Theoretical Fundamentals

of Nonlinear Optics :

Stationary case

H.-H. Ritze (Seminar of Div. A, MBI, 8 November 2000)

Contents

Maxwell equations

$$c \cdot \text{rot}\, \mathbf{H} = 4\pi\, \mathbf{j} + \dot{\mathbf{D}}$$

$$c \cdot \text{rot}\, \mathbf{E} = - \dot{\mathbf{B}}$$

$$\mathbf{j} = \sigma\, \mathbf{E} \quad , \quad \mathbf{D} = \varepsilon\, \mathbf{E} = \mathbf{E} + 4\pi\, \mathbf{P}$$

$$\sigma = 0 \;\rightarrow \mathbf{j} = 0$$

$$\mathbf{B} \cong \mathbf{H}$$

$$\rightarrow \text{rot rot}\, \mathbf{E} = - \ddot{\mathbf{D}} / c^2$$

$$\boxed{\text{rot rot}\, \mathbf{E}(t) + \ddot{\mathbf{E}}(t) / c^2 = 4\pi\, \ddot{\mathbf{P}}(t) / c^2}$$

Response function in the time domain (local response)

Expansion of the polarization of the medium (power series in the field):

$$\mathbf{P}(t) = \mathbf{P}^{(0)}(t) + \mathbf{P}^{(1)}(t) + \mathbf{P}^{(2)}(t) + ...$$

Linear response

$$\mathbf{P}^{(1)}(t) = \int_{-\infty}^{+\infty} d\tau\, \mathbf{T}^{(1)}(t;\tau)\, \mathbf{E}(\tau)$$

or: $$P_\mu^{(1)}(t) = \int_{-\infty}^{+\infty} d\tau\, T_{\mu\alpha}^{(1)}(t;\tau)\, E_\alpha(\tau) \qquad (\mu, \alpha = x, y, z)$$

(Summation over the indices appearing twice, here: α)

$$\rightarrow \quad P_x^{(1)}(t) =$$

$$\int_{-\infty}^{+\infty} d\tau\, [T_{xx}^{(1)}(t;\tau)E_x(\tau) + T_{xy}^{(1)}(t;\tau)E_y(\tau) + T_{xz}^{(1)}(t;\tau)E_z(\tau)]$$

Time invariance:

$$\mathbf{T}^{(1)}(t+t_0;\tau) = \mathbf{T}^{(1)}(t;\tau-t_0)$$

$t = 0,\ t_0 \rightarrow t$: $\mathbf{T}^{(1)}(t;\tau) = \mathbf{T}^{(1)}(0;\tau-t)$

$$\boxed{\mathbf{P}^{(1)}(t) = \int_{0}^{+\infty} d\tau\, \mathbf{R}^{(1)}(\tau)\, \mathbf{E}(t-\tau)} \qquad \text{(causality !)}$$

Quadratic response

$$\mathbf{P}^{(2)}(t) = \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2 \, \mathbf{T}^{(2)}(t;\tau_1,\tau_2) \, \mathbf{E}(\tau_1) \, \mathbf{E}(\tau_2)$$

$$\rightarrow \; P_\mu^{(2)}(t) = \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2 \, T_{\mu\alpha\beta}^{(2)}(t;\tau_1,\tau_2) \, E_\alpha(\tau_1) \, E_\beta(\tau_2)$$

Definition: $T_{\mu\alpha\beta}^{(2)}(t;\tau_1,\tau_2)$ is unique and symmetric,

i.e., $T_{\mu\alpha\beta}^{(2)}(t;\tau_1,\tau_2) = T_{\mu\beta\alpha}^{(2)}(t;\tau_2,\tau_1)$

Time invariance:

$$\mathbf{T}^{(2)}(t;\tau_1,\tau_2) \equiv \mathbf{R}^{(2)}(t-\tau_1,t-\tau_2)$$

$$\boxed{\mathbf{P}^{(2)}(t) = \int_{0}^{+\infty} d\tau_1 \int_{0}^{+\infty} d\tau_2 \, \mathbf{R}^{(2)}(\tau_1,\tau_2) \, \mathbf{E}(t-\tau_1) \, \mathbf{E}(t-\tau_2)}$$

$\mathbf{R}^{(2)}(\tau_1,\tau_2)$ - quadratic polarization response function of the medium

Intrinsic permutation symmetry: Invariance of $R_{\mu\alpha\beta}^{(2)}(\tau_1,\tau_2)$ with respect to $(\alpha,\tau_1) \leftrightarrow (\beta,\tau_2)$

