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

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

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

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),

$$\mathbf{P}^{(1)}(t) = \varepsilon_o \int_{-\infty}^{+\infty} \mathrm{d}\omega \int_{-\infty}^{+\infty} \mathrm{d}\tau\ R^{(1)}(\tau)\cdot\mathbf{E}(\omega)\exp[-i\omega(t-\tau)]$$

$$\mathbf{P}^{(1)}(t) = \varepsilon_o \int_{-\infty}^{+\infty} \mathrm{d}\omega\ \chi^{(1)}(-\omega_\sigma;\omega)\mathbf{E}(\omega)\exp(-i\omega_\sigma t), \qquad (1.7.2.17)$$

where

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

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

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

or

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

with

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

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 $\chi^{(2)}(-\omega_\sigma;\omega_1,\omega_2)^* = \chi^{(2)}(\omega_\sigma^*;-\omega_1^*,-\omega_2^*)$. $\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)$ is invariant under the interchange of the (α,ω_1) and (β,ω_2) pairs.

1.7.2.1.2.3. *nth-order susceptibility*

Substitution of (1.7.2.15) in (1.7.2.14) provides

$$\mathbf{P}^{(n)}(t) = \varepsilon_o \int_{-\infty}^{+\infty} \mathrm{d}\omega_1 \int_{-\infty}^{+\infty} \mathrm{d}\omega_2 \ldots \int_{-\infty}^{+\infty} \mathrm{d}\omega_n\ \chi^{(n)}(-\omega_\sigma;\omega_1,\omega_2,\ldots\omega_n) \cdot \mathbf{E}(\omega_1)\otimes\mathbf{E}(\omega_2)\otimes\ldots\otimes\mathbf{E}(\omega_n)\exp(-i\omega_\sigma t) \qquad (1.7.2.20)$$

where

$$\chi^{(n)}(-\omega_\sigma;\omega_1,\omega_2,\ldots,\omega_n) = \int_{-\infty}^{+\infty} \mathrm{d}\tau_1 \int_{-\infty}^{+\infty} \mathrm{d}\tau_2 \ldots \int_{-\infty}^{+\infty} \mathrm{d}\tau_n\ R^{(n)}(\tau_1,\tau_2,\ldots,\tau_n)\exp\Big(i\sum_{j=1}^{n}\omega_j\tau_j\Big) \qquad (1.7.2.21)$$

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

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

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

Intrinsic permutation symmetry implies that $\chi^{(n)}_{\mu\alpha_1\alpha_2\ldots\alpha_n}(-\omega_\sigma;\omega_1,\omega_2,\ldots,\omega_n)$ 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

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

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

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

leads to

$$\mathbf{P}^{(n)}(\omega) = \varepsilon_o \int_{-\infty}^{+\infty} \mathrm{d}\omega_1 \int_{-\infty}^{+\infty} \mathrm{d}\omega_2 \ldots \int_{-\infty}^{+\infty} \mathrm{d}\omega_n\ \chi^{(n)}(-\omega_\sigma;\omega_1,\omega_2,\ldots\omega_n) \times \mathbf{E}(\omega_1)\mathbf{E}(\omega_2)\ldots\mathbf{E}(\omega_n)\delta(\omega-\omega_\sigma). \qquad (1.7.2.25)$$

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

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

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

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

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

$$I_{\omega'} = \varepsilon_o cn(\omega')\langle\mathbf{E}^2(t)\rangle_t = \tfrac{1}{2}\varepsilon_o cn(\omega')|E_{\omega'}|^2. \qquad (1.7.2.28)$$

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

$$P^{(n)}_\mu(\omega_\sigma) = \varepsilon_o \sum_{\alpha_1\alpha_2\ldots\alpha_n}\sum_{\omega} K(-\omega_\sigma;\omega_1,\omega_2,\ldots,\omega_n) \times \chi^{(n)}_{\mu\alpha_1\alpha_2\ldots\alpha_n}(-\omega_\sigma;\omega_1,\omega_2,\ldots,\omega_n) \times E_{\alpha_1}(\omega_1)E_{\alpha_2}(\omega_2)\ldots E_{\alpha_n}(\omega_n), \qquad (1.7.2.29)$$

references