

# Problem Set 6

## Nonlinear Optics (NLO)

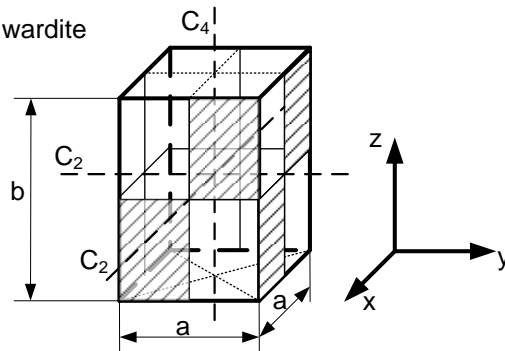
Due: 10. June 2015

### 1) Crystal Symmetry Classes and Susceptibility

In crystallography, by means of group theory, all crystals can be classified according to their point group. A crystallographic point group is a set of symmetry operations, such as rotations or reflections, that leave a central point fixed while moving atoms into position previously occupied by another atom of the same kind, therefore leaving the crystal unchanged.

For crystals in 3-dimensional space, there are 32 possible point groups. Each of them corresponds to one of the 32 “crystal classes”. An example of a point group is the so-called tetragonal-trapezoidal group, also called  $D_4$  in the Schoenflies notation, or 422 in the international (Hermann–Mauguin) notation. This point group belongs to the tetragonal crystal system and therefore the crystal’s Bravais lattice is a cuboid with two equal dimensions whereas the third dimension is shorter or longer than the other two. The  $D_4$  point group of symmetry operations comprises rotation by  $2\pi/4$  about one axis ( $z$ ) and rotations by  $2\pi/2$  about the other two axes ( $x$  and  $y$ ), see Fig. 1.

e.g. cristobalite, wardite  
 $D_4$  crystal class



**Figure 1: Schematic representation of the  $D_4$  symmetry class with its three axes and two- and four-fold axes of rotation ( $C_2$ ,  $C_4$ ).**

In this exercise, we are going to look at the restrictions that the  $D_4$  point group imposes on the form of the second-order nonlinear susceptibility tensor. For simplicity, we are going to analyze the case of second-harmonic generation (SHG).

Let us consider a material with a second-order nonlinear susceptibility tensor  $\chi_{qrs}$ , where the subscripts  $q, r, s \in \{1, 2, 3\}$  denote the vector component of the polarization and the electric field. The component  $q$  of the polarization is then given by

$$P_q = \frac{1}{2} \epsilon_0 \chi_{qrs} E_r E_s, \quad (1)$$

where Einstein summation convention is used so that index repetition on the right hand side of the equation indicates sum over these indices.

Now apply a symmetry operation  $T$  to the entire configuration ( $E$ ,  $P$ , and the material), for example a  $\pi/2$  rotation. In general, the tensor describing the new material has now changed. However, because of crystal symmetry,  $T$  leaves the material physically unchanged. The susceptibility tensor can therefore be replaced by the original susceptibility tensor elements:

$$T_{qq'} P_{q'} = \frac{1}{2} \epsilon_0 \chi_{qrs} (T_{rr'} E_{r'}) (T_{ss'} E_{s'}). \quad (2)$$

Equation (2) can be further simplified by using the contracted notation,  $\underline{\chi}_{qrs}^{(2)} \mapsto d_{ql} = \underline{\chi}_{qrs}^{(2)} / 2$ , with the matrix components  $d_{ij}$  instead of the susceptibility tensor components:

$$\begin{bmatrix} P_{x,2\omega} \\ P_{y,2\omega} \\ P_{z,2\omega} \end{bmatrix} = \epsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_{x,\omega}^2 \\ E_{y,\omega}^2 \\ E_{z,\omega}^2 \\ 2E_{y,\omega} E_{z,\omega} \\ 2E_{x,\omega} E_{z,\omega} \\ 2E_{x,\omega} E_{y,\omega} \end{bmatrix} \quad (3)$$

For finding the implications of the symmetry on the form of the matrix  $d_{ij}$  proceed as follows:

1. Complete the following tables that describe how the elements in Eq. (3) change for each of the three rotations described above. In the table, “ $2\pi/2, x$ ” denotes a rotation by  $2\pi/2$  about the  $x$ -axis. The second column of the first table has been already completed. (Hint: Use the matrix expression for the rotation operations).

	$\frac{2\pi}{2}, x$	$\frac{2\pi}{2}, y$	$\frac{2\pi}{4}, z$
$P'_{x,2\omega}$		$-P_{x,2\omega}$	
$P'_{y,2\omega}$		$P_{y,2\omega}$	
$P'_{z,2\omega}$		$-P_{z,2\omega}$	

(4)

	$\frac{2\pi}{2}, x$	$\frac{2\pi}{2}, y$	$\frac{2\pi}{4}, z$
$E_{x,\omega}^2$			
$E_{y,\omega}^2$			
$E_{z,\omega}^2$			
$2E'_{y,\omega} E'_{z,\omega}$			
$2E'_{x,\omega} E'_{z,\omega}$			
$2E'_{x,\omega} E'_{y,\omega}$			

(5)

Use Eq. (3), (4) and (5) to derive expressions for the  $P'_{x,2\omega}$ ,  $P'_{y,2\omega}$  and  $P'_{z,2\omega}$  components of the polarization after the various rotations and compare them to the original expressions before rotation. Use the concept express by Eq. (2) to compare the coefficients associated with the various products of the electric fields and find the constraints on the matrix components  $d_{ij}$ . Identify the non-zero elements of the matrix.

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