Part 3 :

- Light matter interactions in terms of response functions

- Scattering theory :
  - Applications and basics of Mie theory
  - Electric polarizability theory for small scatterers
  - Radiation reactions

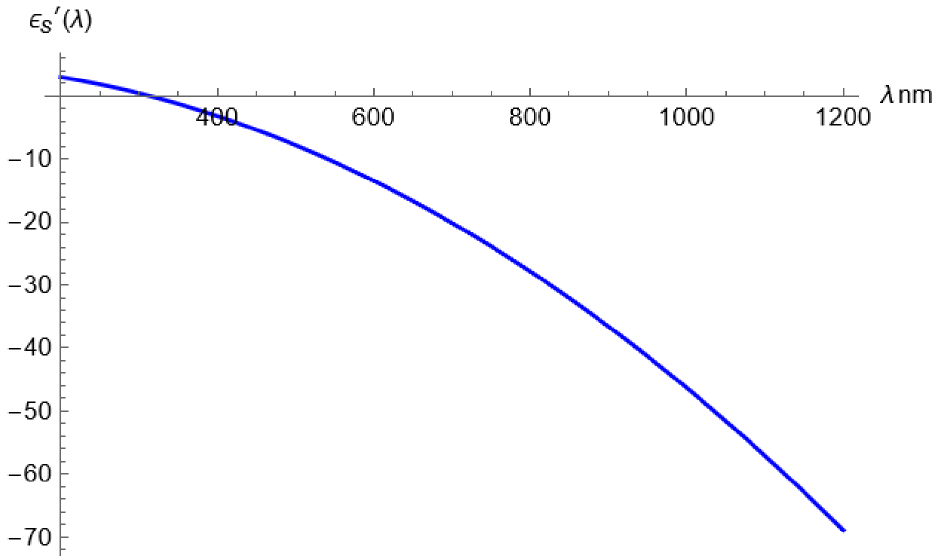
- Multipole theory :
  - Multipole basis functions
  - Expansions of Green’s functions
  - Purcell factor/Decay rate modifications

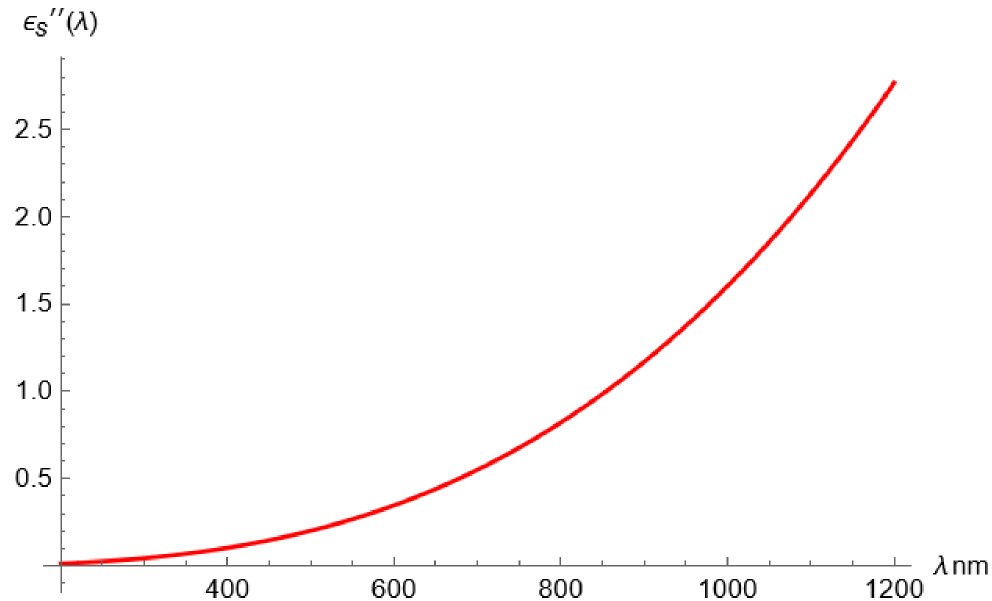
Drude-Lorentz model of material media

Frequency *dispersion* of permittivity

$$\varepsilon(\omega) = \varepsilon_{\mathrm{DC}} - \frac{\omega_{\mathrm{pl}}^2}{\omega(\omega + i\Gamma)} - \sum_{\alpha=1}^{N} \frac{\omega_{\mathrm{pT},\alpha}^2}{\omega^2 + 2i\gamma_\alpha \omega - \omega_{T,\alpha}^2}$$

