Solution to Problem Set 1
Nonlinear Optics (NLO)

Due: 10. May 2016

1) Refractive Index, Extinction Coefficient and Absorption

Express the real and imaginary part of the complex refractive index

$$\underline{n} = n - \mathrm{j}n_i \tag{0.1}$$

using the real and imaginary part of the complex susceptibility

$$\underline{\chi}^{(1)} = \chi + \mathrm{j}\chi_i . \tag{0.2}$$

Simplify the results in the case of low losses, $\chi_i << \chi$, and derive an expression for the power attenuation coefficient α, that is experienced by a plane wave in a homogeneous medium.

Solution

The complex refractive index $\underline{n}$ and the complex dielectric constant $\underline{\varepsilon}_r$ are related by $\underline{\varepsilon}_r = \underline{n}^2$. Using (0.1) we get the relations between real and imaginary part

$$\begin{aligned}\underline{\varepsilon}_r &= \varepsilon_r - \mathrm{j}\varepsilon_{ri} \\ &= \underline{n}^2 = (n - \mathrm{j}n_i)^2 \\ &= n^2 - n_i^2 - \mathrm{j}2nn_i\end{aligned} \tag{0.3}$$

The dielectric constant can itself be expressed by real and imaginary parts of the first order susceptibility

$$\begin{aligned}\underline{\varepsilon}_r &= 1 + \underline{\chi}^{(1)} \\ \varepsilon_r - \mathrm{j}\varepsilon_{ri} &= 1 + \chi + \mathrm{j}\chi_i\end{aligned} \tag{0.4}$$

By comparing the imaginary parts of (0.4) and inserting (0.3) we get

$$\begin{aligned}\varepsilon_{ri} &= 2nn_i = -\chi_i \\ n_i &= -\chi_i / (2n).\end{aligned} \tag{0.5}$$

Comparing the real part of (0.4) and substituting (0.3) we find a quadratic equation for n:

$$\begin{aligned}\varepsilon_r = 1 + \chi &= n^2 - n_i^2 \\ &= n^2 - \left(\frac{\chi_i}{2n}\right)^2 .\end{aligned} \tag{0.6}$$

Equation (0.6) can be solved for n by substituting $N = n^2$:

$$\begin{aligned}1+\chi &= n^2-\left(\frac{\chi_i}{2n}\right)^2\\ 0 &= N^2-N(1+\chi)-\left(\frac{\chi_i}{2}\right)^2\\ N_{1,2} &= \frac{(1+\chi)\pm\sqrt{(1+\chi)^2+\chi_i^2}}{2}\\ n &= \sqrt{\frac{(1+\chi)}{2}+\frac{\sqrt{(1+\chi)^2+\chi_i^2}}{2}}\end{aligned} \tag{0.7}$$

Since $n^2>0$, only $N_1>0$ is a meaningful solution. From Eq. (0.6) one can get the expression for imaginary part n_i by substituting the expression for n obtained in Eq. (0.7)

$$n_i=\sqrt{n^2-(1+\chi)}=\sqrt{-\frac{(1+\chi)}{2}+\frac{\sqrt{(1+\chi)^2+\chi_i^2}}{2}} \tag{0.8}$$

When the condition $|\chi|_i \ll |\chi|$ is fulfilled, the real and imaginary parts of the complex refractive index $\underline{n}$ can be approximated as

$$n=\sqrt{\frac{(1+\chi)}{2}+\frac{(1+\chi)}{2}\sqrt{\left(1+\frac{\chi_i^2}{(1+\chi)^2}\right)}}\approx\sqrt{1+\chi} \tag{0.9}$$

$$\begin{aligned}n_i &= \sqrt{-\frac{(1+\chi)}{2}+\frac{(1+\chi)}{2}\sqrt{\left(1+\frac{\chi_i^2}{(1+\chi)^2}\right)}}\\ &\approx \sqrt{-\frac{(1+\chi)}{2}+\frac{(1+\chi)}{2}\left(1+\frac{1}{2}\frac{\chi_i^2}{(1+\chi)^2}\right)}\\ &= -\frac{\chi_i}{2\sqrt{1+\chi}}\end{aligned} \tag{0.10}$$

as shown in Eq. (0.5), n_i and χ_i exhibit opposite sign, therefore we choose the negative sign when taking the square root.

