Problem Set 2
Nonlinear Optics (NLO)

Due: May 17, 2017, 08:00 AM

1) Lorentz oscillator model for the linear case

In a classical picture, an electron bound to an atom can be considered as a harmonic oscillator in analogy to a mass connected to a spring. This oscillator, when driven by an external electric field E_x, follows the classical equation of motion

$$m_e \frac{d^2 x(t)}{dt^2} = -eE_x(t) - m_e \omega_r^2 x(t) - m_e \gamma_r \frac{dx(t)}{dt}, \tag{1.1}$$

where $x(t)$ is the dislocation of the electron, m_e denotes the electronic mass, $-eE_x(t)$ is the driving force by the external electric field, $-m_e \omega_r^2 x$ is the restoring force of the oscillator and $-m_e \gamma_r \frac{dx}{dt}$ is a damping term, with damping constant γ_r. The parameter ω_r will turn out to be the resonance frequency of the oscillator.

1. Solve the differential equation (1.1) for a time-harmonic electric field of the form $E(t) = \frac{1}{2}\left(\underline{E}(\omega)\exp(j\omega t) + c.c.\right)$ by using a similar ansatz for the dislocation. Derive an expression for $\underline{x}(\omega)$.
2. The electric polarization is the dipole moment per volume,

$$\underline{P}_x(\omega) = \varepsilon_0 \underline{\chi}^{(1)}(\omega) \underline{E}_x(\omega) = -\frac{N}{V} e \cdot \underline{x}(\omega), \tag{1.2}$$

   where $\frac{N}{V}$ is the number density of atoms in the medium and $-e \cdot \underline{x}(\omega)$ is the induced dipole moment per atom. Show that the susceptibility is given by

$$\underline{\chi}^{(1)}(\omega) = \frac{Ne^2}{V m_e \varepsilon_0} \frac{1}{\omega_r^2 - \omega^2 + j\omega\gamma_r}, \tag{1.3}$$

   and separate the susceptibility into real and imaginary part.
3. Sketch the real and imaginary part of the susceptibility $\underline{\chi}^{(1)}$ around the resonance frequency ω_r. What is the consequence of this result for the real and imaginary part of the refractive index at very large frequencies, e.g. X-ray frequencies?

2) Third-order nonlinear polarization

Consider a linearly polarized plane wave $\mathbf{E}(z,t) = E(z,t)\mathbf{e}_x$ propagating in z-direction in a homogeneous medium, in which third-order nonlinear effects dominate over second and higher-order contributions, $\chi^{(3)} \neq 0, \chi^{(m)} = 0$ for $m = 2$ or $m > 3$. Assuming an instantaneous response of the polarization P_{NL} to the applied electric field, we can express P_{NL} as

$$P_{\mathrm{NL}}(z,t) = P^{(3)}(z,t)\mathbf{e}_{\mathrm{x}}\text{, with } P^{(3)}(z,t) = \varepsilon_0 \chi^{(3)} E(z,t)^3 . \qquad (2.1)$$

Calculate $P^{(3)}(z,t)$ considering a field $E(z,t)$ composed of two distinct frequency components ω_1 and ω_2 with their complex amplitudes $\underline{E}_1$ and $\underline{E}_2$

$$E(z,t) = \frac{1}{2}(\underline{E}_1 e^{\mathrm{j}(\omega_1 t - k_1 z)} + \underline{E}_2 e^{\mathrm{j}(\omega_2 t - k_2 z)} + c.c.) . \qquad (2.2)$$

Group the resulting terms with appropriate degeneracy factors according to their frequency and assign them to the following effects:

- Third-Harmonic Generation (THG)
- Third-order Sum-Frequency Generation (TSFG)
- Self-Phase Modulation (SPM)
- Cross-Phase Modulation (XPM)
- (Degenerate) Four-Wave Mixing (FWM)

