

Lecture 8: Polarization in nonlinear optics.

Nonlinear susceptibilities: tensor description. Second-order susceptibility and the nonlinear susceptibility tensor, its symmetry. Phase matching. Effective susceptibility.

Nonlinear optics describes frequency conversion of light, due to the nonlinearity of the electromagnetic response of the matter. In this course, it is assumed that you have some basic knowledge of nonlinear optics; here we will only consider the polarization aspects.

1. Nonlinear susceptibilities: tensor description.

The main equation in nonlinear optics is the dependence of the polarization on the field,

$$\vec{P} = \varepsilon_0 \hat{\chi}^{(1)} \cdot \vec{E} + \varepsilon_0 \hat{\chi}^{(2)} : \vec{E}\vec{E} + \varepsilon_0 \hat{\chi}^{(3)} : \vec{E}\vec{E}\vec{E} + \dots \quad (1)$$

where $\hat{\chi}^{(n)}$ is the n^{th} -order nonlinear susceptibility, a tensor of rank $n+1$.

Alternatively, (1) can be written as

$$P_i = \varepsilon_0 \chi_{ij}^{(1)} E_j + \varepsilon_0 \chi_{ijk}^{(2)} E_j E_k + \varepsilon_0 \chi_{ijkl}^{(3)} E_j E_k E_l + \dots, \quad (1')$$

where repeating indices imply summation.

We already discussed the first-order susceptibility, to which the linear dielectric tensor is related. Indeed, because the displacement is related to the electric field as

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 \hat{\varepsilon} \vec{E}, \quad (2)$$

and the linear polarization is related to the field as

$$\vec{P}^{(1)} = \varepsilon_0 \hat{\chi}^{(1)} \vec{E},$$

we get for the linear dielectric tensor

$$\hat{\varepsilon}^{(1)} = \hat{I} + \hat{\chi}^{(1)}, \quad (3)$$

where \hat{I} is the unity matrix, or

$$\varepsilon_{ij}^{(1)} = \delta_{ij} + \chi_{ij}^{(1)}. \quad (3')$$

We already know that this tensor is symmetric, hence $\chi_{ij}^{(1)}$ is also symmetric,

$$\chi_{ij}^{(1)} = \chi_{ji}^{(1)}.$$

We also know that by choosing the frame of reference appropriately (corresponding to the symmetry of the matter), the number of tensor elements reduces to 3, $\chi_x^{(1)}$, $\chi_y^{(1)}$, $\chi_z^{(1)}$, or less.

Consider now the second-order susceptibility $\chi_{ijk}^{(2)}$. It makes sense to discuss some particular case of frequency conversion, for instance, conversion from frequencies ω_n and ω_m to frequencies $\omega_n + \omega_m$ (all frequencies here can be negative as well). The polarization at frequency $\omega_n + \omega_m$ is

$$P_i^{(2)}(\omega_n + \omega_m) = \varepsilon_0 \sum_{jk} \sum_{nm} \chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m), \quad (4)$$

where we stressed that the value of the susceptibility depends on the process we consider.

For instance, second harmonic generation and parametric down-conversion will correspond to $\chi_{ijk}^{(2)}(2\omega, \omega, \omega)$; optical rectification, to $\chi_{ijk}^{(2)}(0, \omega, -\omega)$; difference frequency generation, to $\chi_{ijk}^{(2)}(\omega_1 - \omega_2, \omega_1, \omega_2)$.

Let us consider the most general case, where all frequencies are different: $\omega_3 = \omega_1 + \omega_2$. How many values of the tensor $\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m)$ are there?

Without any restrictions, there will be 6 tensors, $\chi_{ijk}^{(2)}(\omega_1, \omega_3, -\omega_2)$; $\chi_{ijk}^{(2)}(\omega_1, -\omega_2, \omega_3), \dots, \chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2)$, plus the tensors with all frequencies with opposite signs, and each having 27 permutations w.r.t. i, j, k - altogether $12 \times 27 = 324$ complex tensors.

But fortunately there are restrictions.

1. Because the fields are real,

$$\chi_{ijk}^{(2)}(-\omega_n - \omega_m, -\omega_n, -\omega_m) = [\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m)]^* \quad (5)$$

2. *Permutation symmetry*. From the definition of the process (4), it follows that the frequencies can be interchanged together with the indices, for instance

$$\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m) = \chi_{ikj}^{(2)}(\omega_n + \omega_m, \omega_m, \omega_n). \quad (6)$$

In a lossless medium, additional symmetries appear.

