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Dr. Sándor Bordács (BME, FT) bordacs.sandor@ttk.bme.hu

Optical Spectroscopy in Materials Science:
Classical response functions: Drude model, oscillator model

Classical model of a free electron gas: Drude model

Assumptions in the Drude model:

- electrons obey Newton equation
    1. scattering relaxes momentum
    2. external field accelerates
- electrons are independent
- single ‚band': parameters τ, m, q, n

$$m\dot{v} = -\frac{mv}{\tau} + qE$$

$$-i\omega v = -\frac{v}{\tau} + \frac{q}{m}E$$

$$v = \frac{q}{m}\frac{1}{\frac{1}{\tau} - i\omega}E$$

Current response in the Drude model:

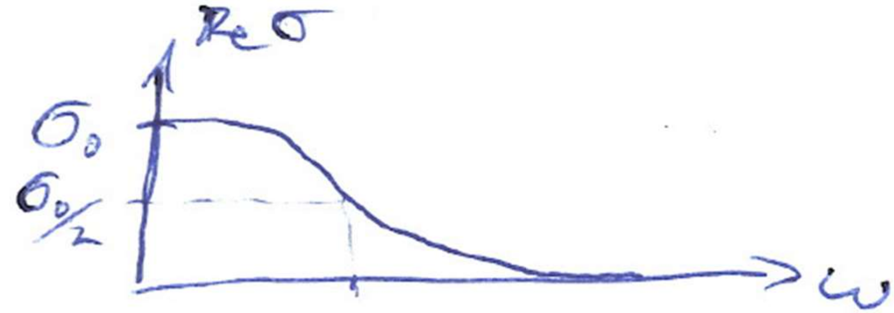
$$j = qnv = \boxed{\frac{q^2 n}{m}\tau \cdot \frac{1}{1 - i\omega\tau}} E$$

Optical/frequency dependent conductivity in the Drude model:

$$\boxed{\sigma(\omega) = \frac{\sigma_0}{1 - i\omega\tau}} \qquad \sigma_0 = \frac{q^2 n}{m}\tau$$

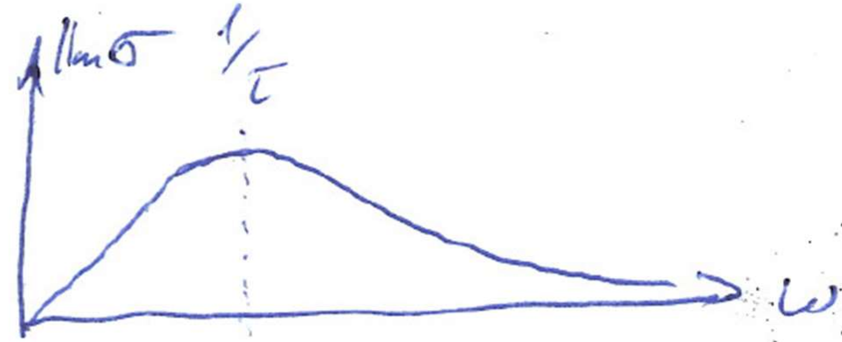
Response functions in Drude model

Parameters for a typical metal:
m, q ~ free electron
n ~ $1/3^3$ Å^{-3} ~ 10^{23} cm^{-3}
τ ~ 100 fs .. 1 ps (e.g. from resistivity)



$$\sigma_0 = \frac{q^2 n}{m}\tau$$

$$\sigma = \sigma_0\left(\frac{1}{1+\omega^2\tau^2} + i\,\frac{\omega\tau}{1+\omega^2\tau^2}\right)$$



Sum rule for conductivity:

$$\int_0^\infty \mathrm{Re}\,\sigma(\omega)\cdot d\omega = \int \frac{nq^2}{m}\frac{1}{1+\omega^2\tau^2}\tau\, d\omega = \frac{nq^2}{m}\int_0^\infty \frac{dx}{1+x^2} = \frac{nq^2}{m}\,\mathrm{arctg}(x)\Big|_0^\infty = \frac{nq^2}{m}\cdot\frac{\pi}{2}$$

Response functions in Drude model

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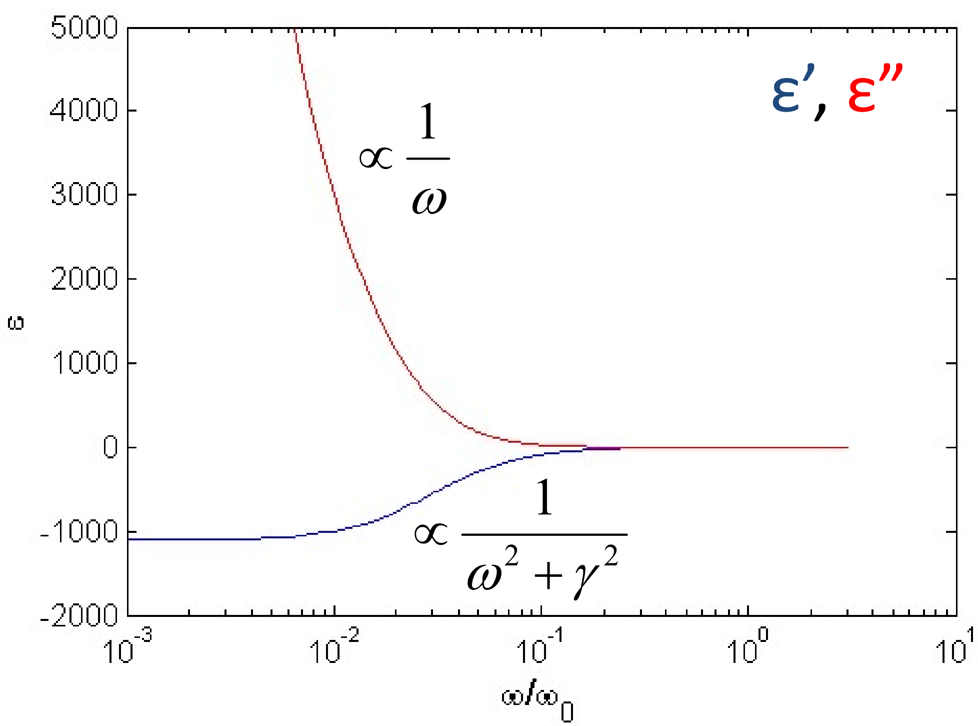
m, q ~ free electron
n ~ $1/3^3$ Å^{-3} ~ 10^{23} cm^{-3}
τ ~ 100 fs .. 1 ps (e.g. from resistivity

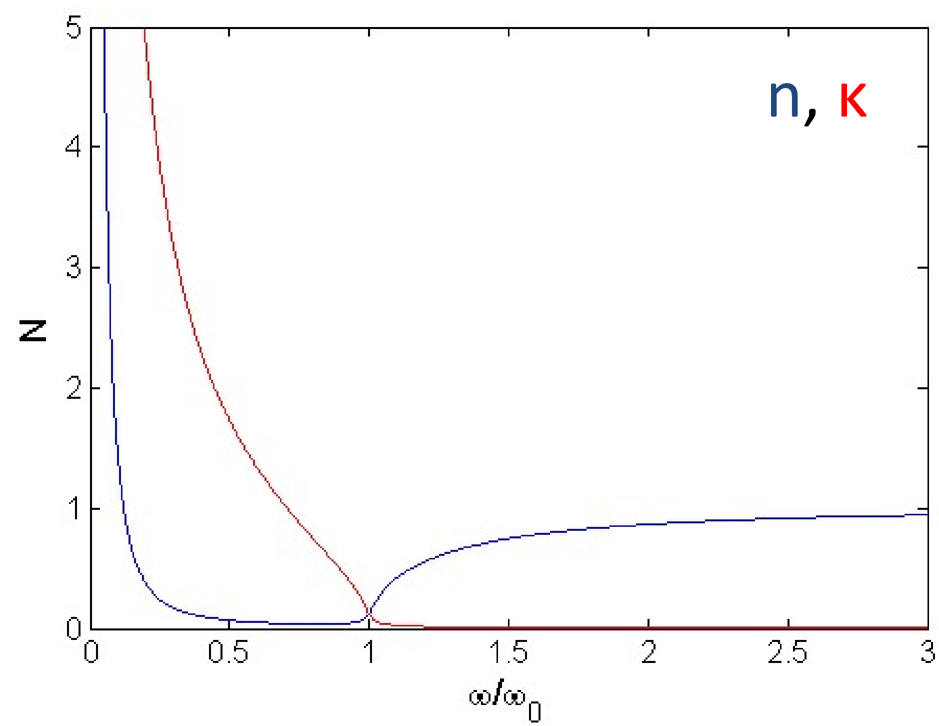
$\hbar\omega_p$ ~ few eV

$$\varepsilon = 1 + \frac{i\sigma}{\varepsilon_0\omega} = 1 + \frac{i}{\varepsilon_0\omega}\cdot\frac{q^2 n}{m}\tau\,\frac{1}{1-i\omega\tau}$$

$$\varepsilon = 1 - \underbrace{\frac{nq^2}{\varepsilon_0 m}}_{\omega_p^2}\cdot\frac{1}{\omega(\omega+i\gamma)} \qquad \gamma = \frac{1}{\tau}$$

$$\varepsilon = 1 - \omega_p^2\left(\frac{1}{\omega^2+\gamma^2} - \frac{i\gamma}{\omega}\,\frac{1}{\omega^2+\gamma^2}\right)$$



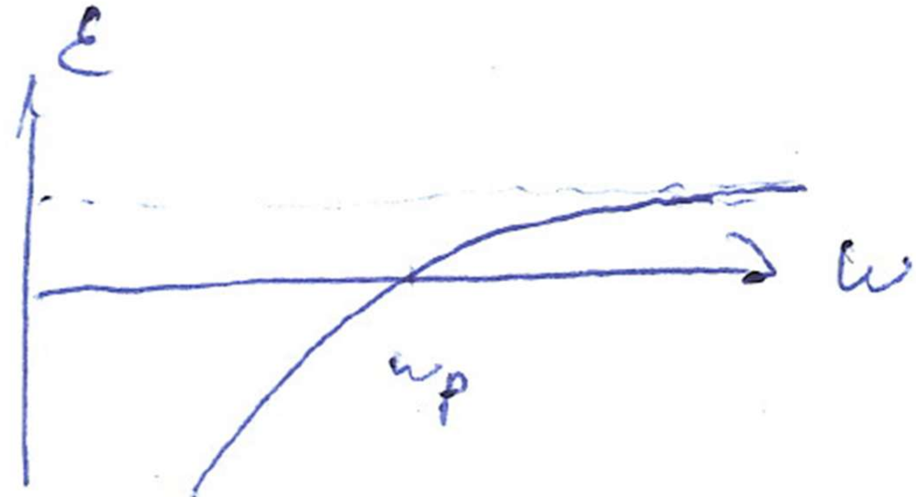


Drude model at high frequencies

High frequency limit: $\omega >> \gamma$

$$\varepsilon = 1 - \omega_p^2 \left(\frac{1}{\omega^2 + \gamma^2} - \frac{i\gamma}{\omega} \frac{1}{\omega^2 + \gamma^2} \right)$$

$$\boxed{\varepsilon = 1 - \frac{\omega_p^2}{\omega^2}}$$



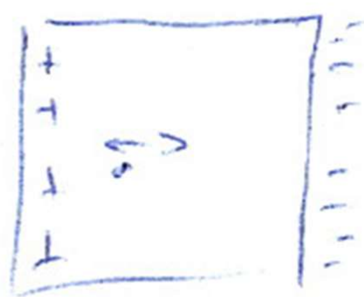
At $\omega=\omega_p$ $\varepsilon=0$, plasma oscillations:

$$D = 0$$

$$-P = \varepsilon E$$

$$m\ddot{x} = -nqx \cdot q$$

$$\omega^2 = \frac{nq^2}{\varepsilon_0 m}$$



Magnetic field component cannot couple to such oscillations:

$$\mathrm{q} \cdot \varepsilon_0 \varepsilon \mathrm{E} = 0$$

$$\mathrm{q} \times \mathrm{E} = \omega \mu_0 H$$

$$\left.\begin{aligned} \mathrm{q} \cdot \mu_0 H &= 0 \\ \mathrm{q} \times \mathrm{H} &= -\omega \varepsilon_0 \varepsilon \mathrm{E} \end{aligned}\right\} \quad \begin{aligned} \mathrm{q} \cdot H &= 0 \\ \mathrm{q} \times \mathrm{H} &= 0 \end{aligned}$$

