Introduction to Nonlinear Optics

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Outline

- General picture of nonlinear optical susceptibilities
- Qualitative description of nonlinear optical processes
- Anharmonic oscillator model for nonlinear susceptibilities
- Formal properties of nonlinear optical susceptibilities
- Conservation laws in nonlinear media

Linear optical response:

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

Critical electric field:

- E_c ranges from 10^9 to 10^{10} V/cm.
- available laser field: 10^6 to 10^7 V/cm

No ionization; small electron displacement from its equilibrium!

$$P = \varepsilon_0(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \ldots),$$

$\chi^{(n)} \Longleftrightarrow$ nth order susceptibility

Estimating order-of magnitude of $\chi^{(n)}$

Atomic radius:

$$a_0 = \hbar^2/me^2 \simeq 5 \times 10^{-9} cm$$

Atomic electric field:

$$E_{at} = e/4\pi\varepsilon_0 a_0^2 \simeq 5 \times 10^{11} V/m.$$

- linear susceptibility, $\chi^{(1)} \sim 1$;
- second-order,

$$\chi^{(2)} \sim E_{at}^{-1} \sim 10^{-12}, \text{ m/V}.$$

- third-order,

$$\chi^{(3)} \sim 10^{-21} \text{ to } 10^{-22}, \text{ m}^2/\text{V}^2.$$

conclusion:

The lowest non-vanishing order always dominates!

Qualitative picture of nonlinear processes

Second-harmonic generation & Optical Rectification

Driving fundamental field:

$$E(t) = \frac{1}{2}(\mathscr{E} e^{-i\omega t} + c.c),$$

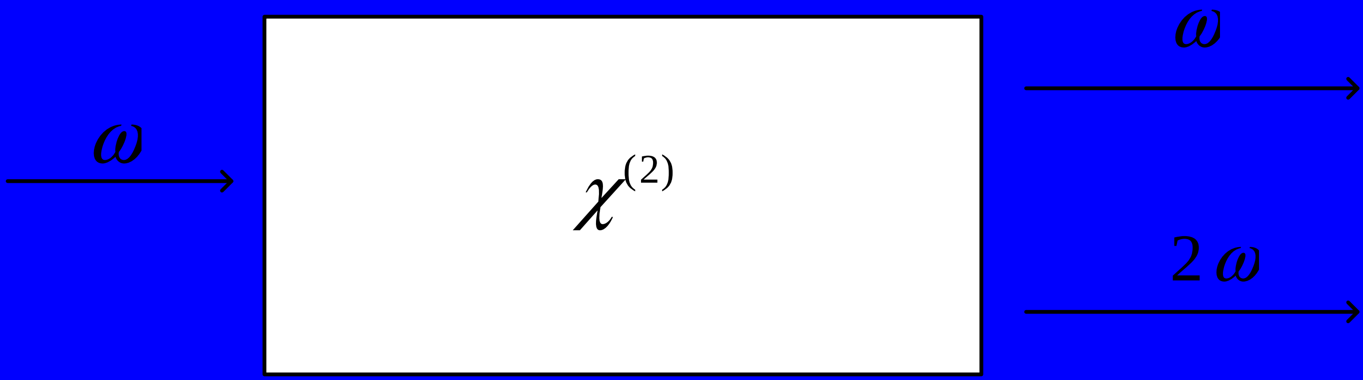
Nonlinear polarization:

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E^2(t)$$

$$P^{(2)}(t) = \mathscr{P}_{OR}(0) + \mathscr{P}_{SHG}(2\omega_j) e^{-i2\omega t} + c.c$$

$$\mathscr{P}_{SHG}(2\omega_j) = \frac{1}{2}\varepsilon_0\chi^{(2)}\mathscr{E}_j^2,$$

$$\mathscr{P}_{OR}(0) = \varepsilon_0\chi^{(2)}(|\mathscr{E}_1|^2 + |\mathscr{E}_2|^2).$$