$$\varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega)$$



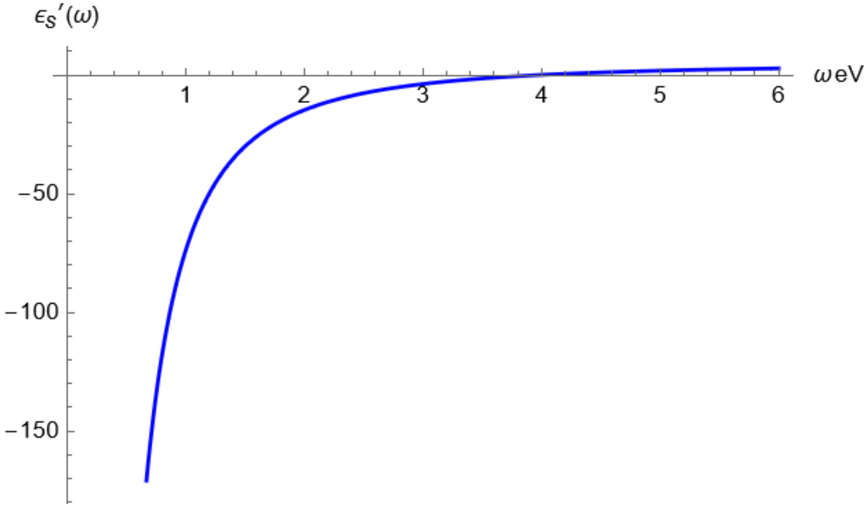


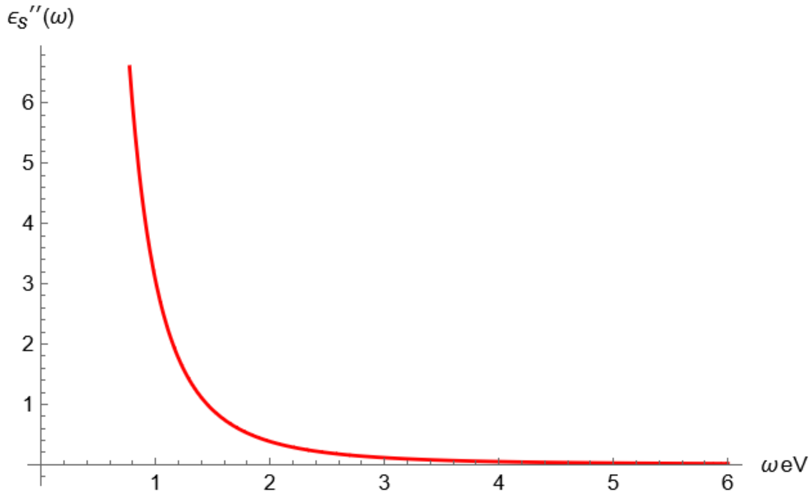
Drude model for the permittivity of silver (Ag)

Drude-Lorentz model of material media

$$\varepsilon(\omega) = \varepsilon_{\mathrm{DC}} - \frac{\omega_{\mathrm{pl}}^2}{\omega(\omega + i\Gamma)} - \sum_{\alpha=1}^{N} \frac{\omega_{\mathrm{pT},\alpha}^2}{\omega^2 + 2i\gamma_\alpha\omega - \omega_{T,\alpha}^2}$$

$$\varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega)$$





Drude model for the permittivity of silver (Ag)

Electric constitutive relations (adimensioned units)

$$\boldsymbol{D}(\boldsymbol{r},t) = \int_{-\infty}^{t} \overleftrightarrow{\varepsilon}(\boldsymbol{r},t-t')\cdot\boldsymbol{E}(\boldsymbol{r},t')dt'$$

Homogeneous media

$$\boldsymbol{D}(\boldsymbol{r},t) = \int_{-\infty}^{t} \overleftrightarrow{\varepsilon}(t-t')\cdot\boldsymbol{E}(\boldsymbol{r},t')dt'$$

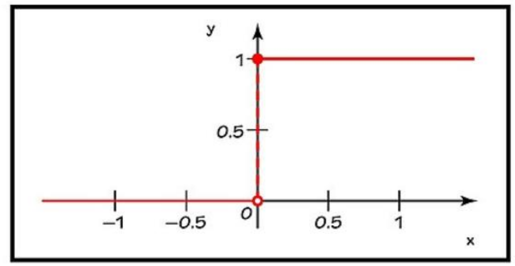
Isotropic media

$$\boldsymbol{D}(\boldsymbol{r},t) = \int_{-\infty}^{t} \varepsilon(t-t')\,\boldsymbol{E}(\boldsymbol{r},t')dt'$$

Convolution integral
(with Heaviside $\Theta(t)$)

$$\boldsymbol{D}(\boldsymbol{r},t) = \int_{-\infty}^{\infty} \Theta(t)\varepsilon(t-t')\,\boldsymbol{E}(\boldsymbol{r},t')dt'$$

$\Theta(t)$



The Heaviside Function

Fourier transform

$$\tilde{\varepsilon}(\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t}\Theta(t)\varepsilon(t) \qquad \widetilde{\boldsymbol{D}}(\boldsymbol{r},\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t}\boldsymbol{D}(\boldsymbol{r},t) \qquad \widetilde{\boldsymbol{E}}(\boldsymbol{r},\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t}\boldsymbol{E}(\boldsymbol{r},t)$$

$$\boldsymbol{D}(t) = \int_{-\infty}^{\infty} \Theta(t)\varepsilon(t-t')\,\boldsymbol{E}(\boldsymbol{r},t')dt' \;\Rightarrow\; \widetilde{\boldsymbol{D}}(\boldsymbol{r},\omega) = \tilde{\varepsilon}(\omega)\widetilde{\boldsymbol{E}}(\boldsymbol{r},\omega) = \{1+\tilde{\chi}_e(\omega)\}\widetilde{\boldsymbol{E}}(\boldsymbol{r},\omega)$$

