Optical Properties of Plasmas Based on an Average-Atom Model

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- Linear Response $\Rightarrow$ Kubo-Greenwood formula for $\sigma(\omega)$
- Kramers-Kronig Dispersion Relation $\Rightarrow$ Dielectric Function $\epsilon(\omega)$
- Index of refraction $n(\omega) + i\kappa(\omega) = \sqrt{\epsilon(\omega)}$

Motivation

Free electron formula for index of refraction is used to determine electron densities.

$$n = \sqrt{1 - \frac{\omega_0^2}{\omega^2}} \approx 1 - \frac{\omega_0^2}{2\omega^2} < 1 \quad \text{where} \quad \omega_0^2 = 4\pi \frac{e^2}{m} \frac{N_{\text{free}}}{\Omega}$$

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Reason: Effect of bound electrons on optical properties.

- LLNL COMET laser facility[1] (14.7 nm Ni-like Pd laser)
- Advanced Photon Research Center JAERI[2] (13.9 nm Ni-like Ag laser)

[1] J. Filevich et al. *Proceedings of the 9th International Conference on X-Ray Lasers*, May 23-28 (2004)
[2] H. Tang et al., Appl. Phys. B**78**, 975 (2004)

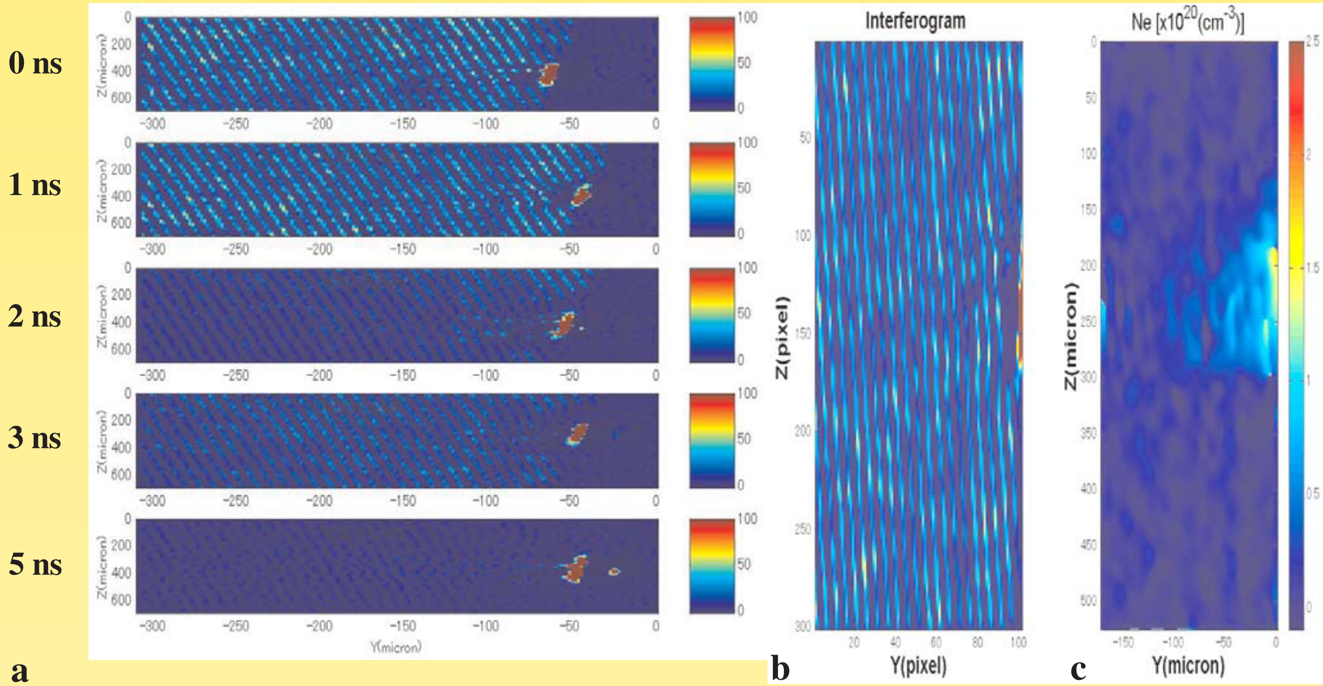

FIGURE 2 **a** Interference fringes after removing self-emission of the Al plasma. The fringes evolve with the increasing of diagnosing delay (0 ~ 5 ns). Only the fringes very close to the target surface cannot be resolved due to the over intense self-emission. **b** Interference fringe pattern **c** Electron density map for the 1 ns-delay and for an irradiance of 1.4×10^{11} Wcm^{-2}

Average-Atom Model

QM version of a model proposed by Feynman, Metropolis, and Teller[3]

Inside a neutral Wigner-Seitz cell: $\Omega = A/$(Avagadro No.$\times$ density)

$$\left[\frac{p^2}{2m} - \frac{Z}{r} + V\right]\psi_i(\boldsymbol{r}) = \epsilon_i\psi_i(\boldsymbol{r}) \tag{1}$$

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$V = V_{\text{dir}}(r) + V_{\text{exc}}(r)$ for $r \leq R$ and $V = 0$ otherwise.

$$\nabla^2 V_{\text{dir}} = -4\pi\rho \tag{2}$$

$V_{\text{exc}}(\rho)$ is given in the local density approximation

[3] R. P. Feynman, N. Metropolis and E. Teller, Phys. Rev. **75** 1561 (1949)

