Notes on the "Butcher and Cotter convention" in nonlinear optics

Convention for description of nonlinear optical polarization

As a "recipe" in theoretical nonlinear optics, Butcher and Cotter provide a very useful convention which is well worth to hold on to. For a superposition of monochromatic waves, and by invoking the general property of the intrinsic permutation symmetry, the monochromatic form of the nth order polarization density can be written as

$$(P^{(n)}_{\omega_\sigma})_\mu = \varepsilon_0 \sum_{\alpha_1} \cdots \sum_{\alpha_n} \sum_{\omega} K(-\omega_\sigma; \omega_1, \ldots, \omega_n) \chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma; \omega_1, \ldots, \omega_n)(E_{\omega_1})_{\alpha_1} \cdots (E_{\omega_n})_{\alpha_n}. \tag{1}$$

The first summations in Eq. (1), over $\alpha_1, \ldots, \alpha_n$, is simply an explicit way of stating that the Einstein convention of summation over repeated indices holds. The summation sign $\sum_\omega$, however, serves as a reminder that the expression that follows is to be summed over *all distinct sets of* $\omega_1, \ldots, \omega_n$. Because of the intrinsic permutation symmetry, the frequency arguments appearing in Eq. (1) may be written in arbitrary order.

By "all distinct sets of $\omega_1, \ldots, \omega_n$", we here mean that the summation is to be performed, as for example in the case of optical Kerr-effect, over the single set of nonlinear susceptibilities that contribute to a certain angular frequency as $(-\omega; \omega, \omega, -\omega)$ *or* $(-\omega; \omega, -\omega, \omega)$ *or* $(-\omega; -\omega, \omega, \omega)$. In this example, each of the combinations are considered as *distinct*, and it is left as an arbitary choice which one of these sets that are most convenient to use (this is simply a matter of choosing notation, and does not by any means change the description of the interaction).

In Eq. (1), the degeneracy factor K is formally described as

$$K(-\omega_\sigma; \omega_1, \ldots, \omega_n) = 2^{l+m-n} p$$

where

$$\begin{aligned} p &= \text{the number of } \textit{distinct} \text{ permutations of } \omega_1, \omega_2, \ldots, \omega_1, \\ n &= \text{the order of the nonlinearity}, \\ m &= \text{the number of angular frequencies } \omega_k \text{ that are zero, and} \\ l &= \begin{cases} 1, & \text{if } \omega_\sigma \neq 0, \\ 0, & \text{otherwise.} \end{cases} \end{aligned}$$

In other words, m is the number of DC electric fields present, and $l = 0$ if the nonlinearity we are analyzing gives a static, DC, polarization density, such as in the previously (in the spring model) described case of optical rectification in the presence of second harmonic fields (SHG).

A list of frequently encountered nonlinear phenomena in nonlinear optics, including the degeneracy factors as conforming to the above convention, is given in Butcher and Cotters book, Table 2.1, on page 26.

Note on the complex representation of the optical field

Since the observable electric field of the light, in Butcher and Cotters notation taken as

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{2} \sum_{\omega_k \geq 0} [\mathbf{E}_{\omega_k} \exp(-i\omega_k t) + \mathbf{E}^*_{\omega_k} \exp(i\omega_k t)],$$

is a real-valued quantity, it follows that negative frequencies in the complex notation should be interpreted as the complex conjugate of the respective field component, or

$$\mathbf{E}_{-\omega_k} = \mathbf{E}^*_{\omega_k}.$$

Example: Optical Kerr-effect

Assume a monochromatic optical wave (containing forward and/or backward propagating components) polarized in the xy-plane,

$$\mathbf{E}(z,t) = \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)] \in \mathbb{R}^3,$$

with all spatial variation of the field contained in

$$\mathbf{E}_\omega(z) = \mathbf{e}_x E^x_\omega(z) + \mathbf{e}_y E^y_\omega(z) \in \mathbb{C}^3.$$

Optical Kerr-effect is in isotropic media described by the third order susceptibility

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega),$$

with nonzero components of interest for the xy-polarized beam given in Appendix 3.3 of Butcher and Cotters book as