Sum- and difference-frequency generation

Driving field

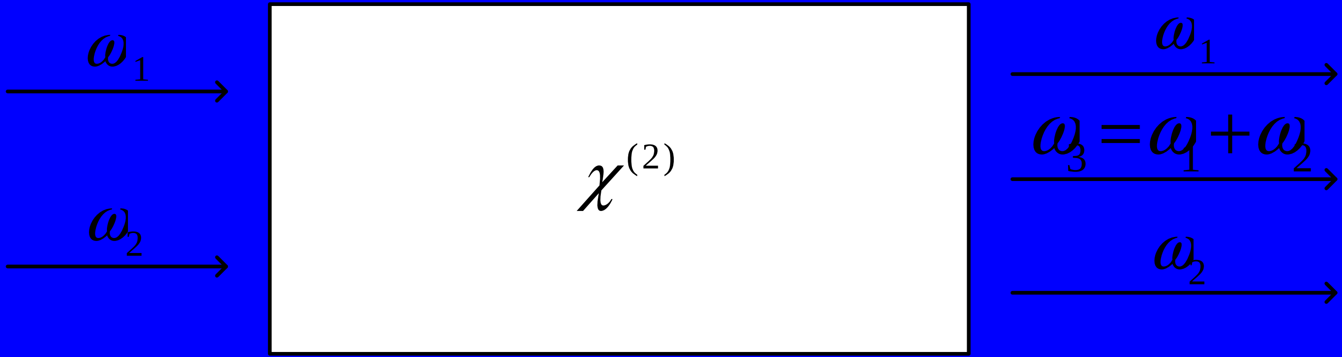
$$E(t) = \frac{1}{2}(\mathscr{E}_1 e^{-i\omega_1 t} + \mathscr{E}_2 e^{-i\omega_2 t} + c.c)$$

Nonlinear polarization:

$$\begin{aligned} P^{(2)}(t) &= \mathscr{P}_{SFG}(\omega_1 + \omega_2) e^{i(\omega_1+\omega_2)t} \\ &+ \mathscr{P}_{DFG}(\omega_1 - \omega_2) e^{i(\omega_1-\omega_2)t} + c.c. \end{aligned} \tag{1}$$

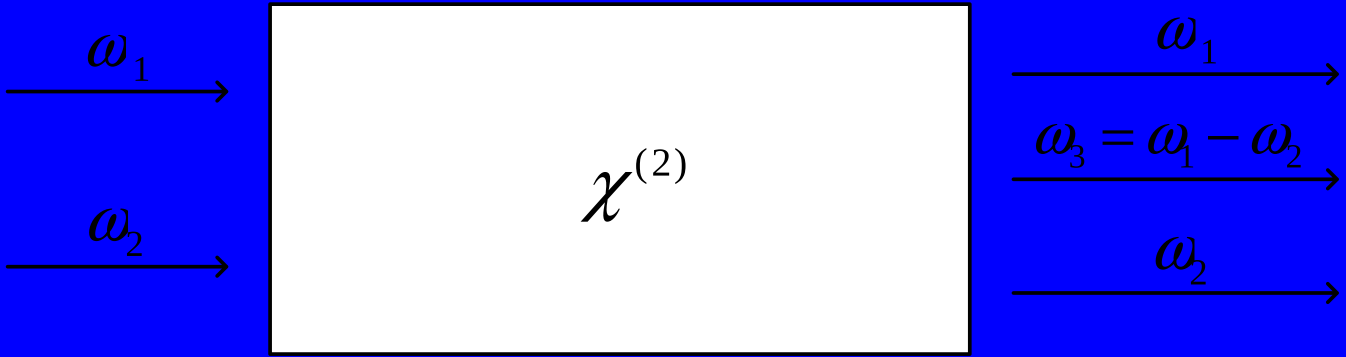
$$P^{(2)}(t) = \mathscr{P}_{SFG}(\omega_1+\omega_2)e^{i(\omega_1+\omega_2)t} + \mathscr{P}_{DFG}(\omega_1-\omega_2)e^{i(\omega_1-\omega_2)t} + c.c. \qquad (2)$$

$$\mathscr{P}_{SFG}(\omega_1+\omega_2) = \varepsilon_0 \chi^{(2)} \mathscr{E}_1 \mathscr{E}_2,$$



$$P^{(2)}(t) = \mathscr{P}_{SFG}(\omega_1 + \omega_2)e^{i(\omega_1+\omega_2)t} + \mathscr{P}_{DFG}(\omega_1 - \omega_2)e^{i(\omega_1-\omega_2)t} + c.c. \qquad (3)$$

$$\mathscr{P}_{DFG}(\omega_1 - \omega_2) = \varepsilon_0 \chi^{(2)} \mathscr{E}_1 \mathscr{E}_2^*,$$



