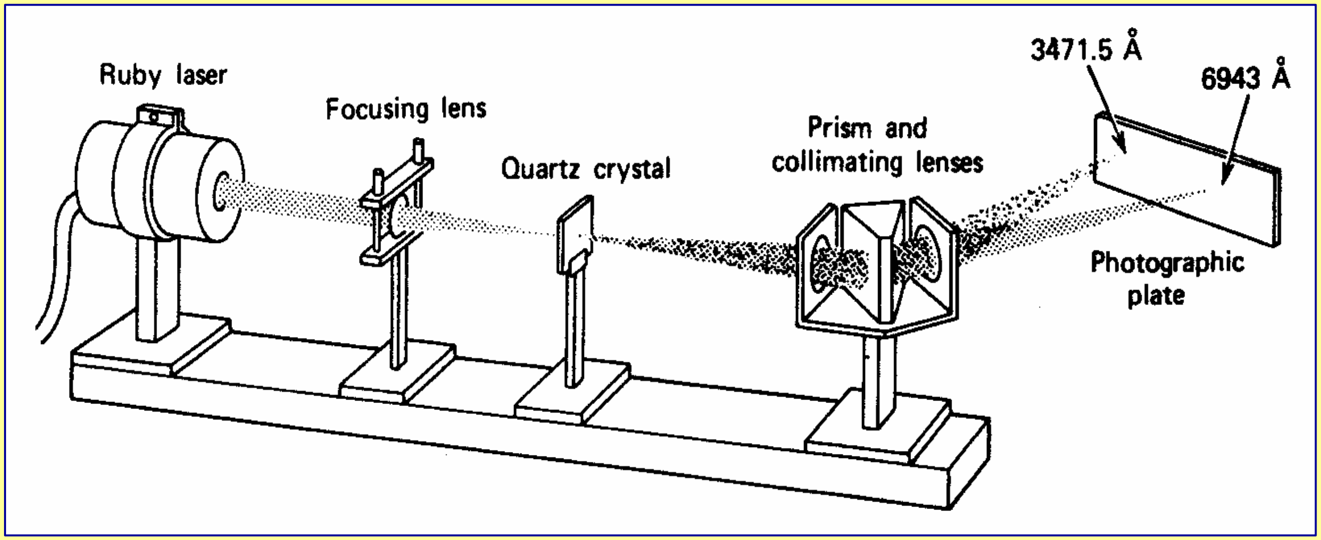
Nonlinear Optics

Nicolaas Bloembergen
Nobel prize 1981

The first non-linear optical laser experiment



P.A. Franken, A.E. Hill, C.W. Peters and G. Weinreich, Phys. Rev. Lett. **7** (1961) 118

The Nonlinear Susceptibility

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

The $\chi_{ij}^{(n)}$ are tensors even for lowest order

$$P_i = \chi_{ij}^{(n)} E_j$$

Polarization not directed along electric field vector

In media with inversion symmetry:

$$I_{\mathrm{op}}\vec{P} = -\vec{P} = -\chi^{(1)}\vec{E} - \chi^{(2)}\vec{E}\vec{E} - \chi^{(3)}\vec{E}\vec{E}\vec{E} - \ldots$$

$$I_{\mathrm{op}}\vec{E} = -\vec{E}$$

Hence:

$$I_{\mathrm{op}}\vec{P} = -\chi^{(1)}\vec{E} + \chi^{(2)}\vec{E}\vec{E} - \chi^{(3)}\vec{E}\vec{E}\vec{E} + \ldots$$

Conclude for centro-symmetric media:

$$\chi^{(2n)} = 0$$

Note that in principle there exist also nonlinear magnetic susceptibilities

Nonlinear Optics; graphically

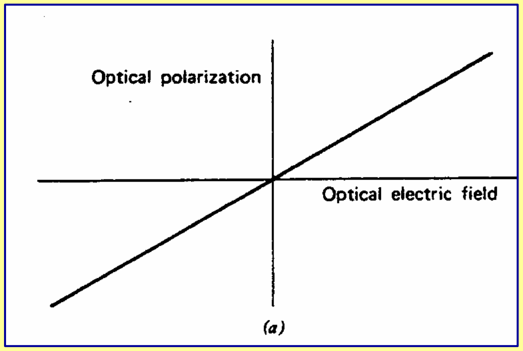
Linear response:

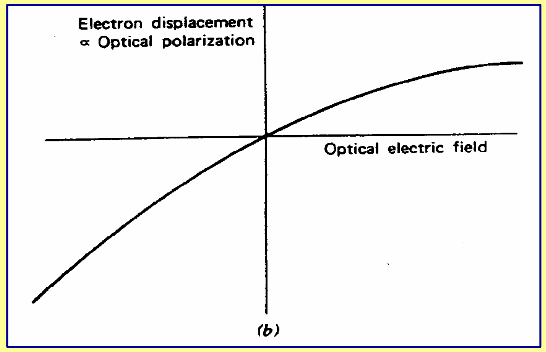
$$\vec{P} = \chi^{(1)}\vec{E}$$

Nonlinear response:

$$\vec{P} = \chi^{(1)}\vec{E} + \chi^{(2)}\vec{E}\vec{E}$$

DC





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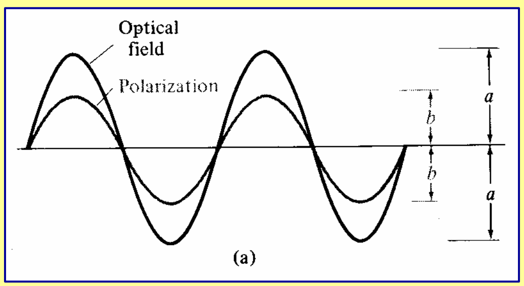
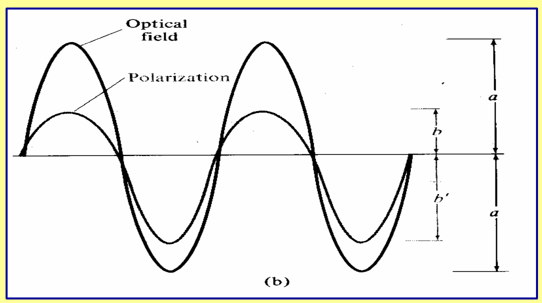
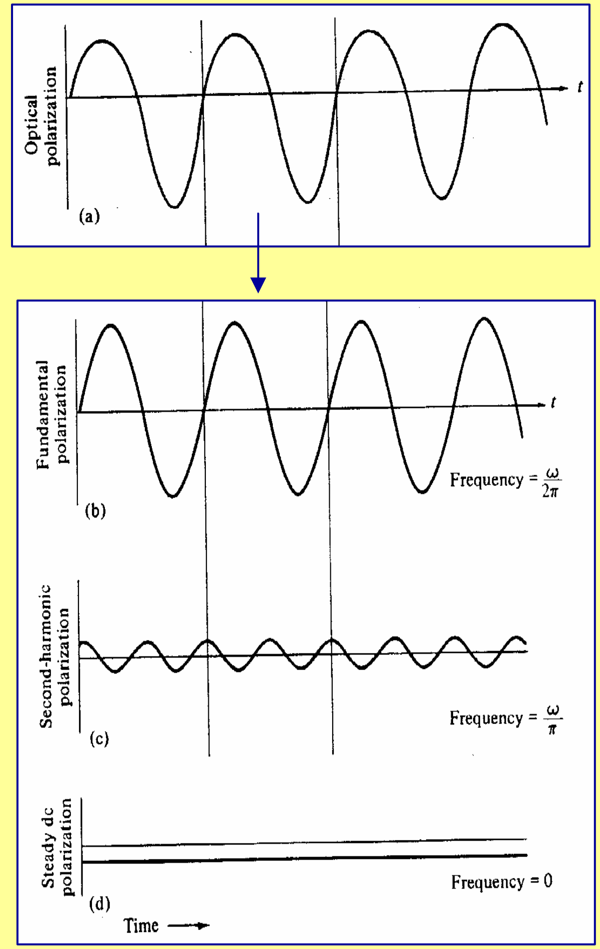




Nonlinear response evaluated in terms of Fourier series



$$P = \sum a_n \sin(n\omega t + \phi_n)$$

Lorentz model of linear optics: classical oscillator

Equation of motion for a damped electronic oscillator in one dimension

$$\frac{d^2}{dt^2}\mathbf{r} + 2\gamma\frac{d}{dt}\mathbf{r} + \omega_0{}^2\mathbf{r} = -\frac{e}{m}\mathbf{E}$$

Write electric field and position vector:

$$\mathbf{E} = \mathrm{Re}\left[Ee^{i\omega t}\right] \qquad \mathbf{r} = \mathrm{Re}\left[re^{i\omega t}\right]$$

$$\longrightarrow \quad \left(\omega_0^2 - \omega^2\right)r + 2i\omega\gamma r = -\frac{e}{m}E$$

Solution

$$r = \frac{-eE}{m\left[\omega_0{}^2 - \omega^2 + 2i\omega\gamma\right]} \approx \frac{-eE}{2m\left[\omega_0\left(\omega_0 - \omega\right) + i\omega\gamma\right]}$$

Near resonance $\omega = \omega_0$

$$\longrightarrow \quad r = \frac{Ne^2}{2m\left[\omega_0\left(\omega_0 - \omega\right) + i\omega\gamma\right]}E = \varepsilon_0\chi(\omega)E$$