Thermal Average Electron Density

Contributions to the density are

$$\rho_b(r) = \frac{1}{4\pi r^2} \sum_l 2(2l+1) \sum_n f(\epsilon_{nl})\, P_{nl}(r)^2 \quad (3)$$

$$\rho_c(r) = \frac{1}{4\pi r^2} \sum_l 2(2l+1) \int_0^\infty d\epsilon\, f(\epsilon)\, P_{\epsilon l}(r)^2 \quad (4)$$

where

$$f(\epsilon) = \frac{1}{1 + \exp[(\epsilon - \mu)/kT]}$$

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$$Z = \int_{r<R} \rho(r)\, d^3r \equiv \int_0^R 4\pi r^2 \rho(r)\, dr \ . \tag{5}$$

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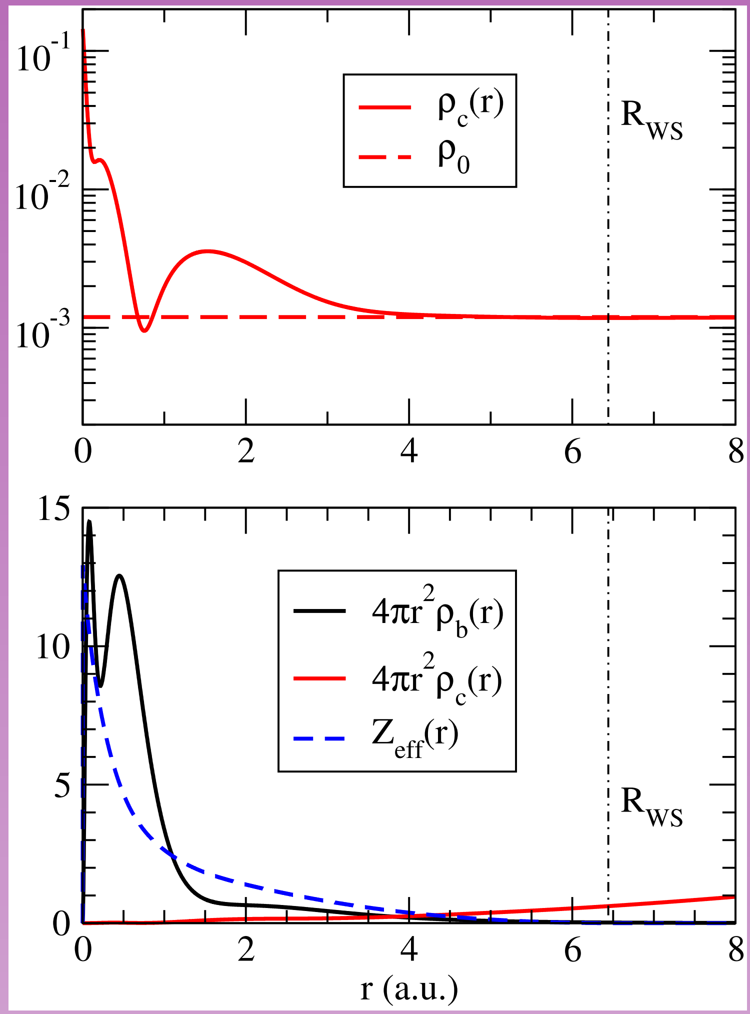
Eqs. (1-5) are solved self-consistently for ρ, V, and μ.

Example

Al: density 0.27 gm/cc, $T = 5$ eV, $R = 6.44$ a.u., $\mu = -0.3823$ a.u.

Bound States			Continuum States			
State	Energy	$n(l)$	l	$n(l)$	$n_0(l)$	$\Delta n(l)$
$1s$	-55.189	2.0000	0	0.1090	0.1975	-0.0885
$2s$	-3.980	2.0000	1	0.2149	0.3513	-0.1364
$2p$	-2.610	6.0000	2	0.6031	0.3192	0.2839
$3s$	-0.259	0.6759	3	0.2892	0.2232	0.0660
$3p$	-0.054	0.8300	4	0.1514	0.1313	0.0201
			5	0.0735	0.0674	0.0061
			6	0.0326	0.0308	0.0018
			7	0.0132	0.0127	0.0005
			8	0.0049	0.0048	0.0001
			9	0.0017	0.0016	0.0001
			10	0.0005	0.0005	0.0000
	Nbound	11.5059	Nfree	1.4941	1.3404	0.1537



10^{-1}
10^{-2}
10^{-3}
$\rho_c(r)$
ρ_0
R_{WS}
0
2
4
6
8
15
10
5
0
$4\pi r^2 \rho_b(r)$
$4\pi r^2 \rho_c(r)$
$Z_{eff}(r)$
R_{WS}
r (a.u.)

Pressure

$$
\begin{aligned}
P &= \frac{1}{3}\left[T_{xx} + T_{yy} + T_{zz}\right]\Big|_{r=R} \\
&\approx \frac{(2mkT)^{5/2}}{6m\pi^2} I_{3/2}(\mu/kT),
\end{aligned}
$$

where $I_k(x)$ is a “Fermi-Dirac” integral

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I_k(x) = \int_0^\infty \frac{y^k dy}{1 + e^{(y-x)}}
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$P = P(\Omega, T)$ provides an **equation of state** for the plasma

n.b. atomic unit of $[P]$: 294.21 Mbar.

Kinetic and Potential Energies

$$
\begin{aligned}
E_{\rm kin} &= \int d^3r \sum_i \langle \psi_i | \frac{p^2}{2m} | \psi_i \rangle f(\epsilon_i) \\
&= \frac{3}{2} P\,\Omega - \frac{1}{2} E_{\rm pot}
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$$

This is the **generalized virial theorem**.