First, the susceptibility should be a real tensor, then, from (5),

$$\chi_{ijk}^{(2)}(-\omega_n - \omega_m, -\omega_n, -\omega_m) = \chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m). \quad (7)$$

Second, there is a *full permutation symmetry*. Indeed, we can exchange according to (6), the frequencies with the indices, so that the first one is the sum of the other two,

$$\chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2) = \chi_{jik}^{(2)}(-\omega_1, -\omega_3, \omega_2).$$

But the sign of the frequencies can be changed together with complex conjugation, and the susceptibilities are real; therefore,

$$\chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2) = \chi_{jik}^{(2)}(\omega_1, \omega_3, -\omega_2) = \chi_{kij}^{(2)}(\omega_2, \omega_3, -\omega_1). \quad (8)$$

Kleinman's symmetry. This symmetry follows from a very strong assumption, namely that there is no dispersion. Usually, this approximation is valid, *roughly*, in the middle of the visible – NIR range (Fig.1).

Then, it does not matter which frequency stands on which place. One can write

$$\begin{aligned} \chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2) &= \chi_{jik}^{(2)}(\omega_3, \omega_1, \omega_2) = \dots = \\ &= \chi_{kij}^{(2)}(\omega_3, \omega_1, \omega_2), \end{aligned} \quad (9)$$

i.e., all possible permutations of the indices.

This assumption is really too strong; but for effects like second harmonic generation, it is always possible to permute the last two indices. In this case, as well as in the presence of Kleinman's symmetry, contracted notation is used.

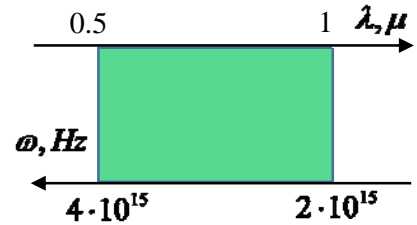


Fig.1

Contracted notation. It is convenient to pass from the $\chi_{ijk}^{(2)}$ tensor to another tensor, where the last two indices are replaced by one:

$$d_{il}(\omega_3, \omega_1, \omega_2) = \frac{1}{2} \chi_{jik}^{(2)}(\omega_3, \omega_1, \omega_2), \quad (10)$$

where the factor $\frac{1}{2}$ is added historically and there is a correspondence between the l index and the combination of j, k indices:

$$\begin{array}{l} jk : 11 \quad 22 \quad 33 \quad 23,32 \quad 13,31 \quad 12,21 \\ l : 1 \quad 2 \quad 3 \quad 4 \quad 5 \quad 6 \end{array}$$

Accordingly, the *nonlinear susceptibility tensor* has the form

$$d_{il} = \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix}. \quad (11)$$

Now, if the Kleinman symmetry is valid, some of these 18 components turn out to be equal. For instance,

$$\begin{aligned} d_{12} &= \frac{1}{2} \chi_{122}^{(2)} = \frac{1}{2} \chi_{212}^{(2)} = d_{26}; \\ d_{13} &= \frac{1}{2} \chi_{133}^{(2)} = \frac{1}{2} \chi_{313}^{(2)} = d_{35}; \\ d_{23} &= \frac{1}{2} \chi_{233}^{(2)} = \frac{1}{2} \chi_{323}^{(2)} = d_{34}, \dots \end{aligned}$$

Then, we find that there are only 10 components of the tensor:

$$d_{il} = \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix}. \quad (12)$$

Furthermore, for a certain crystal some components of this tensor will be zero because of symmetry. (And of course for crystals with center of symmetry, there is no quadratic susceptibility.)

For instance, for crystals belonging to the 3m point group (for instance, lithium niobate crystal), the tensor has the form

$$d_{LNB} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}. \quad (13)$$

In the general case of a nonlinear interaction, all waves can have different polarization states. For instance, in the simplest case of the second harmonic generation, Eq. (4) becomes

$$P_i^{(2)}(2\omega) = 2\varepsilon_0 \sum_{l=j,k} \chi_{ijk}^{(2)}(2\omega, \omega, \omega) E_j(\omega) E_k(\omega),$$

or, using the nonlinear susceptibility tensor,

$$\begin{pmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{pmatrix} = 2\varepsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix} \begin{pmatrix} E_x^2(\omega) \\ E_y^2(\omega) \\ E_z^2(\omega) \\ 2E_y(\omega)E_z(\omega) \\ 2E_x(\omega)E_z(\omega) \\ 2E_x(\omega)E_y(\omega) \end{pmatrix}. \quad (14)$$

As always in nonlinear optics, nonlinear polarization will generate the field at frequency 2ω according to the Helmholtz equation. This field will have the same direction as $\vec{P}(2\omega)$.