Classical polarization of the medium

$$P(\omega) = -Ner(\omega)$$

and the complex susceptibility

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$$

yields expressions for the susceptibility

Real part,
connected to the index of refraction

$$\chi'(\omega) = \frac{Ne^2}{2m\omega_0\gamma\varepsilon_0}\frac{(\omega_0 - \omega)/\gamma}{\left[1 + (\omega_0 - \omega)^2/\gamma^2\right]}$$

Imaginary part,
connected to the absorption coefficient

$$\chi''(\omega) = \frac{Ne^2}{2m\omega_0\gamma\varepsilon_0}\frac{1}{\left[1 + (\omega_0 - \omega)^2/\gamma^2\right]}$$

Lorentz model of nonlinear optics: classical oscillator

Motion of electron with anharmonic term:

$$\frac{d^2}{dt^2}r + 2\gamma\frac{d}{dt}r + \omega_0{}^2 r - \xi r^2 = -\frac{e}{m}E$$

Try a solution in power series

$$r = r_1 + r_2 + r_{3+\dots}$$

with: $r_i = a_i E^i$

Collect terms in same order of E

First order $\frac{d^2}{dt^2}r_1 + 2\gamma\frac{d}{dt}r_1 + \omega_0{}^2 r_1 = -\frac{e}{m}E$ (*)

Second order $\frac{d^2}{dt^2}r_2 + 2\gamma\frac{d}{dt}r_2 + \omega_0{}^2 r_2 = \xi r_1^2$ (**)

General form of the field:

$$E = \sum E(\omega_n)e^{-i\omega_n t}$$

Calculate: $\frac{d}{dt}r_1$ $\frac{d^2}{dt^2}r_1$

Insert in (*)

$$r_1 = -\frac{e}{m}\frac{\sum E(\omega_n)e^{-i\omega_n t}}{(\omega_0-\omega_n)^2 - 2i\gamma\omega_n}$$

Calculate r_1 and insert in (**); use

$$\left(\sum E(\omega_n)e^{i\omega_n t}\right)^2 = \sum\sum E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}$$

$$r_2 = -\frac{e\xi}{m^2}\frac{\sum\sum E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}}{\left[\omega_0^2-\omega_n^2-2i\gamma\omega_n\right]\left[\omega_0^2-\omega_m^2-2i\gamma\omega_m\right]\left[\omega_0^2-(\omega_n+\omega_m)^2-2i\gamma(\omega_n+\omega_m)\right]}$$

Write polarization;

$$P = \sum P_k \qquad P_k = -Ner_k$$

$$P_{linear} = \sum \chi^{(1)}(\omega_n)E(\omega_n)e^{-i\omega_n t}$$

$$P_{second} = \sum\sum \chi^{(2)}(\omega_n,\omega_m)E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}$$

Linear and nonlinear sucseptibilities:

$$\chi^{(1)}(\omega_n) = \frac{Ne^2}{m}\frac{1}{(\omega_0-\omega_n)^2 - 2i\gamma\omega_n}$$

$$\chi^{(2)}(\omega_n,\omega_m) = \frac{Ne^3\xi}{m^2}\frac{1}{\left[(\omega_0-\omega_n)^2-2i\gamma\omega_n\right]\left[(\omega_0-\omega_m)^2-2i\gamma\omega_m\right]} \times \frac{1}{\left[(\omega_0-(\omega_n+\omega_m))^2-2i\gamma(\omega_n+\omega_m)\right]}$$

Verify: $\chi^{(2)}(\omega_n,\omega_m) = \frac{-m\xi}{N^2e^3}\chi^{(1)}(\omega_n)\chi^{(1)}(\omega_m)\chi^{(1)}(\omega_n+\omega_m)$

Maxwell's equations for nonlinear optics

Starting point:

$$\vec{\nabla}\times\vec{E} = -\frac{\partial \vec{B}}{\partial t} \qquad \vec{\nabla}\times\vec{H} = \vec{j} + \frac{\partial \vec{D}}{\partial t}$$

$$\vec{\nabla}\cdot\vec{D} = \rho \qquad \vec{\nabla}\cdot\vec{B} = 0$$

with

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} \qquad \vec{j} = \sigma\vec{E}$$

Induced polarization:

$$\vec{P} = \varepsilon_0 \chi \vec{E} + \vec{P}^{NL}$$

Insert in Maxwells equation

$$\varepsilon = \varepsilon_0(1+\chi)$$

$$\vec{\nabla}\times\vec{H} = \sigma\vec{E} + \varepsilon\frac{\partial \vec{E}}{\partial t} + \frac{\partial \vec{P}^{NL}}{\partial t}$$