Self-focusing and third-harmonic generation

Driving field

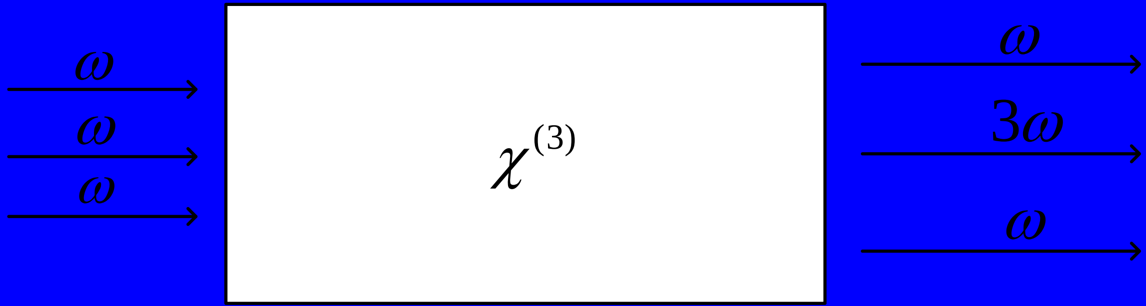
$$E(t) = \tfrac{1}{2}(\mathscr{E} e^{-i\omega t} + c.c),$$

Nonlinear polarization

$$P^{(3)}(t) = \varepsilon_0 \chi^{(3)} E^3(t)$$

$$P^{(3)}(t) = \tfrac{1}{2}[\mathscr{P}_{THG}(3\omega)e^{-i3\omega t} + \mathscr{P}_{SF}(\omega)e^{-i\omega t} + c.c.],$$

$$\mathscr{P}_{THG}(3\omega) = \frac{1}{2}\varepsilon_0\chi^{(3)}\mathscr{E}^3,$$



$$\mathscr{P}_{SF}(\omega) = \frac{3\varepsilon_0}{2}\chi^{(3)}|\mathscr{E}|^2\mathscr{E}.$$

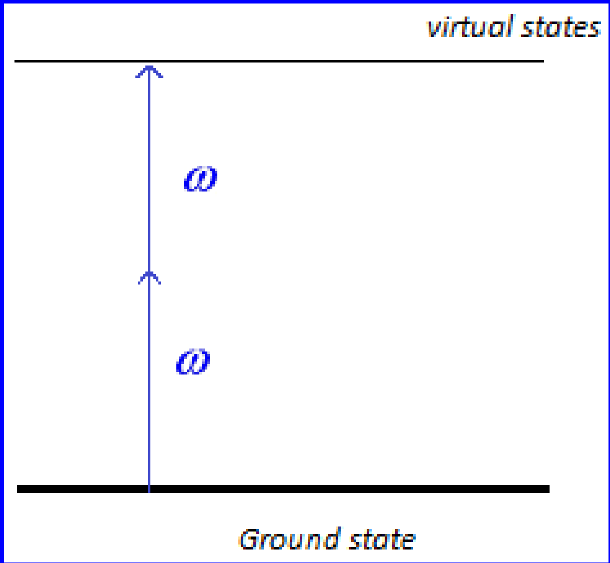
The SF $\Longleftrightarrow$ the refractive index modification:

$$n = n_0 + n_2|\mathscr{E}|^2,$$

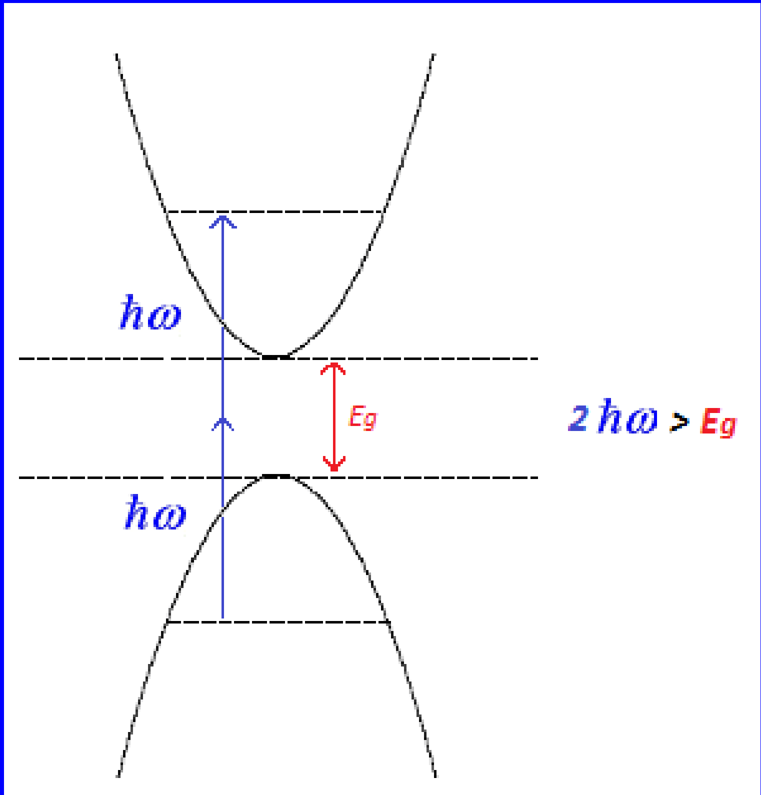
Self-lensing of a light beam!