Drude model at high frequencies

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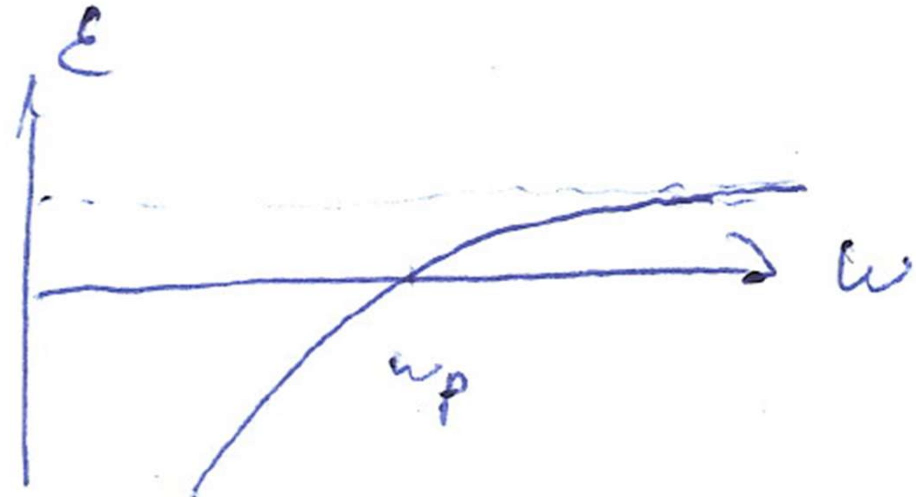
$$\varepsilon = 1 - \omega_p^2 \left(\frac{1}{\omega^2 + \gamma^2} - \frac{i\gamma}{\omega} \frac{1}{\omega^2 + \gamma^2} \right)$$

$$\boxed{\begin{gathered} \varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2} \\ \Omega_p = \frac{\omega_p}{\sqrt{\varepsilon_\infty}} \end{gathered}}$$



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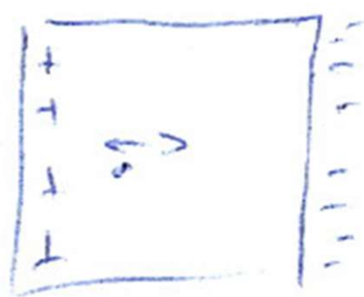

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Drude model at high frequencies

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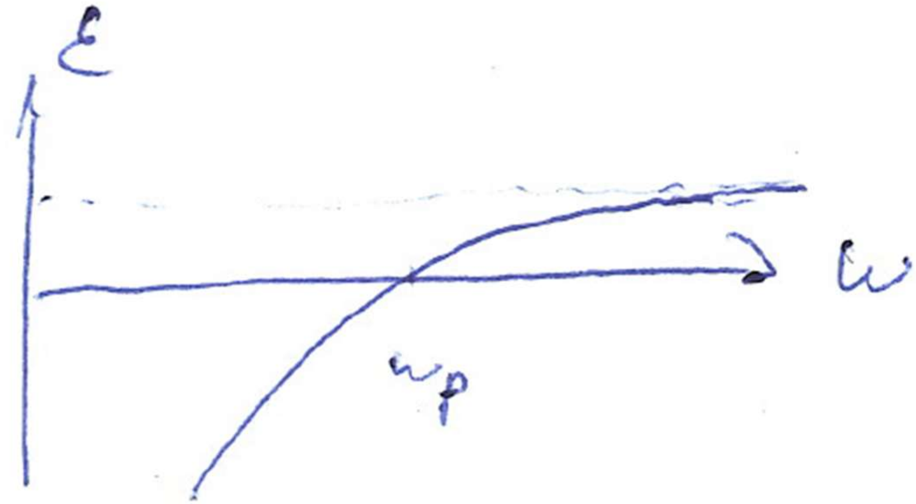
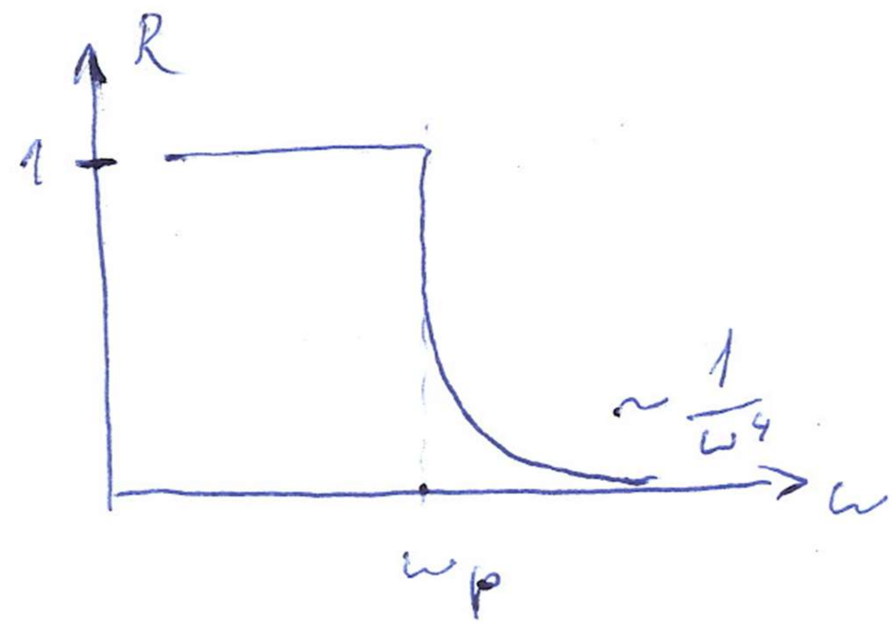
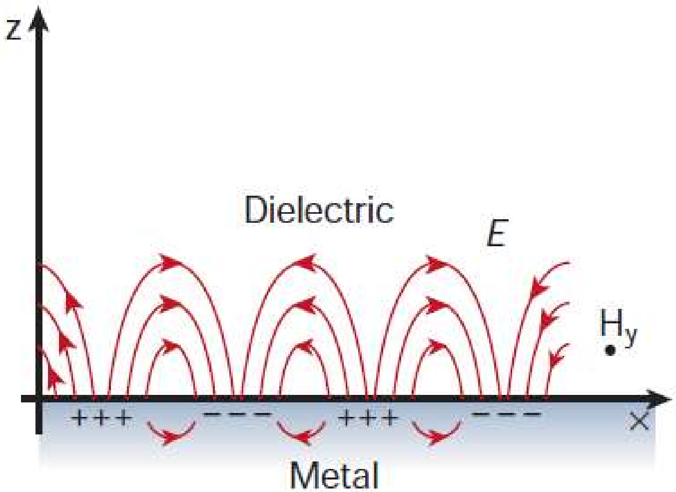


Above $\omega > \omega_p$ $\sqrt{\varepsilon} = n$ $R = \left| \frac{1-N}{1+N} \right|^2 = \left| \frac{1-n}{1+n} \right|^2$

Below $\omega < \omega_p$ $\sqrt{\varepsilon} = i\kappa$ $R = \left| \frac{1-i\kappa}{1+i\kappa} \right|^2 = 1$



Surface plasmon



Boundary conditions:

$$D_{z1} = D_{z2}$$
$$E_{x1,y1} = E_{x2,y2}$$
$$B_{z1} = B_{z2}$$
$$H_{x1,y1} = H_{x2,y2}$$

$$\boxed{\begin{aligned} D_{z1} &= D_{z2} \\ E_{x1,y1} &= E_{x2,y2} \\ \mu_0 H_{z1} &= \mu_0 H_{z2} \\ H_{x1,y1} &= H_{x2,y2} \end{aligned}}$$

$$E_{z1} \neq E_{z2}$$

H fields are the same

Maxwell eqs.

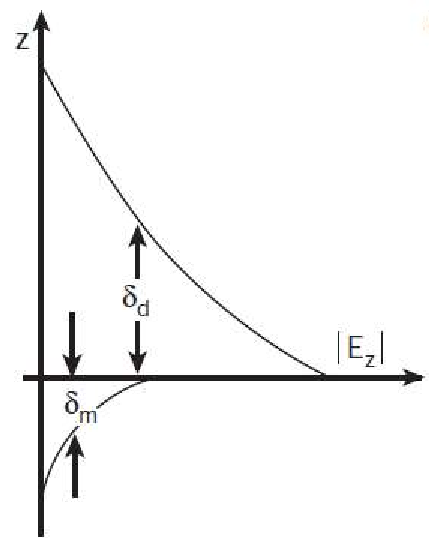
$$\mathrm{q} \cdot \varepsilon_0 \varepsilon \mathrm{E} = 0$$
$$\mathrm{q} \times \mathrm{E} = \omega \mu_0 H$$
$$\mathrm{q} \cdot \mu_0 H = 0$$
$$\mathrm{q} \times \mathrm{H} = -\omega \varepsilon_0 \varepsilon \mathrm{E}$$

Propagation along x

$$\mathrm{q_x} \varepsilon_1 \mathrm{E_{x1}} + \mathrm{q_{z1}} \varepsilon_1 \mathrm{E_{z1}} = 0$$
$$\mathrm{q_x} \varepsilon_2 \mathrm{E_{x2}} + \mathrm{q_{z2}} \varepsilon_2 \mathrm{E_{z2}} = 0$$

(Due to surface charges $E_z \neq 0$)

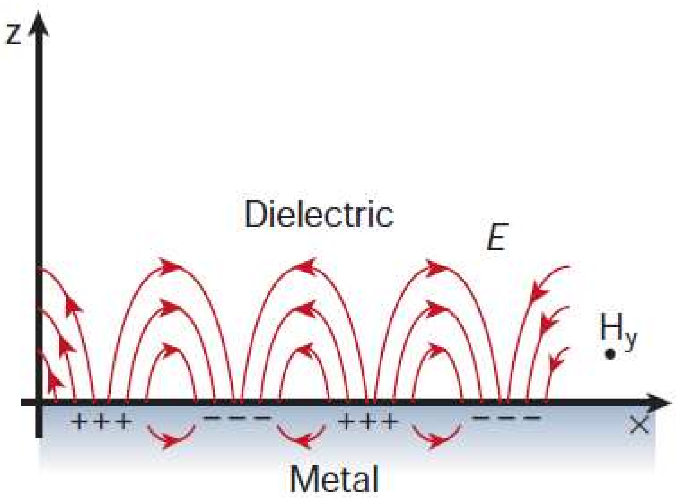
$$\boxed{\begin{aligned} \mathrm{E_{z1}} &= -\frac{\mathrm{q_x}}{\mathrm{q_{z1}}} \mathrm{E_{x1}} \\ \mathrm{E_{z2}} &= -\frac{\mathrm{q_x}}{\mathrm{q_{z2}}} \mathrm{E_{x2}} \end{aligned}}$$