Use the equation for $\vec{E}$

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E} = -\frac{\partial}{\partial t}\left(\vec{\nabla}\times\vec{B}\right) = -\mu\frac{\partial}{\partial t}\left(\vec{\nabla}\times\vec{H}\right)$$

$$= -\mu\frac{\partial}{\partial t}\left(\sigma\vec{E} + \varepsilon\frac{\partial \vec{E}}{\partial t} + \frac{\partial}{\partial t}\vec{P}^{NL}\right)$$

Use the vector relation:

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E} = \vec{\nabla}\left(\vec{\nabla}\cdot\vec{E}\right) - \nabla^2\vec{E}$$

And (no charges in medium) $\vec{\nabla}\cdot\vec{E} = 0$

$$\nabla^2\vec{E} = \mu\sigma\frac{\partial \vec{E}}{\partial t} + \mu\varepsilon\frac{\partial^2 \vec{E}}{\partial t^2} + \mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

Maxwells wave equation in nonlinear optics

This equation for SI units

$$\vec{P}^{(n)} = \varepsilon_0 \chi^{(n)} \vec{E}^{(n)}$$

in C/m²

Often used esu units

$$\vec{P}^{(n)} = \chi^{(n)} \vec{E}^{(n)}$$

in statvolt/cm

$$\frac{\chi_{SI}^{(n)}}{\chi_{esu}^{(n)}} = 4\pi / \left(10^{-4} c\right)^{n-1}$$

$$\frac{P_{SI}^{(n)}}{P_{esu}^{(n)}} = \frac{10^3}{c}$$

Coupled Wave Equations

Input waves, *plane waves*, at frequencies

$$\omega_1 \qquad \omega_2$$

$$\vec{E}(t) = \mathrm{Re}\left[E(\omega_1)\exp(i\omega_1 t) + E(\omega_2)\exp(i\omega_2 t)\right]$$

Polarization at the <u>sum</u>-frequency:

$$P_i(\omega_1+\omega_2) = \mathrm{Re}\left[\chi_{ijk}(\omega=\omega_1+\omega_2)E_j(\omega_1)E_k(\omega_2)\exp[i(\omega_1+\omega_2)t]\right]$$

and at the <u>difference</u>-frequency:

$$P_i(\omega_1-\omega_2) = \mathrm{Re}\left[\chi_{ijk}(\omega=\omega_1-\omega_2)E_j(\omega_1)E_k{}^*(\omega_2)\exp[i(\omega_1-\omega_2)t]\right]$$

Notation: $E_k(-\omega_2) = E_k^*(\omega_2)$

$\chi_{ijk}(\omega=\omega_1+\omega_2)$ and $\chi_{ijk}(\omega=\omega_1-\omega_2)$

are material properties of the medium

Use Maxwell's equation

$$\nabla^2\vec{E} = \mu\sigma\frac{\partial\vec{E}}{\partial t} + \mu\varepsilon\frac{\partial^2\vec{E}}{\partial t^2} + \mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

- take one component of linear polarization
- propagate plane wave along z-axis

$$E_1(z,t) = E_1(z)\exp(i\omega_1 t - ik_1 z)$$
$$E_2(z,t) = E_2(z)\exp(i\omega_2 t - ik_2 z)$$

Producing a non-linear polarization at sum.

$$P_{NL}(z,t) = dE_1(z)E_2(z) \times \exp[i(\omega_1+\omega_2)t - i(k_1+k_2)z]$$

A new field is created at $\omega_3 = \omega_1 + \omega_2$

$$E_3(z,t) = E_3(z)\exp(i\omega_3 t - ik_3 z)$$

All this is subsituted into Maxwell's equation

and $\nabla^2 E_3(z,t) = \frac{d^2}{dz^2}E_3(z)$

Coupled Wave Equations - 2

Again

$$\nabla^2 \vec{E} - \mu\sigma \frac{\partial \vec{E}}{\partial t} - \mu\varepsilon \frac{\partial^2 \vec{E}}{\partial t^2} = \mu \frac{\partial^2}{\partial t^2} \vec{P}^{NL}$$

Substitute left side:

$$\frac{d^2}{dz^2} E_3(z,t) - \mu\sigma \frac{d}{dt} E_3(z,t) - \mu\varepsilon \frac{d^2}{dt^2} E_3(z,t) =$$

$$\frac{d^2}{dz^2} E_3(z,t) + 2ik_3 \frac{d}{dz} E_3(z,t) - k_3^2 E_3(z,t)$$

$$+ i\omega_3 \mu\sigma E_3(z,t) + \mu\varepsilon\omega_3^2 E_3(z,t)$$

Slowly varying amplitude approximation

$$\left| \frac{d^2}{dz^2} E_3(z,t) \right| << \left| 2ik_3 \frac{d}{dz} E_3(z,t) \right|$$