Self-focusing (SF) vs two-photon absorption (TPA)

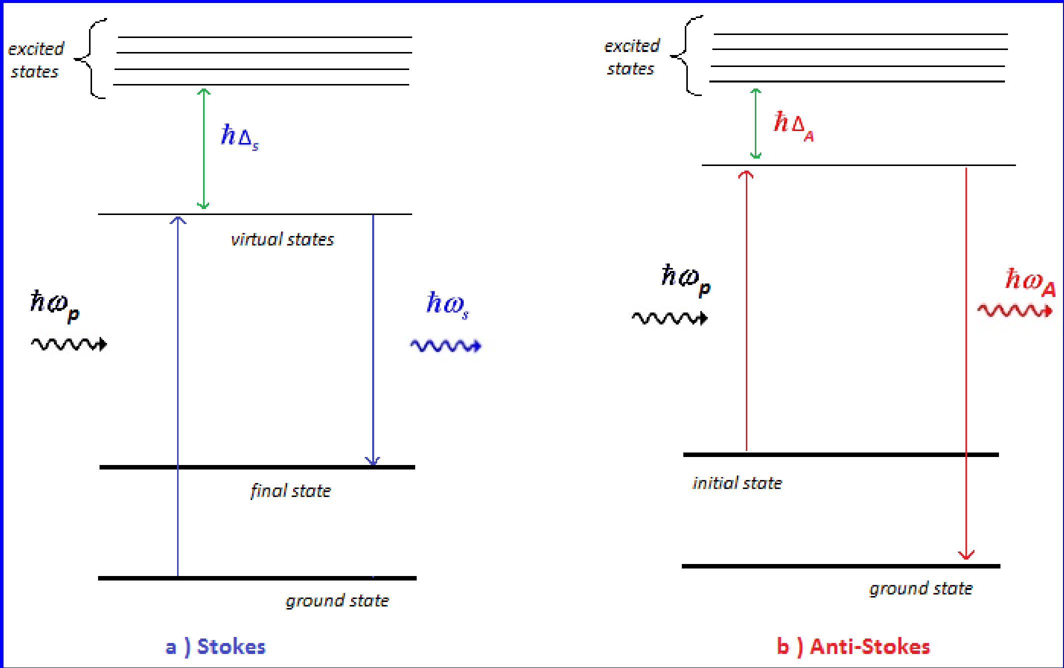
$$\chi^{(3)} = \underbrace{\mathrm{Re}\{\chi^{(3)}\}}_{SF} + i\,\underbrace{\mathrm{Im}\{\chi^{(3)}\}}_{TPA}$$



TPA in semiconductors $\Longrightarrow$ carrier photo-excitation



Stimulated Raman & Brillouin scattering



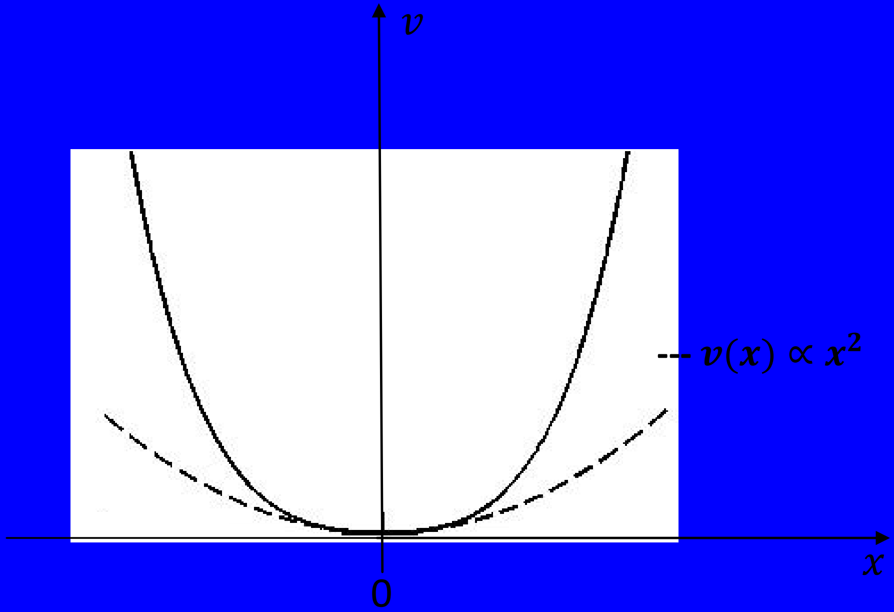
$$\omega_\Delta = \omega_p - \omega_s = \omega_{as} - \omega_p$$

$\hbar\omega_{\Delta} \Longrightarrow$ single molecule vibration/rotation

Raman scattering $\Longrightarrow$ individual molecule excitations, i. e., usually gas molecule rotational/vibrational degrees of freedom

Brillouin scattering $\Longrightarrow$ collective medium vibrations, i. e., sound generation in gases or condensed matter!

