Lecture 10: Non-local response

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Nonlocal response:

- in time (frequency dispersion)
  - ☞ Fourier transform
  - ☞ Causality, Kramers-Kronig relations
- in space (spatial dispersion)
  - ☞ Optical activity
    - phenomenological description
    - connection to the spatial dispersion

Consecutive relations

In the first lecture we have discussed the consecutive relations:

$$\boldsymbol{D} = \varepsilon \boldsymbol{E} = \varepsilon_0 \boldsymbol{E} + \boldsymbol{P}$$
$$\boldsymbol{P} = \varepsilon_0 \chi \boldsymbol{E}$$
$$\boldsymbol{B} = \mu \boldsymbol{H} = \mu_0 \boldsymbol{H} + \boldsymbol{M}$$

It is valid only if

- The electromagnetic wave is monochromatic
- The material shows no dispersion within spectrum of the wave
- The field vectors represent the spectral components of the wave:

$$\boldsymbol{D}(\omega) = \varepsilon(\omega) \boldsymbol{E}(\omega)$$
$$\boldsymbol{P}(\omega) = \varepsilon_0\, \chi(\omega) \boldsymbol{E}(\omega)$$

Response non-local in time

Let us study the meaning of these relations in the time domain:

$$\boldsymbol{D}(t) = \int_{-\infty}^{\infty} \boldsymbol{D}(\omega) e^{i\omega t} \, d\omega = \int_{-\infty}^{\infty} \varepsilon(\omega) \boldsymbol{E}(\omega) e^{i\omega t} \, d\omega$$

$$\tilde{\varepsilon}(t) = \int_{-\infty}^{\infty} \varepsilon(\omega) e^{i\omega t} \, d\omega$$

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

⇨ Fourier transform

The time-domain consecutive relation is a convolution of the field with the response function:

$$\boldsymbol{D}(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \, \boldsymbol{E}(\omega) \int_{-\infty}^{\infty} \tilde{\varepsilon}(t') e^{-i\omega(t'-t)} \, dt' = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt' \tilde{\varepsilon}(t') \int_{-\infty}^{\infty} d\omega \, \boldsymbol{E}(\omega) e^{-i\omega(t'-t)} =$$

$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} dt' \tilde{\varepsilon}(t') \boldsymbol{E}(t - t')$$

Fourier transform: definitions

The Fourier pairs of functions can have several possible definitions:

1. The natural conjugated variables are ν and *t*:

$$F(\nu)=\int_{-\infty}^{\infty} f(t)e^{-2\pi i\nu t}\,dt \qquad f(t)=\int_{-\infty}^{\infty} F(\nu)e^{2\pi i\nu t}\,d\nu$$

2. In optics we think usually in terms of the angular frequency $\omega = 2\pi\nu$:

$$F'(\omega)=\int_{-\infty}^{\infty} f(t)e^{-i\omega t}\,dt \qquad f(t)=\frac{1}{2\pi}\int_{-\infty}^{\infty} F'(\omega)e^{i\omega t}\,d\omega$$

3. In order to work with the spectral density in ω, we substitute: $F''(\omega) = F'(\omega)/2\pi$:

$$F''(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty} f(t)e^{-i\omega t}\,dt \qquad f(t)=\int_{-\infty}^{\infty} F''(\omega)e^{i\omega t}\,d\omega$$