Entropy

The entropy S of a collection of fermions is given by the expression

$$TS = -kT \sum_i \{f(\epsilon_i) \ln f(\epsilon_i) + [1 - f(\epsilon_i)] \ln [1 - f(\epsilon_i)]\}$$

where $f(\epsilon_i) = 1/\left[1 + \exp{(\epsilon_i - \mu)/kT}\right]$

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where $f(\epsilon_i) = 1/\left[1 + \exp{(\epsilon_i - \mu)/kT}\right]$ This can be manipulated to give

$$\begin{aligned} TS &= \frac{5}{3} E_{\rm kin} + E_{e-n} + 2E_{e-e} - \mu N \\ &= \frac{5}{2} P\,\Omega + \frac{1}{6} E_{e-n} + \frac{7}{6} E_{e-e} - \mu N \end{aligned}$$

Other Thermodynamic Quantities

The internal energy U and the Helmholtz free energy F are given by

$$U = E_{\rm kin} + E_{\rm pot} = \frac{3}{2}P\,\Omega + \frac{1}{2}E_{e-n} + \frac{1}{2}E_{e-e}$$

$$F = U - TS = -P\,\Omega + \frac{1}{3}E_{e-n} - \frac{2}{3}E_{e-e} + N\mu$$

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$$dU = dQ - Pd\Omega = TdS - Pd\Omega$$

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The Helmholtz free energy is a thermodynamic function of the "natural" variables of the problem, Ω and T:

$$S = -\frac{\partial F}{\partial T}\bigg|_{\Omega} \qquad P = -\frac{\partial F}{\partial \Omega}\bigg|_{T}$$

Application: Plasma Conductivity

The Ziman formula[4] for the *static* conductivity of a many-particle system is

$$\sigma = -\frac{2e^2}{3}\int \frac{d^3p}{(2\pi)^3}\, v^2\, \tau(p)\, \frac{\partial f}{\partial E},$$

where $\tau(p)$ is the mean collision time and where $f(E)$ is the Fermi function.

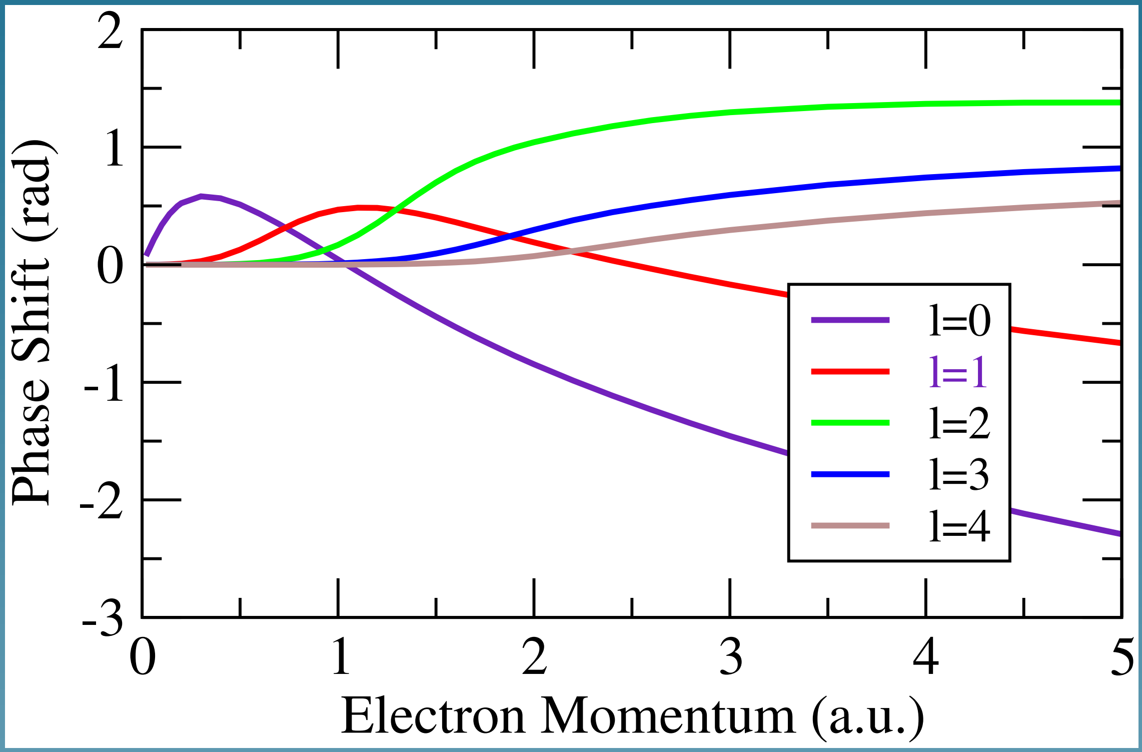
$$\tau(p) = \frac{\Lambda(p)}{v} \qquad \Lambda(p) = \frac{\Omega}{\sigma_{\mathrm{tr}}(p)}$$

$$\sigma_{\mathrm{tr}}(p) = \frac{4\pi}{p^2}\sum_{l=0}^{\infty}(l+1)\sin^2(\delta_{l+1}-\delta_l) \qquad \tau(p) = \frac{\Omega}{v\,\sigma_{\mathrm{tr}}(p)}$$

$$\sigma = -\frac{\Omega}{3\pi^2}\int_0^{\infty} dE \left[\frac{v^2}{\sigma_{\mathrm{tr}}(p)}\right]\frac{\partial f}{\partial E}.$$