Susceptibility : $\tilde{\chi}_e(\omega)$ $\quad \tilde{\varepsilon}(\omega) = 1 + \tilde{\chi}_e(\omega)$

Fourier transform

$$\tilde{\chi}_e(\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t}\Theta(t)\chi_e(t)$$

$$\tilde{\boldsymbol{D}}(\boldsymbol{r},\omega) = \tilde{\varepsilon}(\omega)\tilde{\boldsymbol{E}}(\boldsymbol{r},\omega) = \{1 + \tilde{\chi}_e(\omega)\}\tilde{\boldsymbol{E}}(\boldsymbol{r},\omega)$$

$\chi_e(t)$ is a real − valued function $\Rightarrow$ $\tilde{\chi}_e(-\omega^*) = \tilde{\chi}_e^*(\omega)$

Inverse Fourier transform

$$\chi_e(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega\, e^{-i\omega t}\tilde{\chi}_e(\omega)$$

$$\boldsymbol{E}(\boldsymbol{r},t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, e^{-i\omega t}\tilde{\boldsymbol{E}}(\boldsymbol{r},\omega)$$

Time harmonic formalism : $\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{E}_0(\boldsymbol{r})\cos(\omega t - \varphi) = \frac{1}{2}\left\{\boldsymbol{E}_0(\boldsymbol{r})e^{i\varphi}e^{-i\omega t} + \boldsymbol{E}_0(\boldsymbol{r})e^{-i\varphi}e^{i\omega t}\right\}$

$$= \mathrm{Re}\left\{\boldsymbol{E}_0(\boldsymbol{r})e^{i\varphi}e^{-i\omega t}\right\}$$

Magnetic constitutive relations

(adimensioned units)

$$\boldsymbol{B}(\boldsymbol{r},t) = \int_{-\infty}^{t} \overleftrightarrow{\mu}(\boldsymbol{r},t-t')\cdot\boldsymbol{H}(\boldsymbol{r},t')dt'$$

Homogeneous media

$$\boldsymbol{B}(\boldsymbol{r},t) = \int_{-\infty}^{t} \overleftrightarrow{\mu}(t-t')\cdot\boldsymbol{H}(\boldsymbol{r},t')dt'$$

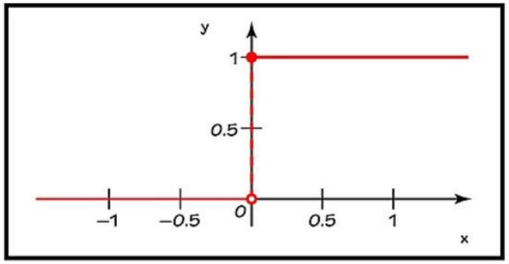
Isotropic media

$$\boldsymbol{B}(\boldsymbol{r},t) = \int_{-\infty}^{t} \mu(t-t')\,\boldsymbol{H}(\boldsymbol{r},t')dt'$$

Convolution integral
(with Heaviside $\Theta(t)$)

$$\boldsymbol{B}(\boldsymbol{r},t) = \int_{-\infty}^{\infty} \Theta(t)\mu(t-t')\,\boldsymbol{H}(\boldsymbol{r},t')dt'$$

$\Theta(t)$



The Heaviside Function

Fourier transform

$$\tilde{\mu}(\omega) = \int_{-\infty}^{\infty} d\omega\, e^{i\omega t}\Theta(t)\mu(t) \qquad \widetilde{\boldsymbol{B}}(\boldsymbol{r},\omega) = \int_{-\infty}^{\infty} d\omega\, e^{i\omega t}\boldsymbol{B}(\boldsymbol{r},t) \qquad \widetilde{\boldsymbol{H}}(\boldsymbol{r},\omega) = \int_{-\infty}^{\infty} d\omega\, e^{i\omega t}\boldsymbol{H}(\boldsymbol{r},t)$$

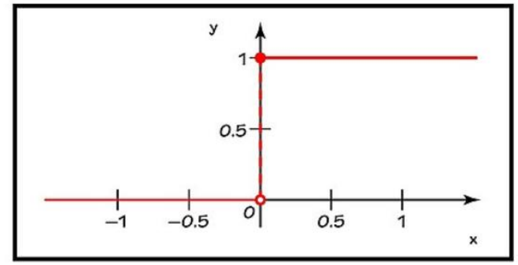
$$\widetilde{\boldsymbol{B}}(\boldsymbol{r},\omega) = \tilde{\mu}(\omega)\widetilde{\boldsymbol{H}}(\boldsymbol{r},\omega) = \{1+\tilde{\chi}_m(\omega)\}\widetilde{\boldsymbol{H}}(\boldsymbol{r},\omega)$$