Anharmonic oscillator model



Actual interaction potential deviates from harmonic oscillator model!

Quantitative model:

Newton's law for anharmonic oscillator

$$m\ddot{x} = -2\gamma m\dot{x} - eE + F_r,$$

restoring "spring" force:

$$F_r = -\frac{\partial V}{\partial x},$$

The potential:

$$V(x) = \frac{1}{2}m\omega_0^2 x^2 + \frac{1}{3}max^3 + \frac{1}{4}mbx^4 + \dots$$

Second-order processes

$$\ddot{x} + \omega_0^2 x + 2\gamma\dot{x} + ax^2 = -\lambda\frac{eE}{m}$$

$\lambda \Longleftrightarrow$ bookkeeping parameter;

Bi-chromatic driving field:

$$E(t) = \tfrac{1}{2}(\mathscr{E}_1 e^{-i\omega_1 t} + \mathscr{E}_2 e^{-i\omega_2 t} + c.\ c.).$$

The oscillator response:

$$x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + \ldots.$$

Seeking first-order solution:

$$x^{(1)} = \frac{1}{2}(x_{\omega_1} e^{-i\omega_1 t} + x_{\omega_2} e^{-i\omega_2 t} + c.\ c.),$$

$$x_{\omega_j} = -\frac{e\mathscr{E}_j}{m\mathscr{D}(\omega_j)}$$

with the resonant denominator being

$$\mathscr{D}(\omega_j) \equiv -\omega_j^2 + \omega_0^2 - 2i\gamma\omega_j$$

First-order is just a linear harmonic oscillator solution!

Second-order solution

$$
\begin{aligned}
x^{(2)} = \frac{1}{2}\Big[& x_{2\omega_1} e^{-i2\omega_1 t} + x_{2\omega_2} e^{-i2\omega_2 t} \\
& + x_{\omega_1+\omega_2} e^{-i(\omega_1+\omega_2)t} \\
& + x_{\omega_1-\omega_2} e^{-i(\omega_1-\omega_2)t} \\
& + x_{\omega_2-\omega_1} e^{-i(\omega_2-\omega_1)t} + c.\ c.\Big]
\end{aligned} \tag{4}
$$

SHG:

$$
x_{2\omega_j} = -\frac{ae^2\mathcal{E}_j^2}{2m^2\mathcal{D}^2(\omega_j)\mathcal{D}(2\omega_j)}
$$

where $j = 1, 2$

SFG:

$$x_{\omega_1+\omega_2} = -\frac{ae^2\mathscr{E}_1\mathscr{E}_2}{m^2\mathscr{D}(\omega_1)\mathscr{D}(\omega_2)\mathscr{D}(\omega_1+\omega_2)}$$

DFG

$$x_{\omega_j-\omega_{3-j}} = -\frac{ae^2\mathscr{E}_j\mathscr{E}^*_{3-j}}{m^2\mathscr{D}(\omega_j)\mathscr{D}^*(\omega_{3-j})\mathscr{D}(\omega_j-\omega_{3-j})}$$

Polarization contributions:

$$\mathscr{P}(2\omega_j) = -Nex_{2\omega_j},$$

$$\mathscr{P}(\omega_1+\omega_2) = -Nex_{\omega_1+\omega_2},$$

$$\mathscr{P}(\omega_j-\omega_{3-j}) = -Nex_{\omega_j-\omega_{3-j}}$$

Macroscopic definitions of polarizations:

SHG:

$$\mathscr{P}(2\omega_j) = \frac{1}{2}\varepsilon_0\chi^{(2)}(-2\omega_j;\omega_j,\omega_j)\mathscr{E}_j^2,$$

SFG

$$\mathscr{P}(\omega_1+\omega_2) = \varepsilon_0\chi^{(2)}(-\omega_1-\omega_2;\omega_1,\omega_2)\mathscr{E}_1\mathscr{E}_2,$$