TE mode ($E_{1x}=E_{2x}=0$, but $E_z \neq 0$) localized on the surface cannot exist

J. R. Sambles, Contemporary Physics **32**, **3**, *173-183* (1991)

Surface plasmon



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Maxwell eqs.

$$q \cdot \varepsilon_0 \varepsilon \mathrm{E} = 0$$
$$q \times \mathrm{E} = \omega \mu_0 H$$
$$q \cdot \mu_0 H = 0$$
$$q \times \mathrm{H} = -\omega \varepsilon_0 \varepsilon \mathrm{E}$$

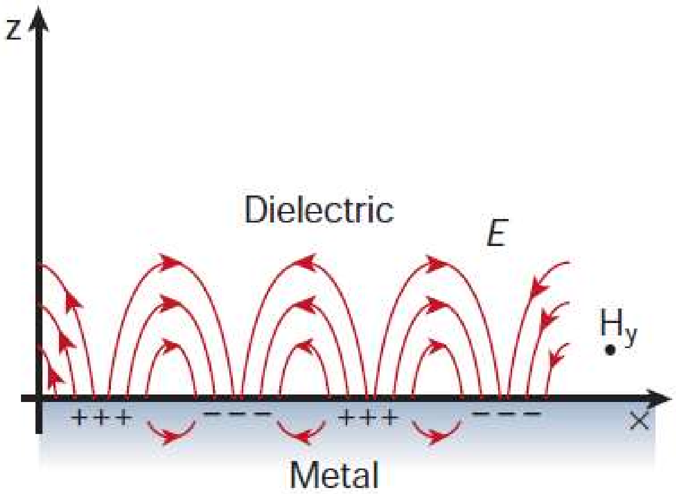
TM mode (satisfies Maxwell eqs.)

$$\boldsymbol{E}_1 = (E_{x1}, 0, E_{z1}) \exp[\mathrm{i}(k_x x - \omega t)] \exp(\mathrm{i} k_{z1} z),$$
$$\boldsymbol{H}_1 = (0, H_{y1}, 0) \exp[\mathrm{i}(k_x x - \omega t)] \exp(\mathrm{i} k_{z1} z),$$
$$\boldsymbol{E}_2 = (E_{x2}, 0, E_{z2}) \exp[\mathrm{i}(k_x x - \omega t)] \exp(\mathrm{i} k_{z2} z)$$
$$\boldsymbol{H}_2 = (0, H_{y2}, 0) \exp[\mathrm{i}(k_x x - \omega t)] \exp(\mathrm{i} k_{z2} z).$$

9 parameters: 1 intensity, 2 $E_{z1}(E_{x1})$&$E_{z2}(E_{x2})$, 2 $H_{y1}(E_{x1})$&$H_{y2}(E_{x2})$, 3 boundary conditions for E_z, E_x, H_y

J. R. Sambles, Contemporary Physics **32**, **3**, *173-183* (1991)

Surface plasmon



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TM mode (satisfies Maxwell eqs.)

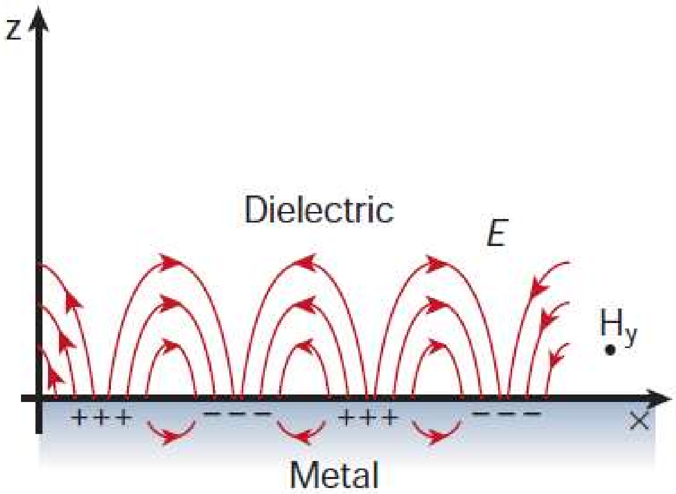
$$\boxed{\begin{aligned} \mathrm{q}_{z1}\mathrm{H}_{y1} &= \omega \varepsilon_0 \varepsilon_1 \mathrm{E}_{x1} \\ \mathrm{q}_{z2}\mathrm{H}_{y2} &= \omega \varepsilon_0 \varepsilon_2 \mathrm{E}_{x2} \end{aligned}}$$

$$\xrightarrow[H_{y1} = H_{y2}]{E_{x1} = E_{x2}}$$

$$\boxed{\frac{\varepsilon_1}{\mathrm{q}_{z1}} = \frac{\varepsilon_2}{\mathrm{q}_{z2}}}$$

J. R. Sambles, Contemporary Physics **32**, **3**, *173-183* (1991)

Surface plasmon



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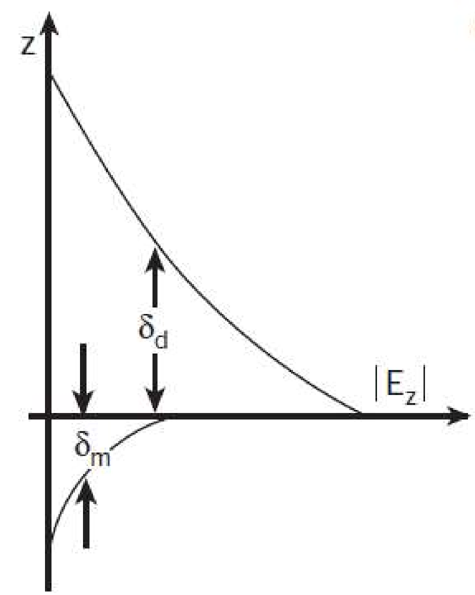
TM mode (satisfies Maxwell eqs.)

solution localized
on the surface if

$$\mathrm{iq}_{z1} < 0$$
$$\mathrm{iq}_{z2} > 0$$

$\longrightarrow$ ε_1 and ε_2 should have opposite signs



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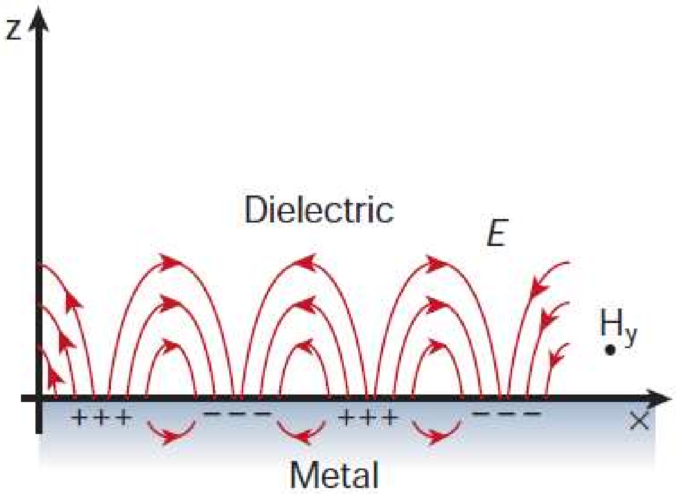
Surface plasmon



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TM mode (satisfies Maxwell eqs.)

$$\frac{\varepsilon_1}{\mathrm{q}_{z1}} = \frac{\varepsilon_2}{\mathrm{q}_{z2}}$$

$$\mathrm{q}_x^2 + \mathrm{q}_{z1}^2 = \varepsilon_1 \left(\frac{\omega}{c}\right)^2$$

$$\mathrm{q}_x^2 + \mathrm{q}_{z2}^2 = \varepsilon_2 \left(\frac{\omega}{c}\right)^2$$

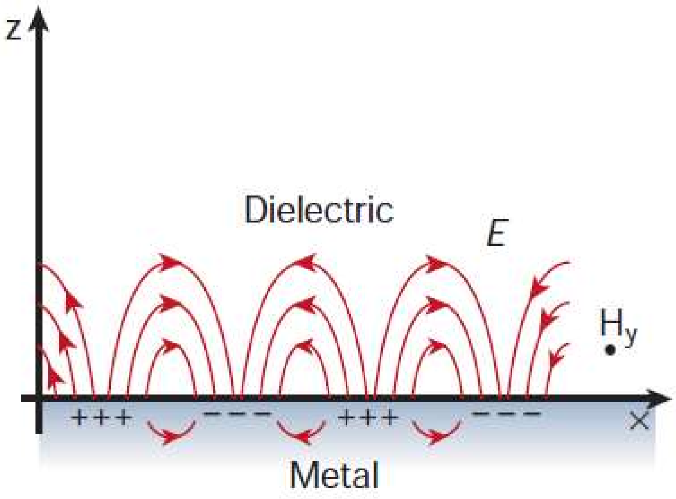
$$\longrightarrow \boxed{\mathrm{q}_x = \frac{\omega}{c} \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}}$$

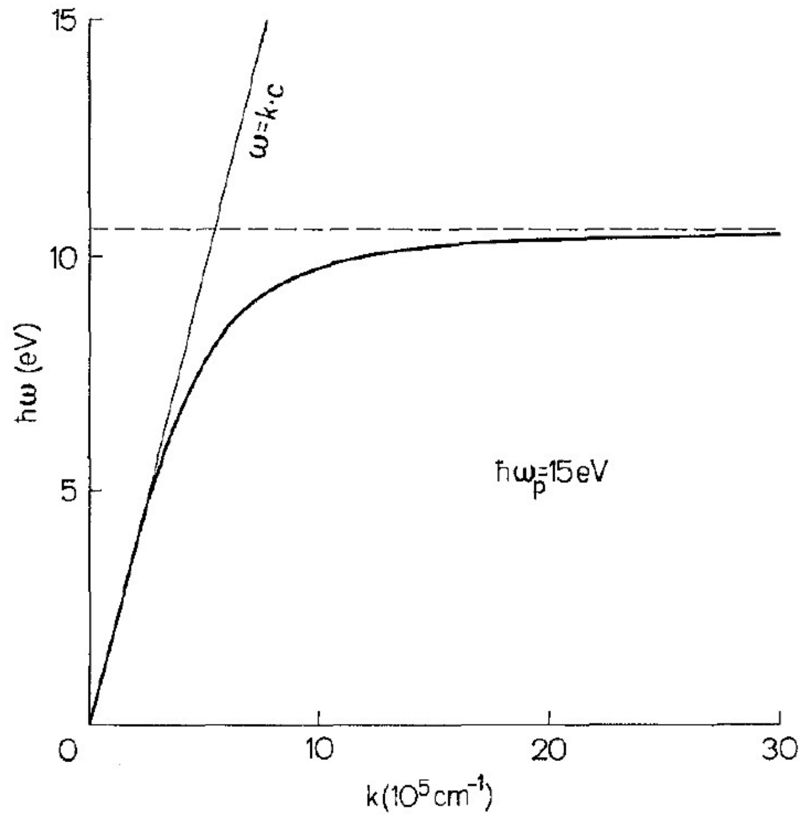
Propagating solution exists if (limit $\varepsilon_1 = -\varepsilon_2$)

$$\frac{1}{1 + \frac{\varepsilon_1}{\varepsilon_2}} > 0$$

J. R. Sambles, Contemporary Physics **32**, **3**, *173-183* (1991)

Surface plasmon





$$q_x = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}$$

$$\varepsilon_2 = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2}$$

$$\boxed{\omega_{limit} = \frac{\omega_p}{\sqrt{\varepsilon_1 + \varepsilon_\infty}}}$$

$$\omega_{limit} = \frac{\omega_p}{\sqrt{2}}$$

$$\varepsilon_1 = \varepsilon_\infty = 1$$

Momentum along the surface at grazing angles: $q_x^{max} = \frac{\omega}{c}\sqrt{\varepsilon_1}$