The Intensity profile of a beam propagating in z-direction changes with

$$I(z)\sim\left|e^{-\mathrm{j}\cdot k_n z}\right|^2=e^{-2n_i k_0 z}=e^{-\alpha z}$$

with $k_n=\underline{n}\cdot k_0=n\cdot k_0-\mathrm{j}\cdot n_i\cdot k_0$ and k_0 being the wavenumber in vacuum. For the power attenuation coefficient follows: $\alpha=2n_i k_0=-\frac{\chi_i}{\sqrt{1+\chi}}\cdot\frac{2\pi}{\lambda_0}$.

2) Kramers-Kronig Relations

The polarization **P**(t) of a medium does not only depend on the interaction with a field **E**(t) at one particular point in time t, but it also depends on the history of the interaction. For a linear time-invariant medium, this can be expressed as a convolution with the impulse response $\chi(t)$ in the time domain. In the frequency domain this corresponds to a multiplication with the frequency dependent complex susceptibility $\underline{\chi}(\omega) = F\left[\chi(t)\right]$:

$$\mathbf{P}(t) = \varepsilon_0 \int_{-\infty}^{+\infty} \chi(\tau)\mathbf{E}(t-\tau)d\tau \tag{0.11}$$

$$\tilde{\mathbf{P}}(\omega) = \varepsilon_0 \underline{\chi}(\omega)\tilde{\mathbf{E}}(\omega). \tag{0.12}$$

1. The reaction of a medium to an electric field is causal, as there cannot be any polarization prior to the application of the electric field to the medium. Explain why for this case the following identity holds, where $H(t)$ is the Heaviside function.

$$\chi(t) = \chi(t)\, H(t) \quad \text{with} \quad H(t) = \begin{cases} 1 & t > 0 \\ 0 & t < 0. \end{cases} \tag{0.13}$$

2. Causality in time domain corresponds to an equivalent relation in frequency domain. Transform (0.13) to the frequency domain. Use the Fourier transform of the Heaviside function:

$$\tilde{H}(\omega) = \frac{1}{j\omega} + \pi\delta(\omega) \quad \text{with} \quad \tilde{H}(t) = \begin{cases} 1 & t \geq 0 \\ 0 & t < 0. \end{cases} \tag{0.14}$$

Note: In this course the following definitions of the Fourier transform are used:

$$\tilde{x}(\omega) = F\left[x(t)\right] = \int_{-\infty}^{+\infty} x(t)e^{-i\omega t}dt$$

$$x(t) = F^{-1}\left[\tilde{x}(\omega)\right] = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \tilde{x}(\omega)e^{i\omega t}d\omega$$

$$F\left[x(t)\cdot y(t)\right] = \frac{1}{2\pi}\tilde{x}(\omega) * \tilde{y}(\omega)$$

In order to calculate the convolution of $f(x)$ and $\frac{1}{x}$, the Cauchy principal value has to be introduced: $f(x) * \frac{1}{x} = \mathcal{P}\int_{-\infty}^{+\infty} \frac{f(x')}{x-x'}dx'$.

3. The susceptibility is complex, $\underline{\chi}(\omega) = \chi(\omega) + \mathrm{j}\chi_i(\omega)$. Use the previous result to derive a general relation between the real part $\chi(\omega)$ and the imaginary part $\chi_i(\omega)$ of the susceptibility. This relation is known as the "Kramers-Kronig relation" (after the

discoverers H. A. Kramers und R. de Laer Kronig.). Note that $\tilde{\chi}(\omega)$ is an even, $\tilde{\chi}_i(\omega)$ an odd function, since $\chi(t)$ is a real function.