Higher-order nonlinearities

$$\mathbf{P}^{(n)}(t) = \int_{-\infty}^{+\infty} d\tau_1 \ldots \int_{-\infty}^{+\infty} d\tau_n \, \mathbf{T}^{(n)}(t,\tau_1,\ldots,\tau_n)\, \mathbf{E}(\tau_1) \ldots \mathbf{E}(\tau_n)$$

Removal of the arbitrariness in the definition of $\mathbf{T}^{(n)}$:

$$T_{\mu\alpha_1 \ldots \alpha_n}^{(n)}(t;\tau_1,\ldots,\tau_n) = (1/n!)\, \mathbf{S}\, T_{\mu\alpha_1 \ldots \alpha_n}^{(n)}(t;\tau_1,\ldots\tau_n)$$

S indicates a summation over all tensors obtained by making the n! permutations of the pairs (α_1,τ_1), (α_2,τ_2),...,(α_n,τ_n).

Time invariance: $\mathbf{T}^{(n)}(t;\tau_1,\ldots,\tau_n) \equiv \mathbf{R}^{(n)}(t-\tau_1,\ldots,t-\tau_n)$

$$P_\mu^{(n)}(t)=\int_0^{+\infty} d\tau_1 \ldots \int_0^{+\infty} d\tau_n R_{\mu\alpha_1 \ldots \alpha_n}^{(n)}(\tau_1,\ldots,\tau_n) E_{\alpha_1}(t-\tau_1) \ldots E_{\alpha_n}(t-\tau_n)$$

$\mathbf{R}^{(n)}(\tau_1,\ldots,\tau_n)$ is a tensor of rank (n+1) and is a real function of the n time variables $\tau_1,\ldots,\tau_n$.

Susceptibility tensors

Complex frequency plane

$$\mathbf{E}(t) = \int_{-\infty}^{+\infty} d\omega\, \mathbf{E}(\omega) \exp(-i\omega t) \; ; \qquad \mathbf{E}(t) \text{ is real}$$

$$\downarrow$$

$$\mathbf{E}(\omega) = (1/2\pi) \int_{-\infty}^{+\infty} d\omega\, \mathbf{E}(\tau) \exp(i\omega\tau); \quad [\mathbf{E}(\omega)]^{*} = \mathbf{E}(-\omega^{*})$$

Linear susceptibility

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

$$\boxed{\chi^{(1)}(-\omega_\sigma;\omega) = \int_{0}^{+\infty} d\tau\, \mathbf{R}^{(1)}(\tau) \exp(i\omega\tau)} \quad (\omega_\sigma = \omega)$$

Reality condition (**E**(t) and **P**(t) are real) for the linear susceptibility tensor:

$$[\chi^{(1)}(-\omega_\sigma;\omega)]^{*} = \chi^{(1)}(\omega_\sigma^{*}; -\omega^{*})$$

Second-order susceptibility

$$\mathbf{P}^{(2)}(t)=\int_{-\infty}^{+\infty}d\omega_1 \int_{-\infty}^{+\infty}d\omega_2\ \chi^{(2)}(-\omega_\sigma;\omega_1,\omega_2)\ \mathbf{E}(\omega_1)\mathbf{E}(\omega_2)\ \exp(-i\omega_\sigma t)$$

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

$$(\omega_\sigma=\omega_1+\omega_2)$$

$$\chi_{\mu\alpha\beta}^{(2)}(-\omega_\sigma;\omega_1,\omega_2) = \chi_{\mu\beta\alpha}^{(2)}(-\omega_\sigma;\omega_2,\omega_1)$$

nth-order susceptibility

$$\chi^{(n)}(-\omega_\sigma;\omega_1,\ldots,\omega_n) = \ \ldots$$

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

Reality condition:

$$[\chi^{(n)}(-\omega_\sigma;\omega_1,\ldots,\omega_n)]^* = \chi^{(n)}(\omega_\sigma^*;-\omega_1^*,\ldots,-\omega_n^*)$$

Intrinsic permutation symmetry: Invariance of $\chi_{\mu\alpha_1\ldots\alpha_n}^{(n)}(-\omega_\sigma;\omega_1,\ldots,\omega_n)$ under all n! permutations of the n pairs $(\alpha_1,\omega_1), (\alpha_2,\omega_2),\ldots, (\alpha_n,\omega_n)$.