↓

transverse EM wave
cannot exite surface plasmons

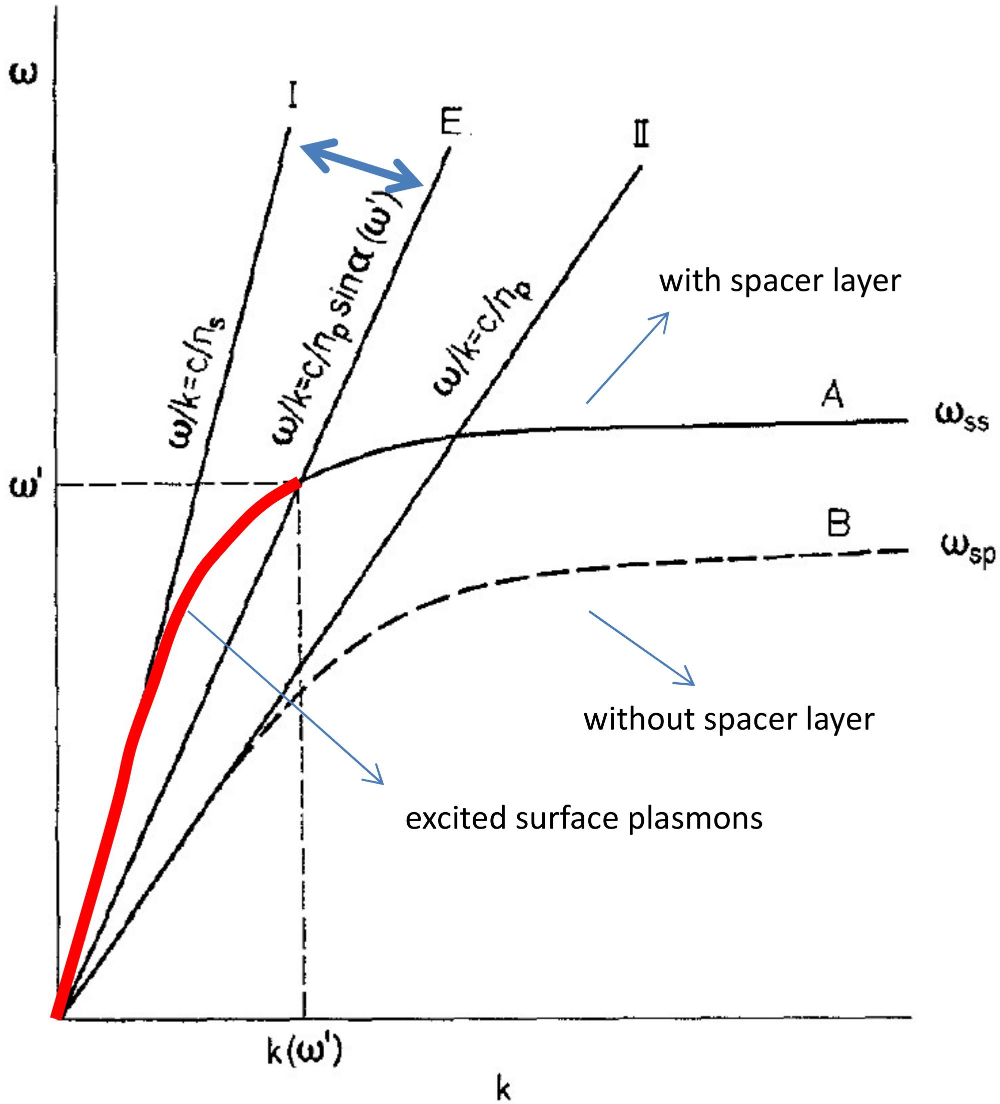
J. R. Sambles, Contemporary Physics **32**, **3**, *173-183* (1991)

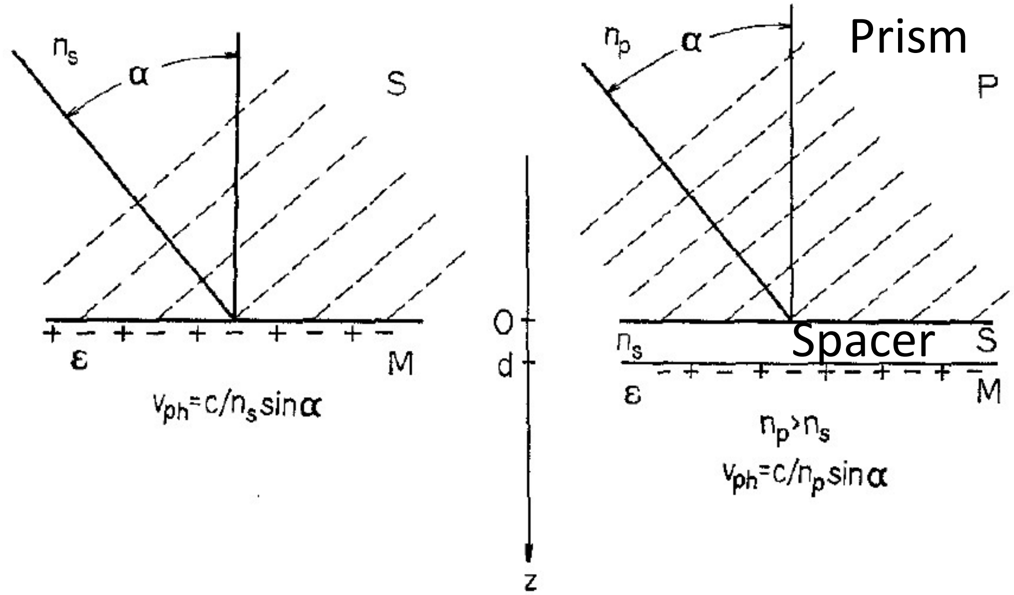
Surface plasmon

Momentum conservation along the surface (Snell's-law)

$$q_{x1} = q_{x2}$$

$$\frac{\omega}{c} n_P \sin\alpha = \frac{\omega}{c} n_S \sin\beta$$



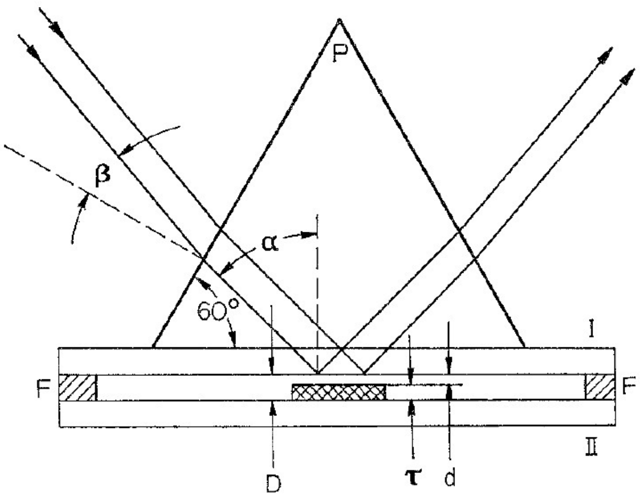


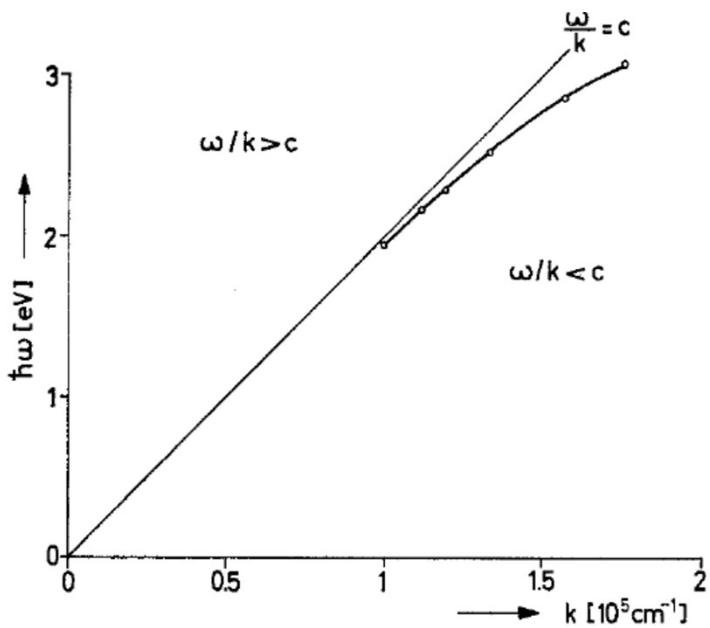
Evenescent waves above the critical angle:

$$\sin\alpha_c = \frac{n_S}{n_P}$$

A. Otto, Zeitschrift für Physik **216**, 398-410 (1968)

Surface plasmon





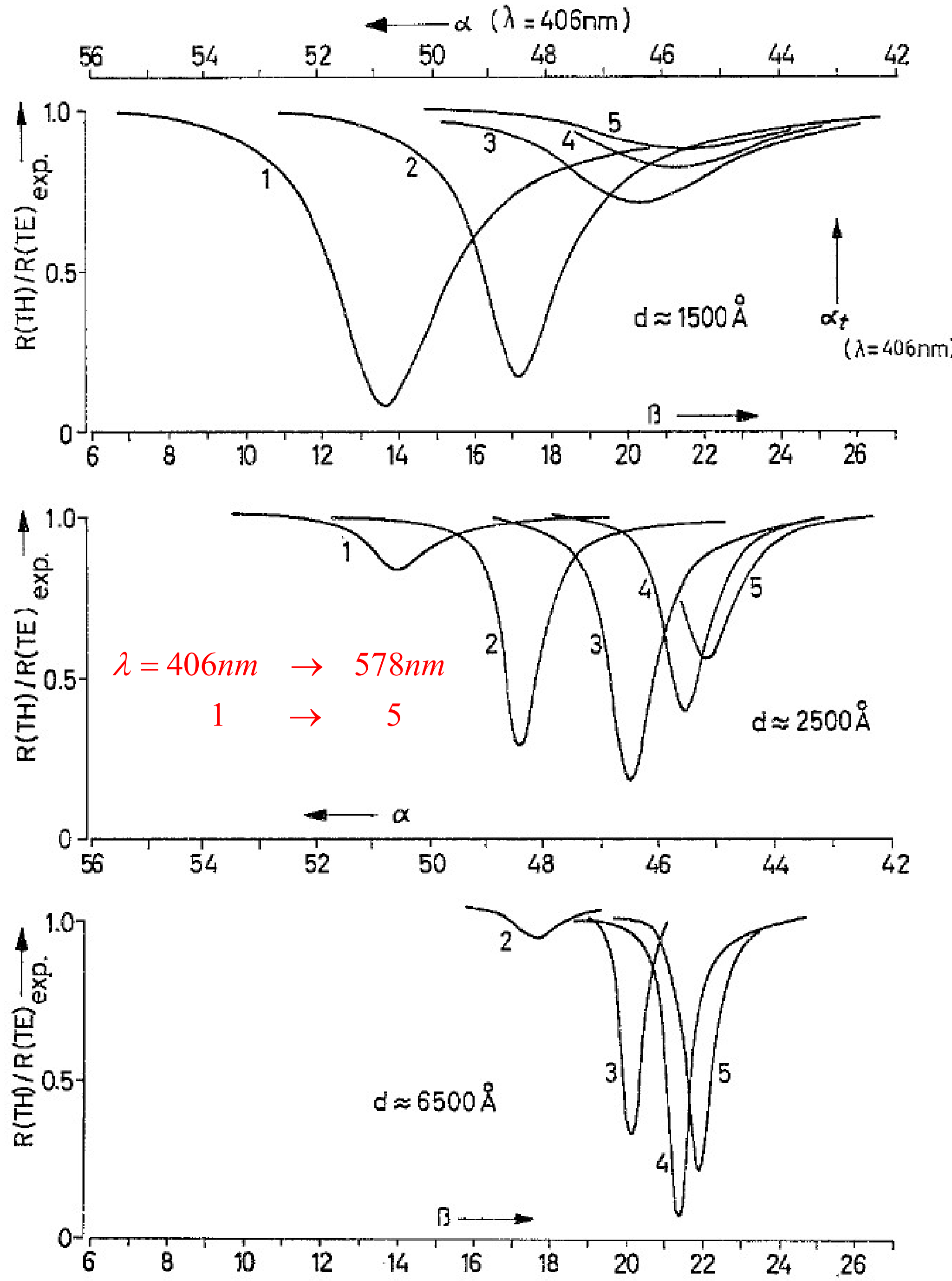


Fig. 5

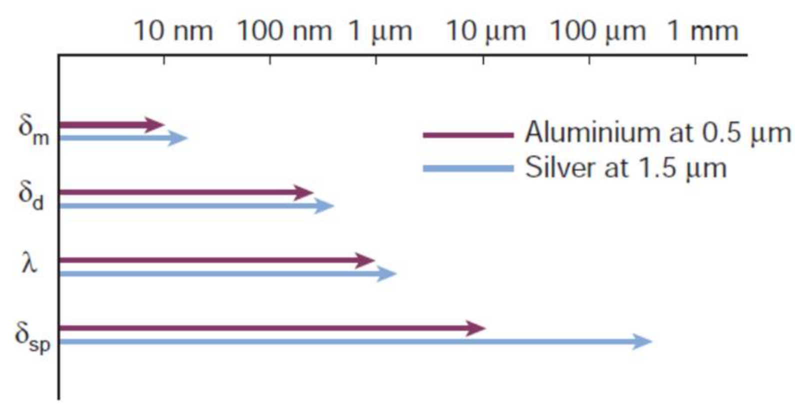
A method to measure the dielectric function

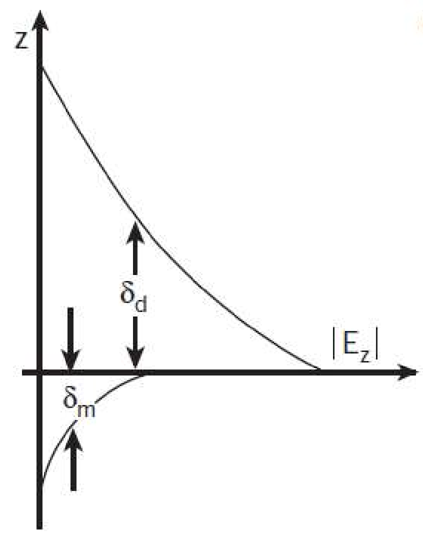
A. Otto, Zeitschrift für Physik **216**, 398-410 (1968)

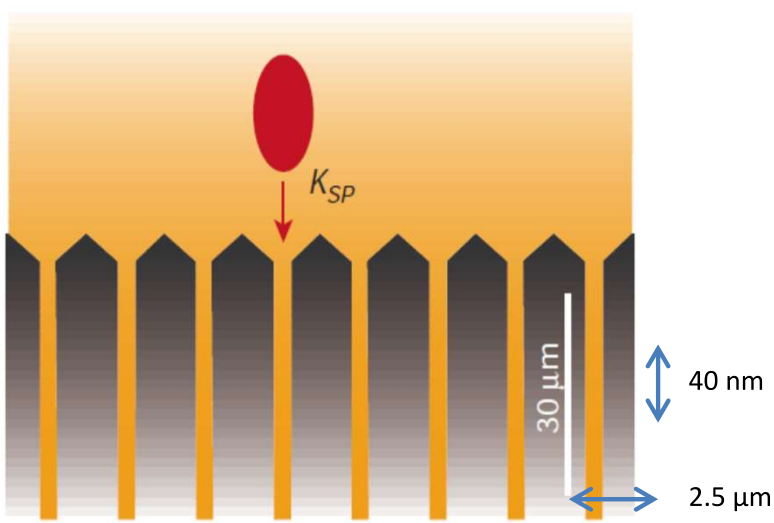
Surface plasmon

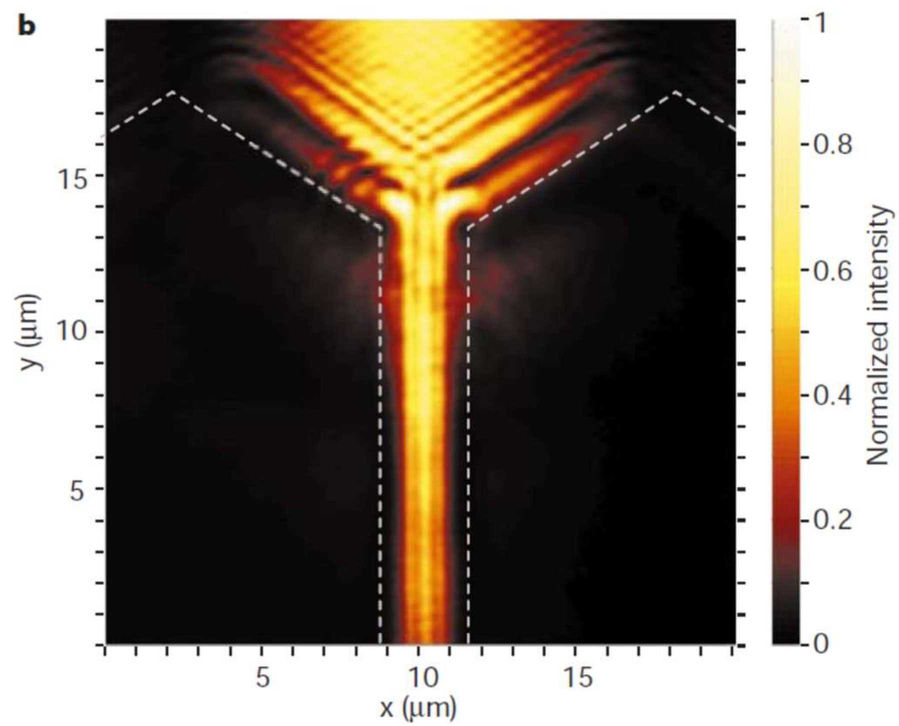
δ_m - decay length in metal (feature size)
δ_d - decay length in dielectric

$$\delta_{SP} = \frac{1}{2k''_{SP}} = \frac{c}{\omega}\left(\frac{\varepsilon'_m+\varepsilon_d}{\varepsilon'_m\varepsilon_d}\right)^{\frac{3}{2}}\frac{(\varepsilon'_m)^2}{\varepsilon''_m}$$









W. L. Barnes, Nature 424, 824 (2003)

Surface plasmon

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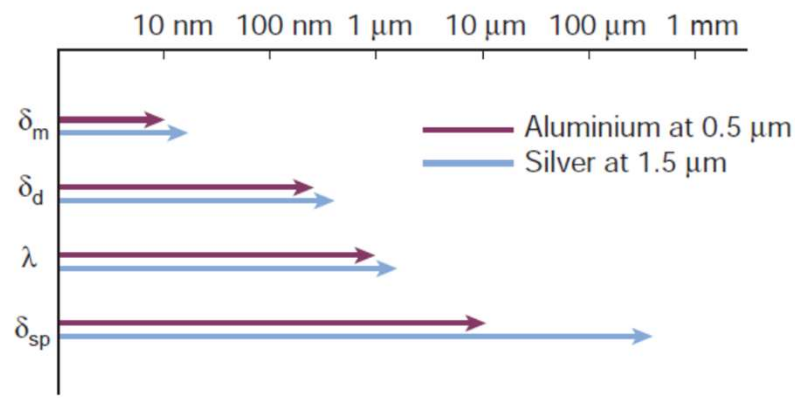
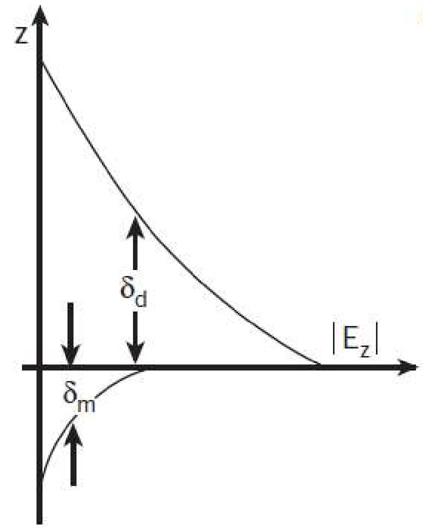
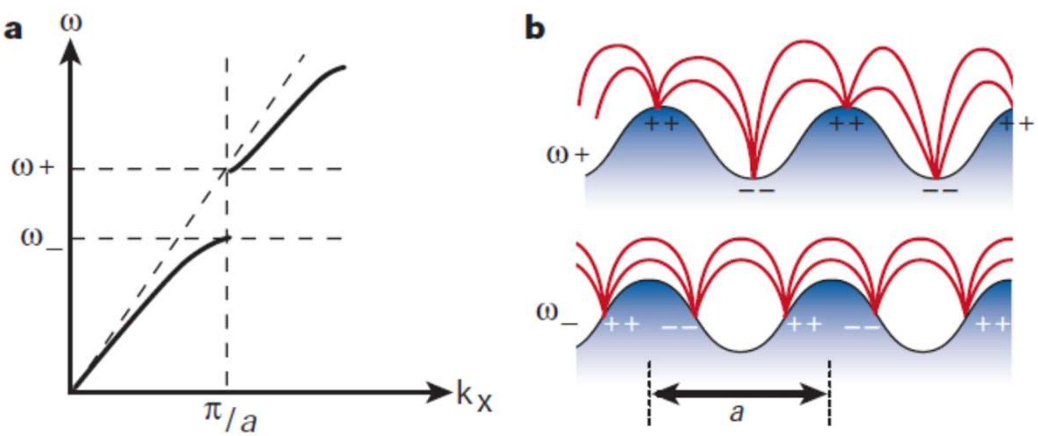
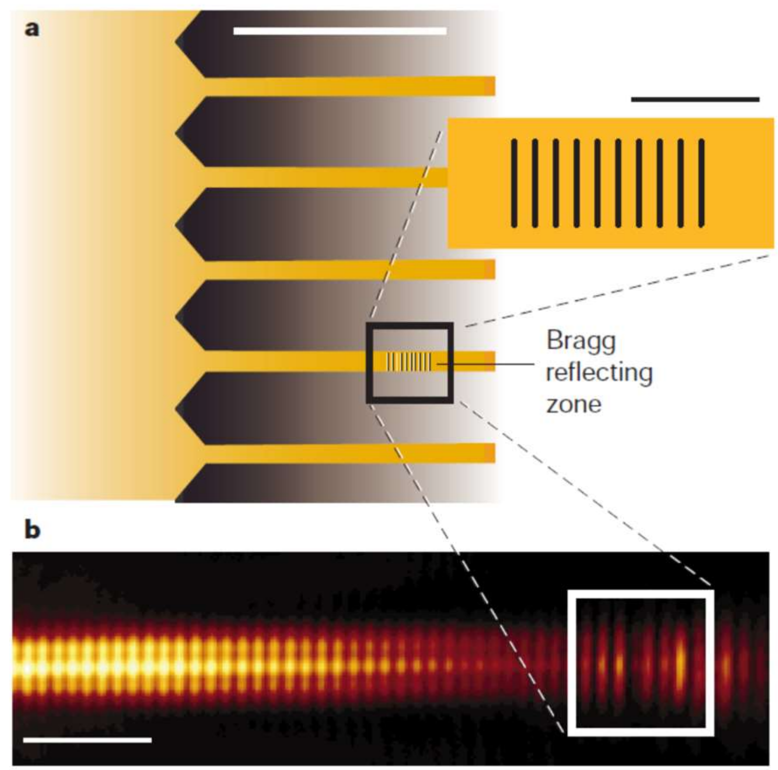