DFG

$$\mathscr{P}(\omega_j-\omega_{3-j}) = \varepsilon_0\chi^{(2)}(-\omega_j+\omega_{3-j};\omega_j,-\omega_{3-j}) \times \mathscr{E}_j\mathscr{E}_{3-j}^* \quad (5)$$

$j = 1,2$

Thus

$$\chi^{(2)}_{SHG} = \frac{Nae^3}{\varepsilon_0 m^2 \mathscr{D}^2(\omega_j)\mathscr{D}(2\omega_j)},$$

and

$$\chi^{(2)}_{SFG} = \frac{Nae^3}{\varepsilon_0 m^2 \mathscr{D}(\omega_1)\mathscr{D}(\omega_2)\mathscr{D}(\omega_1+\omega_2)},$$

as well as

$$\chi^{(2)}_{DFG} = \frac{Nae^3}{\varepsilon_0 m^2 \mathscr{D}(\omega_j)\mathscr{D}^*(\omega_{3-j})\mathscr{D}(\omega_j-\omega_{3-j})},$$

Order-of-magnitude estimates:

$$\chi^{(2)} \simeq \frac{Ne^3 a}{m^2 \varepsilon_0 \omega_0^6}.$$

Using

- $\omega_0 \simeq 10^{16}$ rad/s,
- $N \simeq 10^{28}$, m^{-3}
- $e \simeq 10^{-19}$ C, and $m \simeq 10^{-30}$ kg,

$$\chi^{(2)} \sim 10^{-12} \text{m/V}.$$

Nonlinear processes induced by cw fields

Nonlinear polarization:

$$P_i^{(2)}(t) = \tfrac{1}{2}\textstyle\sum_s \mathscr{P}_i(\omega_s)e^{-i\omega_s t} + c.c,$$

Example: second-order processes

$$\mathscr{P}_i(\omega_3) \;=\; \varepsilon_0 c^{(2)}(\omega_1,\omega_2)\sum_{jk}\chi^{(2)}_{ijk}(-\omega_3;\omega_1,\omega_2) \times \mathscr{E}_j(\omega_1)\mathscr{E}_k(\omega_2) \tag{6}$$

Notations & Conventions

- $$\chi^{(2)}_{ijk}(-\omega_3;\omega_1,\omega_2)$$

  where

  $$\omega_3 = \omega_1 + \omega_2$$

- symmetry coefficient:

  $$c^{(2)}(\omega_1,\omega_2) = \begin{cases} 1, & \omega_1 \neq \omega_2; \\ 1/2, & \omega_1 = \omega_2. \end{cases}$$

Nonlinearities induced by arbitrary fields

linear response

$$\mathbf{P}^{(1)}(\mathbf{r},t) = \varepsilon_0 \int d\mathbf{r}' \int_{-\infty}^{\infty} dt' \chi^{(1)}(\mathbf{r}-\mathbf{r}', t-t') \vdots \mathbf{E}(\mathbf{r}', t')$$

second-order response

$$\mathbf{P}^{(2)}(\mathbf{r},t) = \varepsilon_0 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \times \chi^{(2)}(\mathbf{r}-\mathbf{r}_1, \mathbf{r}-\mathbf{r}_2; t-t_1, t-t_2) \vdots \mathbf{E}(\mathbf{r}_1, t_1)\mathbf{E}(\mathbf{r}_2, t_2), \quad (7)$$

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Notes:

- Medium isotropy & stationarity $\Longrightarrow$ translational invariance in space-time
- $\chi = \chi(\mathbf{r}, t) \Longleftrightarrow$ spatial & temporal dispersion
- No spatial dispersion:

$$\chi^{(1)} = \delta(\mathbf{r} - \mathbf{r}')\chi_t^{(1)}(t - t')$$

$$\chi^{(2)} = \delta(\mathbf{r} - \mathbf{r}_1)\delta(\mathbf{r} - \mathbf{r}_2)\chi_t^{(2)}(t - t_1, t - t_2)$$

Space-frequency representation:

$$\tilde{\chi}^{(1)}(\omega) = \int_{-\infty}^{\infty} dt\, \chi^{(1)}(t) e^{i\omega t}$$

and

$$\tilde{\chi}^{(2)}(\omega_1, \omega_2) = \prod_{s=1}^{2} \int_{-\infty}^{\infty} dt_s \chi^{(2)}(t_1, t_2) e^{i \sum_{s=1}^{2} \omega_s t_s}$$

