Extra exercises for Nonlinear Optics course 2018

E1. Spatial symmetry

Triclinic crystals may possess two types of spatial symmetry. They have the identity operation as only symmetry element or the identity operator together with an improper reflection around 180 degrees. The respective corresponding matrices C_1 and S_2 are given here:

$$C_1 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad ; \quad S_2 = \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$$

a. What are the non-zero tensor elements for the first-order susceptibility for both crystals?
b. What are the non-zero tensor elements for the second-order susceptibility for both crystals?

Consider now 2 types of monoclinic crystals: first one with a vertical mirror plane; the second one with a vertical mirror plane and C_2 axis. *NB: Here the provided matrices have y-axis as the main axis.*

$$\sigma_{xz} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad ; \quad C_2 = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$$

c. What are the non-zero tensor elements for the first-order susceptibility for both crystals?
d. What are the non-zero tensor elements for the second-order susceptibility for both crystals?

Lastly, consider a tetragonal crystal with a C_4 axis. The corresponding matrix is:

$$C_4 = \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

e. What are the non-zero tensor elements for the first-order susceptibility?
f. What are the non-zero tensor elements for the second-order susceptibility?

E2. Anharmonic nonlinear oscillator

For a material that is non-centrosymmetric, we would like to understand the nonlinear optical response. To do this in the simplest form possible we consider a one-dimensional material, represented by an anharmonic oscillator experiencing a force $F = -eE(t)$, with $E(t) = E_1 e^{-i\omega t}$, an optical driving field at single frequency ω.

a. Assuming there is a damping force $F_d = -2\gamma m\dot{x}$, give the equation of motion and provide the physical meaning of all the constants as well as their relative signs (you can rename the harmonic and anharmonic oscillator constants in the equation as ω_0^2 and a).
b. What will be the potential energy corresponding to the answer found in question a?

If we assume that the nonlinear restoring force is much smaller than that linear restoring force we can use a perturbative approach to solve the equation of motion. Write the driving field as $\lambda E_1 e^{i\omega t}$ (with λ being a dimensionless factor ranging from 0 to 1 and describing the strength of the perturbation). Then write the displacement as a power series expansion around λ. You will assume that λ is 1 at the end of the computation.

c. Give the general expression for the displacement in terms of λ. Then solve the equation of motion for the linear displacement. From the result derive the induced linear polarizability and the first order susceptibility.

d. Derive the solution for the second-order polarization and susceptibility for the case of second harmonic generation.

e. Assuming non-resonant conditions, show that the second-order susceptibility becomes:
$\chi^{(2)}(2\omega;\omega,\omega) = \frac{-Nae^3}{\varepsilon_0 m^2 \omega_0^6}$

f. For $KNbO_3$, which is one of the most efficient nonlinear optical materials and used in many devices, the second-order susceptibility is given in the table below. Compare the magnitude of the linear and non-linear force constants in the equation of motion, using $\omega_0 = 1 \cdot 10^{16}$ rad/s and the average value of lattice constant in most crystals $d \sim 3\text{Å}$. What does that mean?

Crystal System—Orthorhombic—*continued*

Orthorhombic material	Symmetry class	d_{im} (pm/V)	Wavelength λ (µm)
$KNbO_3$	mm2	$d_{33} = -19.58 \pm 1.03$	1.064
		$d_{32} = +11.34 \pm 1.03$	1.064
		$d_{31} = -12.88 \pm 1.03$	1.064

Table 1. Second-order susceptibility values for potassium niobate crystal. The susceptibility values are given using contracted notation. Note also that symmetry class ***mm2*** *in Hermann-Mauguin notation corresponds to* $\mathbf{C_{2v}}$ *in Schoenflies notation. This table was found in Weber, Handbook of Optical Materials, 2003.*