Coupling by grating (surface roughness) is also possible
A grating on the surface: crystal for plasmons





W. L. Barnes, Nature 424, 824 (2003)

Drude model: low frequency limit

Low frequency limit: ω<<γ $\quad \varepsilon \approx \varepsilon_\infty + \dfrac{i\sigma_0}{\varepsilon_0 \omega} \approx \dfrac{i\sigma_0}{\varepsilon_0 \omega}$

$$\varepsilon' \approx 0 \qquad n \approx k \approx \sqrt{\frac{\varepsilon''}{2}} \qquad n \approx k \gg 1$$

very high refractive index:
slow phase velocity in metals

$$R = \left|\frac{1-N}{1+N}\right|^2 = \frac{(1-n)^2+k^2}{(1+n)^2+k^2} \approx \frac{n^2+k^2-2n}{n^2+k^2+2n}$$

$$R \approx \left(1-\frac{2n}{n^2+k^2}\right)^2 \approx 1-\frac{4n}{n^2+k^2}$$

$$R \approx 1-2\sqrt{\frac{2}{\varepsilon''}} = 1-2\sqrt{\frac{2\varepsilon_0\omega}{\sigma_0}}$$

Hagen-Rubens law:

$$\boxed{R \approx 1-\sqrt{\frac{8\varepsilon_0}{\sigma_0}}\cdot\sqrt{\omega}}$$

Drude model: low frequency limit

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very high refractive index:
slow phase velocity in metals

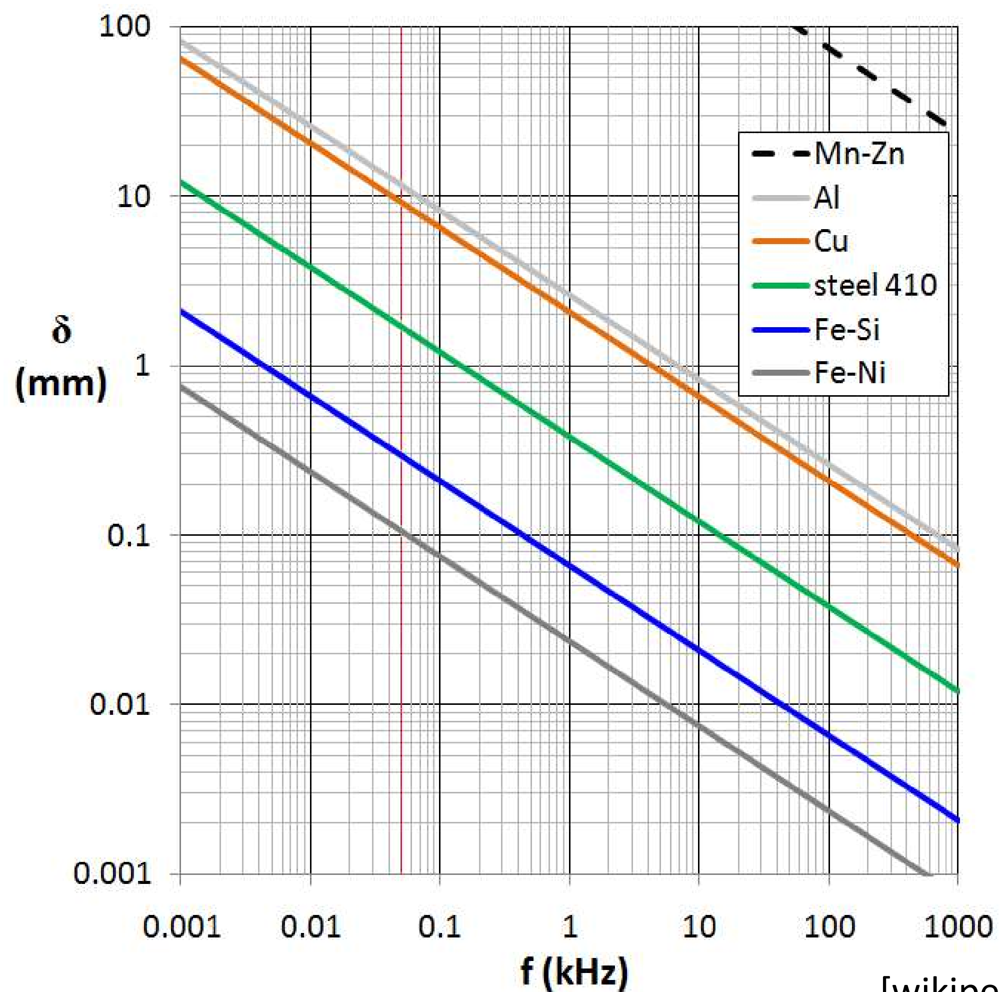
Skin-depth: $$\delta = \frac{1}{\Im m(q)} = \frac{c}{\omega \kappa}$$

$$\delta \approx \frac{c}{\omega} \frac{1}{\sqrt{\frac{\sigma_0}{2\varepsilon_0 \omega} \mu'}}$$

$$\boxed{\delta \approx \sqrt{\frac{2}{\omega \sigma_0 \mu_0 \mu'}}}$$

High freq. resistance of a wire:
(δ thick layer carries current
and wire radius, r>> δ)

$$R \approx \frac{\rho L}{2\pi r \delta}$$



[wikipedia]

Drude model: intermediate frequencies

Intermediate frequencies: $\gamma << \omega << \omega_p$

$$\varepsilon = 1 - \omega_p^2 \left(\frac{1}{\omega^2 + \gamma^2} - \frac{i\gamma}{\omega} \frac{1}{\omega^2 + \gamma^2} \right)$$

$$\varepsilon \approx \varepsilon_\infty - \frac{\omega_p^2}{\omega^2} \left(1 - \frac{i\gamma}{\omega} \right)$$

$$\varepsilon \approx -\frac{\omega_p^2}{\omega^2} \left(1 - \frac{i\gamma}{\omega} \right)$$

$$N = \sqrt{\varepsilon} \approx \sqrt{-\frac{\omega_p^2}{\omega^2} \left(1 - \frac{i\gamma}{\omega} \right)} \approx i \frac{\omega_p}{\omega} \left(1 - \frac{i\gamma}{2\omega} \right) = \frac{\omega_p}{2\omega} \frac{\gamma}{\omega} + i \frac{\omega_p}{\omega}$$

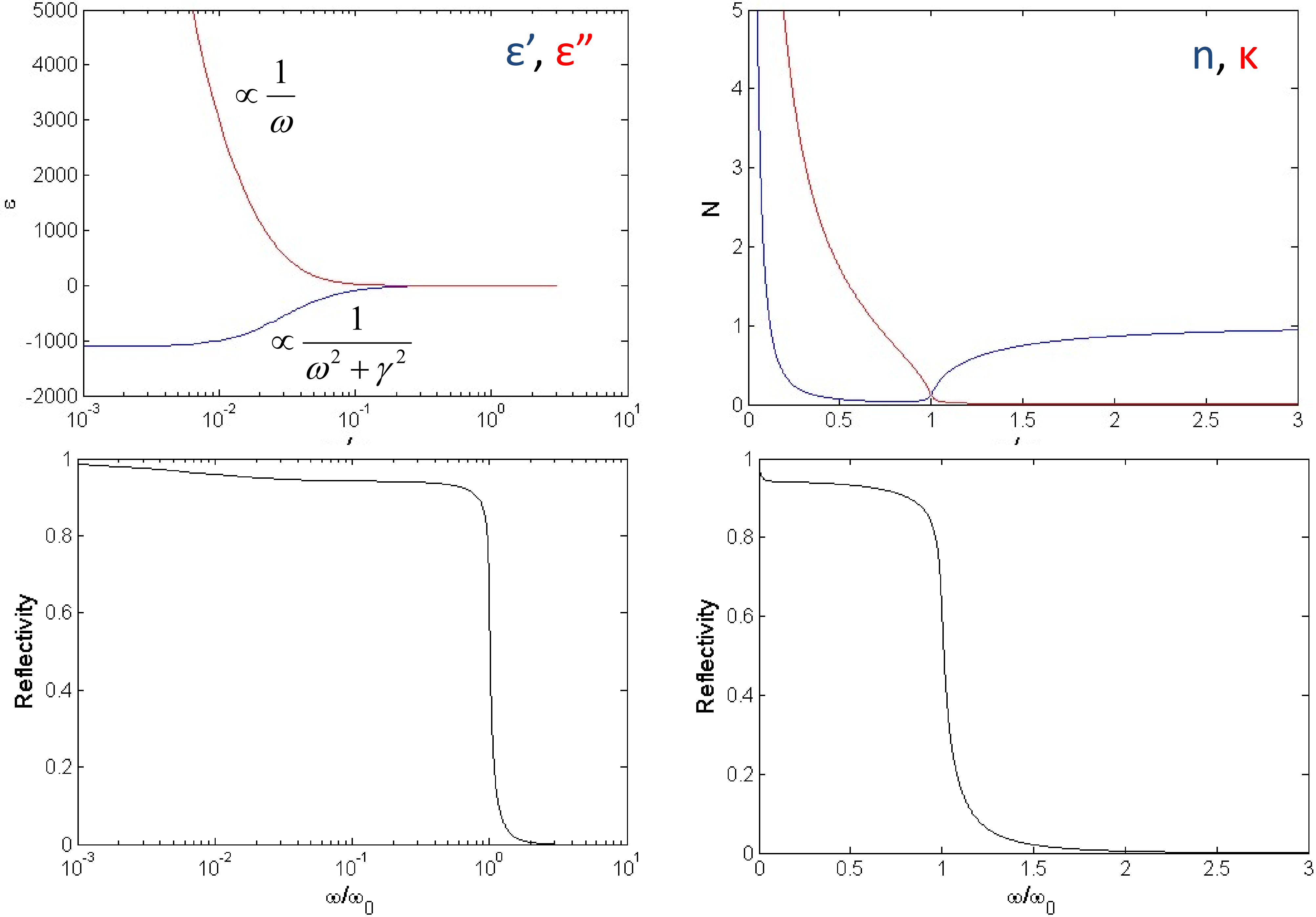
As $n << \kappa$

$$R = \left| \frac{1 - N}{1 + N} \right|^2 = \frac{(1-n)^2 + \kappa^2}{(1+n)^2 + \kappa^2} = \frac{1 + \frac{(1-n)^2}{\kappa^2}}{1 + \frac{(1+n)^2}{\kappa^2}} \approx \left(1 + \frac{(1-n)^2}{\kappa^2} \right) \left(1 - \frac{(1+n)^2}{\kappa^2} \right) \approx 1 - \frac{4n}{\kappa^2}$$

