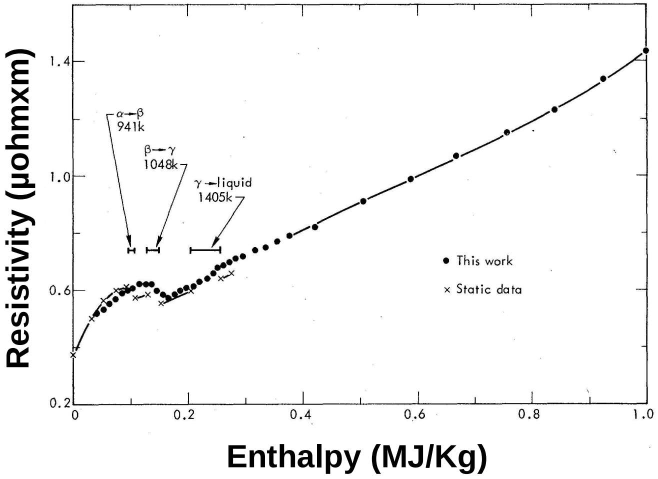
Isobaric Expansion Experiments at LLNL used a pressure cell capable to hold rare gas at 0.4 GPa

Gathers, Int Journal of Thermophys 11 (1990)



Gathers *et al*., Phys Rev Lett 33 (1974)

- **Current, voltage, sample cross section are measured as function of time**
- **The EOS data are the pressure, volume, and enthalpy of the hot liquid (temperature up to 9000 K)**

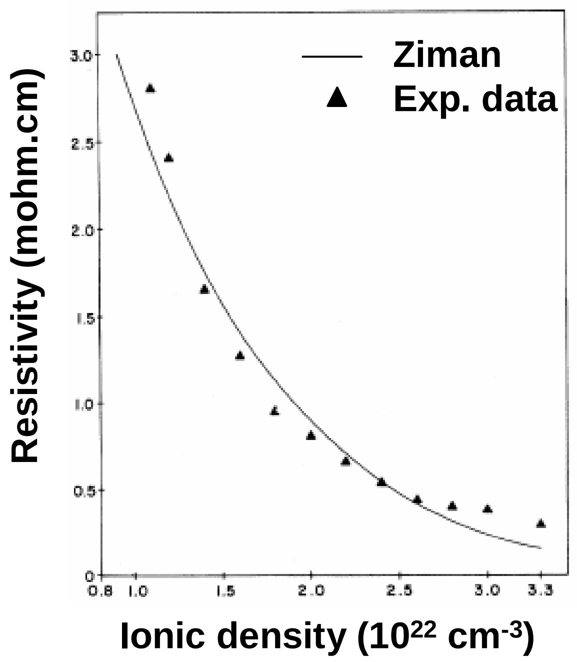


Transport phenomena in liquid metals were studied in exploding-wire experiments

« The Ziman theory of transport properties for liquid metals is remarkably successful when both the interference function and the electron-ion interaction are known »

Ashcroft & Lekner, Phys Rev 145, 83 (1966)

Liquid-copper resistivity



- **Resistance can be measured** from the time-resolved electrical parameters of the circuit
- **Density can be measured** from the time-resolved measurements of the wire radii found by x-ray or optical shadowgraphs

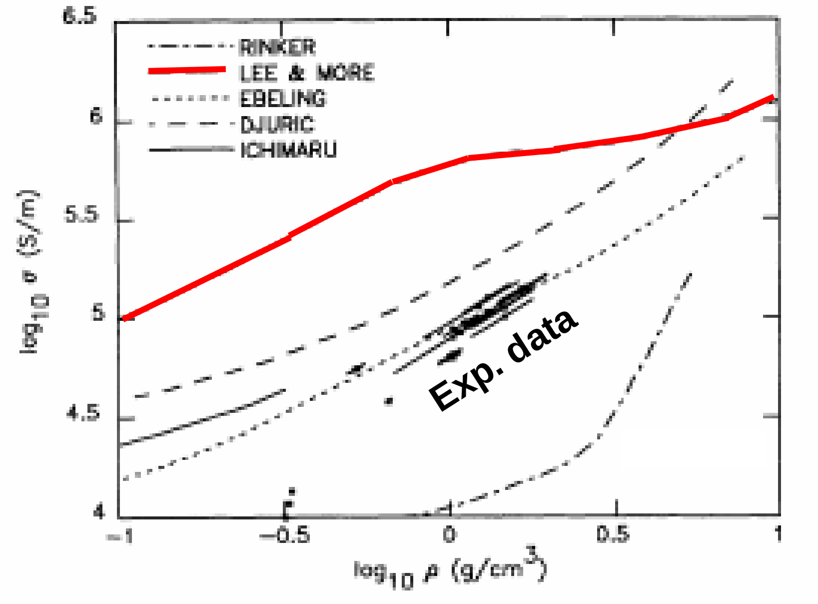
Ben-Yosef & Rubin,
Phys Rev Lett 23, 289 (1969)