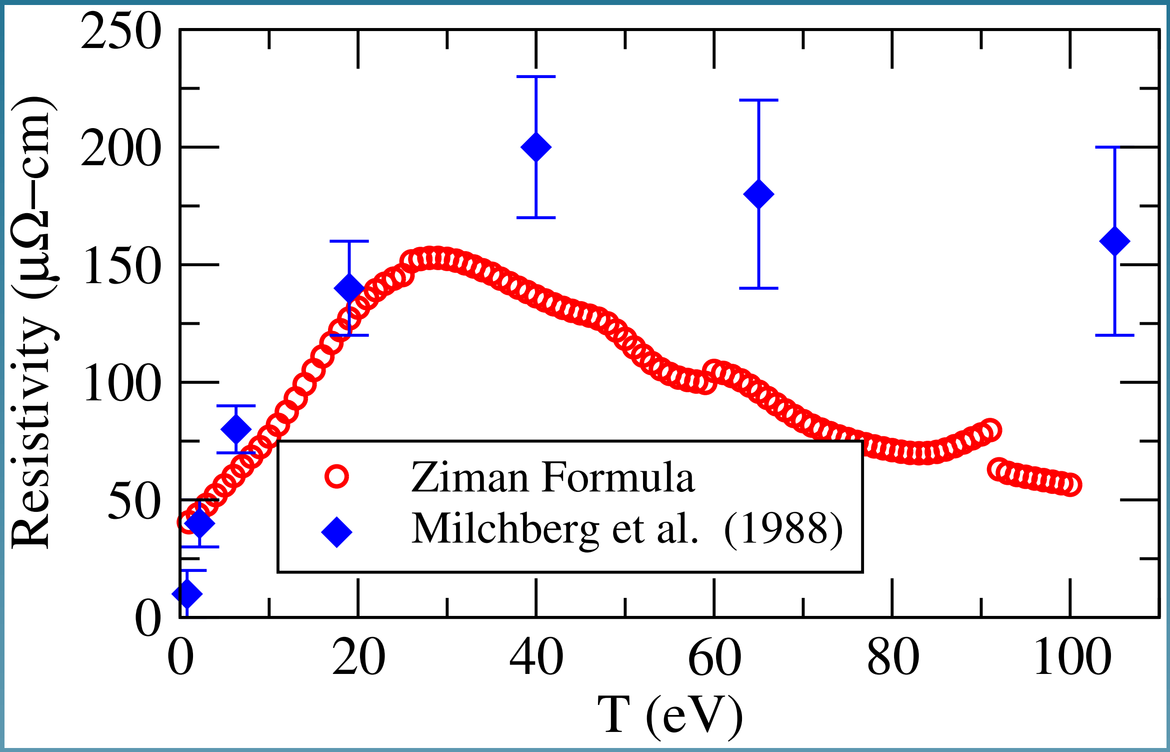
[4] G. D. Mahan, *Many-Particle Physics*, Plenum, 2000

Phase Shifts



$T = 10$ eV and metallic density

Resistivity of Aluminum



Metallic density

Linear Response and the Kubo-Greenwood Formula

Apply an electric field to the average atom:

$$\boldsymbol{E}(t) = F\hat{\boldsymbol{z}}\,\sin\omega t \qquad \boldsymbol{A}(t) = \frac{F}{\omega}\,\hat{\boldsymbol{z}}\,\cos\omega t$$

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The time dependent Schrödinger equation becomes

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The current density is

$$J_z(t) = \frac{2e}{\Omega}\sum_i f_i\,\langle\psi_i(t)|v_z|\psi_i(t)\rangle$$

Solution Ansatz

$$\psi_i(\boldsymbol{r},t) = u_i(\boldsymbol{r})e^{-i\epsilon_i t} + w_i^+(\boldsymbol{r})e^{-i(\epsilon_i+\omega)t} + w_i^-(\boldsymbol{r})e^{-i(\epsilon_i-\omega)t}$$

$$[T_0 + V(n,r)]\,u_i(\boldsymbol{r}) = \epsilon_i u_i(\boldsymbol{r})$$

$$[T_0 + V(n,r) - (\epsilon_i \pm \omega)]\,w_i^\pm(\boldsymbol{r}) = \frac{eF}{2\omega}\,v_z\,u_i(\boldsymbol{r})$$

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Eigenvalue Expansion

$$w_i^+(\boldsymbol{r}) = \sum_j X_i^j u_j(\boldsymbol{r}) \qquad w_i^-(\boldsymbol{r}) = \sum_j Y_i^j u_j(\boldsymbol{r})$$

$$X_i^j = \frac{eF}{2\omega}\frac{\langle j|v_z|i\rangle}{\epsilon_j - i\eta - \epsilon_i - \omega} \qquad Y_i^j = \frac{eF}{2\omega}\frac{\langle j|v_z|i\rangle}{\epsilon_j - i\eta - \epsilon_i + \omega}$$

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The response current may be written

$$\begin{aligned} J \quad = \quad & \frac{4e}{\Omega}\sum_{ij} f_i \, \Big[\Re\Big(\langle i|v_z|j\rangle X_i^j + \langle j|v_z|i\rangle Y_i^{j\star}\Big)\cos\omega t \\ & + \Im\Big(\langle i|v_z|j\rangle X_i^j + \langle j|v_z|i\rangle Y_i^{j\star}\Big)\sin\omega t\Big] \end{aligned}$$

Kubo-Greenwood

- Linearize $\psi_i(\boldsymbol{r}, t)$ in F

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Result:

$$\sigma(\omega) = \frac{2\pi e^2}{\omega\Omega} \sum_{ij} (f_i - f_j)\; |\langle j|v_z|i\rangle|^2\, \delta(\epsilon_j - \epsilon_i - \omega),$$

which is an average-atom version of the Kubo[5]-Greenwood[6] formula.