Frequency independent reflectivity:

$$\boxed{R \approx 1 - \frac{2\gamma}{\omega_p}}$$

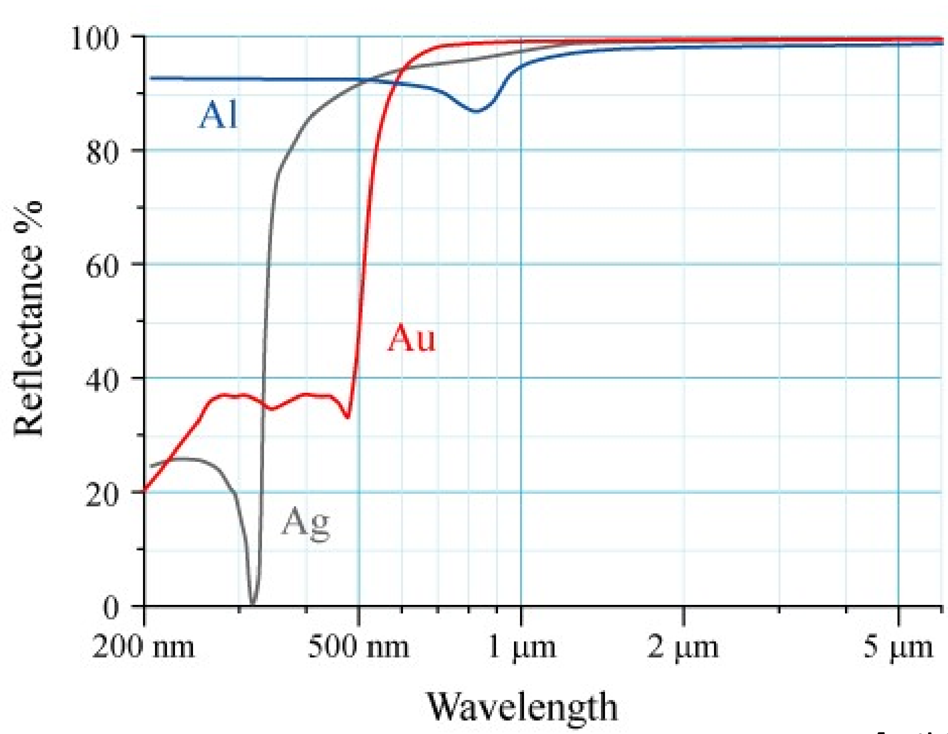
Reflectivity of metals in the Drude model



Reflectivity of metals in real life

„All that glisters is not gold” - William Shakespeare, Merchant of Venice

... but it may have free electrons!



[wikipedia]

[investopedia]

Human vision: 400-700 nm (2-3 eV)

Classical model of bound charges: Lorentz oscillator model

Assumptions in the Lorentz model:

- electrons obey Newton equation
  1. restoring force
  2. damping proportional to velocity
  3. external field accelerates
- oscillators are independent (dilute limit)
- parameters τ, m, q, n, D

$$m\ddot{x} = -Dx - m\frac{1}{\tau}\dot{x} + qE$$

$$\omega_0 = \sqrt{\frac{D}{m}} \qquad \gamma = \frac{1}{\tau}$$

$$\left((-i\omega)^2 + (-i\omega)\gamma + \omega_0^2\right)x = \frac{q}{m}E$$

$$x = \frac{q}{m}\frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}\cdot E$$

Polarization response to the driving field, E:

$$P = nqx = \boxed{n\frac{q^2}{m}\frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}}\,E$$

Charge susceptibility in the Lorentz oscillator model:

$$\boxed{\chi = \frac{nq^2}{\varepsilon_0 m}\frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}}$$

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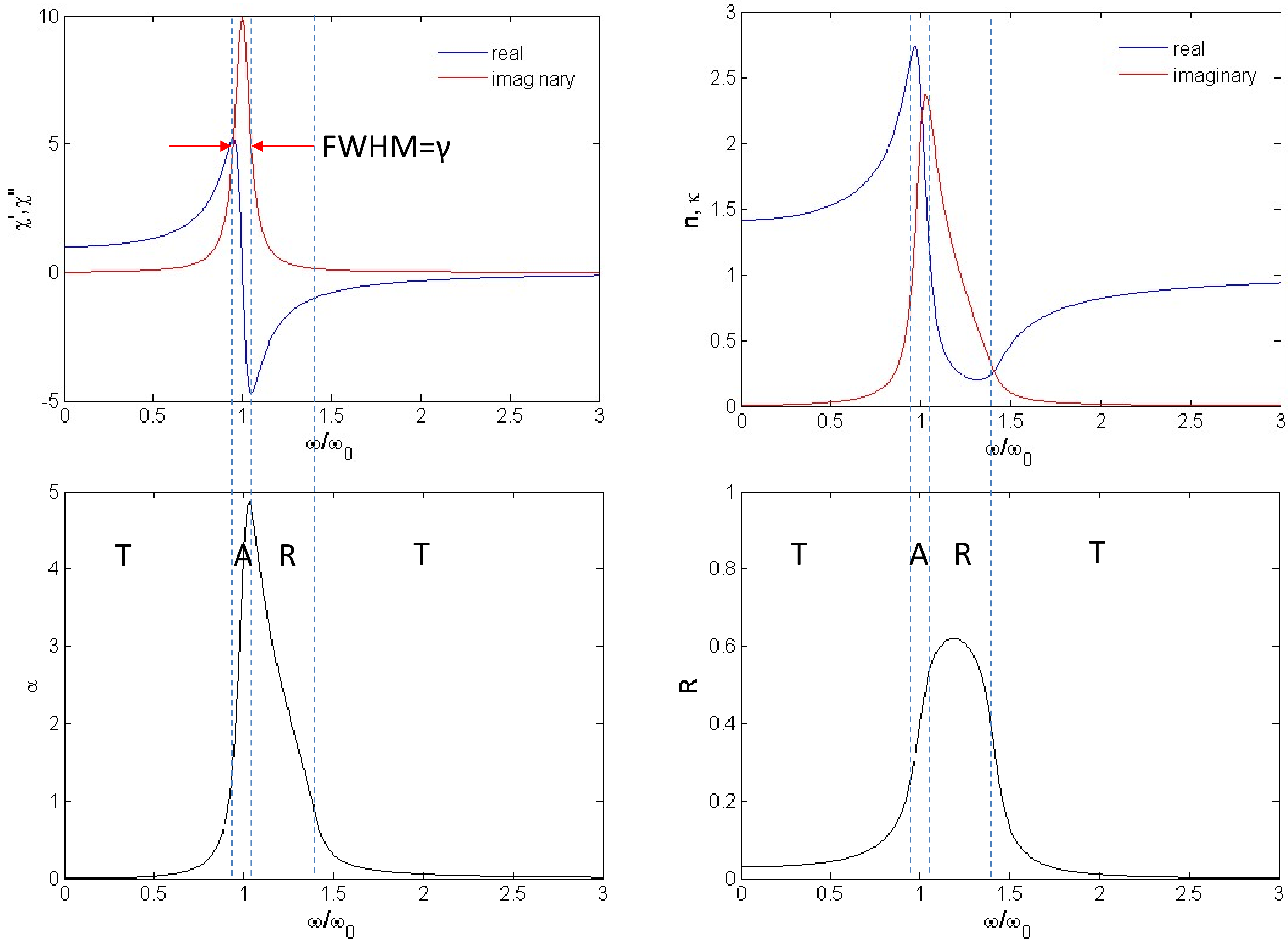
Polarization response to the driving field, E:

$$P = nqx = \boxed{n\frac{q^2}{m}\frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}}\,E$$

Dielectric response in the Lorentz oscillator model:

$$\boxed{\varepsilon = \varepsilon_\infty + \frac{nq^2}{\varepsilon_0 m}\frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}}$$

Classical model of bound charges: Lorentz oscillator model



Classical model of bound charges: Lorentz oscillator model

Static dielectric response (under damped case $\gamma \ll \omega_0$):

$$\varepsilon(0) = \varepsilon_\infty + \frac{nq^2}{\varepsilon_0 m} \cdot \frac{1}{\omega_0^2} \qquad \omega_p = \sqrt{\frac{nq^2}{\varepsilon_0 m}}$$

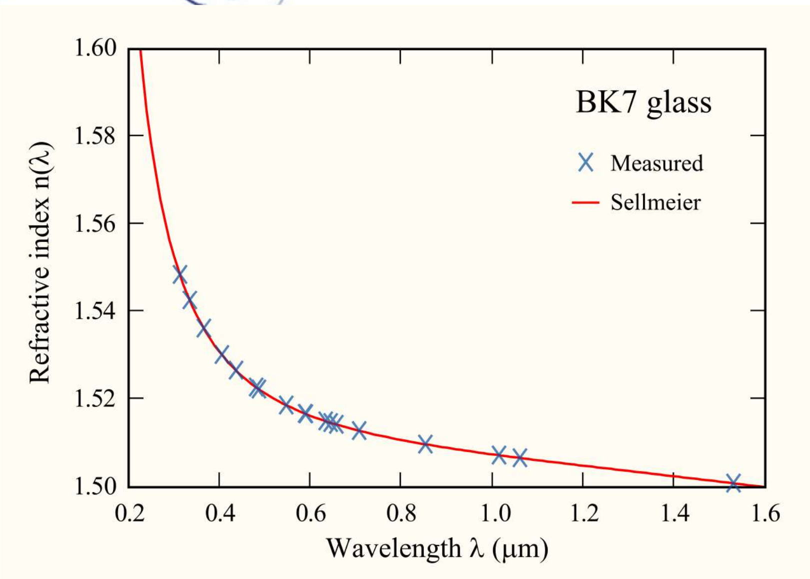
Low frequency limit, $\omega \ll \omega_0$ (under damped case $\gamma \ll \omega_0$):

$$\chi = \frac{nq^2}{\varepsilon_0 m} \frac{\omega_0^2 - \omega^2 + i\omega\gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2} = \frac{nq^2}{\varepsilon_0 m\omega_0^2} \frac{1 - \left(\frac{\omega}{\omega_0}\right)^2 + i\frac{\omega\gamma}{\omega_0^2}}{\left[1 - \left(\frac{\omega}{\omega_0}\right)^2\right]^2 + \left(\frac{\omega\gamma}{\omega_0^2}\right)^2}$$

$$\chi \approx \frac{nq^2}{\varepsilon_0 m\omega_0^2} \frac{1}{1 - \left(\frac{\omega}{\omega_0}\right)^2}$$