2. Phase matching.

Consider now the simplest nonlinear process, second-harmonic generation, in a uniaxial crystal (Fig. 2). The wavevectors for both first and second harmonic waves are at some angle ϑ to the optic axis z . Then, we can consider only the principal plane, the one formed by the wavevector \vec{k} and the optic axis z . We know that only two polarization directions are allowed for any \vec{k} direction, corresponding to the ordinary wave (green vector) and to the extraordinary wave (blue vector).

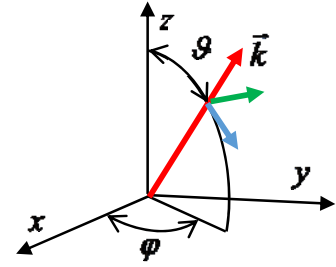


Fig. 2

Depending on the possible polarization states of the pump at frequency ω and the second-harmonic field at frequency 2ω , different types of interaction are distinguished:

- (i) oo->e: the pump is ordinary and the SH extraordinary.
- (ii) ee->o: the pump is extraordinary and the SH ordinary.
- (iii) ee->e: all extraordinary
- (iv) oo->o: all ordinary
- (v) eo->e: one pump photon is ordinary, the other extraordinary
- (vi) eo->o, similarly.

The cases (i) and (ii) are called type-I phase matching, the cases (v) and (vi), type-II, and the case (iii) and (iv), type-0.

Not all these types can be used in practice. This is because there should be phase matching satisfied for the waves of the first and the second harmonic,

$$k(2\omega) = 2k(\omega). \quad (15)$$

This follows from the Helmholtz equation and is required for the interaction to be accumulated as the waves propagate through the nonlinear material. Using the relation between the wavevector and frequency,

$$k = \frac{n(\omega)\omega}{c}, \quad (16)$$

we get the condition

$$n(2\omega) = n(\omega). \quad (17)$$

(Note that both (15) and (17) become more complicated in the case of other nonlinear effects – here we only consider the second harmonic generation.)

Due to the dispersion, this condition cannot be satisfied if the pump and the second harmonic are both ordinary or both extraordinary waves. For instance, consider SHG from 1064 nm to 532 nm in lithium niobate crystal. The refractive indices are given in the table, and the dispersion dependence is in Fig. 3. From Fig. 3, we see that the dispersion in the visible range is somewhat smaller than birefringence. We also see that $n_o(\omega) < n_o(2\omega)$, so both second and first harmonics cannot be o-waves (or both e-waves) but $n_e(2\omega) < n_o(\omega) < n_o(2\omega)$ (see the table and two blue points in the figure), so by choosing the angle ϑ to the optic axis one can make the effective index [see Eq. (6) of Lecture 6],

$$\frac{1}{n^2(2\omega)} = \frac{\sin^2 \vartheta}{n_e^2(2\omega)} + \frac{\cos^2 \vartheta}{n_o^2(2\omega)}, \quad (18)$$

take any value between $n_e(2\omega)$ and $n_o(2\omega)$.

wavelength	n_o	n_e
532 nm (2ω)	2.324	2.219
1064 nm (ω)	2.234	2.146

Then $n(2\omega) = n_o(\omega)$, and the type-I oo->e phase matching is satisfied. Similarly, the type I phase matching ee->o can be satisfied in a positive uniaxial crystal by choosing the angle \mathcal{G} .