In most materials at optical frequencies $\chi_m \sim 0$

Kramers-Krönig relations : causality

Theory of distributions

$$\Theta(\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t}\Theta(t) = \int_{0}^{\infty} dt\, e^{i\omega t}\Theta(t) = \pi\delta(\omega) + \mathrm{P.V.}\frac{i}{\omega}$$



The Heaviside Function

$\Theta(t)$

Causality: $\chi(t) = \chi(t)\Theta(t)$ $\Rightarrow$ $$\chi(\omega) = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi}\chi(\omega')\Theta(\omega-\omega')$$

$\Rightarrow$ $$\chi(\omega) = \frac{1}{i\pi}\mathrm{P.V.}\int_{-\infty}^{\infty}\frac{\chi(\omega')}{\omega'-\omega}d\omega'$$

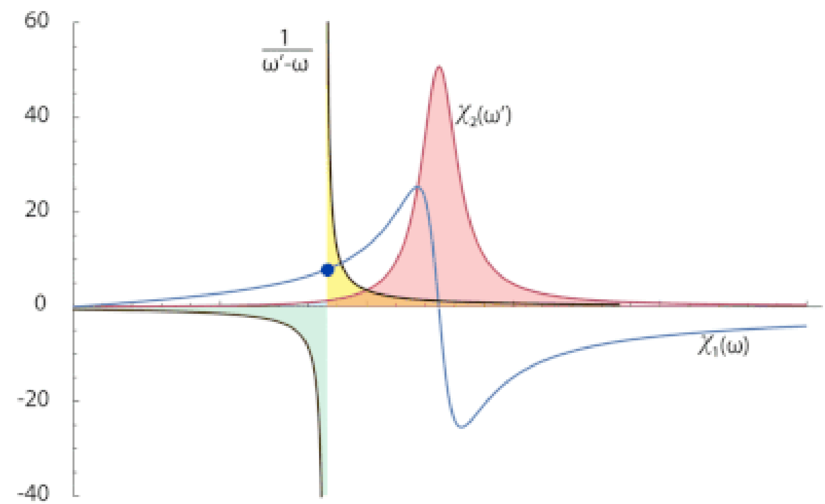
$$\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$$

Kramers-Krönig relations : causality

$$\varepsilon(\omega) = 1 + \chi(\omega) \qquad \chi(\omega) = \chi'(\omega) + i\chi''(\omega) = \chi_1(\omega) + i\chi_2(\omega)$$

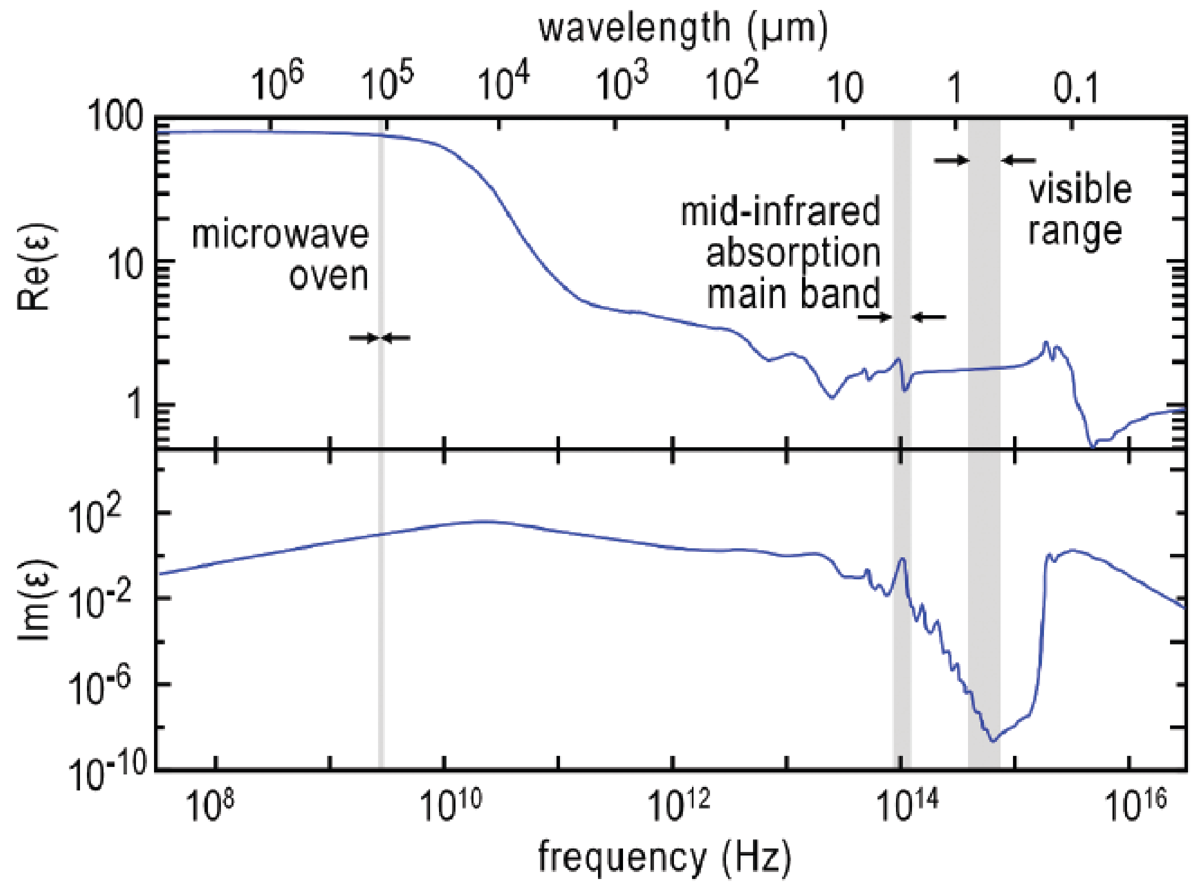
$$\chi'(\omega) = \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi''(\omega')}{\omega' - \omega}d\omega'$$