DC conductivity of dense plasmas has been measured using confined exploded-wire experiments

« The capillary confines the plasma in a uniform & measurable volume for a short time, during which it is possible to make accurate measurements of the conductivity, density and input energy »

DeSilva & Kunze, Phys Rev E 49, 4448 (1994)

Conductivity of Cu at 20 000 K



- **Large discrepancies between models**
- **Lee-More model disagrees more than expected with the experimental data ?**

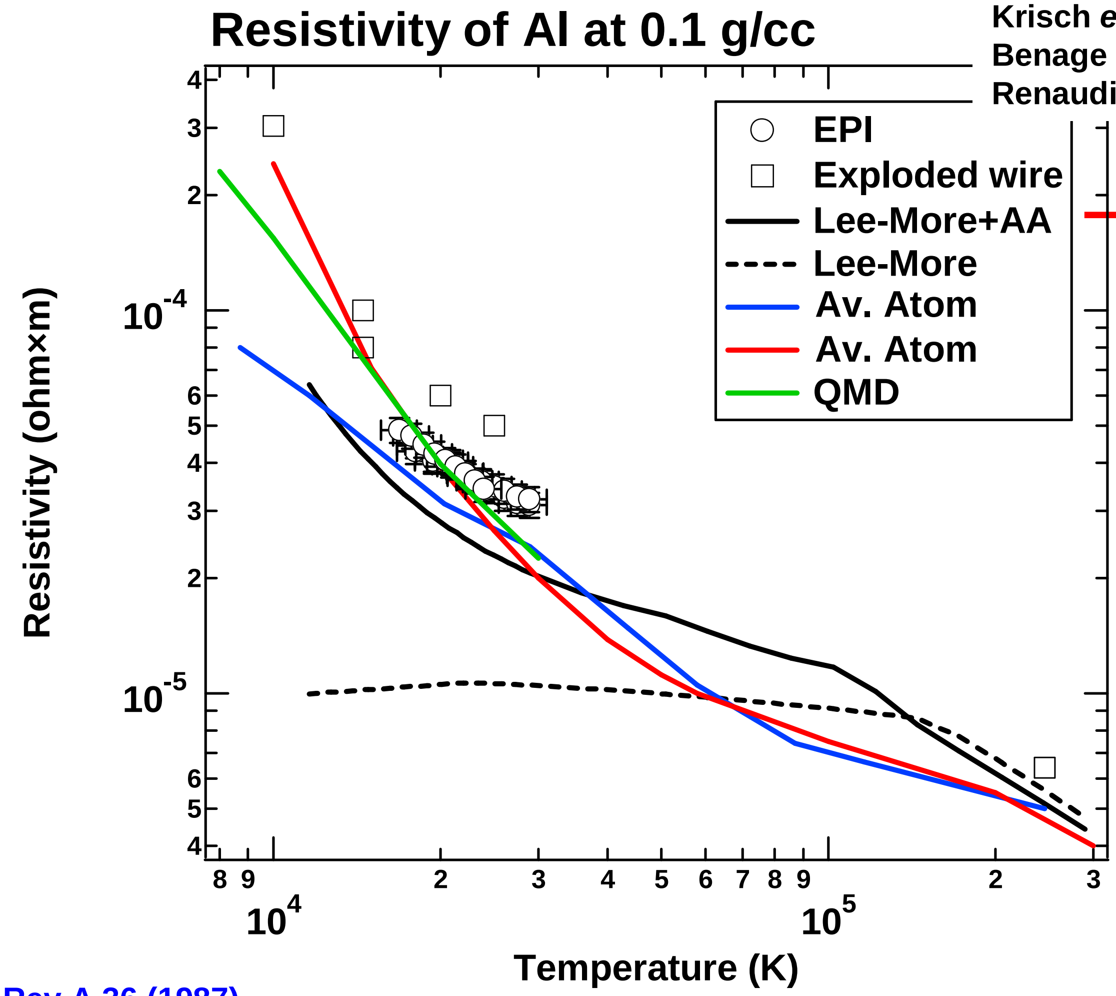
Lee-More model:

Drude formula
+ Effects of magnetic field
+ Dense plasma rules for log(Λ)
+ Minimum mean-free-path

Lee & More, Phys. Fluids 27 (1984)

The agreement is much better with theoretical models that include quantum mechanical effects

DeSilva *et al*., Phys Rev E 57 (1998)
Krisch *et al*., Phys Rev E 58 (1998)
Benage *et al*., Phys Rev Lett 83 (1999)
Renaudin *et al*., Phys Rev Lett 88 (2002)

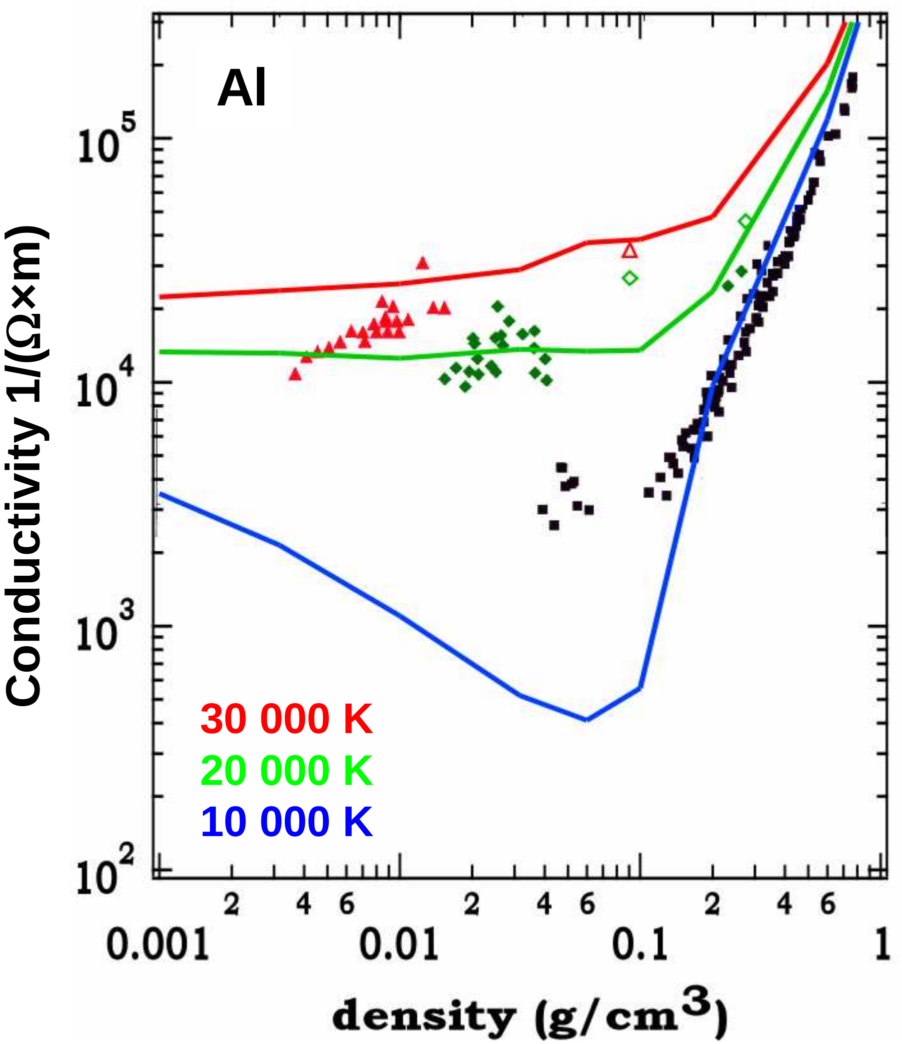


→ or modified LM formula by Desjarlais, Contrib Plasma Phys 41 (2001)