Variation of the amplitude of the distance of a wavelength is small

$$\frac{d^2}{dz^2} E_3(z,t) + 2ik_3 \frac{d}{dz} E_3(z,t) - k_3^2 E_3(z,t)$$

$$+ i\omega_3 \mu\sigma E_3(z,t) + \mu\varepsilon\omega_3^2 E_3(z,t)$$

For plane waves in a medium;

$$\mu\varepsilon\omega_3^2 - k_3^2 = \frac{\omega_3^2}{c^2} - k_3^2 = 0$$

So left side of wave equation;

$$2ik_3 \frac{d}{dz} E_3(z,t) + i\omega_3 \mu\sigma E_3(z,t)$$

Right side of wave equation

$$\mu \frac{\partial^2}{\partial t^2} \vec{P}^{NL} =$$

$$\mu \frac{d^2}{dt^2} dE_1(z) E_2(z) \exp[i(\omega_1 + \omega_2)t - i(k_1 + k_2)z] =$$

$$-\mu(\omega_1 + \omega_2)^2 dE_1(z) E_2(z) \exp[i(\omega_1 + \omega_2)t - i(k_1 + k_2)z]$$

Coupled Wave Equations - 3

Equate left and right side and use:

$$\omega_3 = ck_3 \qquad \omega_3 = \omega_1 + \omega_2$$

Then:

$$\frac{d}{dz}E_3(z) = -\frac{\sigma}{2}\sqrt{\frac{\mu}{\varepsilon_3}}E_3(z) - \frac{i\omega_3}{2}\sqrt{\frac{\mu}{\varepsilon_3}}dE_1(z)E_2(z)\exp\left[-i(k_1+k_2-k_3)z\right]$$

This is a coupled-wave equation.
Also reverse processes occur: $\omega_3 - \omega_2 \to \omega_1$

Leading to other coupled equations

$$\frac{d}{dz}E_1(z) = -\frac{\sigma}{2}\sqrt{\frac{\mu}{\varepsilon_1}}E_1(z) - \frac{i\omega_1}{2}\sqrt{\frac{\mu}{\varepsilon_1}}dE_3(z)E_2(z)^*\exp\left[-i(k_3-k_2-k_1)z\right]$$

$$\frac{d}{dz}E_2(z)^* = -\frac{\sigma}{2}\sqrt{\frac{\mu}{\varepsilon_2}}E_2(z)^* + \frac{i\omega_2}{2}\sqrt{\frac{\mu}{\varepsilon_2}}dE_1(z)E_3(z)^*\exp\left[-i(k_1+k_2-k_3)z\right]$$

Three differential equations describe the couplings of the fields

Note that we used cancellation of the frequency terms via:

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

But this does not hold for the spatial phase factors, because:

$$\omega_i = \frac{k_i}{\sqrt{\mu\varepsilon(\omega_i)}} = \frac{ck_i}{n(\omega_i)}$$

Hence:

$$k_1 + k_2 - k_3 \neq 0$$

There is a phase-mismatch because of dispersion in the medium.

Define the <u>wave vector mismatch</u>:

$$\Delta\vec{k} = \vec{k}_3 - \vec{k}_1 - \vec{k}_2$$

This relation pertains to <u>plane</u> waves;
Later we will use focused beams.

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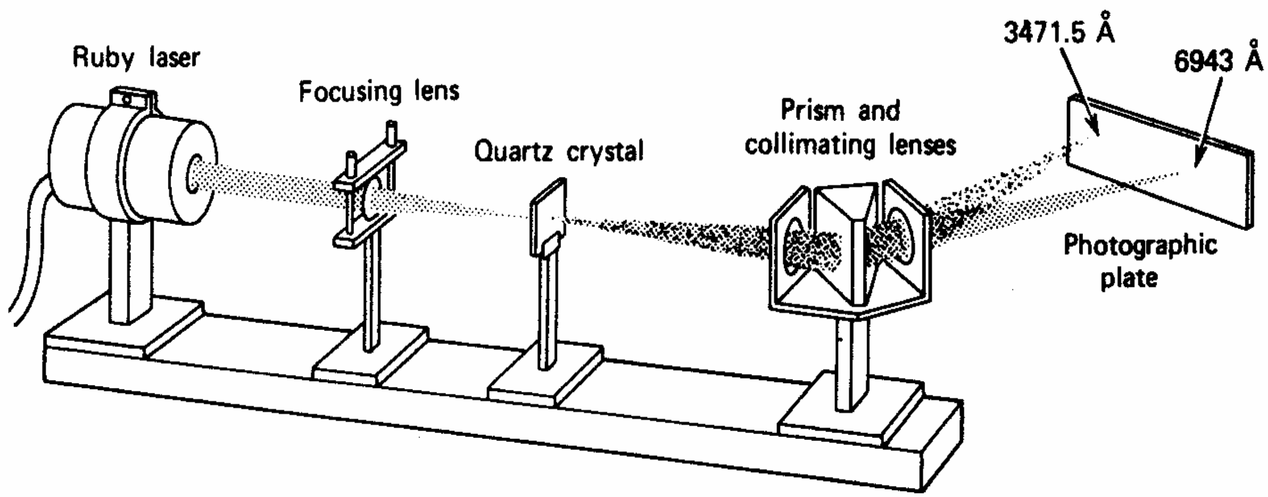