$$\chi''(\omega) = -\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi'(\omega')}{\omega' - \omega}d\omega'$$



$$\tilde{\chi}_e(-\omega^*) = \tilde{\chi}_e^*(\omega) \quad \Rightarrow \quad \begin{aligned} \chi'(\omega) &= \frac{2}{\pi}\mathcal{P}\int_0^{\infty}\frac{\omega'\chi''(\omega')}{\omega'^2 - \omega^2}d\omega' \\ \chi''(\omega) &= -\frac{2\omega}{\pi}\mathcal{P}\int_0^{\infty}\frac{\chi'(\omega')}{\omega'^2 - \omega^2}d\omega' \end{aligned}$$

Permittivity of actual materials is complicated

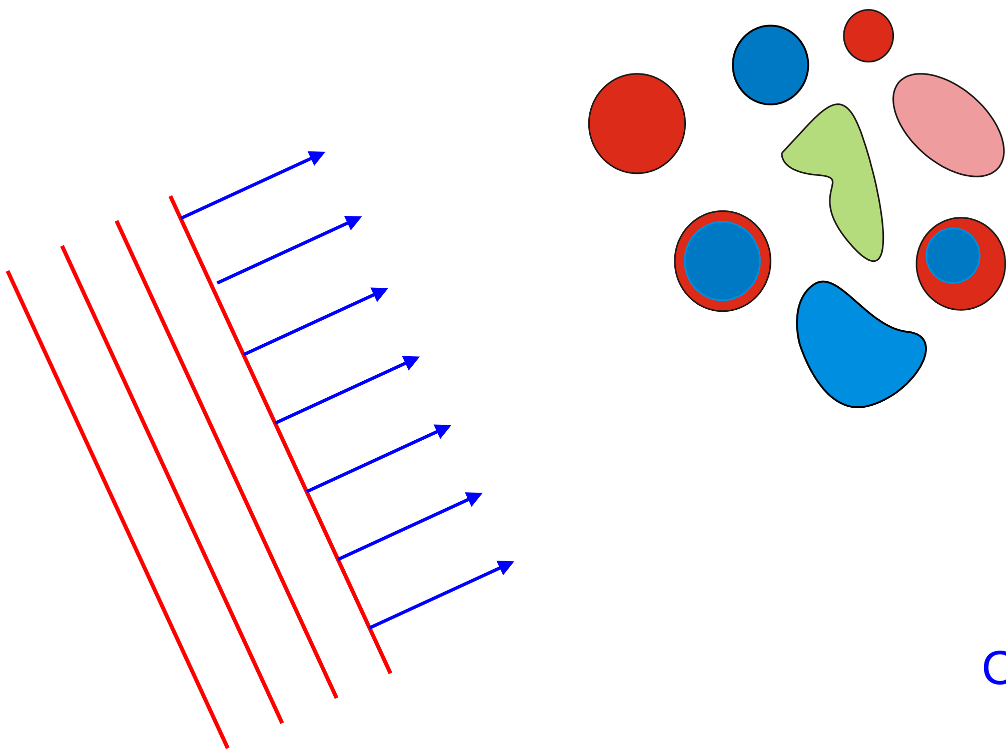


permittivity of water

How do simulate light scattering in the "real world" ?