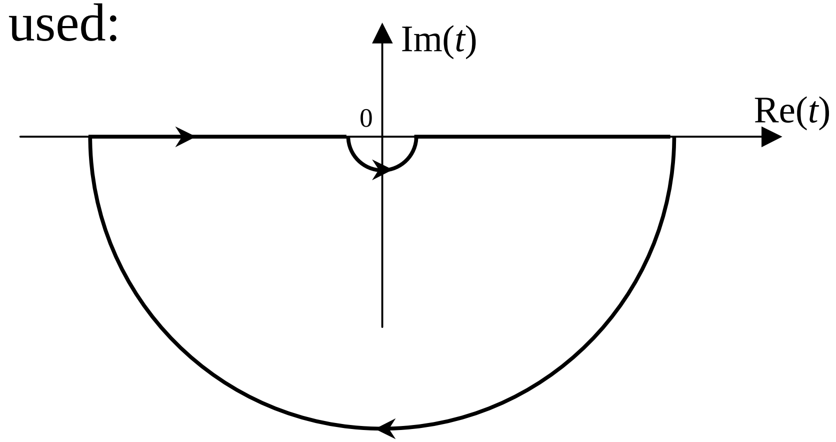
Properties of Fourier transform

Properties of FT			
function	Fourier transform		
	Definition 1	Definition 2	Definition 3
$f * g$	$F(\nu)\cdot G(\nu)$	$F'(\omega)\cdot G'(\omega)$	$2\pi F''(\omega)\cdot G''(\omega)$
$f \cdot g$	$F * G(\nu)$	$\frac{1}{2\pi} F' * G'(\omega)$	$F'' * G''(\omega)$
df/dt	$2\pi i\nu F(\nu)$	$i\omega F'(\omega)$	$i\omega F''(\omega)$
$\delta(t_0)$	$e^{-2\pi i\nu t_0}$	$e^{-i\omega t_0}$	$e^{-i\omega t_0}/2\pi$
$e^{-\alpha t^2}$	$\sqrt{\frac{\pi}{\alpha}}\, e^{-\frac{\pi^2\nu^2}{\alpha}}$	$\sqrt{\frac{\pi}{\alpha}}\, e^{i\frac{\omega^2}{4\alpha}}$	$\sqrt{\frac{1}{4\pi\alpha}}\, e^{-\frac{\nu^2}{4\alpha}}$
vp $1/t$	$-\pi i\, \mathrm{sign}(\nu)$	$-\pi i\, \mathrm{sign}(\omega)$	$-\frac{i}{2}\, \mathrm{sign}(\omega)$
sign t	$-\frac{i}{\pi}\, \mathrm{vp}\frac{1}{\nu}$	$-2i\, \mathrm{vp}\frac{1}{\omega}$	$-\frac{i}{\pi}\, \mathrm{vp}\frac{1}{\omega}$

Fourier pair vp(1/t) — sign(ω)

$$FT\left(\mathrm{vp}\frac{1}{t}\right)=\mathrm{vp}\int_{-\infty}^{\infty}\frac{\mathrm{e}^{-2\pi i\nu t}}{t}\,dt=\ldots$$

For ν > 0 the following contour is used:



$$\mathrm{vp}\int_{-\infty}^{\infty}\frac{e^{-2\pi i\nu t}}{t}\,dt$$

$$=-\pi i\,\mathrm{Res}_{t=0}\left(\frac{e^{-2\pi i\nu t}}{t}\right)=-\pi i$$

For ν < 0 a similar contour with Im(t) > 0 is used.

One finds the result: πi

$$FT\left(\mathrm{vp}\frac{1}{t}\right)=-\pi i\,\mathrm{sign}(\nu)$$

Causality

$$\boldsymbol{D}(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt' \tilde{\varepsilon}(t') \boldsymbol{E}(t - t')$$

In a dispersive material $\boldsymbol{D}(t_0)$ depends, in principle, on $\boldsymbol{E}(t \leq t_0)$, i.e. on all the previous values of the electric field.

D and ***P*** express the reaction of the matter to the applied field and should not depend on the future values of ***E***:

$$\boldsymbol{D}(t) = \frac{1}{2\pi} \int_{0}^{\infty} dt' \tilde{\varepsilon}(t') \boldsymbol{E}(t - t')$$

The above relations are equivalent if $\varepsilon(t < 0) = 0$: this relation will then automatically ensure the causality.

Causality: continued

Let us define:

$$\lim_{\omega\to\infty} \varepsilon(\omega) = \varepsilon_\infty \qquad \tilde{\varepsilon}_1(t) = \int_{-\infty}^{\infty} \left(\varepsilon(\omega) - \varepsilon_\infty\right) e^{i\omega t}\, d\omega$$

Then we can decompose the time response into 2 parts:

$$\tilde{\varepsilon}(t) = \tilde{\varepsilon}_1(t) + 2\pi\varepsilon_\infty\, \delta(0)$$

The causality condition is then fulfilled just when

$$\tilde{\varepsilon}_1(t) = \tilde{\varepsilon}_1(t)\,\mathrm{sign}(t)$$

Finally:

$$\tilde{\varepsilon}(t) = \tilde{\varepsilon}_1(t)\,\mathrm{sign}(t) + 2\pi\varepsilon_\infty\, \delta(0)$$

This is a general form of the response function which obeys the causality relation

Kramers-Kronig relations

The time-domain form of the response function

$$\tilde{\varepsilon}(t) = \tilde{\varepsilon}_1(t)\,\mathrm{sign}(t) + 2\pi\varepsilon_\infty\,\delta(0)$$