Sellmeier equation:

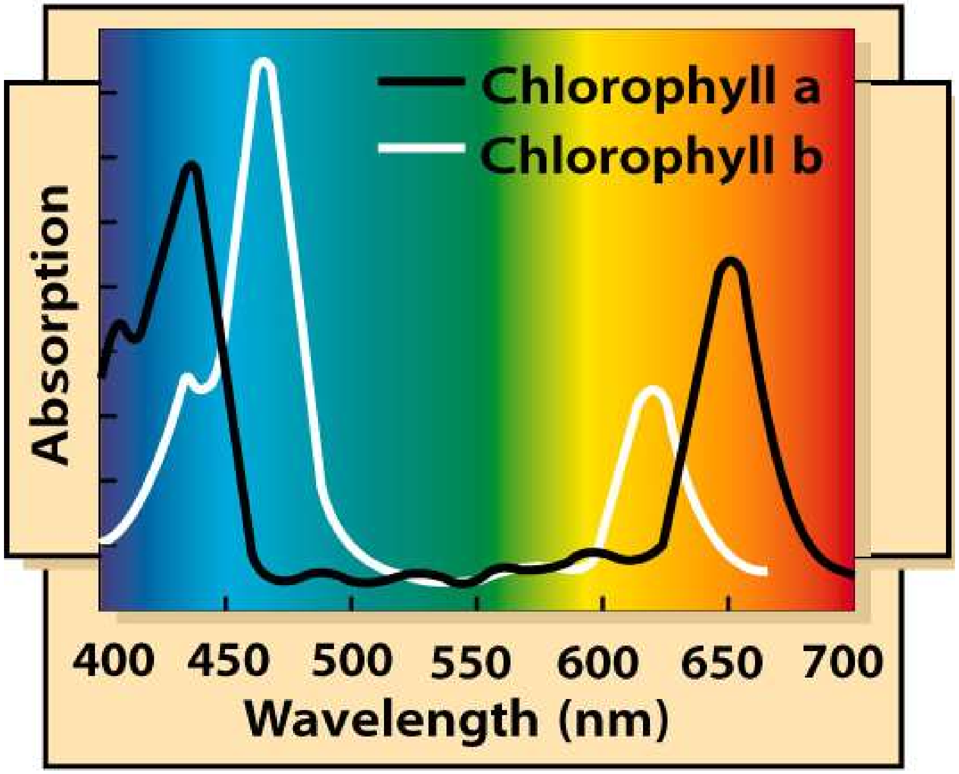
$$n^2(\lambda) = 1 + \sum_i \frac{B_i \lambda^2}{\lambda^2 - C_i}$$

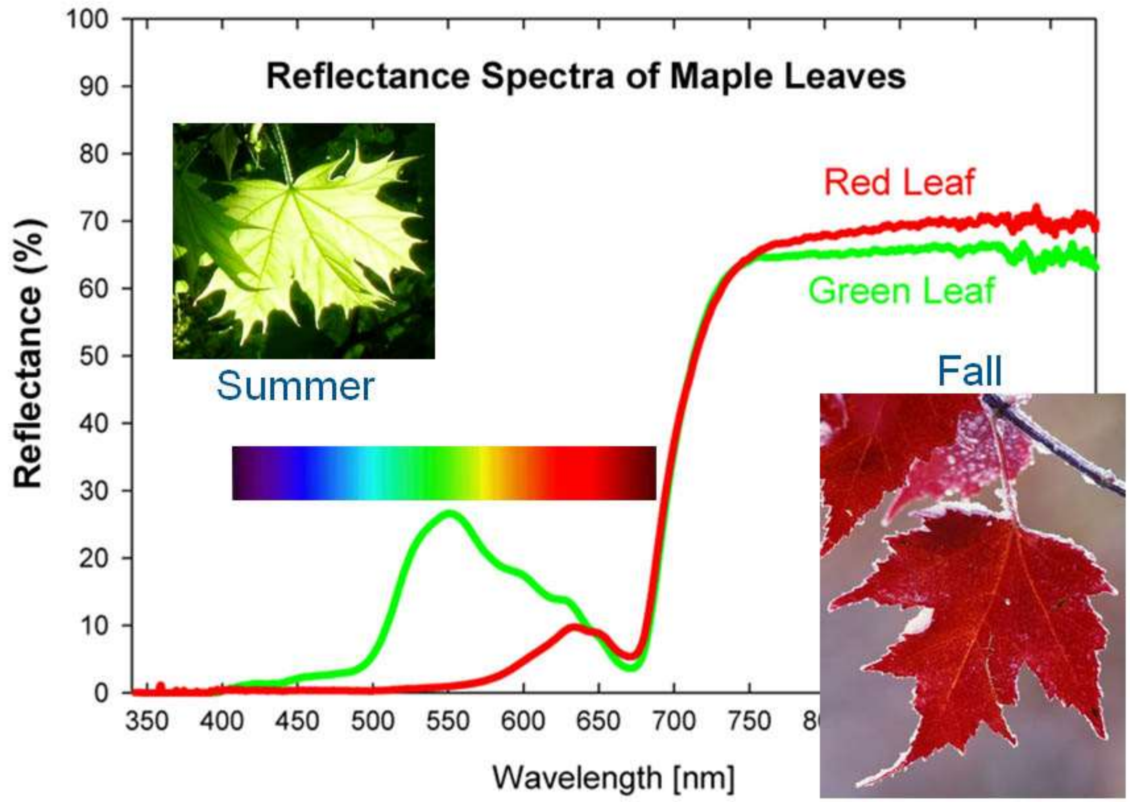


High frequency limit, $\omega \gg \omega_0$:

$$\varepsilon \approx \varepsilon_\infty + \frac{\omega_p^2}{\omega_0^2 - \omega^2} \qquad \varepsilon = 0 \Rightarrow \Omega_{pl}^2 = \omega_0^2 + \frac{\omega_p^2}{\varepsilon_\infty}$$

Applications of Lorentz oscillator model

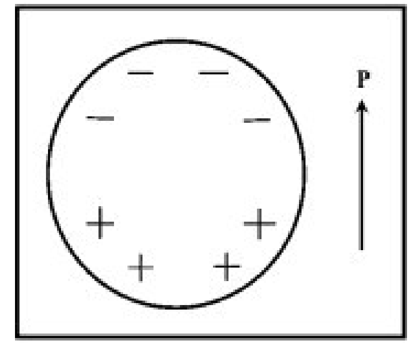




Lorentz oscillator model of phonons

Depolarizing fields in a dense material (e.g. crystal):

$$E_{loc} = E + E_{neighbor} + E_{medium}$$



Polarizing dipoles by local fields: $d = \alpha E_{loc}$

Field in a cavity: $$E_{medium} = \frac{P}{3\varepsilon_0}$$

For cubic crystals the sum of $E_{neigbor}$=0

$$P = nd = \frac{\frac{n\alpha}{\varepsilon_0}}{1 - \frac{n\alpha}{3\varepsilon_0}} \varepsilon_0 E \longrightarrow$$

Clausius-Mossotti relation:

$$\frac{n\alpha}{3\varepsilon_0} = \frac{\varepsilon - 1}{\varepsilon + 2}$$

$$\alpha = \frac{q^2}{m} \frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}$$

$$\chi = \frac{\omega_p^2}{\omega_0^2 - \frac{1}{3}\omega_p^2 - \omega^2 - i\omega\gamma} = \frac{\omega_p^2}{\omega_{d0}^2 - \omega^2 - i\omega\gamma}$$

Resonance frequency smaller due to local field corrections:

$$\boxed{\omega_{d0} = \sqrt{\omega_0^2 - \frac{1}{3}\omega_p^2}}$$

Lorentz oscillator model of phonons

Classical equation of motions for phonons:

$$\mu\ddot{x} = -\mu\omega_0 x - \mu\gamma\dot{x} + ZeE_{loc}$$

where μ is the reduced mass, Z is the effective ionic charge

Oscillator model for a phonon:

$$\boxed{\varepsilon = \varepsilon_\infty + \frac{\omega_p^2}{\omega_T^2 - \omega^2 - i\omega\gamma}}$$

oscillator strength

$$\omega_p = \sqrt{\frac{nZ^2e^2}{\varepsilon_0\mu}} = S\omega_T$$

$$\omega_T = \sqrt{\omega_0^2 - \frac{1}{3}\omega_p^2}$$

(phonon frequencies are renormalized in an ionic crystal)

Longitudinal (~'plasma mode') mode at ε=0

$$\omega_L^2 = \frac{\omega_p^2}{\varepsilon_\infty} + \omega_T^2$$

$$\varepsilon = \varepsilon_\infty \frac{\omega_L^2 - \omega^2 - i\omega\gamma}{\omega_T^2 - \omega^2 - i\omega\gamma} \xrightarrow[\omega \to 0]{}$$

Lyddane-Sachs-Teller

$$\boxed{\varepsilon = \varepsilon_\infty \frac{\omega_L^2}{\omega_T^2}}$$

Lorentz oscillator model of phonons

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$$\omega_p = \sqrt{\frac{nZ^2e^2}{\varepsilon_0 \mu}}$$

$$\omega_T = \sqrt{\omega_0^2 - \frac{1}{3}\omega_p^2}$$

(phonon frequencies are renormalized in an ionic crystal)

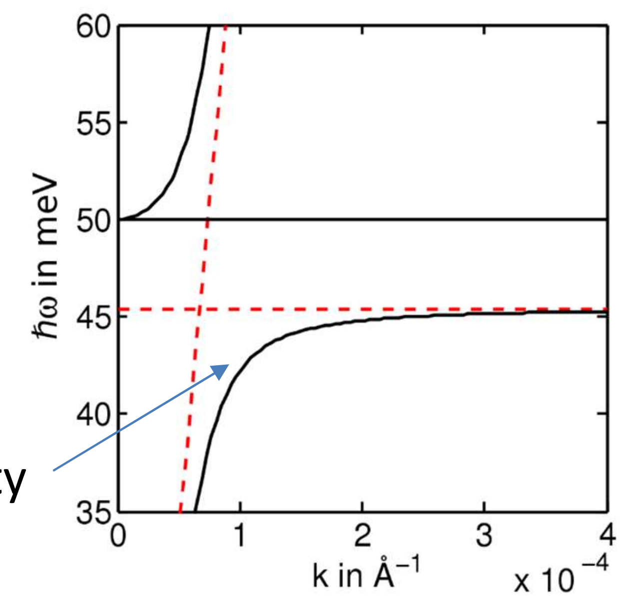
Multiple IR active phonon modes:

$$\varepsilon = \varepsilon_\infty + \sum_j \frac{\omega_{pj}^2}{\omega_{Tj}^2 - \omega^2 - i\omega\gamma_j}$$

Polaritons:
coupled polarization and electromagnetic wave

$$q = \frac{\omega}{c}\sqrt{\varepsilon(\omega)}$$

reduced group velocity



[wikipedia]

Free electrons in a magnetic field