"Seeing is believing and all we see is scattered light" J.C. Stover

Simplification : piecewise continuous media



Constitutive parameters

$$\boldsymbol{D}_\omega(\vec{\mathbf{r}}) = \overleftrightarrow{\varepsilon}(\omega) \cdot \boldsymbol{E}_\omega(\vec{\mathbf{r}})$$

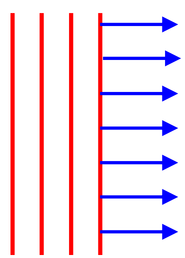
$$\boldsymbol{H}_\omega(\vec{\mathbf{r}}) = \overleftrightarrow{\mu}(\omega) \cdot \boldsymbol{B}_\omega(\vec{\mathbf{r}})$$

Scattering by a spherically symmetric object

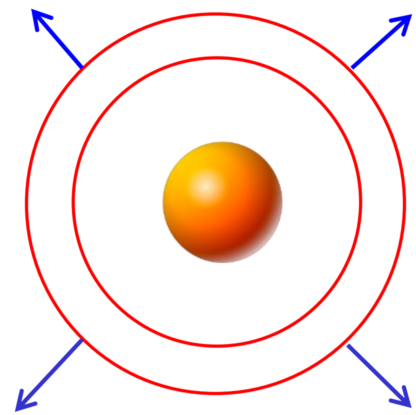
Lorenz - Mie - Debye theory

(1890) (1908) (1909)

Incident -`excitation' field :

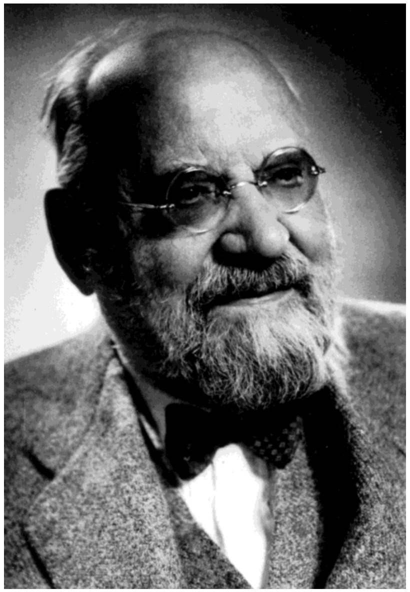


Outgoing scattered field



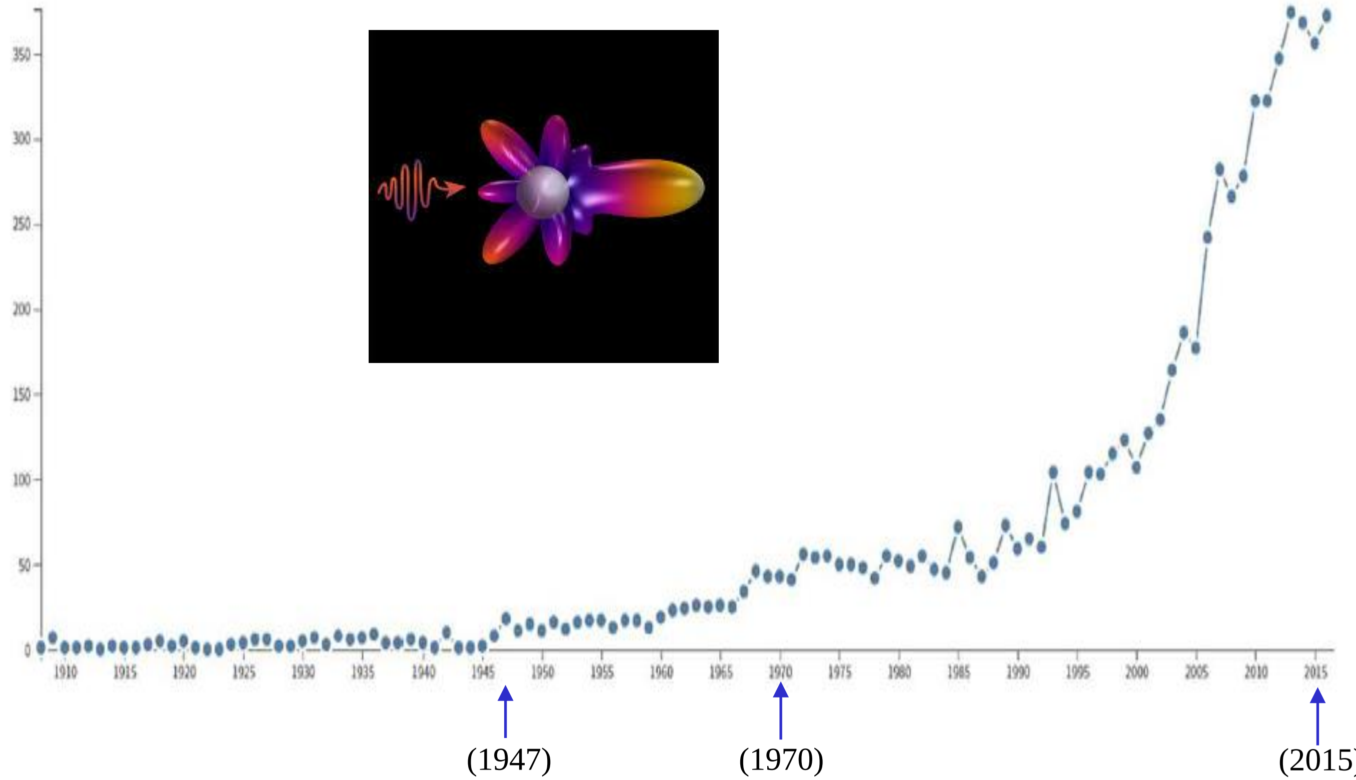
Exact solution !