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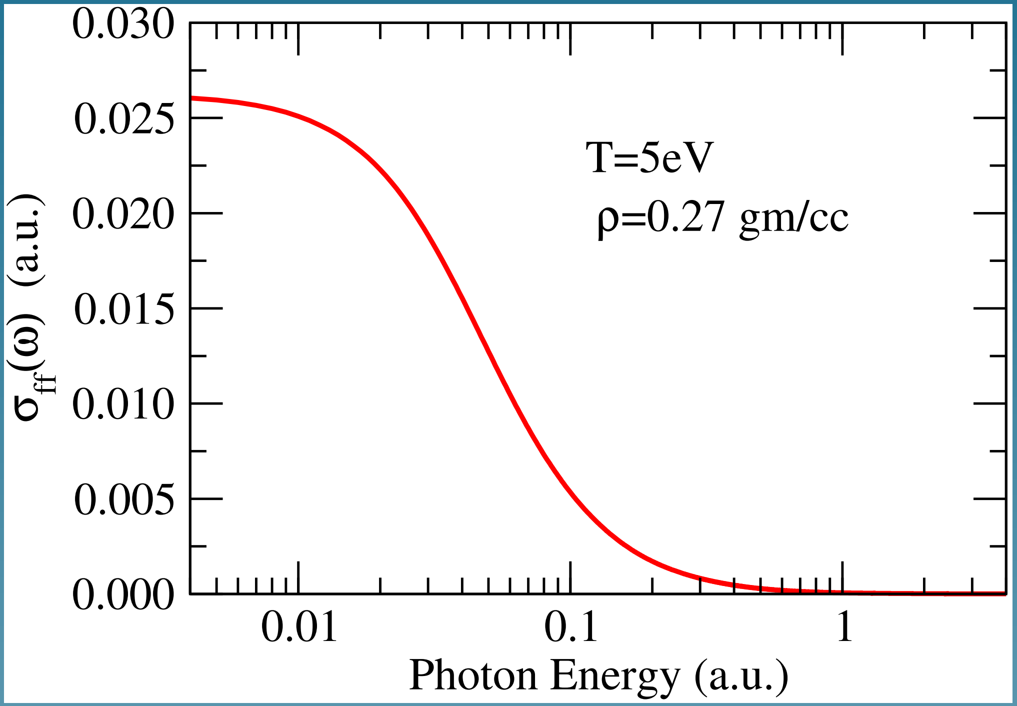
[5] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957)

[6] D. A. Greenwood, Proc. Phys. Soc. London **715**, 585 (1958)