P.A. Franken, A.E. Hill, C.W. Peters and G. Weinreich, Phys. Rev. Lett. **7** (1961) 118

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The $\chi_{ij}^{(n)}$ are tensors even for lowest order

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Polarization not directed along electric field vector

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$$I_{\text{op}}\vec{E} = -\vec{E}$$

Hence:

$$I_{\text{op}}\vec{P} = -\chi^{(1)}\vec{E} + \chi^{(2)}\vec{E}\vec{E} - \chi^{(3)}\vec{E}\vec{E}\vec{E} + \ldots$$

Conclude for centro-symmetric media:

$$\chi^{(2n)} = 0$$

Note that in principle there exist also nonlinear magnetic susceptibilities

Nonlinear Optics; graphically

Linear response:

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

Nonlinear response:

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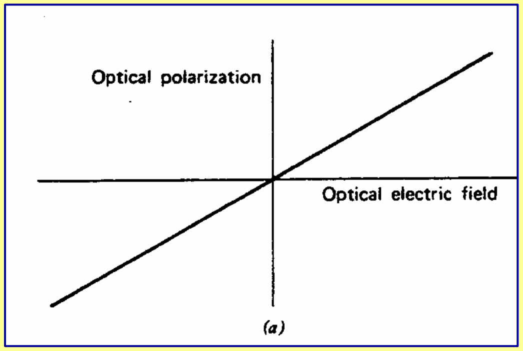

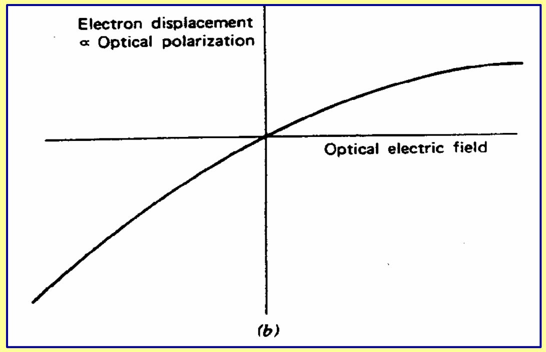

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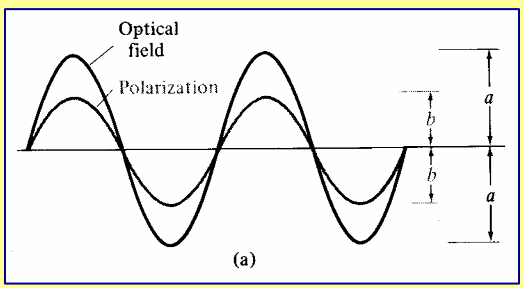
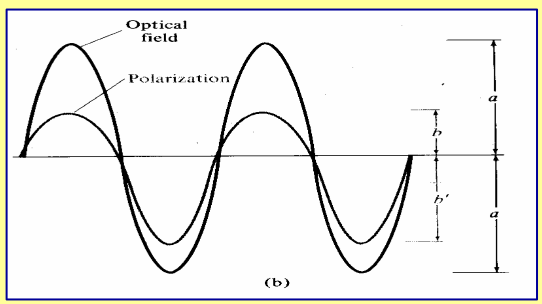
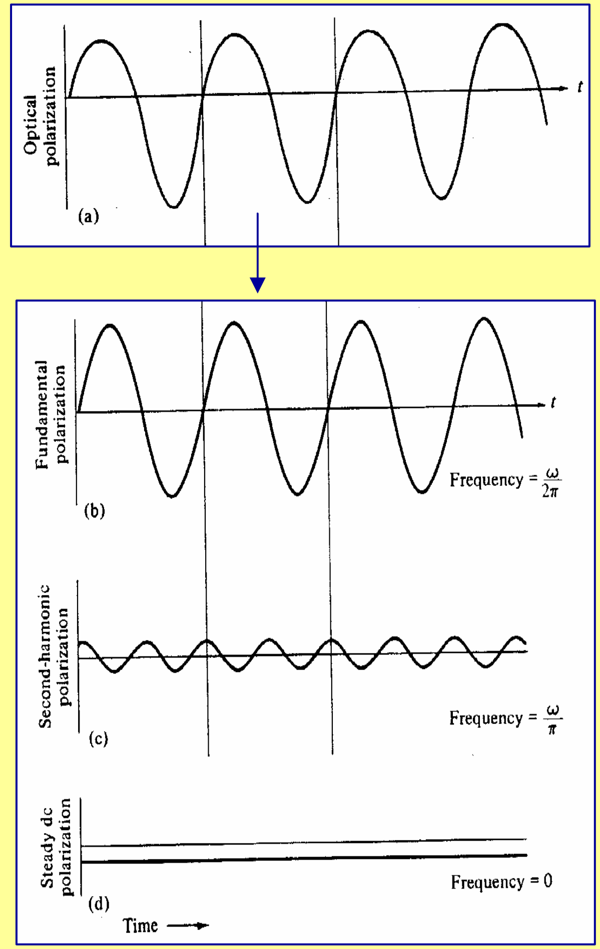