Lorenz(1890)-Mie(1908)-Debye(1909) theory ?



Gustav Mie

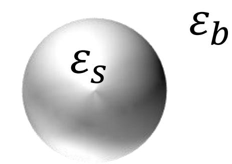
Citations per year



Gustav Mie (1868-1957) **"Contributions to the Optics of Turbid Media, particularly of colloidal metal solutions"**
Translation (Royal Aircraft Establishment (1976). **(1908)**

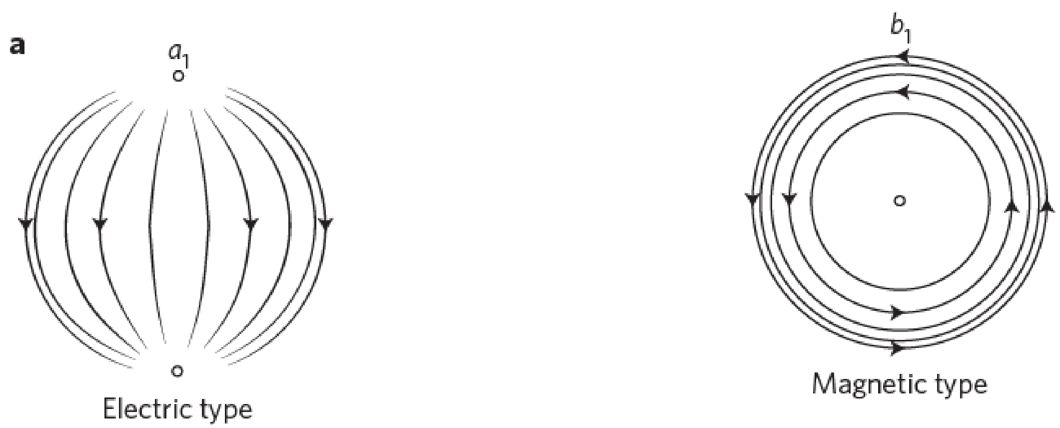
Ludvig Lorenz (1829–91) **"Light scattering and reflection by a transparent sphere (surface)"**
in Oeuvres scientifiques de **L. Lorenz**. 1898, p 403-529 **(1890)**.

Mie theory : multipole expansion



Dielectric scatterers are often called 'Mie resonators' but Mie theory is applicable to arbitrary temporal dispersion.
(In fact, Gustav Mie developed his theory to describe dispersive particles)

Fields expanded on the **multipolar** basis



Mie, G. *Ann. Phys.* **25,** 377 (1908).

A lot of physics is hidden in the Mie coefficients

Long winded to derive but easy to use!

$$a_n = \frac{\frac{\varepsilon_s}{\varepsilon_b} j_n(k_s R)\psi'_n(kR) - \psi'_n(k_s R) j_n(kR)}{\frac{\varepsilon_s}{\varepsilon_b} j_n(k_s R)\xi'_n(kR) - \psi'_n(k_s R) h_n(kR)}$$

$$b_n = \frac{\frac{\mu_s}{\mu_b} j_n(k_s R)\psi'_n(kR) - \psi'_n(kR) j_n(kR)}{\frac{\mu_s}{\mu_b} \psi_n(k_s R)\xi'_n(kR) - \psi'_n(k_s R) h_n(kR)}$$

$$\psi_n(x) \equiv x j_n(x)$$

$$\xi_n(x) \equiv x h_n(x)$$

$$\frac{k_s}{k} \equiv \frac{N_s}{N}$$

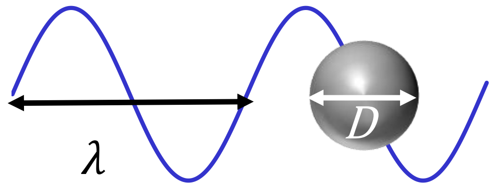
Cross sectios : σ

Extinction : $$\sigma_e = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \mathrm{Re}\{a_n + b_n\}$$

Scattering : $$\sigma_s = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1)(|a_n|^2 + |b_n|^2)$$

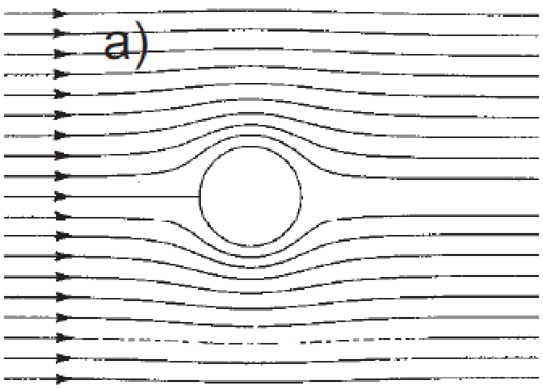
Absorption : $$\sigma_a = \sigma_e - \sigma_s$$