Free-Free Contribution to Conductivity



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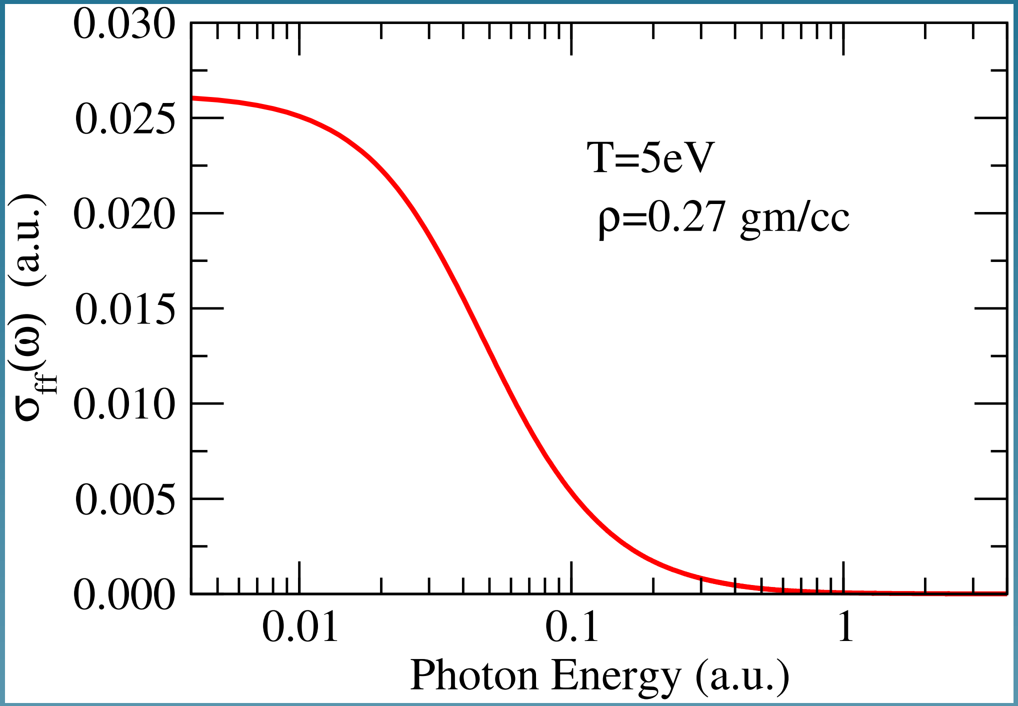
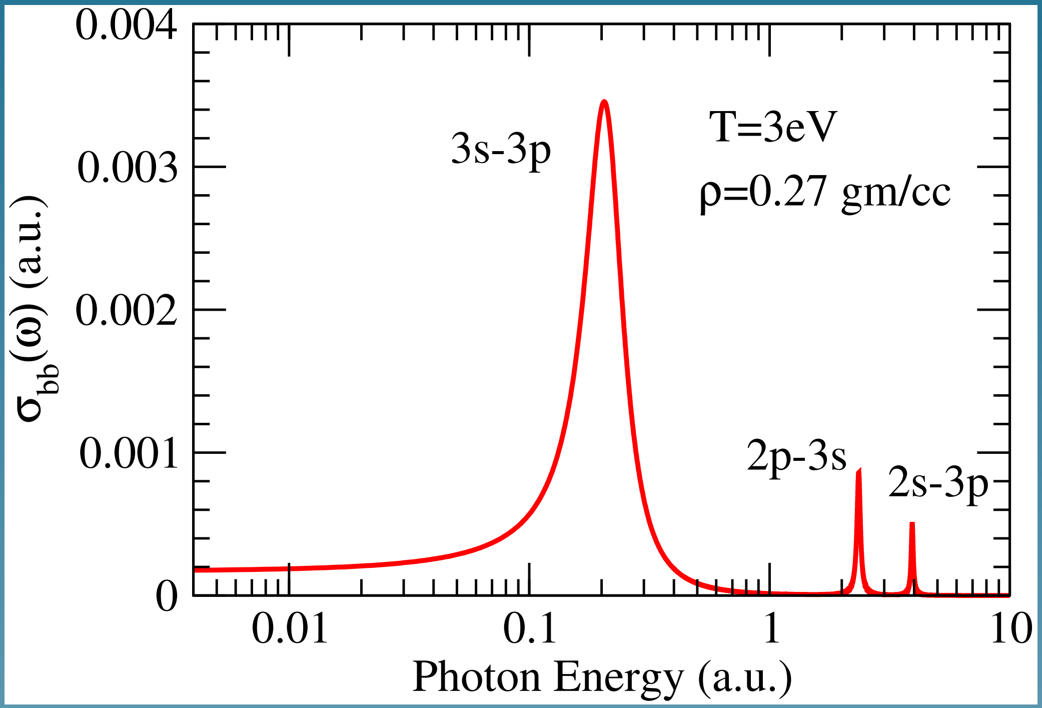
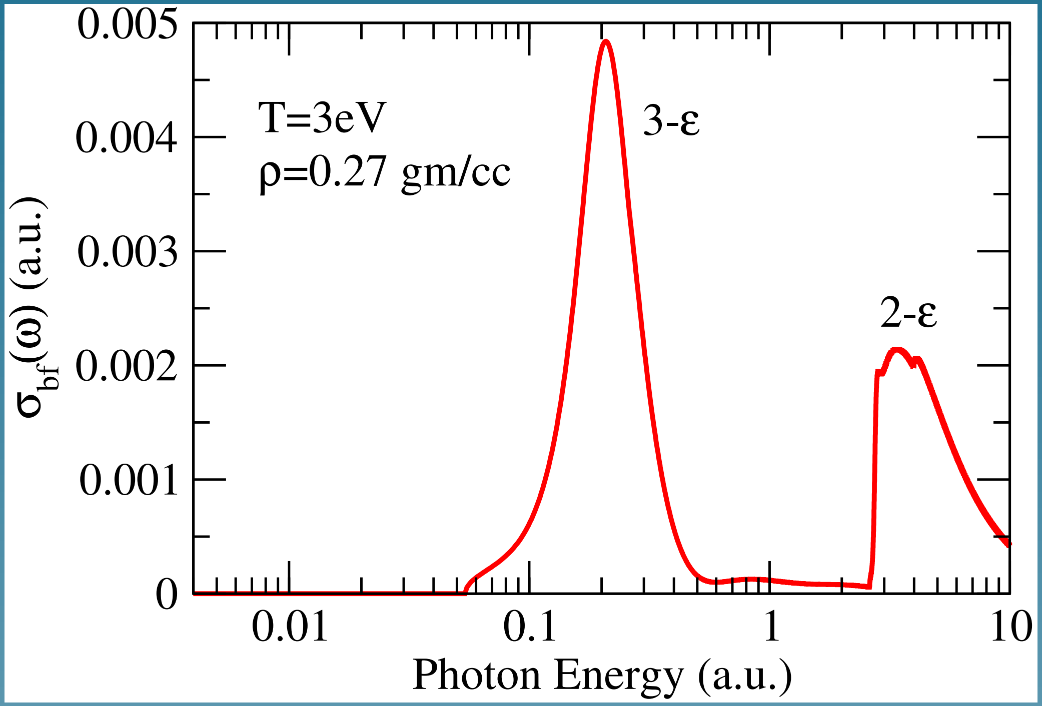


Michael Kuchiev has prepared an elegant note on the low-frequency conductivity explaining the origin of the infrared divergence and proposing a remedy.