Fourier components of the polarization

$$\mathbf{P}^{(n)}(\omega) = \int_{-\infty}^{+\infty} d\omega_1 \ldots \int_{-\infty}^{+\infty} d\omega_2\ \chi^{(n)}(-\omega_\sigma;\omega_1,\ldots,\omega_n)\ \mathbf{E}(\omega_1)\ldots\mathbf{E}(\omega_n)$$

$$\cdot\ \delta(\omega-\omega_\sigma)$$

Superposition of monochromatic waves:

$$\mathbf{E}(t) = ½ \sum_{\omega'\geq 0} [\ \mathbf{E}_{\omega'} \exp(-i\omega' t) + \mathbf{E}_{-\omega'} \exp(i\omega' t)]$$

$$\rightarrow \mathbf{P}^{(n)}(t) = ½ \sum_{\omega\geq 0} [\mathbf{P}_{\omega}^{(n)} \exp(-i\omega t) + \mathbf{P}_{-\omega}^{(n)} \exp(i\omega t)]$$

$$\mathbf{P}_{\omega}^{(n)} = \sum_{\omega} K(-\omega_\sigma;\omega_1,\ldots,\omega_n)\ \chi^{(\mathbf{n})}(-\omega_\sigma;\omega_1,\ldots,\omega_n)\ \mathbf{E}_{\omega 1}\ldots\mathbf{E}_{\omega n}$$

Summation over all distinct sets $\omega_1,\ldots,\omega_n$.

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

p - number of distinct permutations $\omega_1,\ldots,\omega_n$
n - order of nonlinearity
m - number of the d.c. fields
l = 1 if $\omega_\sigma \neq 0$, otherwise l = 0.

Process	Order n	$-\omega_\sigma;\omega_1,\ldots,\omega_n$	K
Linear absorption/emission and refractive index	1	$-\omega;\omega$	1
Optical rectification	2	$0;\omega,-\omega$	1/2
Linear electrooptic effect	2	$-\omega;0,\omega$	2
Second-harmonic generation	2	$-2\omega;\omega,\omega$	1/2
Sum- and difference frequency mixing, parametric amplification and oscillation	2	$-\omega_3;\omega_1,\pm\omega_2$	1
Quadratic electrooptic effect	3	$-\omega;0,0,\omega$	3
d.c.-induced second-harmonic generation	3	$-2\omega;0,\omega,\omega$	3/2
Third-harmonic generation	3	$-3\omega;\omega,\omega,\omega$	1/4
General four-wave mixing	3	$-\omega_4;\omega_1,\omega_2,\omega_3$	3/2
Third-order sum- and difference frequency mixing	3	$-\omega_3;\pm\omega_1,\omega_2,\omega_2$	3/4
Coherent anti-Stokes Raman scattering	3	$-\omega_{AS};\omega,\omega,-\omega_S$	3/4
Optical Kerr effect (optically induced birefringence), stimulated Raman scattering)	3	$-\omega_S;\omega,-\omega,\omega_S$	3/2
Intensity-dependent refractive index,optical Kerr effect, degenerate four-wave mixing, self-focusing	3	$-\omega;\omega,-\omega,\omega$	3/4
Two-photon aborption or emission	3	$-\omega_1;-\omega_2,\omega_2,\omega_1$ or $-\omega;-\omega,\omega,\omega$	3/2 3/4

Spatial dispersion

Polarization at a point is determined by the electric field in the *neighbourhood* of that point (spatial invariance, e.g., homogeneity).

$$\mathbf{P}^{(n)}(t,\mathbf{r}) = \int_{-\infty}^{+\infty} d\tau_1 \ldots \int_{-\infty}^{+\infty} d\tau_n \int_{-\infty}^{+\infty} d\mathbf{r}_1 \ldots \int_{-\infty}^{+\infty} d\mathbf{r}_n$$

$$\cdot\ \mathbf{R}^{(n)}(\tau_1,\mathbf{r}_1,\ldots,\tau_n,\mathbf{r}_n)\ \mathbf{E}(t-\tau_1,\mathbf{r}_1)\ldots\ \mathbf{E}(t-\tau_n,\mathbf{r}_n)$$

$$\mathbf{E}(t,\mathbf{r}) = \int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\mathbf{k}\ \mathbf{E}(\omega,\mathbf{k})\ \exp[-i(\omega t - \mathbf{kr})]$$