4. Sketch the frequency dependence of the real and imaginary part of the susceptibility if the medium has a sharp, symmetric absorption line at a frequency ω_0. To do so, assume that $n_i(\omega)$ is affected mostly by $\chi_i(\omega)$.

Solution

1. If we demand, that the polarization is causal, this means that only values of the electric field $E(t-\tau)$ may enter after a certain point of time *t*, that means for $\tau' > 0$, where $\tau' = t - \tau$. As a conclusion, the susceptibility for previous times must be identically zero:

$$\chi(\tau') = \chi(\tau')H(\tau').$$

2. In order to transform (0.14) to frequency domain, we need to know the individual Fourier transformed values as well as the rule that multiplications in time domain become convolutions in frequency domain.

$$\begin{aligned}
\underline{\chi}(\omega) &= F\left[\chi(t)\; H(t)\right] \\
&= \frac{1}{2\pi} F\left[\chi(t)\right] * F\left[H(t)\right] \\
&= \frac{1}{2\pi} \underline{\chi}(\omega) * \left[\frac{1}{j\omega} + \pi\delta(\omega)\right] \qquad (0.15)\\
&= \frac{1}{2\pi} \mathcal{P}\int \underline{\chi}(\omega') \frac{1}{j(\omega-\omega')} d\omega' + \frac{1}{2}\underline{\chi}(\omega) \\
\frac{1}{2}\underline{\chi}(\omega) &= -j\frac{1}{2\pi}\mathcal{P}\int_{-\infty}^{\infty} \frac{\underline{\chi}(\omega')}{\omega-\omega'} d\omega'
\end{aligned}$$

The principal value $\mathcal{P}$ can be interpreted as the value of a 'symmetric' integration, the sum of the right and left limit values of the divergent integral.

This result shows that the value of the susceptibility at one frequency ω is given by an infinite integral over all other values of the susceptibility, weighted by the frequency difference.

3. We consider real and imaginary part of the complex susceptibility independently:

$$
\begin{aligned}
\underline{\chi}(\omega) &= \chi(\omega) + j\chi_i(\omega) \\
&= -\frac{j}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\underline{\chi}(\omega')}{\omega-\omega'}d\omega' \\
&= -\frac{j}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi(\omega)+j\chi_i(\omega)}{\omega-\omega'}d\omega' \\
&= -\frac{j}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi(\omega')}{\omega-\omega'}d\omega' + \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi_i(\omega')}{\omega-\omega'}d\omega'
\end{aligned}
$$

This result is known as the Kramers-Kronig (KK) relations that relate real and imaginary part of the complex linear susceptibility:

$$
\begin{aligned}
\chi(\omega) &= +\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi_i(\omega')}{\omega-\omega'}d\omega' \\
\chi_i(\omega) &= -\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi(\omega')}{\omega-\omega'}d\omega'
\end{aligned}
\tag{0.16}
$$

Note that in order to derive this result, only *causality was required*. In fact, the same result can be obtained for all analytic functions that are linear and causal!

By using the parity of real and imaginary parts of the real susceptibility the KK-relations can be rewritten to only use positive frequencies in the so called second form:

$$
\begin{aligned}
\chi(\omega) &= +\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi_i(\omega')}{\omega-\omega'}\cdot\frac{\omega+\omega'}{\omega+\omega'}d\omega' \\
&= +\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\omega\cdot\chi_i(\omega')}{\omega^2-\omega'^2}d\omega' + \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\omega'\cdot\chi_i(\omega')}{\omega^2-\omega'^2}d\omega' \\
&= 0 + \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{0}\frac{\omega'\cdot\chi_i(\omega')}{\omega^2-\omega'^2}d\omega' + \frac{1}{\pi}\mathcal{P}\int_{0}^{\infty}\frac{\omega'\cdot\chi_i(\omega')}{\omega^2-\omega'^2}d\omega' \\
&= \frac{2}{\pi}\mathcal{P}\int_{0}^{\infty}\frac{\omega'\cdot\chi_i(\omega')}{\omega^2-\omega'^2}d\omega'
\end{aligned}
$$