Bound-Bound Contribution to Conductivity



Bound-Free Contribution to Conductivity



Optical Properties

For a conducting medium, the dielectric function is related to the *complex* conductivity by

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

We know $\Re\sigma(\omega)$; we must evaluate $\Im\sigma(\omega)$

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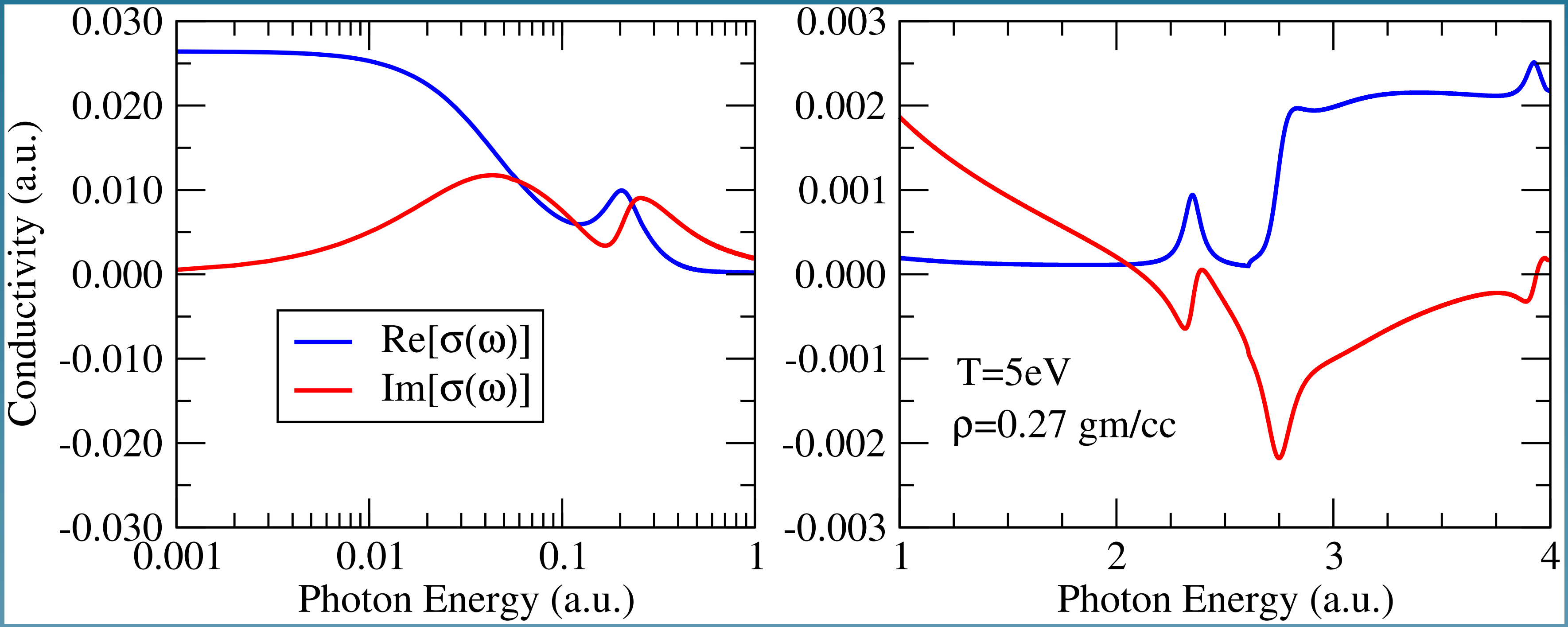
From analytic properties of $\sigma(\omega)$ one infers the dispersion relation[7]

$$\Im\,\sigma(\omega_0) = \frac{2\omega_0}{\pi} \int_0^{\infty} \frac{\Re\,\sigma(\omega)}{\omega_0^2 - \omega^2}\, d\omega.$$

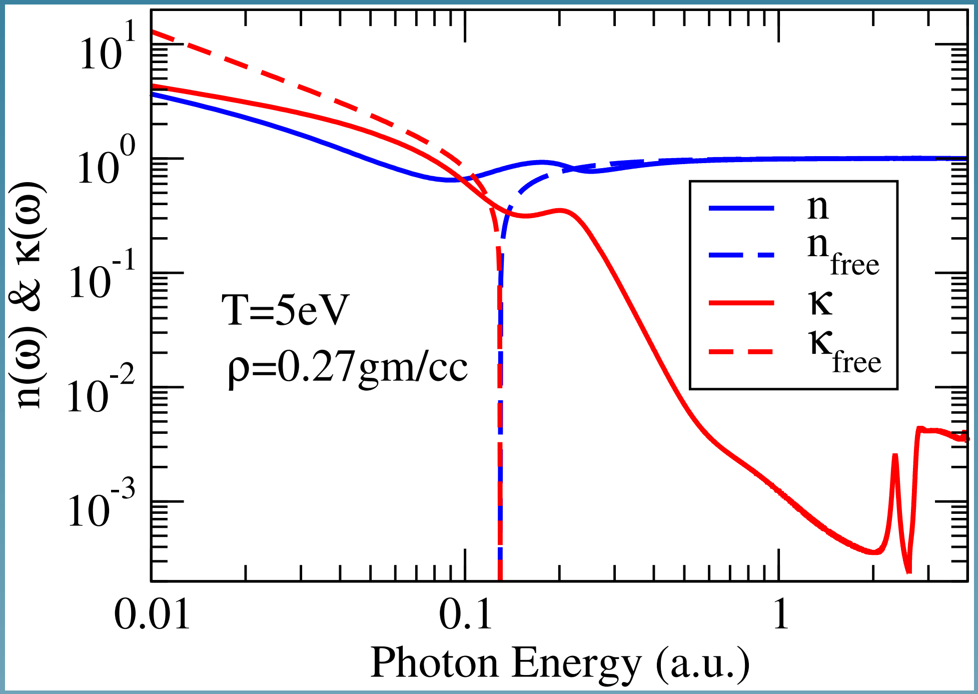
[7] R. de L. Kronig and H. A. Kramers, Atti Congr. Intern. Fisici, **2**, 545 (1927)

