

1.7. NONLINEAR OPTICAL PROPERTIES

The direct and inverse Fourier transforms of the field are defined as

$$\mathbf{E}(t) = \int_{-\infty}^{+\infty} d\omega \mathbf{E}(\omega) \exp(-i\omega t) \quad (1.7.2.15)$$

$$\mathbf{E}(\omega) = (1/2\pi) \int_{-\infty}^{+\infty} dt \mathbf{E}(t) \exp(i\omega t), \quad (1.7.2.16)$$

where $\mathbf{E}(\omega)^* = \mathbf{E}(-\omega)$ as $\mathbf{E}(t)$ is real.

1.7.2.1.2.1. Linear susceptibility

By substitution of (1.7.2.15) in (1.7.2.7),

$$\begin{aligned} \mathbf{P}^{(1)}(t) &= \varepsilon_o \int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\tau R^{(1)}(\tau) \cdot \mathbf{E}(\omega) \exp[-i\omega(t - \tau)] \\ \mathbf{P}^{(1)}(t) &= \varepsilon_o \int_{-\infty}^{+\infty} d\omega \chi^{(1)}(-\omega_\sigma; \omega) \mathbf{E}(\omega) \exp(-i\omega_\sigma t), \end{aligned} \quad (1.7.2.17)$$

where

$$\chi^{(1)}(-\omega_\sigma; \omega) = \int_{-\infty}^{+\infty} d\tau R^{(1)}(\tau) \exp(i\omega\tau).$$

In these equations, $\omega_\sigma = \omega$ to satisfy the energy conservation condition that will be generalized in the following. In order to ensure convergence of $\chi^{(1)}$, ω has to be taken in the upper half plane of the complex plane. The reality of $R^{(1)}$ implies that $\chi^{(1)}(-\omega_\sigma; \omega)^* = \chi^{(1)}(\omega_\sigma^*; -\omega^*)$.

1.7.2.1.2.2. Second-order susceptibility

Substitution of (1.7.2.15) in (1.7.2.12) yields

$$\begin{aligned} \mathbf{P}^{(2)}(t) &= \varepsilon_o \int_{-\infty}^{+\infty} d\omega_1 \int_{-\infty}^{+\infty} d\omega_2 \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2 R^{(2)}(\tau_1, \tau_2) \\ &\cdot \mathbf{E}(\omega_1) \otimes \mathbf{E}(\omega_2) \exp\{-i[\omega_1(t - \tau_1) + \omega_2(t - \tau_2)]\} \end{aligned} \quad (1.7.2.18)$$

or

$$\begin{aligned} \mathbf{P}^{(2)}(t) &= \varepsilon_o \int_{-\infty}^{+\infty} d\omega_1 \int_{-\infty}^{+\infty} d\omega_2 \chi^{(2)}(-\omega_\sigma; \omega_1, \omega_2) \cdot \mathbf{E}(\omega_1) \otimes \mathbf{E}(\omega_2) \\ &\times \exp(-i\omega_\sigma t) \end{aligned} \quad (1.7.2.19)$$

with

$$\begin{aligned} \chi^{(2)}(-\omega_\sigma; \omega_1, \omega_2) &= \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2 R^{(2)}(\tau_1, \tau_2) \\ &\times \exp[i(\omega_1\tau_1 + \omega_2\tau_2)] \end{aligned}$$

and $\omega_\sigma = \omega_1 + \omega_2$. Frequencies ω_1 and ω_2 must be in the upper half of the complex plane to ensure convergence. Reality of $R^{(2)}$ implies $\chi^{(2)}(-\omega_\sigma; \omega_1, \omega_2)^* = \chi^{(2)}(\omega_\sigma^*; -\omega_1^*, -\omega_2^*)$. $\chi_{\mu\alpha\beta}^{(2)}(-\omega_\sigma; \omega_1, \omega_2)$ is invariant under the interchange of the (α, ω_1) and (β, ω_2) pairs.

 1.7.2.1.2.3. n th-order susceptibility

Substitution of (1.7.2.15) in (1.7.2.14) provides

$$\begin{aligned} \mathbf{P}^{(n)}(t) &= \varepsilon_o \int_{-\infty}^{+\infty} d\omega_1 \int_{-\infty}^{+\infty} d\omega_2 \dots \int_{-\infty}^{+\infty} d\omega_n \chi^{(n)}(-\omega_\sigma; \omega_1, \omega_2, \dots, \omega_n) \\ &\cdot \mathbf{E}(\omega_1) \otimes \mathbf{E}(\omega_2) \otimes \dots \otimes \mathbf{E}(\omega_n) \exp(-i\omega_\sigma t) \end{aligned} \quad (1.7.2.20)$$

where

$$\begin{aligned} \chi^{(n)}(-\omega_\sigma; \omega_1, \omega_2, \dots, \omega_n) &= \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2 \dots \int_{-\infty}^{+\infty} d\tau_n R^{(n)}(\tau_1, \tau_2, \dots, \tau_n) \exp\left(i \sum_{j=1}^n \omega_j \tau_j\right) \end{aligned} \quad (1.7.2.21)$$

and $\omega_\sigma = \omega_1 + \omega_2 + \dots + \omega_n$.