$$
\begin{aligned}
\chi(\omega) &= +\frac{2}{\pi}\mathcal{P}\int_{0}^{\infty}\frac{\omega'\chi_i(\omega')}{\omega^2-\omega'^2}d\omega' \\
\chi_i(\omega) &= -\frac{2}{\pi}\mathcal{P}\int_{0}^{\infty}\frac{\omega\chi(\omega')}{\omega^2-\omega'^2}d\omega'
\end{aligned}
\tag{0.17}
$$

Note that the sign of our Kramer Kronig relation is different from other resources as e.g. Boyd. This is due to the different definition of the Fourier transform. With Boyd's definition the Fourier transformation of the Heaviside function would be the complex conjugate and hence result in opposite signs for Eq. (0.16) and (0.17).

4. As shown in the first part of this problem set, the power extinction coefficient, which is responsible for losses, is proportional to the imaginary part of the refractive index. This in turn can be related to the imaginary part of the susceptibility devided by the real part of the refractive index:

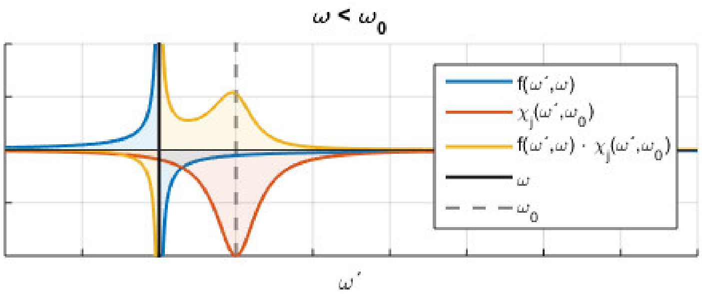
$$n_i = -\chi_i / (2n) \quad \begin{cases} >0 & \text{loss} \\ <0 & \text{gain} \end{cases} \tag{0.18}$$

That means, the imaginary part $\chi_i(\omega)$ is always negative and exhibits a peak at ω_0.

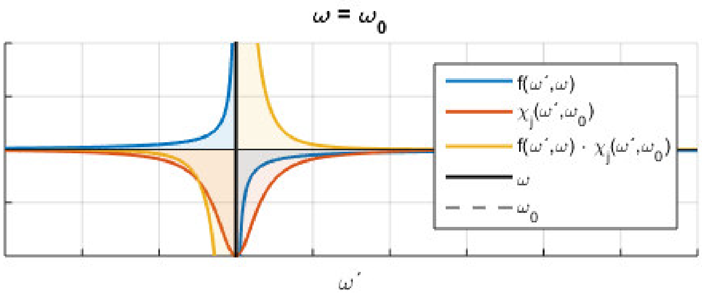
In order to derive the behavior of $\chi(\omega)$ around the resonance, various cases of the relative position of ω with respect to ω_0 are analyzed by comparing the left and the right side of the Cauchy principal value of the integral in equation (0.17):

$$\chi(\omega) = +\frac{2}{\pi}\mathcal{P}\int_0^\infty \frac{\omega'\chi_i(\omega')}{\omega^2-\omega'^2}d\omega' = +\frac{2}{\pi}\left(\lim_{\varepsilon\to 0^+}\int_0^{\omega-\varepsilon}\frac{\omega'}{\omega^2-\omega'^2}\cdot\chi_i(\omega')d\omega' + \lim_{\varepsilon\to 0^+}\int_{\omega+\varepsilon}^{\infty}\frac{\omega'}{\omega^2-\omega'^2}\cdot\chi_i(\omega')d\omega'\right)$$
$$= +\frac{2}{\pi}\left(\lim_{\varepsilon\to 0^+}\int_0^{\omega-\varepsilon} f(\omega',\omega)\cdot\chi_i(\omega',\omega_0)d\omega' + \lim_{\varepsilon\to 0^+}\int_{\omega+\varepsilon}^{\infty} f(\omega',\omega)\cdot\chi_i(\omega',\omega_0)d\omega'\right)$$