Solution

1. We insert the ansatz for the electric field $E(t)=\frac{1}{2}\left(\underline{E}(\omega)\exp(j\omega t)+c.c.\right)$ and the dislocation $x(t)=\frac{1}{2}\left(\underline{x}(\omega)\exp(j\omega t)+c.c.\right)$ into the differential equation (1.1). We only take into account the terms oscillating at the same angular frequency and obtain:

$$-\omega^2 m_{\mathrm{e}}\underline{x}(\omega)=-e\underline{E}(\omega)-m_{\mathrm{e}}\omega_{\mathrm{r}}^2\underline{x}(\omega)-j\omega m_{\mathrm{e}}\gamma_{\mathrm{r}}\underline{x}(\omega)$$

We solve for $\underline{x}(\omega)$ and get the frequency dependent dislocation of the oscillator:

$$\underline{x}(\omega)=\frac{-e\underline{E}(\omega)}{m_{\mathrm{e}}(\omega_{\mathrm{r}}^2-\omega^2+j\omega\gamma_{\mathrm{r}})}$$

2. The polarization of the charged system (atom and electron) is given by $-e\cdot\underline{x}(\omega)$. As the total electric polarization of the medium is given by all of its atomic dipoles, we multiply with the number density $\frac{N}{V}$ of atoms in the medium. We then use Eq. **Error! Reference source not found.** to relate the result to the 1st order susceptibility:

$$\underline{P}_{\mathrm{x}}(\omega)=\varepsilon_0\underline{\chi}^{(1)}(\omega)\underline{E}_{\mathrm{x}}(\omega)=-\frac{N}{V}e\cdot\underline{x}(\omega)\Rightarrow\underline{\chi}^{(1)}(\omega)=\frac{Ne^2}{Vm_{\mathrm{e}}\varepsilon_0}\frac{1}{\omega_{\mathrm{r}}^2-\omega^2+j\omega\gamma_{\mathrm{r}}}$$

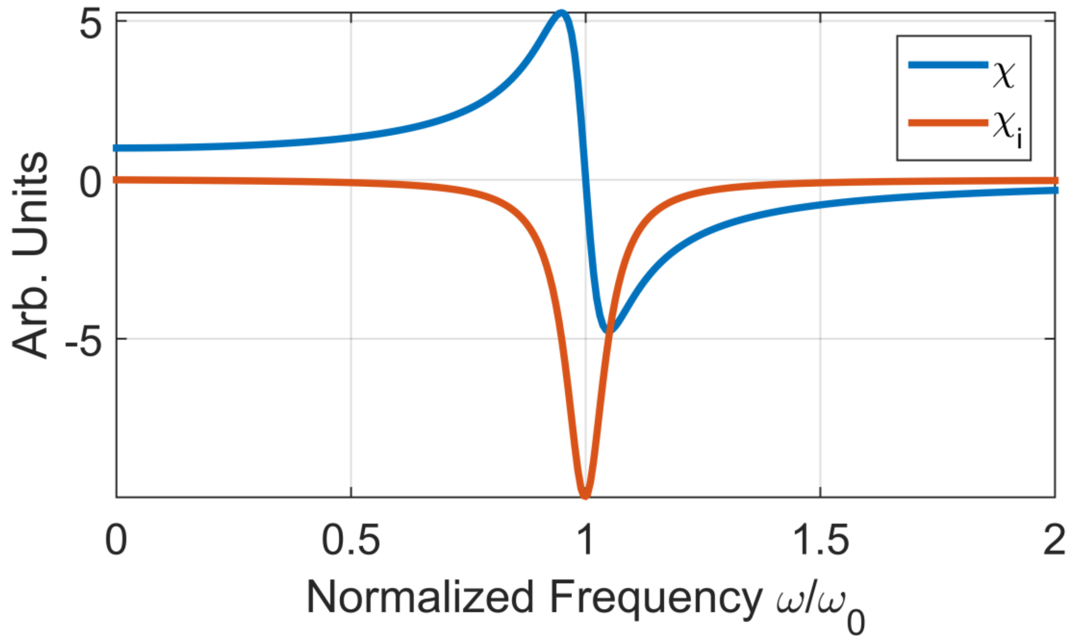
We separate the susceptibility into real and imaginary part by expanding the fraction:

$$\underline{\chi}^{(1)}(\omega)=\frac{Ne^2}{Vm_{\mathrm{e}}\varepsilon_0}\frac{1}{\omega_{\mathrm{r}}^2-\omega^2+j\omega_{\mathrm{r}}\gamma_{\mathrm{r}}}\cdot\frac{\omega_{\mathrm{r}}^2-\omega^2-j\omega\gamma_{\mathrm{r}}}{\omega_{\mathrm{r}}^2-\omega^2-j\omega\gamma_{\mathrm{r}}}=\frac{Ne^2}{Vm_{\mathrm{e}}\varepsilon_0}\frac{\omega_{\mathrm{r}}^2-\omega^2-j\omega_{\mathrm{r}}\gamma_{\mathrm{r}}}{\left(\omega_{\mathrm{r}}^2-\omega^2\right)^2+\left(\omega\gamma_{\mathrm{r}}\right)^2}$$

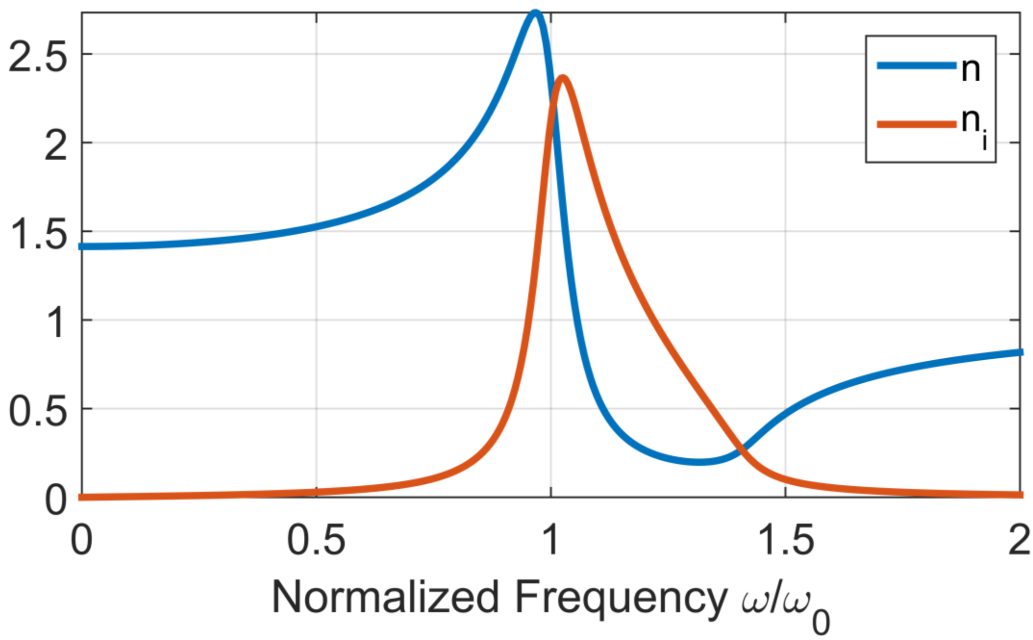
$$\chi^{(1)}=\frac{Ne^2}{Vm_{\mathrm{e}}\varepsilon_0}\frac{\omega_{\mathrm{r}}^2-\omega^2}{\left(\omega_{\mathrm{r}}^2-\omega^2\right)^2+\left(\omega\gamma_{\mathrm{r}}\right)^2}$$

$$\chi_i^{(1)}=-\frac{Ne^2}{Vm_{\mathrm{e}}\varepsilon_0}\frac{\omega\gamma_{\mathrm{r}}}{\left(\omega_{\mathrm{r}}^2-\omega^2\right)^2+\left(\omega\gamma_{\mathrm{r}}\right)^2}$$

3. Sketch of the susceptibility around the resonance frequency:



As derived in Problem Set 1, the real part of the refractive index $n \approx \sqrt{1+\chi^{(1)}}$. Sketching this, it can be seen that the refractive index is smaller than one for frequencies above the resonance frequency (e.g. X-ray). Hence for building lenses for X-ray frequencies one has to use concave instead of convex structures.