High-Te
Treatment of exchange-correlation and ion-ion interaction have a weak influence

Perrot *et al*., Phys Rev A 36 (1987)
Blancard & Faussurier, Phys Rev E 69 (2004)
Desjarlais *et al*., Phys Rev E 66 (2002)

A minimum mean-free path translates into a minimum of DC conductivity



DeSilva *et al*. (■) *vs* Ziman formula in average atom model

At low density ionization is driven by temperature

At low T a minimum of conductivity indicates where the ionization is driven by the density

DeSilva *et al*., Phys Rev E (1998)

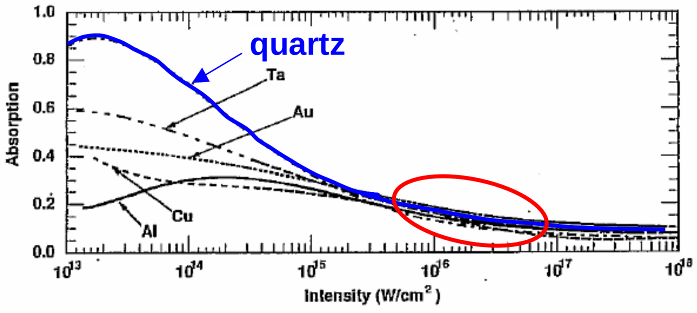
USP laser experiments approximately agree with the Drude theory and the solutions of the Maxwell equations

Ultrashort-pulse laser pump-probe experiments in the UV-IR range

Heat-pulse reflectivity “measures” AC conductivity and observe a minimal mean-free path during the metal to plasma transition
Milchberg *et al*., Phys Rev Lett 61 (1988)

High-Te

Heat-pulse reflectivity showing “black glass” in the WDM regime & excellent agreement with Drude formula for all materials at high temperature
Price *et al*., Phys Rev Lett 75 (1995)



Low-Te
Low-ρ

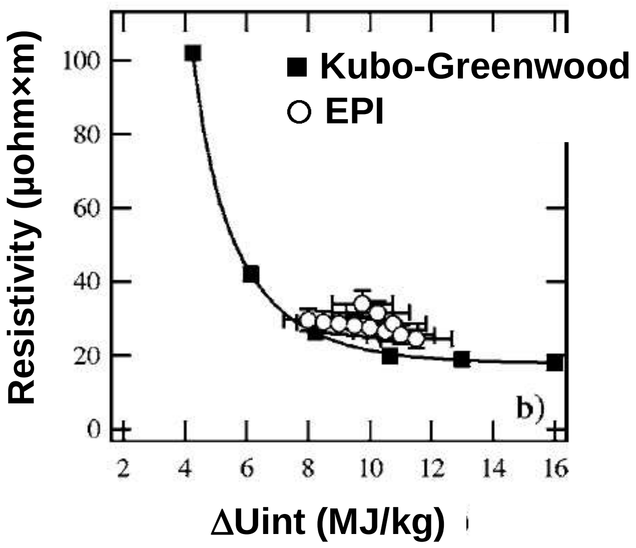
Ellipsometry experiments of s, p reflectivity and phase-difference of s- and p-polarized waves in low temperature gold plasma are well described by a Drude model that include atomic polarizability
Yoneda *et al*., Phys Rev Lett 91 (2003)

These experiments give a sensitive measure of optical properties in the WDM regime