Nonlinear response evaluated in terms of Fourier series



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Lorentz model of linear optics: classical oscillator

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Write electric field and position vector:

$$\mathbf{E} = \mathrm{Re}\left[Ee^{i\omega t}\right] \qquad \mathbf{r} = \mathrm{Re}\left[re^{i\omega t}\right]$$

$$\longrightarrow \quad \left(\omega_0^2 - \omega^2\right)r + 2i\omega\gamma r = -\frac{e}{m}E$$

Solution

$$r = \frac{-eE}{m\left[\omega_0{}^2 - \omega^2 + 2i\omega\gamma\right]} \approx \frac{-eE}{2m\left[\omega_0\left(\omega_0 - \omega\right) + i\omega\gamma\right]}$$

Near resonance $\omega = \omega_0$

$$\longrightarrow \quad r = \frac{Ne^2}{2m\left[\omega_0\left(\omega_0 - \omega\right) + i\omega\gamma\right]}E = \varepsilon_0\chi(\omega)E$$

Classical polarization of the medium

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and the complex susceptibility

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$$

yields expressions for the susceptibility

Real part,
connected to the index of refraction

$$\chi'(\omega) = \frac{Ne^2}{2m\omega_0\gamma\varepsilon_0}\frac{(\omega_0 - \omega)/\gamma}{\left[1 + (\omega_0 - \omega)^2/\gamma^2\right]}$$

Imaginary part,
connected to the absorption coefficient

$$\chi''(\omega) = \frac{Ne^2}{2m\omega_0\gamma\varepsilon_0}\frac{1}{\left[1 + (\omega_0 - \omega)^2/\gamma^2\right]}$$

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Motion of electron with anharmonic term:

$$\frac{d^2}{dt^2}r + 2\gamma\frac{d}{dt}r + \omega_0^2 r - \xi r^2 = -\frac{e}{m}E$$

Try a solution in power series

$$r = r_1 + r_2 + r_{3+...}$$

with: $r_i = a_i E^i$

Collect terms in same order of E

First order $\frac{d^2}{dt^2}r_1 + 2\gamma\frac{d}{dt}r_1 + \omega_0^2 r_1 = -\frac{e}{m}E$ (*)

Second order $\frac{d^2}{dt^2}r_2 + 2\gamma\frac{d}{dt}r_2 + \omega_0^2 r_2 = \xi r_1^2$ (**)

General form of the field:

$$E = \sum E(\omega_n)e^{-i\omega_n t}$$

Calculate: $\frac{d}{dt}r_1$ $\frac{d^2}{dt^2}r_1$

Insert in (*)

$$r_1 = -\frac{e}{m}\frac{\sum E(\omega_n)e^{-i\omega_n t}}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n}$$

Calculate r_1 and insert in (**); use

$$\left(\sum E(\omega_n)e^{i\omega_n t}\right)^2 = \sum\sum E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}$$

$$r_2 = -\frac{e\xi}{m^2}\frac{\sum\sum E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}}{\left[\omega_0^2 - \omega_n^2 - 2i\gamma\omega_n\right]\left[\omega_0^2 - \omega_m^2 - 2i\gamma\omega_m\right]\left[\omega_0^2 - (\omega_n + \omega_m)^2 - 2i\gamma(\omega_n + \omega_m)\right]}$$

Write polarization;

$$P = \sum P_k \qquad P_k = -Ner_k$$

$$P_{linear} = \sum \chi^{(1)}(\omega_n)E(\omega_n)e^{-i\omega_n t}$$

$$P_{second} = \sum\sum \chi^{(2)}(\omega_n, \omega_m)E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}$$