3) Third-order nonlinear polarization

Consider a linearly polarized plane wave $\mathbf{E}(z,t)=E(z,t)\mathbf{e}_x$ propagating in z-direction in a homogeneous medium, in which third-order nonlinear effects dominate over second and higher-order contributions, $\chi^{(3)}\neq 0$, $\chi^{(m)}=0$ for m = 2 or m > 3. Assuming an instantaneous response of the polarization P_{NL} to the applied electric field, we can express P_{NL} as

$$P_{\mathrm{NL}}(z,t)=P^{(3)}(z,t)\mathbf{e}_{\mathrm{x}}\text{, with } P^{(3)}(z,t)=\varepsilon_0\chi^{(3)}E(z,t)^3 . \tag{3.1}$$

Calculate $P^{(3)}(z,t)$ considering a field $E(z,t)$ composed of two distinct frequency components ω_1 and ω_2 with their complex amplitudes $\underline{E}_1$ and $\underline{E}_2$

$$E(z,t)=\frac{1}{2}(\underline{E}_1 e^{\mathrm{j}(\omega_1 t-k_1 z)}+\underline{E}_2 e^{\mathrm{j}(\omega_2 t-k_2 z)}+c.c.) . \tag{3.2}$$

Group the resulting terms with appropriate degeneracy factors according to their frequency and assign them to the following effects:

- Third-Harmonic Generation (THG)
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- Self-Phase Modulation (SPM)
- Cross-Phase Modulation (XPM)
- (Degenerate) Four-Wave Mixing (FWM)

Solution:

As we will calculate the third power of four factors, we expect a total number of 4^3=64 terms, which can be expressed as

$$\begin{aligned}P^{(3)}(z,t)=\frac{1}{8}\varepsilon_0\chi^{(3)}\cdot\\
&[E_1^3 e^{j(3\omega_1 t-3k_1 z)}+E_2^3 e^{j(3\omega_2 t-3k_2 z)}+c.c.\\
&+3E_1^2E_2 e^{j((2\omega_1+\omega_2)t-(2k_1+k_2)z)}+3E_2^2E_1 e^{j((\omega_1+2\omega_2)t-(k_1+2k_2)z)}+c.c.\\
&+3\left|E_1\right|^2 E_1 e^{j(\omega_1 t-k_1 z)}+3\left|E_2\right|^2 E_2 e^{j(\omega_2 t-k_2 z)}+c.c.\\
&+6\left|E_1\right|^2 E_2 e^{j(\omega_2 t-k_2 z)}+6\left|E_2\right|^2 E_1 e^{j(\omega_1 t-k_1 z)}+c.c.\\
&+3E_1^2E_2^* e^{j((2\omega_1-\omega_2)t-(2k_1-k_2)z)}+3E_2^2E_1^* e^{j((-\omega_1+2\omega_2)t-(-k_1+2k_2)z)}+c.c.].\end{aligned}$$

The resulting terms can be grouped by frequencies:

Frequency	Degeneracy	Name
$3\omega_1,\ 3\omega_2$	1	THG – third harmonic generation
$2\omega_1+\omega_2,\ 2\omega_2+\omega_1$	3	TSFG – third order sum-frequency generation
$2\omega_1-\omega_2,\ 2\omega_2-\omega_1$	3	FWM – (degenerate) four-wave mixing
ω_1 from $(\omega_2;-\omega_2;\omega_1)$, ω_2 from $(\omega_1;-\omega_1;\omega_2)$	6	XPM – cross-phase modulation
ω_1 from $(\omega_1;\omega_1;-\omega_1)$, ω_2 from $(\omega_2;\omega_2;-\omega_2)$	3	SPM – self-phase modulation

For three frequencies the general rule is that all frequencies $\omega=\left[\pm\omega_l\pm\omega_m\pm\omega_n\right]$ for $l,m,n\in\{1,2,3\}$ are generated. The degeneracy factor is given by the number of possible permutations of those frequencies. For each 'negative' frequency the complex conjugate of the respective field component has to be taken, e.g. $2\omega_1-\omega_2\rightarrow E_1E_1E_2^*$.

Note: Here TSFG and DFWM are classified as different processes but FWM embraces also TSFG and, in fact, all third order nonlinear processes can be considered as FWM processes. Different authors could name in slightly different way some of the corresponding third order nonlinear processes.

Questions and Comments:

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