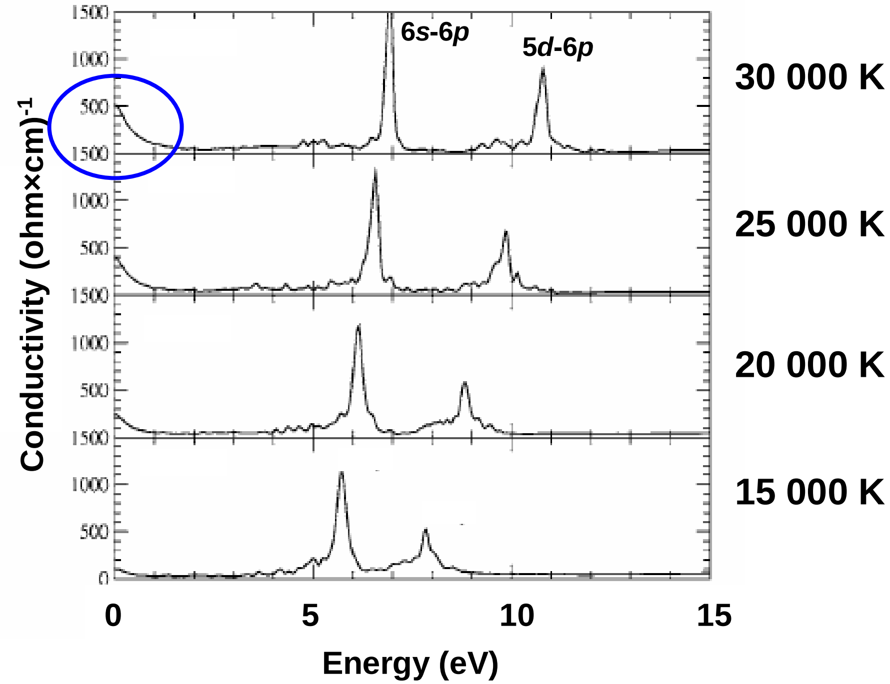
Kubo-Greenwood formulas with *ab initio* simulations show how the atomic-scale plasma conditions influence the conductivity and the spectra

Superposition of a Drude central peak with a few isolated peaks

Au at 0.5 g/cm^3



Kubo-Greenwood σ(ω)



The optical conductivity must be computed using both the Drude model + atomic polarizability