All frequencies must lie in the upper half complex plane and reality of $\chi^{(n)}$ imposes

$$\chi^{(n)}(-\omega_\sigma; \omega_1, \omega_2, \dots, \omega_n)^* = \chi^{(n)}(\omega_\sigma^*; -\omega_1^*, -\omega_2^*, \dots, -\omega_n^*). \quad (1.7.2.22)$$

Intrinsic permutation symmetry implies that $\chi_{\mu\alpha_1\alpha_2\dots\alpha_n}^{(n)}(-\omega_\sigma; \omega_1, \omega_2, \dots, \omega_n)$ is invariant with respect to the $n!$ permutations of the (α_i, ω_i) pairs.

1.7.2.1.3. Superposition of monochromatic waves

Optical fields are often superpositions of monochromatic waves which, due to spectral discretization, will introduce considerable simplifications in previous expressions such as (1.7.2.20) relating the induced polarization to a continuous spectral distribution of polarizing field amplitudes.

The Fourier transform of the induced polarization is given by

$$\mathbf{P}^{(n)}(\omega) = (1/2\pi) \int_{-\infty}^{+\infty} dt \mathbf{P}^{(n)}(t) \exp(i\omega t). \quad (1.7.2.23)$$

Replacing $\mathbf{P}^{(n)}(t)$ by its expression as from (1.7.2.20) and applying the well known identity

$$(1/2\pi) \int_{-\infty}^{+\infty} dt \exp[i(\omega - \omega_\sigma)t] = \delta(\omega - \omega_\sigma) \quad (1.7.2.24)$$

leads to

$$\begin{aligned} \mathbf{P}^{(n)}(\omega) &= \varepsilon_o \int_{-\infty}^{+\infty} d\omega_1 \int_{-\infty}^{+\infty} d\omega_2 \dots \int_{-\infty}^{+\infty} d\omega_n \chi^{(n)}(-\omega_\sigma; \omega_1, \omega_2, \dots, \omega_n) \\ &\times \mathbf{E}(\omega_1) \mathbf{E}(\omega_2) \dots \mathbf{E}(\omega_n) \delta(\omega - \omega_\sigma). \end{aligned} \quad (1.7.2.25)$$

In practical cases where the applied field is a superposition of monochromatic waves

$$\mathbf{E}(t) = (1/2) \sum_{\omega'} [E_{\omega'} \exp(-i\omega' t) + E_{-\omega'} \exp(i\omega' t)] \quad (1.7.2.26)$$

with $E_{-\omega'} = E_{\omega'}^*$. By Fourier transformation of (1.7.2.26)

$$\mathbf{E}(\omega) = (1/2) \sum_{\omega'} [E_{\omega'} \delta(\omega - \omega') + E_{-\omega'} \delta(\omega + \omega')]. \quad (1.7.2.27)$$

The optical intensity for a wave at frequency ω' is related to the squared field amplitude by

$$I_{\omega'} = \varepsilon_o c n(\omega') (\mathbf{E}^2(t))_t = \frac{1}{2} \varepsilon_o c n(\omega') |E_{\omega'}|^2. \quad (1.7.2.28)$$

The averaging as represented above by brackets is performed over a time cycle and $n(\omega')$ is the index of refraction at frequency ω' .

1.7.2.1.4. Conventions for nonlinear susceptibilities

1.7.2.1.4.1. Classical convention

Insertion of (1.7.2.26) in (1.7.2.25) together with permutation symmetry provides

$$\begin{aligned} P_{\mu}^{(n)}(\omega_\sigma) &= \varepsilon_o \sum_{\alpha_1\alpha_2\dots\alpha_n} \sum_{\omega} K(-\omega_\sigma; \omega_1, \omega_2, \dots, \omega_n) \\ &\times \chi_{\mu\alpha_1\alpha_2\dots\alpha_n}^{(n)}(-\omega_\sigma; \omega_1, \omega_2, \dots, \omega_n) \\ &\times E_{\alpha_1}(\omega_1) E_{\alpha_2}(\omega_2) \dots E_{\alpha_n}(\omega_n), \end{aligned} \quad (1.7.2.29)$$