Thus,

$$\tilde{P}_i^{(1)}(\mathbf{r}, \omega) = \varepsilon_0 \sum_j \tilde{\chi}_{ij}^{(1)}(\omega) \tilde{E}_j(\mathbf{r}, \omega)$$

and

$$\tilde{P}_i^{(2)}(\mathbf{r},\omega_3) = \varepsilon_0 \sum_{jk} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \tilde{\chi}_{ijk}^{(2)}(-\omega_3,\omega_1,\omega_2) \times \tilde{E}_j(\mathbf{r},\omega_1)\tilde{E}_k(\mathbf{r},\omega_2) \quad (8)$$

where $\omega_3 = \omega_1 + \omega_2$

Formal properties of nonlinear susceptibilities

- Generic properties, always satisfied
- Symmetry properties depending on particular media symmetries

- *Intrinsic permutational symmetry*:

Example:

$$\tilde{\chi}^{(2)}_{ijk}(-\omega,\omega_1,\omega_2)=\tilde{\chi}^{(2)}_{ikj}(-\omega,\omega_2,\omega_1).$$

- *Reality of χ in the time-domain*:

Example:

$$\tilde{\chi}^{(2)*}_{ijk}(-\omega,\omega_1,\omega_2)=\tilde{\chi}^{(2)}_{ijk}(\omega,-\omega_1,-\omega_2).$$

- *Causality*:

$$\chi^{(n)}_{jj_1\ldots j_n}(t-\tau_1,\ldots t-\tau_n)=0, \qquad \text{for any} \quad \tau_s>t.$$

Cause precedes the effect!

Consequences of causality: Kramers-Kronig relations:

$$\mathrm{Re}\,\tilde{\chi}^{(1)}(\boldsymbol{\omega}) = \frac{1}{\pi}\mathscr{P}\int_{-\infty}^{\infty} d\boldsymbol{\omega}' \frac{\mathrm{Im}\,\tilde{\chi}^{(1)}(\boldsymbol{\omega}')}{\boldsymbol{\omega}-\boldsymbol{\omega}'}$$

and

$$\mathrm{Im}\,\tilde{\chi}^{(1)}(\boldsymbol{\omega}) = -\frac{1}{\pi}\mathscr{P}\int_{-\infty}^{\infty} d\boldsymbol{\omega}' \frac{\mathrm{Re}\,\tilde{\chi}^{(1)}(\boldsymbol{\omega}')}{\boldsymbol{\omega}-\boldsymbol{\omega}'}$$

- Makes it possible to determine real part (dispersion) from measurable imaginary (absorption) part!

Medium symmetry specific properties of χ:

- Isotropic media $\Longleftrightarrow$ invariance with respect to rotations
- Isotropic and centrosymmetric media $\Longleftrightarrow$ invariance with respect to rotations, translations, and inversions

$$\chi^{(1)}_{ij} = \sum_{kl} \mathcal{T}_{ik} \mathcal{T}_{jl} \chi^{(1)}_{kl},$$

or

$$\chi^{(3)}_{ijkl} = \sum_{mnpq} \mathcal{T}_{im} \mathcal{T}_{jn} \mathcal{T}_{kp} \mathcal{T}_{lq} \chi^{(3)}_{mnpq},$$

Inversion: $\mathbf{r} \longrightarrow -\mathbf{r}$

$$\mathcal{T}_{ij} = -\delta_{ij}$$

Thus for any $n = 2k$

$$\chi^{(2k)}_{ii_1 \ldots i_{2k}} = -\chi^{(2k)}_{ii_1 \ldots i_{2k}} = 0$$

No second-order effects in centrosymmetric media!!

- Lossless media $\Longleftrightarrow$ invariance with respect to time reversal

Mathematically

$$\chi^{(n)}(\tau_1 \ldots \tau_n) = \chi^{(n)}(-\tau_1 \ldots -\tau_n)$$

Consequences:

- Overall permutational symmetry of χ:

Example: $\tilde{\chi}^{(3)}_{ijkl}(-\omega_4, \omega_1, \omega_2, \omega_3) = \tilde{\chi}^{(3)}_{jlik}(\omega_1, \omega_3, -\omega_4, \omega_2)$.

Kleinmann symmetry for all ω far below resonance

Example: $\tilde{\chi}^{(2)}_{ijk}(-\omega_3, \omega_1, \omega_2) = \tilde{\chi}^{(2)}_{jki}(-\omega_3, \omega_1, \omega_2) = \tilde{\chi}^{(2)}_{kij}(-\omega_3, \omega_1, \omega_2)$.

