Additional exercises Nonlinear Optics course 2023

0. Tensors

The Levi-Cevita tensor is a useful tool to derive vector identities that are useful in nonlinear optics. To practice using the tensor derive the following identities:

a. Show that $\boldsymbol{A} \cdot (\boldsymbol{B} \times \boldsymbol{C}) = \boldsymbol{B} \cdot (\boldsymbol{C} \times \boldsymbol{A}) = \boldsymbol{C} \cdot (\boldsymbol{A} \times \boldsymbol{B})$
b. Show that $\boldsymbol{\nabla} \times \boldsymbol{\nabla}\psi = 0$
c. Show that $\boldsymbol{\nabla} \times (\boldsymbol{\nabla} \times \boldsymbol{A}) = \boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} \cdot \boldsymbol{A}) - \nabla^2 \boldsymbol{A}$

I. 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 (S_2 axis).

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

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 with vertical mirror plane and the second with vertical mirror plane + a C_2 axis.

Note: The provided matrices have the y-axis as the main symmetry axis.

$$_{xz} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad C_2 = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$$

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.

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

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?

II. 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.

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.
b. What will be the potential energy corresponding to the answer found in a?

If we assume that the nonlinear restoring force is much smaller than the linear restoring force we can use a perturbative approach to solve the equation of motion. Write the driving field as $\lambda E_1 e^{i\omega t}$ and assuming that λ is 1 we can write the displacement as a power series expansion around λ.

c. Give the general expression for the displacement in terms of λ and solve the equation of motion for the linear displacement. From this 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 difference frequency generation.

e. Assuming non-resonant conditions, show that the second-order susceptibility for second harmonic generation 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*), $d = 3$ Å.

Table 1. Nonlinear optical properties of $KNbO_3$

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