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Optical functions: Kramers-Kronig relations

- linear response of matter to electromagnetic waves
- time and frequency domains
- dispersion, absorption, attenuation
- causality and complex macroscopic response functions, KK relations

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Electromagnetic waves

Harmonic oscillations at a fixed point in space,

$$\vec{E}(t) = \vec{E_0} e^{-i2\pi ft} = \vec{E_0} e^{-i\omega t}, \tag{2.1}$$

wavy pattern in space-time, like the plane wave propagating along the unit vector k_0:

$$\vec{E}(\vec{r},t) = \vec{E}_0 e^{-i(\omega t - 2\pi \vec{k}_0 \vec{r}/\lambda)} = \vec{E}_0 e^{-i(\omega t - \vec{k}\vec{r})}. \tag{2.2}$$

The basic characteristics:

electric intensity E (V/m),

frequency $f = \omega/2\pi$ (Hz), THz – PHz in the optical range, too high for the temporal dependencies to be detected

wavelength $\lambda = c/f$ (mm, μm, nm),

wavenumber $W = 1/\lambda$ (cm^{-1}).

quantum behavior (Planck + Einstein):

photon energy $\hbar\omega$ (eV),

momentum $\hbar\omega/c$ (eVs/m).

Electromagnetic waves

Wavelength - photon energy - wavenumber conversions:

$$\hbar\omega \text{ (eV)} = \frac{1.239852}{\lambda \text{ (µm)}}, \quad W \text{ (cm}^{-1}\text{)} = 8065.48\hbar\omega \text{ (eV)}. \tag{2.3}$$

The interaction governed by the electric field E of the wave; however, the signals measured by detectors are proportional to intensity, the time-averaged Poynting vector:

$$I = |\langle \vec{E}(t) \times \vec{H}(t) \rangle| = \frac{c\varepsilon_0}{2}|\vec{E}_0|^2. \tag{2.4}$$

A wave with the electric field amplitude of 1V/m has the intensity of 1.33 mW/m^2; a wave with the intensity of 1 mW/µm^2 has the amplitude of the electric intensity of 8.68×10^5 V/m.

This can be compared with the magnitude of the intensity of the field of one elementary charge at the distance of 0.1 nm, which is 1.44×10^{11} V/m.

Electromagnetic waves - quantum behavior

The classical picture of the electromagnetic wave fails on many occasions.
The flow of energy - a train of quanta (photons);
the intensity I and power P of a monochromatic beam are

$$I = \hbar\omega \times \text{(number of photons per units of area and time)},$$

$$P = \hbar\omega \times \text{(number of photons per unit of time)}.$$

Example: red HeNe laser line of λ=632.8 nm is composed of the 1.959 eV quanta. The power of 1 mW - the flux of 3.19×10^{18} photons per second.

State-of-the-art detectors operate at the dark noise level of about 10 elementary charges (that can be produced by a slightly higher number of photons) →
it is fairly easy to observe the linear response of matter (the number of absorbed photons proportional to the number of incoming photons).
Suggestion: find information on "photon-counting devices" (e.g. Hamamatsu).

Polarization of matter

The electric field of the light wave moves the atomic nuclei and electrons (or, change their quantum mechanical states);

the movements are typically asynchronous, with "phase shifts" between the external force (proportional to the electric intensity ***E***) and the induced oscillating dipole moments (or, equivalently, induced currents), with pronounced dependence on the frequency ω (which is called "dispersion");

complex numbers keep track of the amplitudes and phases

(ellipsometry provides two real numbers at each frequency);

in weak fields, the response is linear (***D*** is displacement, ***P*** is polarization, χ is susceptibility, ε is the relative permittivity, usually called "dielectric function", ***j*** is induced current, σ is conductivity;

the "response functions" χ, ε, σ are frequency-dependent, we neglect here the possible spatial dispersion);

the hat indicates possible anisotropy (the response functions need not be scalars) :

