Solution to Problem Set 1
Nonlinear Optics (NLO)

1) Refractive Index, Extinction Coefficient and Absorption

Express the real and imaginary part of the complex refractive index

$$\underline{n} = n - \mathrm{j}n_i \tag{1.1}$$

using the real and imaginary part of the complex susceptibility

$$\underline{\chi}^{(1)} = \chi + \mathrm{j}\chi_i . \tag{1.2}$$

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 (1.1) we get the relations between real and imaginary part:

$$\underline{\varepsilon}_r = \varepsilon_r - \mathrm{j}\varepsilon_{ri} = \underline{n}^2 = (n - \mathrm{j}n_i)^2 = n^2 - n_i^2 - \mathrm{j}2nn_i \tag{1.3}$$

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

$$\underline{\varepsilon}_r = 1 + \underline{\chi}^{(1)}$$

$$\varepsilon_r - \mathrm{j}\varepsilon_{ri} = 1 + \chi + \mathrm{j}\chi_i \tag{1.4}$$

From (1.3) and (1.4) we get:

$$\varepsilon_{ri} = 2nn_i = -\chi_i \text{, and} \tag{1.5}$$

$$\varepsilon_r = 1 + \chi = n^2 - n_i^2 . \tag{1.6}$$

From (1.5), we find $n_i = -\chi_i / (2n)$. By substituting it into (1.6) we find an equation for n:

$$1 + \chi = n^2 - \left(\frac{\chi_i}{2n}\right)^2 \Leftrightarrow n^2 - (1+\chi) - \left(\frac{\chi_i}{2n}\right)^2 = 0 \Leftrightarrow n^4 - (1+\chi)n^2 - \left(\frac{\chi_i}{2}\right)^2 \tag{1.7}$$

By substituting $N = n^2$, equation (1.7) can be solved as a quadratic equation:

$$N^2 - N(1+\chi) - \left(\frac{\chi_i}{2}\right)^2 = 0 \Leftrightarrow N_{1,2} = \frac{(1+\chi) \pm \sqrt{(1+\chi)^2 + \chi_i^2}}{2} \tag{1.8}$$

Since $n^2 > 0$, only $N_1 > 0$ is a meaningful solution. Therefore,

$$n = \sqrt{\frac{(1+\chi)}{2} + \frac{\sqrt{(1+\chi)^2 + \chi_i^2}}{2}}\,. \tag{1.9}$$

From (1.6) it follows that $n_i = \pm\sqrt{n^2 - (1+\chi)}$. By using (1.9), we get:

$$n_i = \pm\sqrt{-\frac{(1+\chi)}{2} + \frac{\sqrt{(1+\chi)^2 + \chi_i^2}}{2}} \tag{1.10}$$

Since the value of the square root in (1.10) is non-negative, the sign of n_i, according to (1.5) will be the opposite of the sign of χ_i.

$$n_i = -\operatorname{sgn}(\chi_i)\sqrt{-\frac{(1+\chi)}{2} + \frac{\sqrt{(1+\chi)^2 + \chi_i^2}}{2}} \tag{1.11}$$

When the condition $|\chi_i| << |\chi|$ is fulfilled, the real and imaginary part of the complex refractive index $\underline{n}$ can be approximated using Taylor series in the following way:

$$\begin{aligned} n &= \sqrt{\frac{(1+\chi)}{2} + \frac{(1+\chi)}{2}\sqrt{\left(1+\frac{\chi_i^2}{(1+\chi)^2}\right)}} \approx \sqrt{\frac{(1+\chi)}{2} + \frac{(1+\chi)}{2}\left(1+\frac{\chi_i^2}{2(1+\chi)^2}\right)} = \\ &= \sqrt{1+\chi+\frac{\chi_i^2}{4(1+\chi)}} \approx \sqrt{1+\chi}, \text{ and} \end{aligned} \tag{1.12}$$

$$\begin{aligned} n_i &= -\operatorname{sgn}(\chi_i)\sqrt{-\frac{(1+\chi)}{2} + \frac{(1+\chi)}{2}\sqrt{\left(1+\frac{\chi_i^2}{(1+\chi)^2}\right)}} \approx \\ &\approx -\operatorname{sgn}(\chi_i)\sqrt{-\frac{(1+\chi)}{2} + \frac{(1+\chi)}{2}\left(1+\frac{\chi_i^2}{2(1+\chi)^2}\right)} = \\ &= -\operatorname{sgn}(\chi_i)\sqrt{\frac{\chi_i^2}{4(1+\chi)}} = -\operatorname{sgn}(\chi_i)\frac{|\chi_i|}{2\sqrt{1+\chi}} = -\frac{\chi_i}{2\sqrt{1+\chi}}. \end{aligned} \tag{1.13}$$