Interaction of light with subwavelength structures



$D < \lambda$

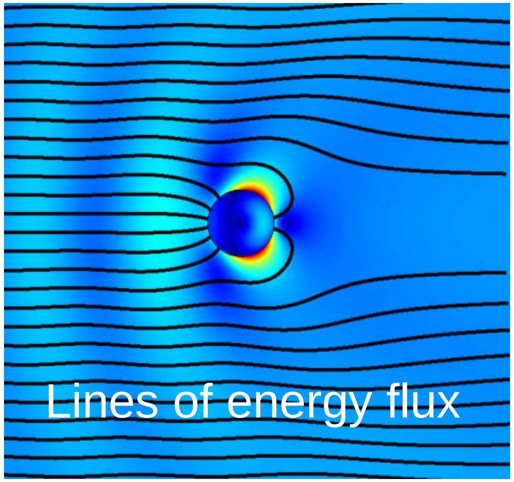
Weak interaction



Bohren, C. F., & Huffman, D. R. (2008). *Absorption and scattering of light by small particles*. John Wiley & Sons.

Extinction cross sections

Resonant optical interaction



$$\mathbf{S}_{\text{ext}} + \mathbf{S}_{\text{inc}} = \mathbf{S}_{\text{tot}} - \mathbf{S}_{\text{scat}}$$

Mie theory from nano to milli-metric particles

Rayleigh scattering

$$\sigma_p = \sigma_e + \frac{4\pi}{k^2}\left[\frac{n(n+2)}{n+1}\mathrm{Re}\{a_n a_{n+1}^* + b_n b_{n+1}^*\} + \frac{2n+1}{n(n+1)}\mathrm{Re}\{a_n b_n^*\}\right]$$

Radiation pressure

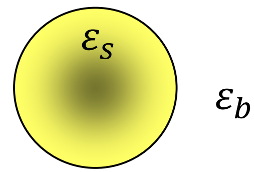
Glory – backscattering

Rainbows, clouds

Photonics : polarizability approach light interacting with a small material sphere)

Electric dipole moment and polarizability, $\alpha(\omega)$

$$\vec{\boldsymbol{p}} = \epsilon_0 \varepsilon_b \alpha(\omega) \vec{\boldsymbol{E}}_{\mathrm{exc}}$$



Quasi-static polarizability

$$\boxed{\alpha_0(\omega) \equiv \lim_{kR \to 0} \alpha(\omega) = 4\pi R^3 \frac{\varepsilon_s - \varepsilon_b}{\varepsilon_s + 2\varepsilon_b}}$$

Quasi-static limit of the electric dipole in Mie theory

$$a_n = -t_n^{(e)} \qquad b_n = -t_n^{(h)} \qquad x = kR$$

$$\lim_{x\to 0} a_n(x) = -\frac{i x^{2n+1}(n+1)}{(2n-1)!!\,(2n+1)!!}\,\frac{\frac{\varepsilon_s}{\varepsilon_b}-1}{\frac{\varepsilon_s}{\varepsilon_b}n+(n+1)}$$

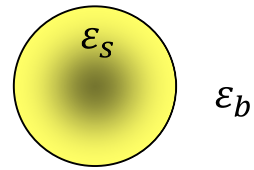
Electric dipole term (n=1) dominates for small particle sizes :

$$\lim_{kR\to 0} t_n^{(e)}(kR) \to i(kR)^3\,\frac{2}{3}\,\frac{\varepsilon_s-\varepsilon_b}{\varepsilon_s+2\varepsilon_b} = \frac{ik^3}{4\pi}\frac{2}{3}4\pi R^3\,\frac{\varepsilon_s-\varepsilon_b}{\varepsilon_s+2\varepsilon_b}$$
$$= \frac{ik^3}{4\pi}\frac{2}{3}\alpha(\omega)$$

Photonics : polarizability approach to cross section (case of a material sphere)

Electric dipole moment and polarizability, $\alpha(\omega)$

$$\boldsymbol{p} = \epsilon_0 \varepsilon_b \alpha(\omega) \boldsymbol{E}_{\mathrm{exc}}$$



$$k = \frac{2\pi}{\lambda_b} = \frac{\omega}{c_b}$$

$$\boxed{\alpha_0(\omega) \equiv \lim_{kR \to 0} \alpha(\omega) = 4\pi R^3 \frac{\varepsilon_s - \varepsilon_b}{\varepsilon_s + 2\varepsilon_b}}$$

$$P_{\mathrm{s,e,a}} = \sigma_{s,e,a} I_{\mathrm{inc}} \qquad I_{\mathrm{inc}} \propto \|\boldsymbol{E}_{\mathrm{inc}}\|^{\mathbf{2}}$$

Cross sections :

$$\sigma_{\mathrm{ext}} = k \mathrm{Im}\{\alpha(\omega)\} \qquad \sigma_{\mathrm{scat}} = \frac{k^4}{6\pi} |\alpha(\omega)|^2 \qquad \sigma_{\mathrm{abs}} \equiv \sigma_{\mathrm{ext}} - \sigma_{\mathrm{scat}}$$