Application of Dispersion Relation



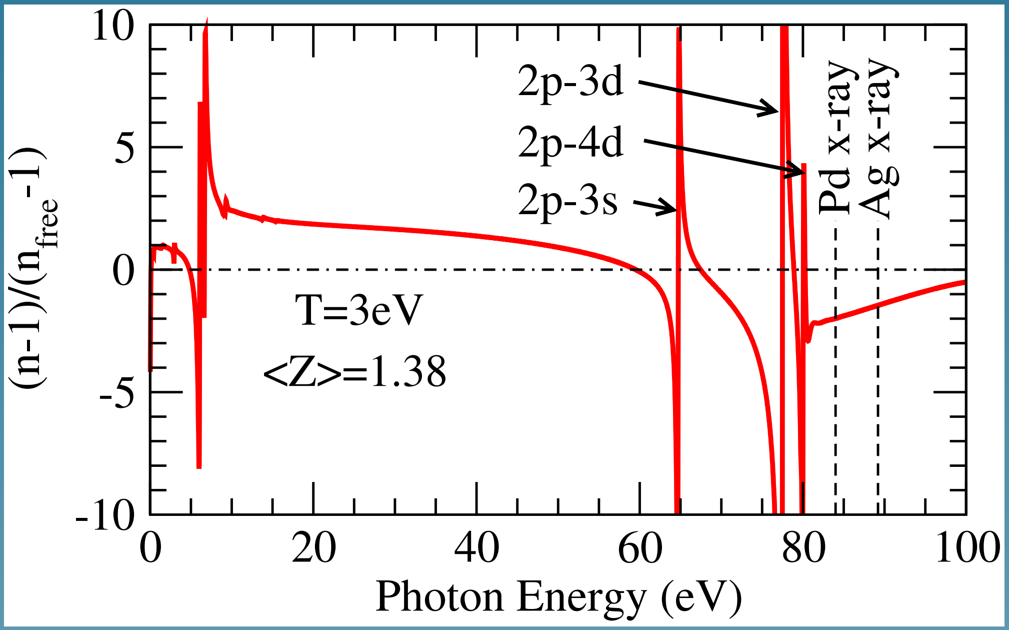
Index of Refraction

$$\Re\epsilon(\omega) = 1 - 4\pi\frac{\Im\sigma(\omega)}{\omega} \qquad \Im\epsilon(\omega) = 4\pi\frac{\Re\sigma(\omega)}{\omega},$$
$$n + i\kappa = \sqrt{\epsilon}.$$



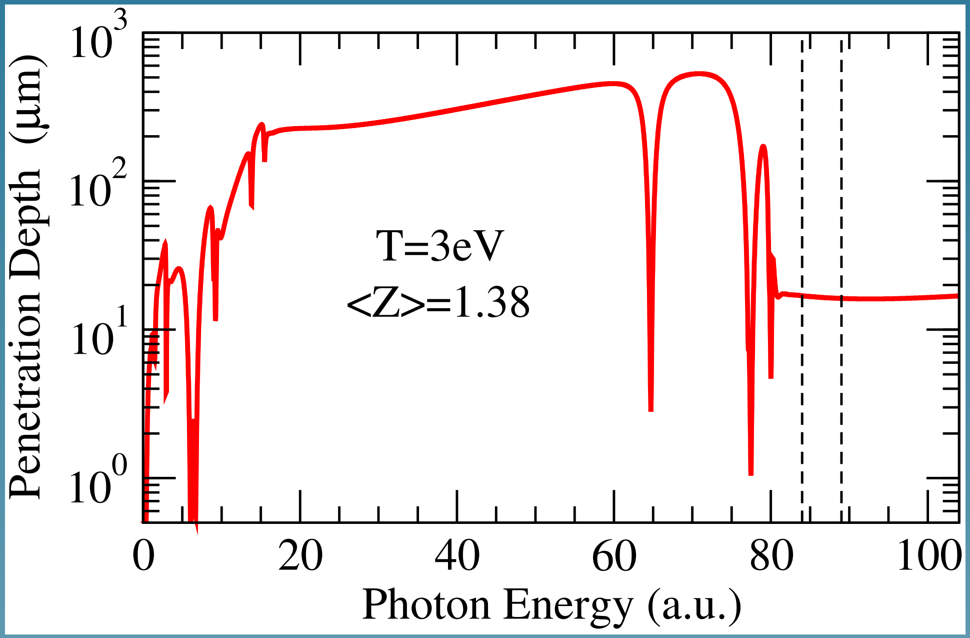
Al: Comparison with Free Electron Model

Plasma with ion density $n_{\text{ion}} = 10^{20}/\text{cc}$



Al: Penetration Depth

Plasma with ion density $n_{\text{ion}} = 10^{20}/\text{cc}$



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- The dielectric function (and index of refraction) can be reconstructed with the aid of a dispersion relation.

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- Average atom model is a simple way to understand the electronic structure of a plasma. (next step - look at neighbors)
- Linear response theory applied to the average atom model. provides a straightforward method for obtaining the frequency-dependent conductivity.
- The dielectric function (and index of refraction) can be reconstructed with the aid of a dispersion relation.
- The model explains "anomalous behavior" of low temperature Al plasmas in the 80-90 eV frequency range.

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- Linear response theory applied to the average atom model. provides a straightforward method for obtaining the frequency-dependent conductivity.
- The dielectric function (and index of refraction) can be reconstructed with the aid of a dispersion relation.
- The model explains "anomalous behavior" of low temperature Al plasmas in the 80-90 eV frequency range.

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