$$\mathbf{P}^{(n)}(t,\mathbf{r}) = \int_{-\infty}^{+\infty} d\omega_1 \ldots \int_{-\infty}^{+\infty} d\omega_n \int_{-\infty}^{+\infty} d\mathbf{k}_1 \ldots \int_{-\infty}^{+\infty} d\mathbf{k}_n\ \exp[-i(\omega_\sigma t - \mathbf{k}_P \cdot \mathbf{r})]$$

$$\cdot\ \chi^{(n)}(-\omega_\sigma;\omega_1,\mathbf{k}_1,\ldots,\ \omega_n,\mathbf{k}_n)\ \mathbf{E}(\omega_1,\mathbf{k}_1)\ldots\mathbf{E}(\omega_n,\mathbf{k}_n)$$

$$\chi^{(n)}(-\omega_\sigma;\omega_1,\mathbf{k}_1,\ldots,\omega_n,\mathbf{k}_n) = \int_{-\infty}^{+\infty} d\tau_1 \ldots \int_{-\infty}^{+\infty} d\tau_n \int_{-\infty}^{+\infty} d\mathbf{r}_1 \ldots \int_{-\infty}^{+\infty} d\mathbf{r}_n$$

$$\cdot \mathbf{R}^{(n)}(\tau_1,\mathbf{r}_1,\ldots,\tau_n,\mathbf{r}_n)\ \exp\left[\ i \sum_{j=1}^{n} (\omega_j \tau_j - \mathbf{k}_j \cdot \mathbf{r}_j)\right]$$

with $\omega_\sigma = \sum_{j=1}^{n} \omega_j$ and $\mathbf{k}_P = \sum_{j=1}^{n} \mathbf{k}_j$.

nth-order susceptibility is invariant under all n! permutations of the triplets $(\alpha_1,\omega_1,\mathbf{k}_1),\ldots,(\alpha_n,\omega_n,\mathbf{k}_n)$.

Overall permutation symmetry

Approximation: loss-free medium at all the relevant optical frequencies (far from any resonances).

$\chi^{(n)}$ is invariant under all (n+1)! permutations of the pairs $(\mu,-\omega_\sigma),(\alpha_1,\omega_1), (\alpha_2,\omega_2),...,(\alpha_n,\omega_n)$.

$\mathbf{S}_T$ indicates summation over (n+1)! permutations.

$$\chi_{\mu\alpha}^{(1)}(-\omega_\sigma;\omega) = (Ne^2/h)\, \mathbf{S}_T \sum_{ab} \rho_0(a)\, [r_{ab}^{\mu}\, r_{ba}^{\alpha}/(\Omega_{ba} -\omega)]$$

$$\chi_{\mu\alpha\beta}^{(2)}(-\omega_\sigma;\omega_1,\omega_2) = (Ne^3/2h^2)\, \mathbf{S}_T \sum_{abc} \rho_0(a)$$

$$\cdot[r_{ab}^{\mu}\, r_{bc}^{\alpha}\, r_{ca}^{\beta} /(\Omega_{ba}-\omega_1-\omega_2)(\Omega_{ca}-\omega_2)]$$

$$\chi_{\mu\alpha\beta\gamma}^{(3)}(-\omega_\sigma;\omega_1,\omega_2,\omega_3) = (Ne^4/6h^3)\, \mathbf{S}_T \sum_{abcd} \rho_0(a)$$

$$\cdot[r_{ab}^{\mu}\, r_{bc}^{\alpha}\, r_{cd}^{\beta}\, r_{da}^{\gamma} /(\Omega_{ba}-\omega_1-\omega_2-\omega_3)(\Omega_{ca}-\omega_2-\omega_3)(\Omega_{da}-\omega_3)]$$

Consequences:
Kleinman symmetry (low frequencies), $\mu\leftrightarrow\alpha\leftrightarrow\beta\leftrightarrow\gamma$

Manley-Rowe power relations (conservation of the *total* number of photons)

Spatial symmetry

The components of the susceptibility tensor are invariant under any transformation of coordinates that is governed by a valid symmetry operation for the medium.

$\rightarrow \chi^{(n)}$ for even n is different from zero only if the medium is *not* characterized by inversion symmetry (e.g., it is impossible to generate even-order harmonics in isotropic media).

Time-reversal symmetry

Operators which are real in the Schrödinger picture are invariant under time-reversal ($t \rightarrow -t$).