Leads to the Kramers-Kronig relations in the frequency domain:

$$\varepsilon(\omega_0) = (\varepsilon(\omega) - \varepsilon_\infty) * \left(-\frac{i}{\pi}\,\mathrm{vp}\,\frac{1}{\omega}\right) + \varepsilon_\infty = -\frac{i}{\pi}\,\mathrm{vp}\int_{-\infty}^{\infty} \frac{\varepsilon(\omega) - \varepsilon_\infty}{\omega_0 - \omega}\,d\omega + \varepsilon_\infty =$$

$$= \varepsilon_\infty - \frac{i}{\pi}\,\mathrm{vp}\int_{-\infty}^{\infty} \frac{\varepsilon(\omega)}{\omega_0 - \omega}\,d\omega$$

$$\varepsilon'(\omega_0) = \varepsilon_\infty - \frac{1}{\pi}\,\mathrm{vp}\int_{-\infty}^{\infty} \frac{\varepsilon''(\omega)\,d\omega}{\omega_0 - \omega}$$

$$\varepsilon''(\omega_0) = \frac{1}{\pi}\,\mathrm{vp}\int_{-\infty}^{\infty} \frac{\varepsilon'(\omega)\,d\omega}{\omega_0 - \omega}$$

$$n(\omega_0) = 1 - \frac{1}{\pi}\,\mathrm{vp}\int_{-\infty}^{\infty} \frac{\kappa(\omega)\,d\omega}{\omega_0 - \omega}$$

$$\kappa(\omega_0) = \frac{1}{\pi}\,\mathrm{vp}\int_{-\infty}^{\infty} \frac{n(\omega)\,d\omega}{\omega_0 - \omega}$$

Note

Which other physical phenomena are connected to vp — sign transformation?

Mathematicians thought useful to introduce so called Hilbert transformation which is closely related to that:

$$H(y) = y * \frac{i}{\pi} \mathrm{vp}\frac{1}{x}$$

$E(t)$, $e(\nu)$ is an electric field Fourier pair of an arbitrary waveform

$$e(\nu) = \int_{-\infty}^{\infty} E(t)\exp(-2\pi i\nu t)dt$$

Frequency components $e(\nu)$ within the pulse bandwidth acquire a constant phase change θ [strictly speaking: $\theta \times \mathrm{sign}(\nu)$ because $e(-\nu) = e^*(\nu)$]

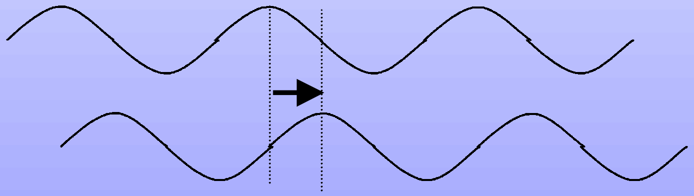
$$E'(t) = \int_{-\infty}^{\infty} e(\nu)\exp(i\theta\,\mathrm{sign}(\nu))\exp(2\pi i\nu t)d\nu$$

Note: continued

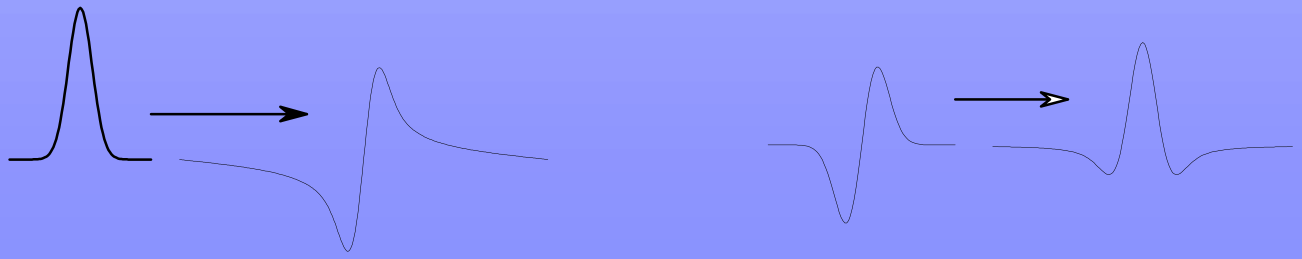
It means in the time domain: $$E'(t) = E(t) * \left[\cos\theta\, \delta(t) - \frac{\sin\theta}{\pi} \mathrm{vp}\frac{1}{t} \right]$$

In particular, for θ=π/2: $$E'(t) = -\frac{E(t)}{\pi} * \mathrm{vp}\frac{1}{t}$$

Monochromatic wave [cos(*t*) becomes sin(*t*)]:



Half-cycle or single-cycle pulse:



Optical activity: description of the phenomenon

Experimental fact: rotation of the polarization plane in some materials (quartz). This rotation can be right- or left-handed.