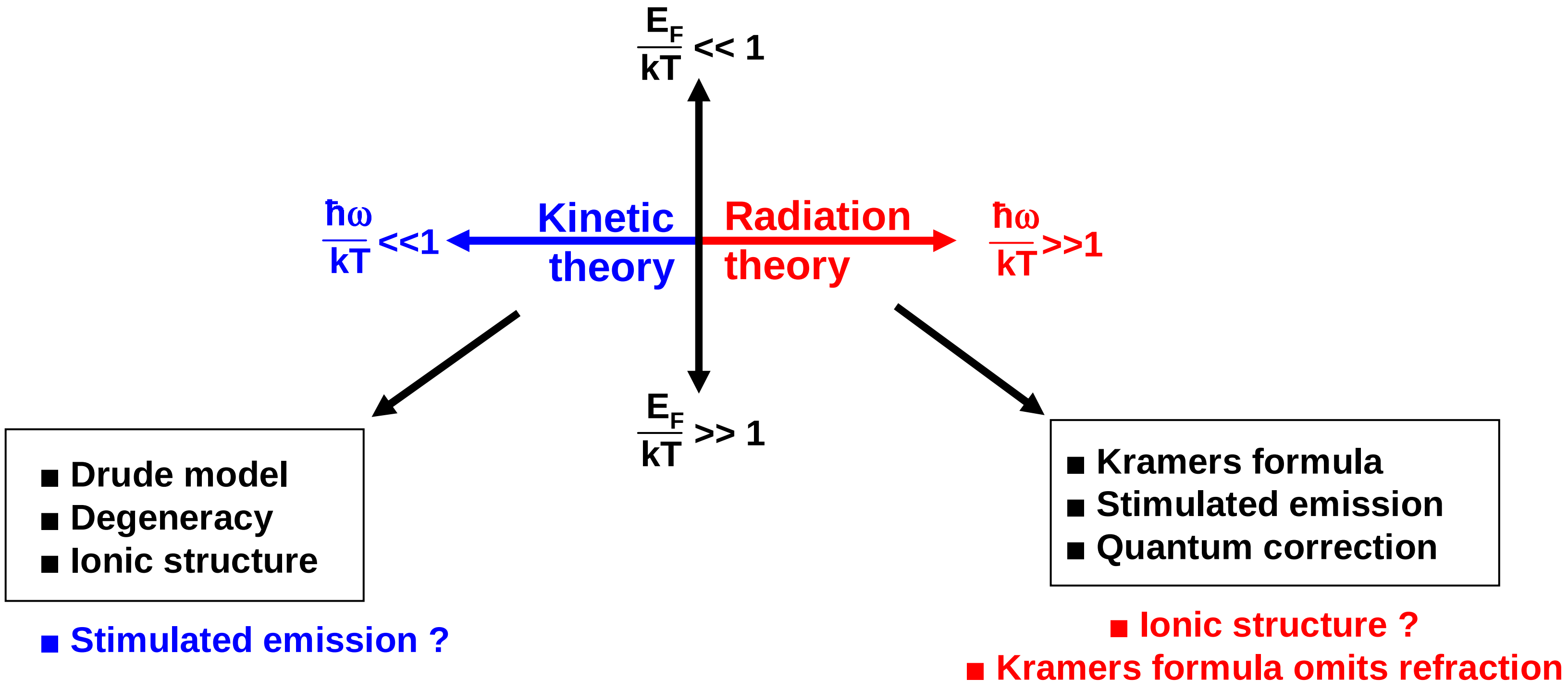
Dielectric function

$$\epsilon(\omega) = 1 + 4\pi n_a \alpha(\omega) + \frac{4\pi i \sigma(\omega)}{\omega}$$

Renaudin *et al.*, Phys Rev E 73 (2006)

Dense plasmas may have very interesting transport regimes

- **Radiation begins to rival electron thermal conduction**
- **Low-frequency opacity & quantum conduction phenomena are difficult to take into account**



$\frac{E_F}{kT} << 1$

$\frac{\hbar\omega}{kT} <<1$

Kinetic theory

Radiation theory

$\frac{\hbar\omega}{kT} >>1$

$\frac{E_F}{kT} >> 1$

■ **Drude model**
■ **Degeneracy**
■ **Ionic structure**

■ **Stimulated emission ?**

■ **Kramers formula**
■ **Stimulated emission**
■ **Quantum correction**

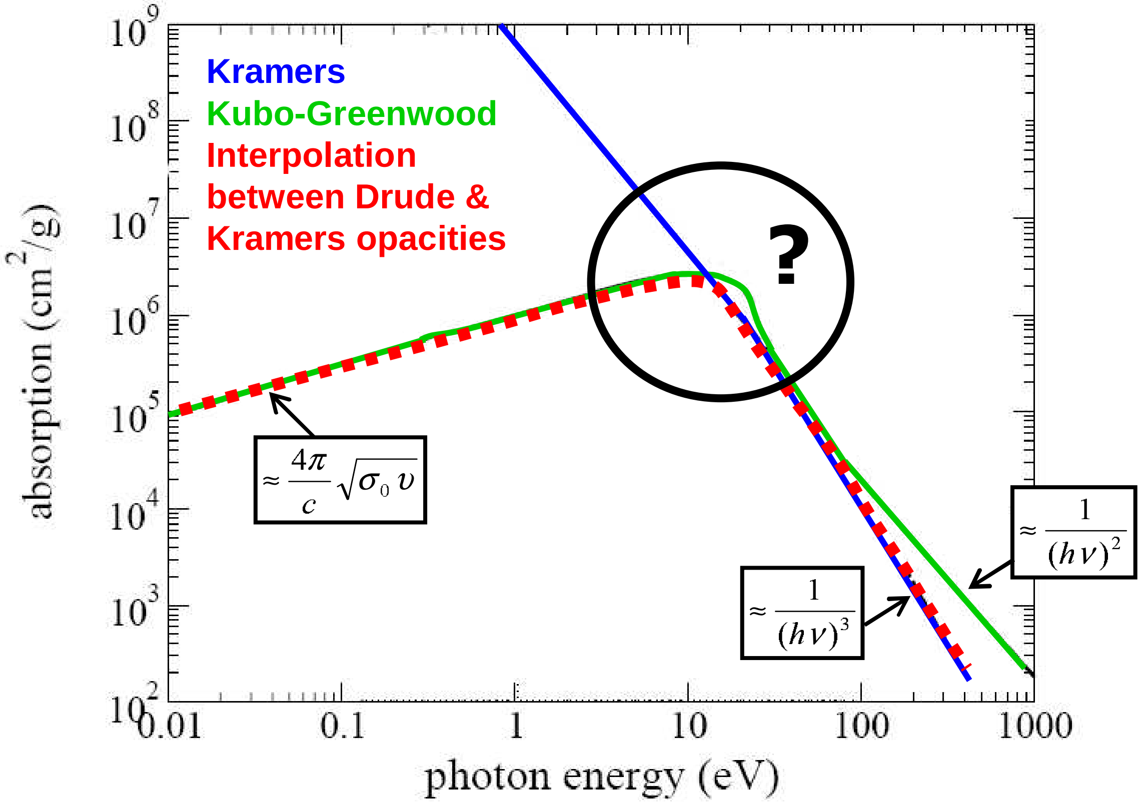
■ **Ionic structure ?**
■ **Kramers formula omits refraction**