3. Effective susceptibility.

For a given phase matching, one can calculate the relation between the polarization and the field and find the effective susceptibility tensor, which will be not just one single tensor element. One uses for this Eq. (14), which we will now write, as an example, for the case of lithium niobate:

$$\begin{pmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{pmatrix} = 2\varepsilon_0 \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_x^2(\omega) \\ E_y^2(\omega) \\ E_z^2(\omega) \\ 2E_y(\omega)E_z(\omega) \\ 2E_x(\omega)E_z(\omega) \\ 2E_x(\omega)E_y(\omega) \end{pmatrix},$$

which yields

$$\begin{aligned} P_x(2\omega) &= 4\varepsilon_0 d_{31} E_x(\omega) E_z(\omega) - 4\varepsilon_0 d_{22} E_x(\omega) E_y(\omega), \\ P_y(2\omega) &= -2\varepsilon_0 d_{22} E_x^2(\omega) + 2\varepsilon_0 d_{22} E_y^2(\omega) + 4\varepsilon_0 d_{31} E_y(\omega) E_z(\omega), \\ P_z(2\omega) &= 2\varepsilon_0 d_{31} E_x^2(\omega) + 2\varepsilon_0 d_{31} E_y^2(\omega) + 2\varepsilon_0 d_{33} E_z^2(\omega). \end{aligned} \quad (19)$$

If oo->e geometry is used, the pump is o, and the SH, e:

$$\vec{E}(\omega) = E(\omega) \begin{pmatrix} -\sin \varphi \\ \cos \varphi \\ 0 \end{pmatrix}, \quad (20)$$

$$\vec{E}(2\omega) = E(2\omega) \begin{pmatrix} \cos \mathcal{G} \cos \varphi \\ \cos \mathcal{G} \sin \varphi \\ -\sin \mathcal{G} \end{pmatrix} \quad (21)$$

(we ignored the walk-off angle here). Substituting (20) into (19), we get

$$\begin{aligned} P_x(2\omega) &= 4\varepsilon_0 d_{22} E^2(\omega) \sin \varphi \cos \varphi = 2\varepsilon_0 d_{22} E^2(\omega) \sin 2\varphi, \\ P_y(2\omega) &= -2\varepsilon_0 d_{22} E^2(\omega) \sin^2 \varphi + 2\varepsilon_0 d_{22} E^2(\omega) \cos^2 \varphi = 2\varepsilon_0 d_{22} E^2(\omega) \cos 2\varphi, \\ P_z(2\omega) &= 2\varepsilon_0 d_{31} E^2(\omega) \sin^2 \varphi + 2\varepsilon_0 d_{31} E^2(\omega) \cos^2 \varphi = 2\varepsilon_0 d_{31} E^2(\omega). \end{aligned} \quad (22)$$

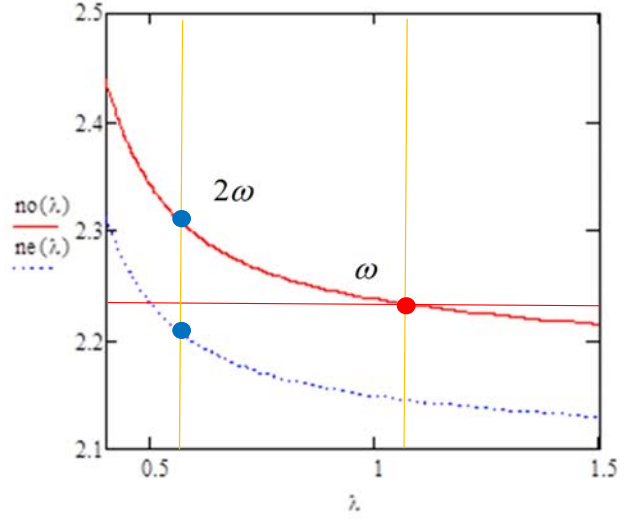


Fig. 3

These are the polarization vector components. They can take any values; but the only possible e-field direction to propagate in the crystal is given by Eq. (21). By projecting the polarization (22) on the unit vector given by (21), we get

$$\begin{aligned} P_e(2\omega) &= 2\varepsilon_0 E^2(\omega)[d_{22} \sin 2\varphi \cos \varphi \cos \mathcal{G} + d_{22} \cos 2\varphi \sin \varphi \cos \mathcal{G} - d_{31} \sin \mathcal{G}] = \\ &= 2\varepsilon_0 E^2(\omega)[d_{22} \sin 3\varphi \cos \mathcal{G} - d_{31} \sin \mathcal{G}] \end{aligned} \quad (23)$$

Then, the effective susceptibility for the oo->e phase matching is

$$d_{eff} = d_{22} \sin 3\varphi \cos \mathcal{G} - d_{31} \sin \mathcal{G}. \quad (24)$$

Literature:

1. R. W. Boyd, Nonlinear Optics, sections 1.5.
2. V. G. Dmitriev, G. G. Gurzadyan, D. N. Nikogosyan, Handbook of Nonlinear Optical Crystals, sections 2.10, 3.1.8.