$$\vec{D} \equiv \varepsilon_0 \vec{E} + \vec{P} = (1 + \hat{\chi})\varepsilon_0 \vec{E} = \hat{\varepsilon}\varepsilon_0 \vec{E} \quad (2.5)$$

$$\vec{j} = -i\omega \vec{P} = \hat{\sigma}\vec{E} \quad (2.6)$$

SI units

D	(F/m)·(V/m)=C/m^2
P	C/m^2
j	A/m^2
ED	(V/m)·(A·s/m^2)=J/m^3
I	(V/m)·(A/m)= W/m^2
χ, ε	dimensionless
σ	(A/m^2)/(V/m)=1/(Ωm)

Optical "constants" and their relationships, see also (1.7.3)

Optical Constant (symbol)	Real part	Imaginary part
conductivity ($\sigma = \sigma_1 + i\sigma_2$)	$\sigma_1 = \omega\varepsilon_0\varepsilon_2$	$\sigma_2 = -\omega\varepsilon_0(\varepsilon_1 - 1)$
dielectric function ($\varepsilon = \varepsilon_1 + i\varepsilon_2$)	$\varepsilon_1 = 1 - \sigma_2/(\omega\varepsilon_0)$ $\varepsilon_1 = n^2 - k^2$	$\varepsilon_2 = \sigma_1/(\omega\varepsilon_0)$ $\varepsilon_2 = 2nk$
refractive index ($N = n + ik$)	$n = \sqrt{(\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2})/2}$ $n = \varepsilon_2/(2k)$	$k = \sqrt{(-\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2})/2}$ $k = \varepsilon_2/(2n)$
negative inverse of dielectric function ($-\varepsilon^{-1}$)	$-\varepsilon_1/(\varepsilon_1^2 + \varepsilon_2^2)$	$\varepsilon_2/(\varepsilon_1^2 + \varepsilon_2^2)$

(2.7)

The real part of conductivity (imaginary part of dielectric function) is a measure of absorbed energy (preferable in physical models);

the imaginary part of the refractive index is a measure of the attenuation of the light wave (preferable in solving the wave equation).

Suggestion: find the correspondence between the energy dissipated in an optical wave and the joule heat produced by a current flowing through a resistor.

Causality of the response - Kramers-Kronig (KK) relations

the optical functions are analytic in the whole upper half-plane of complex frequencies

Landau-Livshits, Electrodynamics of Continuous Media

→

integral relations between their real and imaginary parts on the real axis,

$$\varepsilon_1(\omega) - 1 = \frac{2}{\pi}\int_0^\infty \frac{\Omega\varepsilon_2(\Omega)}{\Omega^2-\omega^2}\mathrm{d}\Omega\ ,\quad \varepsilon_2(\omega) = \frac{\sigma_0}{\omega\epsilon_0} - \frac{2\omega}{\pi}\int_0^\infty \frac{\varepsilon_1(\Omega)-1}{\Omega^2-\omega^2}\mathrm{d}\Omega \qquad (2.8)$$

$$\sigma_1(\omega) = \frac{1}{\pi}\int_{-\infty}^\infty \frac{\sigma_2(\Omega)}{\Omega-\omega}\mathrm{d}\Omega = \frac{2}{\pi}\int_0^\infty \frac{\Omega\sigma_2(\Omega)}{\Omega^2-\omega^2}\mathrm{d}\Omega \qquad (2.9a)$$

$$\sigma_2(\omega) = \frac{-1}{\pi}\int_{-\infty}^\infty \frac{\sigma_1(\Omega)}{\Omega-\omega}\mathrm{d}\Omega = \frac{-2\omega}{\pi}\int_0^\infty \frac{\sigma_1(\Omega)}{\Omega^2-\omega^2}\mathrm{d}\Omega \qquad (2.9b)$$

$$n(\omega) - 1 = \frac{2}{\pi}\int_0^\infty \frac{\Omega k(\Omega)}{\Omega^2-\omega^2}\mathrm{d}\Omega\ ,\quad k(\omega) = \frac{-2\omega}{\pi}\int_0^\infty \frac{n(\Omega)-1}{\Omega^2-\omega^2}\mathrm{d}\Omega \qquad (2.10)$$

Suggestion: check the equivalence of the integration over the whole real axis and its positive part, such as in (2.9a & b).