What is the correct answer between the Fermi energy and a few plasmon energy ?

More, J of Plasma & Fusion Research 76, 623 (2000)

The low-frequency part of the opacity is dominated by the free-free contribution

H @ 48000 K & 0.65 g/cm³



Radiation theory

Kramers Formula
Classical approximation
In the low-density regime

Photon scattering of free electrons in the Coulomb field of an ion

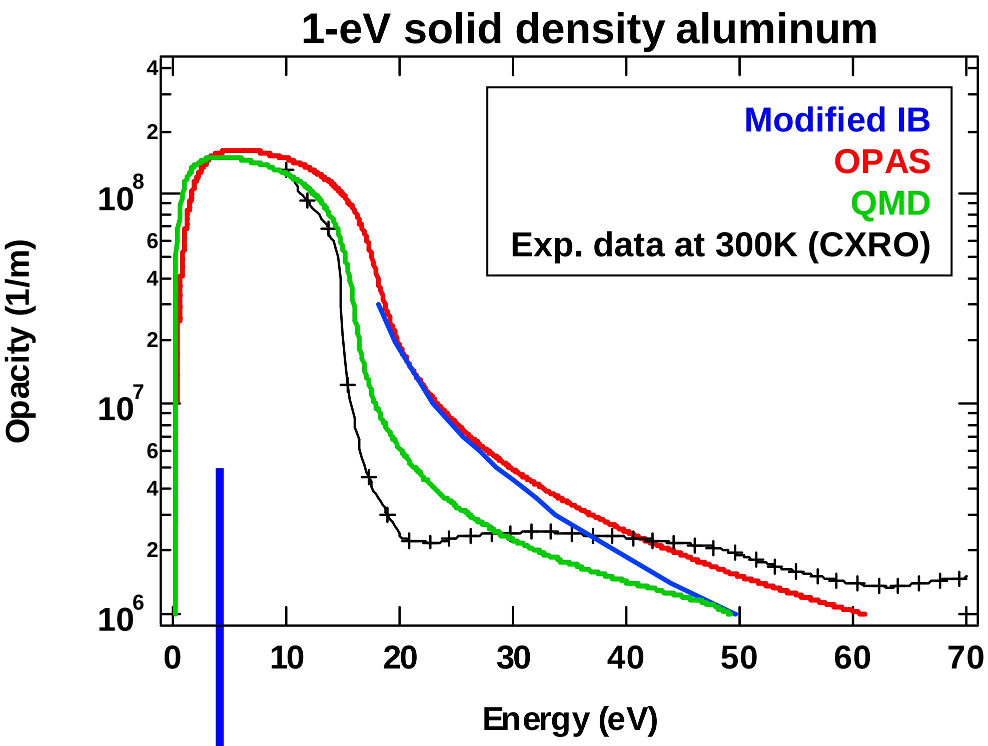
$$K^{ff}(h\nu,\rho,T) = \frac{2}{3}\sqrt{\frac{2mc^2}{3\pi}}(2\pi\eta c)^3 \times \alpha r_e^2 \frac{Z^{*3}\rho\aleph^2}{A^2\sqrt{k_B T}}\frac{G^{ff}(h\nu,T)}{(h\nu)^3}$$

$$K(\omega) \sim \frac{\langle Z\rangle^3 \rho\, G^{ff}}{\sqrt{kT}\,\omega^3}$$

With non-relativistic quantum corrections through the Gaunt factor G^{ff}

From Mazevet *et al.*, A&A 405 (2003)