Angle of the rotation is proportional to the length of the sample: a specific rotation angle (per unit length) can be defined

Direction of the rotation is related to the propagation direction: the total rotation for a propagation back and forth is zero.

Phenomenologically, it can be described as a circular birefringence
Eigenmodes:

$$\boldsymbol{R}\, e^{i(\omega t - k_0 z\, n_R)} \qquad \boldsymbol{L}\, e^{i(\omega t - k_0 z\, n_L)}$$

R and ***L*** are the Jones vectors for the right and left circular polarizations

Optical activity: continued

At $z = 0$ (input face of the sample) the polarization of the beam is linear:

$$\frac{A}{\sqrt{2}} e^{i\omega t}(\boldsymbol{R}+\boldsymbol{L}) = \frac{A}{2} e^{i\omega t}\left[\begin{pmatrix} 1 \\ i \end{pmatrix} + \begin{pmatrix} 1 \\ -i \end{pmatrix}\right] = A\, e^{i\omega t}\begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

At $z = d$ (output face of the sample) the polarization writes:

$$\frac{A}{\sqrt{2}} e^{i\omega t}\left(\boldsymbol{R}\, e^{-ik_0 d\, n_R} + \boldsymbol{L}\, e^{-ik_0 d\, n_L}\right) =$$

$$= A e^{i\omega t - ik_0 d\,(n_R+n_L)/2}\begin{pmatrix} \cos\beta \\ \sin\beta \end{pmatrix}$$

It is a linear polarization in the direction given by the angle β:

$$\beta = k_0 d\,\frac{n_R - n_L}{2} = \pi\frac{d}{\lambda}(n_R - n_L)$$

Quartz at 546 nm: $n_e - n_o = 0.009$, $|n_R - n_L| = 8\times10^{-5}$.

Response non-local in space

An electric field in one place can produce a polarization in the near vicinity

$$\boldsymbol{D}(\boldsymbol{r}) = \frac{1}{(2\pi)^3} \iiint \tilde{\varepsilon}(\boldsymbol{r}') \boldsymbol{E}(\boldsymbol{r} - \boldsymbol{r}') d\boldsymbol{r}'$$

$$\boldsymbol{P}(\boldsymbol{r}) = \frac{\varepsilon_0}{(2\pi)^3} \iiint \tilde{\chi}(\boldsymbol{r}') \boldsymbol{E}(\boldsymbol{r} - \boldsymbol{r}') d\boldsymbol{r}'$$

The above response functions take the form of a Dirac δ in the local response approximation

In the reciprocal space the following relations are obtained

$$\boldsymbol{D}(\omega, \boldsymbol{k}) = \varepsilon(\omega, \boldsymbol{k}) \boldsymbol{E}(\omega, \boldsymbol{k})$$

$$\boldsymbol{P}(\omega, \boldsymbol{k}) = \varepsilon_0 \chi(\omega, \boldsymbol{k}) \boldsymbol{E}(\omega, \boldsymbol{k})$$

Dependence on ***k*** (namely on its direction): spatial dispersion

In the following we will search for the consequences of the spatial dispersion

Spatial dispersion

We take a first-order Taylor development of $\varepsilon(\boldsymbol{k})$: the linear term does not vanish in the non-centrosymmetric materials

$$\varepsilon_{ij}(\boldsymbol{k}) = \varepsilon_{ij}^{0} + \varepsilon_0 \sum_{l=1}^{3} \gamma_{ijl} \boldsymbol{k}_l$$

γ_{ijl} is a 3rd-rank tensor. Its intrinsic symmetry properties can be derived from those of the dielectric constant:

$$\varepsilon_{ij} = \varepsilon_{ji}^{*} \quad \text{and} \quad \varepsilon_{ij}(\boldsymbol{k}) = \varepsilon_{ij}^{*}(-\boldsymbol{k})$$

$$\gamma_{ijl} = i\delta_{ijl}$$

(γ_{ijl} is imaginary, δ_{ijl} is real)

$$\gamma_{iil} = 0$$

$$\delta_{ijl} = -\delta_{jil}$$

An antisymmetric tensor g_{ij} can be defined:

$$g_{ij} = \delta_{ijl} k_l$$

Spatial dispersion: continued

$$g_{ij} = \delta_{ijl} k_l \qquad \boldsymbol{g} = \begin{pmatrix} 0 & g_{12} & g_{13} \\ -g_{12} & 0 & g_{23} \\ -g_{13} & -g_{23} & 0 \end{pmatrix}$$