An alternative to the contour integration in the derivation of KK relations;

let us do this for the susceptibility (after Hu, "Kramers–Kronig in two lines", Am. J. Phys. 57, 821 (1989)).

First, remember two auxiliary results concerning Fourier transform.

1. Fourier transform of the step function,

$$\bar{\theta}(t) = \begin{cases} 1 & \text{for } t > 0 \\ 0 & \text{for } t < 0 \end{cases} , \qquad (2.11)$$

is (in the limit of vanishing $\varepsilon > 0$)

$$\theta(\omega) = \int_{-\infty}^{\infty} \bar{\theta}(t) e^{i(\omega+i\varepsilon)t} dt = \left[\frac{1}{i(\omega+i\varepsilon)} e^{i(\omega+i\varepsilon)t} \right]_0^{\infty} = \frac{i(\omega-i\varepsilon)}{\omega^2+\varepsilon^2} = \pi\delta(\omega) + \frac{i}{\omega} \ .$$

$$(2.12)$$

Note: the real part with finite ε, except for the normalizing factor of $1/\pi$, is the probability density of Cauchy distribution (alternately called Breit-Wigner distribution), or the simplified Lorentzian profile.

2. The convolution theorem; for two functions, *f* and *g*, of time (frequency)

$$f(\omega)=\int_{-\infty}^{\infty}\bar{f}(t)e^{i\omega t}dt\ ,\ \bar{f}(t)=\frac{1}{2\pi}\int_{-\infty}^{\infty}f(\omega)e^{-i\omega t}d\omega\ ,$$

$$g(\omega)=\int_{-\infty}^{\infty}\bar{g}(t)e^{i\omega t}dt\ ,\ \bar{g}(t)=\frac{1}{2\pi}\int_{-\infty}^{\infty}g(\omega)e^{-i\omega t}d\omega\ , \qquad (2.13)$$

the <u>convolution</u> *h* of the two, in the frequency domain, can be obtained as the Fourier transform of their <u>product</u> in the time domain:

$$h(\omega)=\int_{-\infty}^{\infty}f(\omega-\omega')g(\omega')d\omega'=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\bar{f}(t)e^{i(\omega-\omega')t}\bar{g}(t')e^{i\omega't'}dtdt'd\omega'=$$

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\bar{f}(t)\bar{g}(t')e^{i(\omega-\omega')t}e^{i\omega't'}dtdt'd\omega'=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\bar{f}(t)\bar{g}(t')e^{i\omega t}2\pi\delta(t-t')dtdt'=$$

$$2\pi\int_{-\infty}^{\infty}\bar{f}(t)\bar{g}(t)e^{i\omega t}dt\ . \qquad (2.14)$$

Now, the “two lines” in deriving the KK relations:
the causal response requires

$$\bar{\chi}(t) = \bar{\theta}(t)\bar{y}(t) \text{ , where } \bar{y}(t) = \bar{\chi}(t) \quad \text{for } t > 0 \text{ .} \tag{2.15}$$

We are free to choose the values of the auxiliary function y for $t < 0$.
The Fourier transform of (2.15) is

$$\chi(\omega) = \int_{-\infty}^{\infty} \bar{\theta}(t)\bar{y}(t)e^{i\omega t}dt = \frac{1}{2\pi}\int_{-\infty}^{\infty} \theta(\omega-\omega')y(\omega')d\omega' =$$

$$\frac{1}{2\pi}\int_{-\infty}^{\infty}\left[\pi\delta(\omega-\omega')+\frac{i}{\omega-\omega'}\right]y(\omega')d\omega' =$$

$$\frac{y(\omega)}{2}+i\frac{1}{2\pi}P\int_{-\infty}^{\infty}\frac{y(\omega')}{\omega-\omega'}d\omega' \text{ .} \tag{2.16}$$

Here, P denotes the Cauchy principal value of the integral,
the same as integration symbols in (2.8-10).