X-UV opacity of degenerate solid-density plasma highlights the conflict between kinetic theory and quantum mechanics



A minimum mean-free-path translates into a maximum opacity for free-free transitions

Plasma parameters

Nearly free M-shell electrons
L-edge at 73 eV
Plasma frequency at 15 eV
Fermi energy at 11 eV

Modified Inverse Bremsstrahlung
with partially degenerate electrons

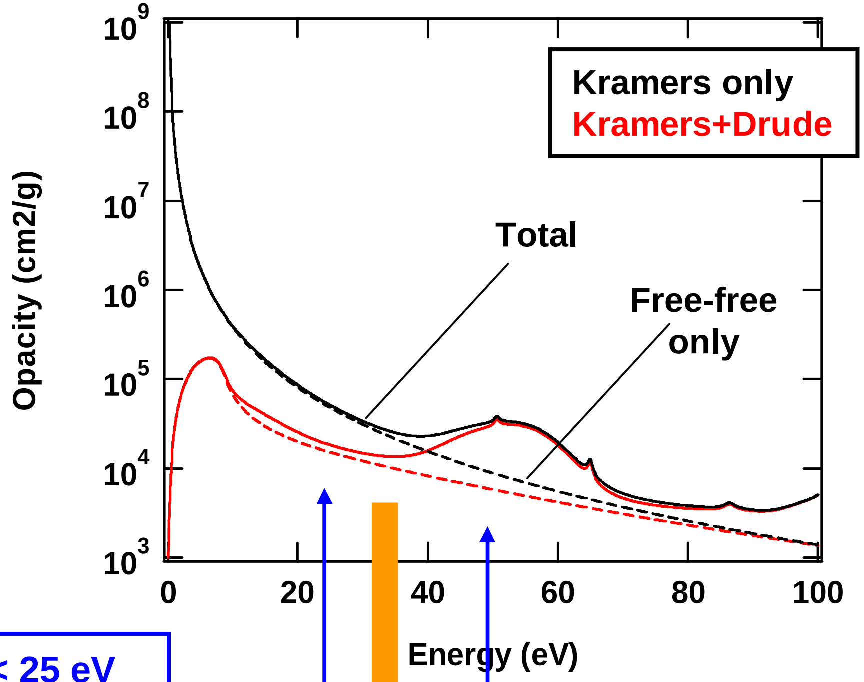
Considers fast and slow electrons with different collision frequencies. Pauli blocking reduces the number of electrons taking part in the absorption process for hν < E_F and T->0

Meyer-ter-Vehn *et al*.,
33th EPS Plasma Conf (2008)

Usual plasma approximations fail to reproduce the behaviour near ω_p

- **What happened to the opacity when the temperature is increasing?**
- **How accurate are the experimental data in this spectral range ?**

Bright sub-ps XUV radiation would allow to measure the low-frequency opacity of dense plasmas



hν < 25 eV
The free-free contribution dominates

hν > 50 eV
The bound-free contribution is not negligible

Al 40 eV - 0.5 g/cm^3

OPAS code

Renaudin *et al.*, Atomic Processes in Plasma, AIP Conf Proc 926 (2007)

Plasma parameters

Γ_{eff} = 1.3
Θ = 7.3
Plasma Frequency = 8.9 eV
Fermi Energy = 5.4 eV

Low-frequency opacity of dense plasmas

- at frequency greater than the plasma frequency,
- with kT~hν,
- with non-negligible contribution of the bound-free opacity

Conclusion

- **Drude model gives the correct answer for a large range of density and temperature.**
- **Ultrashort-pulse laser pump-probe experiments in the UV-IR range approximately agree with the Drude theory and the solutions of the Maxwell equations.**
- **Dense plasmas may have very interesting transport regimes.**
- **Any conductivity and low-frequency opacity data in the dense plasma regime would allow to better quantified 3D and quantum mechanical effects which are challenging to simulate.**
- **Bright sub-ps X-UV radiation (high-order harmonics and XFEL) is the unique way to perform such experiments**
- **Accurate design of sub-ps XFEL experiments need accurate opacities and conductivities**