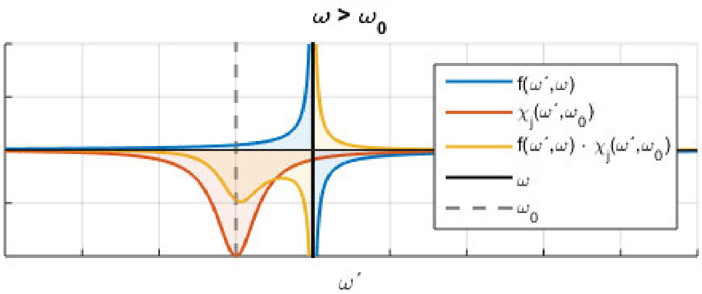
$\omega < \omega_0$: The overlap of the imaginary part of the susceptibility $\chi_i(\omega')$ with the remaining part of the integral $f(\omega')$ is located mostly on the right of ω. This means, that the right integral dominates over the left one, leading to a positive real part of the susceptibility $\chi(\omega) > 0$.



$\omega = \omega_0$: The imaginary part of the susceptibility now contributes equally to both integrals. As a result, the two integrals have the same value, however opposite signs. This leads overall to $\chi(\omega) = 0$.



$\omega > \omega_0$: Now $\chi_i(\omega')$ is contributing mostly to the left integral such that the sum of both, $\chi(\omega)$, becomes smaller than 0.



$\omega \to \infty, \omega \to 0$: For very large values of ω as well as for approaching 0, the overlap between the two functions $\chi_i(\omega')$ and $f(\omega')$ in the two respective integrals vanishes. This leads to decreasing values of the integrals and overall decreasing values of $\chi(\omega)$. For $\omega < \omega_0$, this means $\chi(\omega)$ is approaching 0 from positive values, for $\omega > \omega_0$ $\chi(\omega)$ approaches 0 from negative values.

Sketch:

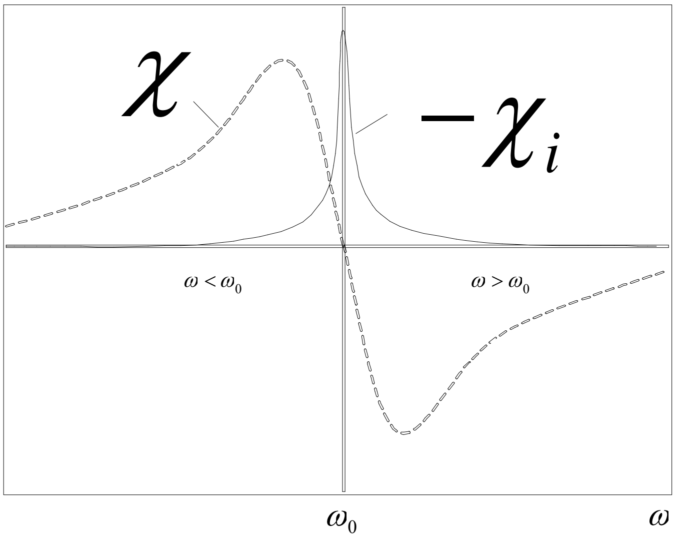


Figure 1. Real and imaginary part of the first order susceptibility.

Bonus Program:

At three randomly chosen tutorials we will collect your solutions before the session starts. The solutions will be marked. If you have 70% or more of each collected problem set completed correctly, your oral examination grade will be upgraded by 0.3 or 0.4 (except grades of 1.0 and 4.7 or worse). If you cannot join a tutorial, you may also hand in your solutions by email to the teaching assistants (see contact details below) **before** the respective session. Please attach all pages in one pdf-file with white background. Students, who handed in a problem set, will be chosen randomly to present their solution.

Questions and Comments:

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