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

$$I(z) \sim \left|e^{-\mathrm{j}\cdot \underline{k}_n z}\right|^2,$$

where $\underline{k}_n = \underline{n}\cdot k_0 = n\cdot k_0 - \mathrm{j}\cdot n_i \cdot k_0$ and where k_0 is the wavenumber in vacuum. From here, it follows: $I(z) \sim \left|e^{-\mathrm{j}\underline{k}_n z}\right|^2 = \left|e^{-\mathrm{j}(n-\mathrm{j}n_i)k_0 z}\right|^2 = \left|e^{-\mathrm{j}nk_0 z - nk_0 z}\right|^2 = \left|e^{-\mathrm{j}nk_0 z}e^{-nk_0 z}\right|^2 = e^{-2n_i k_0 z} = e^{-\alpha z}$. The power attenuation coefficient is therefore: $\alpha = 2n_i k_0 = -\frac{\chi_i}{\sqrt{1+\chi}}\cdot\frac{2\pi}{\lambda_0}$.

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 by the frequency dependent complex susceptibility $\underline{\chi}(\omega) = F\left[\chi(t)\right]$:

$$\mathbf{P}(t) = \varepsilon_0 \int_{-\infty}^{+\infty} \chi(\tau)\mathbf{E}(t-\tau)d\tau \tag{2.1}$$

$$\tilde{\mathbf{P}}(\omega) = \varepsilon_0 \underline{\chi}(\omega)\tilde{\mathbf{E}}(\omega). \tag{2.2}$$

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:

$$\chi(t) = \chi(t)\, H(t) \quad \text{with} \quad H(t) = \begin{cases} 1 & t > 0 \\ 0 & t < 0. \end{cases} \tag{2.3}$$

2. Causality in the time domain corresponds to an equivalent relation in the frequency domain. Transform (2.3) to its frequency domain equivalent. Use the Fourier transform of the Heaviside function:

$$\tilde{H}(\omega) = \frac{1}{j\omega} + \pi\delta(\omega). \tag{2.4}$$

   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 and R. de Laer Kronig). Note that $\tilde{\chi}(\omega)$ is an even, and $\tilde{\chi}_i(\omega)$ 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 predominantly affected by $\chi_i(\omega)$.

Solution

1. The relation (2.1) determines the electric polarization at the moment t, which is the present time. In the same relation, the electric field is a function of $t-\tau$. If t is the present, we can write the following:

$$t-\tau = \begin{cases} \text{past, } \tau > 0 \\ \text{present, } \tau = 0 \\ \text{future, } \tau < 0. \end{cases} \tag{2.5}$$

Since $\mathbf{P}$ depends only on past and present values of $\mathbf{E}$, it follows from (2.5) that only $\tau \geq 0$ makes sense in (2.1). From here, we can write:

$$\mathbf{P}(t) = \varepsilon_0 \int_{-\infty}^{+\infty} \chi(\tau)\mathbf{E}(t-\tau)d\tau = \varepsilon_0 \int_{0}^{+\infty} \chi(\tau)\mathbf{E}(t-\tau)d\tau = \varepsilon_0 \int_{-\infty}^{+\infty} \chi(\tau)H(\tau)\mathbf{E}(t-\tau)d\tau. \tag{2.6}$$

That means that $\chi(\tau) = \chi(\tau)H(\tau)$, which is equivalent to (2.3).

2. In order to transform (2.3) to its frequency domain equivalent, we need to know the individual Fourier transforms of $\chi(t)$ and $H(t)$, as well as the rule that multiplication in the time domain becomes convolution in the frequency domain.

$$\underline{\chi}(\omega) = F\left[\chi(t)\, H(t)\right] = \frac{1}{2\pi} F\left[\chi(t)\right] * F\left[H(t)\right] = \frac{1}{2\pi}\underline{\chi}(\omega) * \left[\frac{1}{j\omega} + \pi\delta(\omega)\right] =$$

$$= \frac{1}{2\pi}\mathcal{P}\int \underline{\chi}(\omega')\frac{1}{j(\omega-\omega')}d\omega' + \frac{1}{2}\underline{\chi}(\omega).$$

Finally: $$\underline{\chi}(\omega) = -\frac{j}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\underline{\chi}(\omega')}{\omega-\omega'}d\omega'. \tag{2.7}$$

The principal value $\mathcal{P}$ can be interpreted as the value of a "symmetric" integration, of the improper integral. It is the sum of two integrals: from $-\infty$ to $\omega-\varepsilon$, and $\omega+\varepsilon$ to $+\infty$, when $\varepsilon \to 0$. 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 the real and imaginary part of the complex susceptibility independently:

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

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

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

$$\chi(\omega) = +\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi_i(\omega')}{\omega-\omega'}d\omega' \tag{2.8a}$$

$$\chi_i(\omega) = -\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\chi(\omega')}{\omega-\omega'}d\omega' \tag{2.8b}$$

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 the real and imaginary part of the real susceptibility, the KK relations can be written in the so called second form, where we use only positive frequencies:

$$\chi(\omega) = +\frac{2}{\pi}\mathcal{P}\int_{0}^{\infty}\frac{\omega'\chi_i(\omega')}{\omega^2-\omega'^2}d\omega' \tag{2.9a}$$

$$\chi_i(\omega) = -\frac{2}{\pi}\mathcal{P}\int_{0}^{\infty}\frac{\omega\chi(\omega')}{\omega^2-\omega'^2}d\omega' \tag{2.9b}$$

Note that the sign of our KK relations is different from what is used in some literature resources, e.g. Boyd. This is due to the different definition of the Fourier transform. By using Boyd's definition, the Fourier transform of the Heaviside function would be the complex conjugate of what we got, and hence result in opposite signs for Eqs. (2.8) and (2.9).

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 divided by the real part of the refractive index:

$$n_i = -\chi_i/(2n) \quad \begin{cases} >0 & \text{loss} \\ <0 & \text{gain} \end{cases} \tag{2.10}$$

That means that the imaginary part $\chi_i(\omega)$ is always negative and exhibits a peak at ω_0. Concerning $\chi(\omega)$, if we have a symmetric absorption line at ω_0, the left and right limit values of the Cauchy principal value in $\chi(\omega_0)$ will have the same magnitude, but opposite signs. The sum of both is then zero. For a frequency $\omega > \omega_0$, the value of the denominator of the fraction in the integral of (2.8a) will be < 0 whenever $\omega' > \omega$, and > 0 whenever $\omega' < \omega$. The numerator of the same fraction, (which is χ_i), is always negative, and together with the denominator determines the sign of the left and right limit values. The left limit value (integral from $-\infty$ to $\omega-\varepsilon$) is negative (negative numerator × positive denominator), and the right limit value (integral from $\omega+\varepsilon$ to $+\infty$) is positive (negative numerator × negative denominator). The left limit value however dominates, since the peak of χ_i is in ω_0, which is on the left of ω (remember that we are considering the case $\omega > \omega_0$). Therefore, the Cauchy principal value (the sum of the left and right limit value) and the value of χ will be negative. For frequencies far from the resonance ω_0, χ approaches a constant value. Analog consideration can be applied to the case $\omega < \omega_0$.

$$\omega < \omega_0 \Rightarrow \mathcal{P}\left\{\frac{\chi_i}{\omega - \omega'}\right\} > 0 \Rightarrow \chi > 0$$

$$\omega > \omega_0 \Rightarrow \mathcal{P}\left\{\frac{\chi_i}{\omega - \omega'}\right\} < 0 \Rightarrow \chi < 0$$

Sketch:

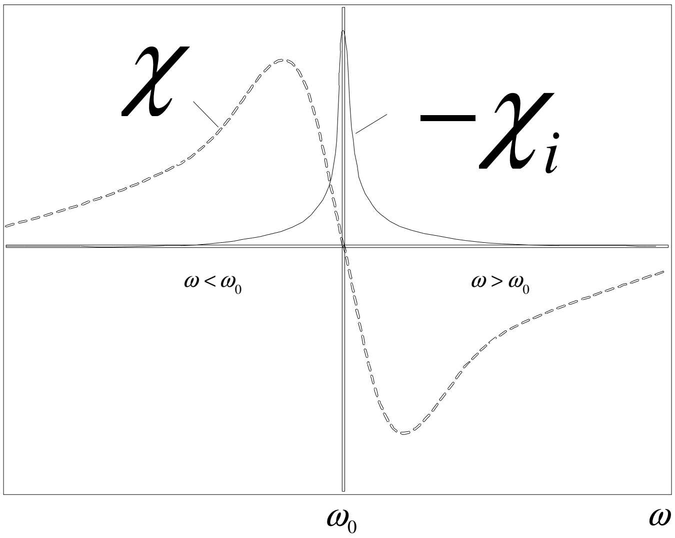

Figure 1. Real and imaginary part of the first order susceptibility.

Questions and Comments:

Aleksandar Nesic
Building: 30.10, Room: 2.32-2
Phone: 0721/608-42480

Matthias Blaicher
Building: 30.10, Room: 2.22
Phone: 0721/608-41934 (26824)

nlo@ipq.kit.edu