University of Hamburg, Department of Physics

Nonlinear Optics

Kärtner/Mücke, WiSe 2018/2019

Problem Set 1

Issued: 26.10.2018 Due : 02.11.2018

1. **Susceptibility tensor and symmetries.**

   Consider a two–dimensional medium with D_4- symmetry. There is a set of symmetry operations, which leave the unit cell invariant:

   E: Identity operation
   $\mathbf{R}_+$: Rotation by $\pi/2$
   $\mathbf{R}_-$: Rotation by -$\pi/2$
   R: Rotation by π
   $\mathbf{M}_x$: Mirror image around the x-axis
   $\mathbf{M}_y$: Mirror image around the y-axis
   $\mathbf{D}_1$: Mirror image around the diagonal D_1
   $\mathbf{D}_2$: Mirror image around the diagonals D_2

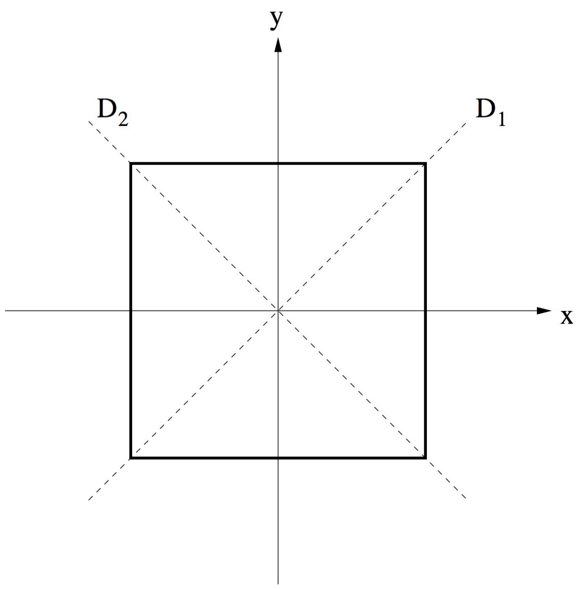

Abbildung 1: D_4-symmetry

   A sequence of symmetry operations (e.g, two) is defined as the concatenation or product of the corresponding (two) operators. For example $\mathbf{M}_x\mathbf{R}_+ = \mathbf{D}_2$.

(a) Construct the multiplication table for the D_4-symmetry group:

	E	R_+	R_-	R	M_x	M_y	D_1	D_2
E								
R_+								
R_-								
R								
M_x								
M_y								
D_1								
D_2								

Tabelle 1: multiplication table for symmetry group D_4

(b) The dielectric tensor of a general two-dimensional medium without symmetry has the form

$$\boldsymbol{\epsilon} = \begin{pmatrix} \varepsilon_{11} & \varepsilon_{12} \\ \varepsilon_{21} & \varepsilon_{22} \end{pmatrix}$$

Show that for a medium with D_4-symmetry the dielectric tensor is isotropic

$$\boldsymbol{\epsilon} = \begin{pmatrix} \varepsilon & 0 \\ 0 & \varepsilon \end{pmatrix}.$$

2. **Coupled oscillator model for the susceptibility.**

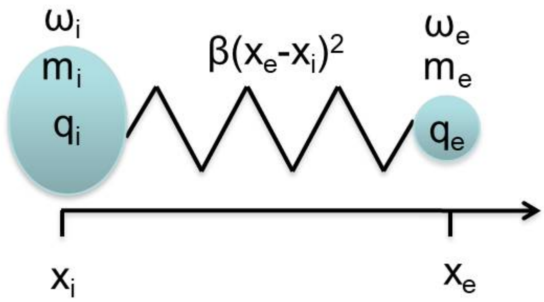

Abbildung 2: Scheme for the anharmonic coupling between ion and electron.

Throughout this problem we want to develop a classical model for a nonlinear optical material, which exhibits optical phonons in the THz frequency range (which spans approximately from 100GHz to 30THz). As a phonon describes the collective motion of the ions, we will try to add the motion of an ion to our classical Lorentz model derived in the previous lecture, where we only considered the electronic motion. As we will later see, including the ions in the process will lead to interesting insights important for THz generation. To start with, we will first derive the linear response of such sort of system in the following.