Medium without losses: $\chi^{(n)}$ is real . From the reality condition (for **E**(t) and **P**(t)) it follows:

$$\chi^{(n)}(\omega_\sigma;-\omega_1,\ldots,-\omega_n) = \chi^{(n)}(-\omega_\sigma;\omega_1,\ldots,\omega_n)$$

Coupled wave equations

$$\mathbf{E}(\omega) = \hat{\mathbf{E}}(\omega) \exp(i\, \mathbf{k}\, \mathbf{r})$$

$$\text{rot rot} [\, \hat{\mathbf{E}}(\omega)\exp(i\, \mathbf{k}\, \mathbf{r})\,] = (\omega^2/c^2)\, \varepsilon(\omega)\, \hat{\mathbf{E}}(\omega)\exp(i\, \mathbf{k}\, \mathbf{r})$$

$$+ 4\pi(\omega^2/c^2)\, \mathbf{P}^{NL}(\omega)$$

Simplification: forward-travelling waves in the z-direction:

$$\text{rot rot}\, \hat{\mathbf{E}} \rightarrow \partial^2 \hat{\mathbf{E}} / \partial z^2$$

Slowly-varying envelope approximation (SVEA)

Slow variation of $\hat{\mathbf{E}}(\omega)$ (both in amplitude and phase) with distance z:

$$\left| \partial^2\, \hat{\mathbf{E}}(\omega) / \partial z^2 \right| << \left| k \cdot \partial\, \hat{\mathbf{E}}(\omega) / \partial z \right|$$

$$\rightarrow \partial\, \hat{\mathbf{E}}(\omega) / \partial z = (2i\pi\omega^2/kc^2)\, \mathbf{P}^{NL}(\omega) \exp(-ikz)$$

$$\partial\, E_\sigma / \partial z = (2i\pi\omega_\sigma/kc^2)\, K(-\omega_\sigma;\omega_1,\ldots,\omega_n)\, \chi^{(n)}(-\omega_\sigma;\omega_1,\ldots,\omega_n)$$

$$\cdot\, E_1 \ldots E_n\, \exp[\, i\, \Delta k\, z\,]\, ; \quad \Delta k = k_P - k_\sigma$$

Parametric processes

Second-harmonic generation

$\partial E_{2\omega} / \partial z = (i\omega / n_{2\omega} c)[½\, \chi^{(2)}(-2\omega;\omega,\omega)\, E_\omega^{\,2}] \exp(i\Delta kz)$

$\partial E_{\omega} / \partial z = (i\omega / 2n_{\omega} c)[\chi^{(2)}(-2\omega;\omega,\omega) E_\omega^{*}\, E_{2\omega}] \exp(-i\Delta kz)$

where $\Delta k = 2k_\omega - k_{2\omega} = 2\omega\,(n_\omega - n_{2\omega})/c$

Neglection of pump depletion (L-interaction length)

$I_{2\omega}(L) \propto I_\omega^{\,2}\; L^2\, [\sin(\Delta kL/2)/(\Delta kL/2)]^2$

Consideration of pump depletion for $\Delta k = 0$:

$I_{2\omega}\,(L) = I_\omega\,(0)\, \tanh^2(\mathrm{const}\; L)$

$I_{\omega}\,(L) = I_\omega\,(0)\, \mathrm{sech}^2(\mathrm{const}\; L)$

Frequency mixing and parametric amplification

Three-wave processes:
Sum-frequency ($\omega_1+\omega_2\rightarrow\omega_3$)
Difference-frequency ($\omega_3 - \omega_1\rightarrow\omega_2$, $\omega_3 - \omega_2\rightarrow\omega_1$)

Phase-matching: $\Delta k = k_3 - k_2 - k_1 = 0$.

$\omega_3 > \omega_2 > \omega_1$: ω_3 - pump , ω_2 - signal , ω_1 - idler

Sum-frequency mixing: energy is transferred from signal and idler waves to the pump wave.

Difference-frequency mixing: *amplification* process (transfer from pump to signal + idler).

Neglection of pump depletion; I_ω (0) = 0 :

$$I_\omega (L) = I_\omega (0) \cosh^2 (\text{const } L)$$

$$I_\omega (L) = (\omega_1/\omega_2) I_\omega (0) \sinh^2 (\text{const } L)$$

Parametric oscillator: generation of coherent light due to parametric amplification.

Parametric fluorescence: spontaneous decay of one pump photon into one signal and one idler photon (Radiation field has to be quantized).

Recommended Literature

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