Let us choose $y(t)$ even,

$\bar{y}(-|t|) = \bar{y}(|t|)$, producing $y(\omega)$ pure real. Then (2.16) implies

$$y(\omega) = 2\mathrm{Re}\{\chi(\omega)\} \ ,$$

$$\mathrm{Im}\{\chi(\omega)\} = -\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\mathrm{Re}\{\chi(\omega')\}}{\omega - \omega'} d\omega' \ . \qquad (2.17)$$

Let us choose $y(t)$ odd,

$\bar{y}(-|t|) = -\bar{y}(|t|)$, producing $y(\omega)$ pure imaginary. Then (2.16) implies

$$iy(\omega) = -2\mathrm{Im}\{\chi(\omega)\} \ ,$$

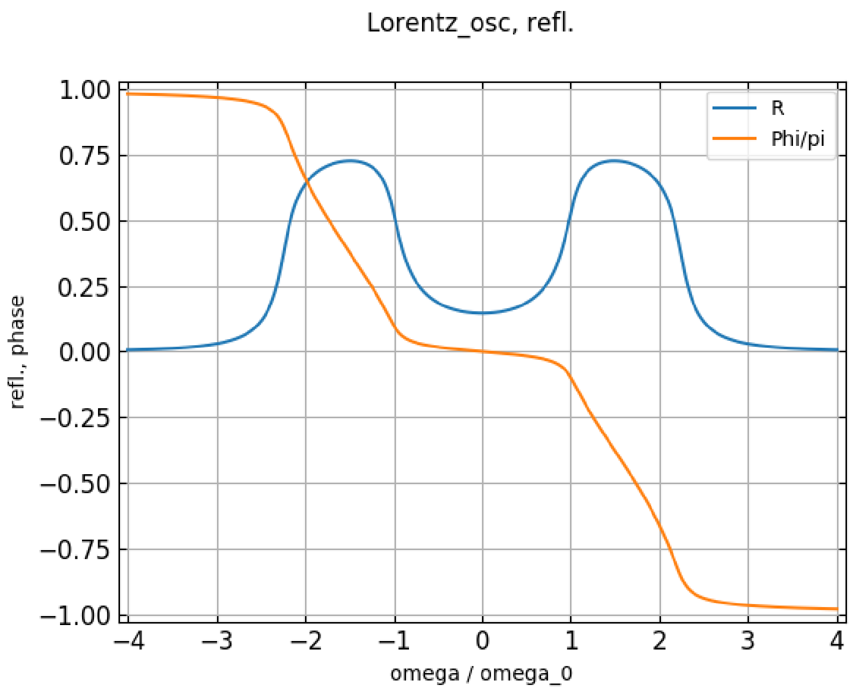
$$\mathrm{Re}\{\chi(\omega)\} = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\mathrm{Im}\{\chi(\omega')\}}{\omega - \omega'} d\omega' \ . \qquad (2.18)$$

We have arrived at the “two lines” of the KK relations, (2.17) and (2.18).

Some useful complex functions of the optical constants also satisfy KK relations. For example, the Fresnel amplitude reflectance *r* for the normal incidence at the interface with vacuum is

$$r = \sqrt{R} e^{i\Phi} = (n - 1 + ik)/(n + 1 + ik) \ ,$$

where *R* is the squared modulus of *r*, i.e., the power reflectivity.
The real and imaginary parts of ln(*r*), are evidently related by the KK relation between the phase $\Phi(\omega)$ and logarithm of reflectance $R(\omega)$.



Normal-incidence reflectivity and the phase angle of *r* (S=4, tau*omega_0=5).