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy}, \quad \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx} = \left\{ \begin{array}{c} \text{intr. perm. symm.} \\ (\alpha,\omega) \rightleftharpoons (\beta,\omega) \end{array} \right\} = \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx}, \quad \chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy},$$

with

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx}.$$

The degeneracy factor $K(-\omega;\omega,\omega,-\omega)$ is calculated as

$$K(-\omega;\omega,\omega,-\omega) = 2^{l+m-n}p = 2^{1+0-3}3 = 3/4.$$

From this set of nonzero susceptibilities, and using the calculated value of the degeneracy factor in the convention of Butcher and Cotter, we hence have the third order electric polarization density at $\omega_\sigma = \omega$ given as $\mathbf{P}^{(n)}(\mathbf{r},t) = \mathrm{Re}[\mathbf{P}^{(n)}_\omega \exp(-i\omega t)]$, with

$$\begin{aligned}
\mathbf{P}^{(3)}_\omega &= \sum_\mu \mathbf{e}_\mu (P^{(3)}_\omega)_\mu \\
&= \{\text{Using the convention of Butcher and Cotter}\} \\
&= \sum_\mu \mathbf{e}_\mu \left[\varepsilon_0 \frac{3}{4} \sum_\alpha \sum_\beta \sum_\gamma \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega)(E_\omega)_\alpha (E_\omega)_\beta (E_{-\omega})_\gamma \right] \\
&= \{\text{Evaluate the sums over } (x,y,z) \text{ for field polarized in the } xy \text{ plane}\} \\
&= \varepsilon_0 \frac{3}{4} \{ \mathbf{e}_x [\chi^{(3)}_{xxxx} E^x_\omega E^x_\omega E^x_{-\omega} + \chi^{(3)}_{xyyx} E^y_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{xyxy} E^y_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{xxyy} E^x_\omega E^y_\omega E^y_{-\omega}] \\
&\qquad + \mathbf{e}_y [\chi^{(3)}_{yyyy} E^y_\omega E^y_\omega E^y_{-\omega} + \chi^{(3)}_{yxxy} E^x_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{yxyx} E^x_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{yyxx} E^y_\omega E^x_\omega E^x_{-\omega}] \} \\
&= \{\text{Make use of } \mathbf{E}_{-\omega} = \mathbf{E}^*_\omega \text{ and relations } \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx}, \text{ etc.}\} \\
&= \varepsilon_0 \frac{3}{4} \{ \mathbf{e}_x [\chi^{(3)}_{xxxx} E^x_\omega |E^x_\omega|^2 + \chi^{(3)}_{xyyx} E^{y\,2}_\omega E^{x*}_\omega + \chi^{(3)}_{xyxy} |E^y_\omega|^2 E^x_\omega + \chi^{(3)}_{xxyy} E^x_\omega |E^y_\omega|^2] \\
&\qquad + \mathbf{e}_y [\chi^{(3)}_{xxxx} E^y_\omega |E^y_\omega|^2 + \chi^{(3)}_{xyyx} E^{x\,2}_\omega E^{y*}_\omega + \chi^{(3)}_{xyxy} |E^x_\omega|^2 E^y_\omega + \chi^{(3)}_{xxyy} E^y_\omega |E^x_\omega|^2] \} \\
&= \{\text{Make use of intrinsic permutation symmetry}\} \\
&= \varepsilon_0 \frac{3}{4} \{ \mathbf{e}_x [(\chi^{(3)}_{xxxx} |E^x_\omega|^2 + 2\chi^{(3)}_{xxyy} |E^y_\omega|^2) E^x_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy}) E^{y\,2}_\omega E^{x*}_\omega \\
&\qquad \mathbf{e}_y [(\chi^{(3)}_{xxxx} |E^y_\omega|^2 + 2\chi^{(3)}_{xxyy} |E^x_\omega|^2) E^y_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy}) E^{x\,2}_\omega E^{y*}_\omega .
\end{aligned}$$

For the optical field being linearly polarized, say in the x-direction, the expression for the polarization density is significantly simplified, to yield

$$\mathbf{P}^{(3)}_\omega = \varepsilon_0 (3/4) \mathbf{e}_x \chi^{(3)}_{xxxx} |E^x_\omega|^2 E^x_\omega,$$

i. e. taking a form that can be interpreted as an intensity-dependent ($\sim |E^x_\omega|^2$) contribution to the refractive index (cf. Butcher and Cotter §6.3.1).