Let's assume the ion and electron are each bound in a harmonic potential each with linear restoring forces and their individual resonance frequencies ω_i and we in that potential. Furthermore their motion is damped by a force linearly dependent on their velocities accounted for by $\gamma_i = \frac{2\omega_i}{Q_i}$ and $\gamma_e = \frac{2\omega_e}{Q_e}$. The nonlinearity we introduce to the system this time via an anharmonic coupling $\beta(x_i - x_e)^2$ between the ion and electron. This can be pictured as the first correction term to a purely quadratic potential and simulates hard-core repulsion between the ion and the electron. This way we arrive at a set of equations describing the motion of the system of ion and electron in the material under the influence of an external field E(t):

$$\ddot{x}_i + \omega_i^2 x_i + \gamma_i \dot{x}_i - \frac{\beta}{m_i}(x_i - x_e)^2 = \frac{q_i E(t)}{m_i} \tag{1}$$

$$\ddot{x}_e + \omega_e^2 x_e + \gamma_e \dot{x}_e + \frac{\beta}{m_e}(x_i - x_e)^2 = \frac{q_e E(t)}{m_e} \tag{2}$$

Equation (1) describes the motion of the ion in external electric field E(t), where q_i, m_i and ω_i denote the charge, mass and resonance frequency of the ion. Equation (2) describes the motion of the electron with the parameters inserted for the electron respectively; β is the coupling constant accounting for the strenght of the anharmonic coupling between ionic and electronic motion. For starters we will only consider a monochromatic driving field such as $E(t) = E_0 e^{i(\omega t)}$.

a) Derive a general frequency-dependent expression for the linear susceptibility $\chi^{(1)}(\omega) = \chi_i^{(1)}(\omega) + \chi_e^{(1)}(\omega)$ of the material due to the ionic $\chi_i^{(1)}$ and electronic $\chi_e^{(1)}$ contribution. Use a perturbative ansatz replacing the external field E(t) by $\epsilon E(t)$ and assuming the solution can be expanded into a power series such as $x(t) = \epsilon x^{(1)} + \epsilon^2 x^{(2)} + \epsilon^3 x^{(3)} + ...$ for both ionic and electronic motion. Remember the relation between the excursion of a charge $x^{(n)}$ relates to the induced polarization field as $P^{(n)} = \epsilon_0 \chi^{(n)} E = qNx^{(n)}$, which relates $x^{(n)}$ and $\chi^{(n)}$, with q representing the charge and N the number density.

You should arrive at:

$$\chi_e^{(1)}(\omega) = \frac{N_e e^2}{m_e \epsilon_0 (\omega_e^2 - \omega^2 + i\gamma_e \omega)} \tag{3}$$

$$\chi_i^{(1)}(\omega) = \frac{N_i q_i^2}{m_i \epsilon_0 (\omega_i^2 - \omega^2 + i\gamma_i \omega)} \tag{4}$$

b) Plot the refractive index n(ω) ($\tilde{n}^2 = 1 + \chi^{(1)}$, $\tilde{n} = n + i\kappa$) and the absorption coefficient $\alpha(\omega)$ ($\alpha = \frac{4\pi\kappa}{\lambda}$) as a function of frequency over the range from 0 to 1500 THz. Add to the same sketch the contributions arising solely from the ionic and the electronic contribution respectively. Which contribution is important for which frequency range? How do the resonances influence absorption and refractive index? Discuss your findings!

For the plot, let's assume the phonon resonance frequency is at $f_i = 2$THz with quality factor $Q_i = 10$. In addition, the atoms in each unit cell of the material show electronic transitions at UV-frequencies $f_e = 1500$THz with a a quality factor of $Q_e = 1000$. Values for the other needed physical constants are given in table 2.

c) Now give an expression for the linear susceptibility of the material at THz and optical frequencies when driving it far off from the resonance frequencies in the THz ($\omega \ll \omega_i$) and optical domain ($\omega_i \ll \omega \ll \omega_e$ and $\omega_e \ll \omega$).

Quantity	ion	electron
resonance frequency	$\omega_i = 2\pi \cdot 2\text{THz}$	$\omega_e = 2\pi \cdot 1500\text{THz}$
quality factor	$Q_i = 10$	$Q_e = 1000$
charge	$q_e = e = -1.6 \cdot 10^{-19}C$	$q_i = 1.6 \cdot 10^{-19}C$
density $\approx a \cdot b \cdot c$	$N_i = 10^{28}m^{-3}$	$N_e = 10^{28}m^{-3}$
mass	$m_i = 1.67 \cdot 10^{-27}kg$	$m_e = 9.11 \cdot 10^{-31}kg$
vacuum permittivity ϵ_0	$8.85 \cdot 10^{-12}F \cdot m^{-1}$	
coupling constant β	$10^9 kg \cdot m^{-1} \cdot s^{-2}$	

Tabelle 2: Table of important physical constants needed for the computation in b).