One can also introduce the gyration vector:

$$\boldsymbol{G} = \begin{pmatrix} g_{23} & -g_{13} & g_{12} \end{pmatrix}$$

The electric induction is then equal to:

$$D_i = \varepsilon_{ij}^0 E_j + i\varepsilon_0 g_{ij} E_j \qquad \boldsymbol{D} = \varepsilon \boldsymbol{E} + i\varepsilon_0 \boldsymbol{G} \wedge \boldsymbol{E}$$

Note: the tensor ***g*** is not characteristic for a medium, it is given for the medium and a specific wave vector. I.e. if we apply a symmetry operation (like a rotation or a mirror) on the tensor ***g*** the crystal sample turns consequently but the radiation wave vector turns as well!

Light waves in non-local media

We have to solve the wave equation:

$$\boldsymbol{s}(\boldsymbol{s}\cdot\boldsymbol{E}) - \boldsymbol{E} + \frac{1}{n^2}\varepsilon_r\cdot\boldsymbol{E} = 0 \qquad \left(\text{ or } \quad \boldsymbol{k}(\boldsymbol{k}\cdot\boldsymbol{E}) - k^2\boldsymbol{E} + \omega^2\mu_0\varepsilon\cdot\boldsymbol{E} = 0\right)$$

where the dielectric constant ε_r is replaced by:

$$\boldsymbol{\varepsilon} = \begin{pmatrix} \varepsilon_{xx}^0 & ig_{12} & ig_{13} \\ -ig_{12} & \varepsilon_{yy}^0 & ig_{23} \\ -ig_{13} & -ig_{23} & \varepsilon_{zz}^0 \end{pmatrix}$$

Let us study the case of the **quartz** (uniaxial crystal) which allows to discuss the most important features. The quartz has only two independent components of the 3rd-rank tensor δ.

$$\delta_{123} = -\delta_{213};\ \delta_{231} = \delta_{312} = -\delta_{321} = -\delta_{132}$$

Propagation // optic axis (//z)

$g_{13} = \delta_{123} k,\ g_{13} = 0,\ g_{23} = 0$

we define: $\Delta_3 = g_{12}\, c^2 / \omega^2$

Wave equation:

$$\begin{pmatrix} n_o^2 - n^2 & i\Delta_3 & 0 \\ -i\Delta_3 & n_o^2 - n^2 & 0 \\ 0 & 0 & n_e^2 \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = 0$$

Eigenvalues (effective refractive index):

$$n_{I,II} \approx n_o \pm \frac{\Delta_3}{2n_o}$$

Eigenvectors (polarization of the waves):

$$\begin{pmatrix} 1 \\ -i \\ 0 \end{pmatrix}, \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix}$$

Propagation ⊥ optic axis (//x)

$g_{23} = \delta_{231} k,\ g_{13} = 0,\ g_{12} = 0$

we define: $\Delta_1 = g_{23} c^2 / \omega^2$

Wave equation:

$$\begin{pmatrix} n_o^2 & 0 & 0 \\ 0 & n_o^2 - n^2 & i\Delta_1 \\ 0 & -i\Delta_1 & n_e^2 - n^2 \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = 0$$

As the birefringence is usually larger than the optical activity, we can assume:

$$\left| n_o^2 - n_e^2 \right| \gg \Delta_1$$

Propagation ⊥ optic axis (//x): continued

Wave equation:

$$\begin{pmatrix} n_o^2 & 0 & 0 \\ 0 & n_o^2 - n^2 & i\Delta_1 \\ 0 & -i\Delta_1 & n_e^2 - n^2 \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = 0$$

Eigenvalues:

$$n_I \approx n_o + \frac{\Delta_1}{2n_o} \frac{\Delta_1}{n_o^2 - n_e^2}$$

$$n_{II} \approx n_e - \frac{\Delta_1}{2n_e} \frac{\Delta_1}{n_o^2 - n_e^2}$$

Eigenvectors:

$$\begin{pmatrix} 0 \\ 1 \\ \dfrac{-i\Delta_1}{n_o^2 - n_e^2} \end{pmatrix}, \quad \begin{pmatrix} 0 \\ \dfrac{i\Delta_1}{n_o^2 - n_e^2} \\ 1 \end{pmatrix}$$