Conservation laws in nonlinear optics

Maxwell's equations for nonlinear nonmagnetic media

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t},$$

$$\nabla \times \mathbf{H} = \mathbf{J} + \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}}{\partial t},$$

$$\nabla \cdot \mathbf{D} = \rho,$$

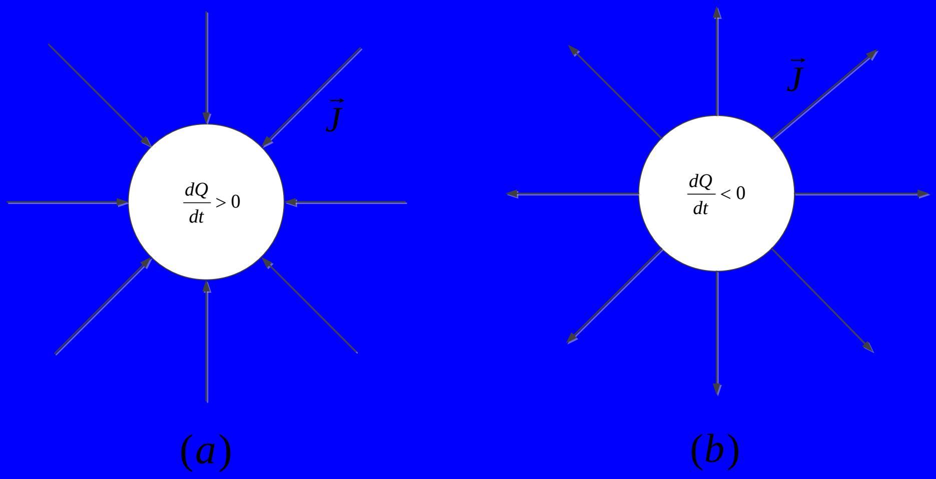
$$\nabla \cdot \mathbf{H} = 0,$$

- Maxwell's equations $\Longrightarrow$ energy conservation for EM fields
- Charge conservation $\Longrightarrow$ fundamental law of nature

Charge conservation

Given a closed volume,

$$\frac{d}{dt}\int dv\rho_v = -\oint d\mathbf{S}\cdot\mathbf{J}$$



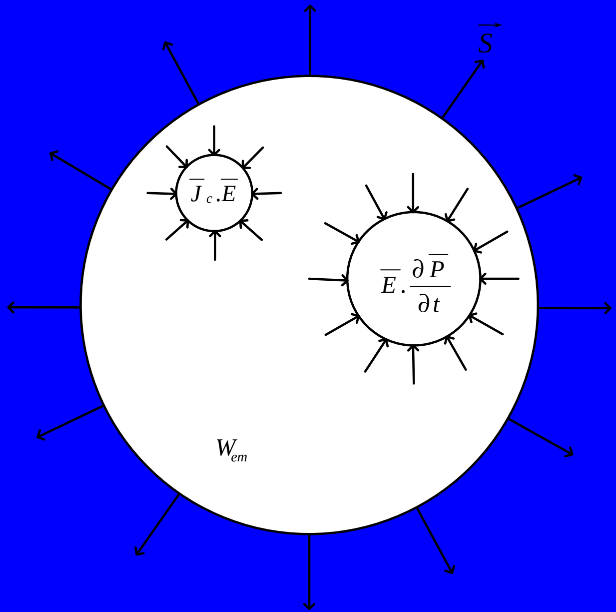
$$\frac{d}{dt}\int dv\rho_v = \int_v dv\frac{\partial \rho_v}{\partial t} = -\oint d\mathbf{S}\cdot\mathbf{J} = -\int dv\nabla\cdot\mathbf{J}$$

implying that

$$\int_v \left(\frac{\partial \rho_v}{\partial t} + \nabla\cdot\mathbf{J}\right) = 0$$

charge conservation in the differential form

$$\frac{\partial \rho_v}{\partial t} + \nabla\cdot\mathbf{J} = 0$$



Integral for of EM energy conservation

$$\frac{dW_{em}}{dt} = -\oint_{\sigma} d\boldsymbol{\sigma}\cdot\mathbf{S} - \int_{v} dv\left(\mathbf{J}\cdot\mathbf{E}+\mathbf{E}\cdot\frac{\partial\mathbf{P}}{\partial t}\right).$$

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Differential form of EM energy conservation

$$\frac{\partial w_{em}}{\partial t} + \nabla \cdot \mathbf{S} = -\mathbf{J} \cdot \mathbf{E} - \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t}$$

electromagnetic energy density:

$$w_{em} = \tfrac{1}{2}\varepsilon_0 E^2 + \tfrac{1}{2}\mu_0 H^2$$

energy flux (Poynting vector):

$$\mathbf{S} = \mathbf{E} \times \mathbf{H}$$

Total EM energy within the volume:

$$W_{em} = \int_v dv w_{em},$$