Linear and nonlinear sucseptibilities:

$$\chi^{(1)}(\omega_n) = \frac{Ne^2}{m}\frac{1}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n}$$

$$\chi^{(2)}(\omega_n, \omega_m) = \frac{Ne^3\xi}{m^2}\frac{1}{\left[(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n\right]\left[(\omega_0 - \omega_m)^2 - 2i\gamma\omega_m\right]} \times \frac{1}{\left[(\omega_0 - (\omega_n + \omega_m))^2 - 2i\gamma(\omega_n + \omega_m)\right]}$$

Verify: $\chi^{(2)}(\omega_n, \omega_m) = \frac{-m\xi}{N^2 e^3}\chi^{(1)}(\omega_n)\chi^{(1)}(\omega_m)\chi^{(1)}(\omega_n + \omega_m)$

Maxwell's equations for nonlinear optics

Starting point:

$$\vec{\nabla}\times\vec{E} = -\frac{\partial\vec{B}}{\partial t} \qquad \vec{\nabla}\times\vec{H} = \vec{j} + \frac{\partial\vec{D}}{\partial t}$$

$$\vec{\nabla}\cdot\vec{D} = \rho \qquad \vec{\nabla}\cdot\vec{B} = 0$$

with

$$\vec{D} = \varepsilon_0\vec{E} + \vec{P} \qquad \vec{j} = \sigma\vec{E}$$

Induced polarization:

$$\vec{P} = \varepsilon_0\chi\vec{E} + \vec{P}^{NL}$$

Insert in Maxwells equation

$$\varepsilon = \varepsilon_0(1+\chi)$$

$$\vec{\nabla}\times\vec{H} = \sigma\vec{E} + \varepsilon\frac{\partial\vec{E}}{\partial t} + \frac{\partial\vec{P}^{NL}}{\partial t}$$

Use the equation for $\vec{E}$

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E} = -\frac{\partial}{\partial t}\left(\vec{\nabla}\times\vec{B}\right) = -\mu\frac{\partial}{\partial t}\left(\vec{\nabla}\times\vec{H}\right)$$

$$= -\mu\frac{\partial}{\partial t}\left(\sigma\vec{E} + \varepsilon\frac{\partial\vec{E}}{\partial t} + \frac{\partial}{\partial t}\vec{P}^{NL}\right)$$

Use the vector relation:

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E} = \vec{\nabla}\left(\vec{\nabla}\cdot\vec{E}\right) - \nabla^2\vec{E}$$

And (no charges in medium) $\vec{\nabla}\cdot\vec{E} = 0$

$$\nabla^2\vec{E} = \mu\sigma\frac{\partial\vec{E}}{\partial t} + \mu\varepsilon\frac{\partial^2\vec{E}}{\partial t^2} + \mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

Maxwells wave equation in nonlinear optics

This equation for SI units

$$\vec{P}^{(n)} = \varepsilon_0\chi^{(n)}\vec{E}^{(n)}$$

in C/m²

Often used esu units

$$\vec{P}^{(n)} = \chi^{(n)}\vec{E}^{(n)}$$

in statvolt/cm

$$\frac{\chi_{SI}^{(n)}}{\chi_{esu}^{(n)}} = 4\pi/\left(10^{-4}c\right)^{n-1}$$

$$\frac{P_{SI}^{(n)}}{P_{esu}^{(n)}} = \frac{10^3}{c}$$

Coupled Wave Equations

Input waves, *plane waves*, at frequencies

$$\omega_1 \qquad \omega_2$$

$$\vec{E}(t) = \mathrm{Re}\left[E(\omega_1)\exp(i\omega_1 t) + E(\omega_2)\exp(i\omega_2 t)\right]$$

Polarization at the <u>sum</u>-frequency:

$$P_i(\omega_1+\omega_2) =$$
$$\mathrm{Re}\left[\chi_{ijk}(\omega=\omega_1+\omega_2)E_j(\omega_1)E_k(\omega_2)\exp[i(\omega_1+\omega_2)t]\right]$$

and at the <u>difference</u>-frequency:

$$P_i(\omega_1-\omega_2) =$$
$$\mathrm{Re}\left[\chi_{ijk}(\omega=\omega_1-\omega_2)E_j(\omega_1)E_k{}^*(\omega_2)\exp[i(\omega_1-\omega_2)t]\right]$$

Notation: $E_k(-\omega_2) = E_k^*(\omega_2)$

$\chi_{ijk}(\omega=\omega_1+\omega_2)$ and $\chi_{ijk}(\omega=\omega_1-\omega_2)$

are material properties of the medium

Use Maxwell's equation

$$\nabla^2\vec{E} = \mu\sigma\frac{\partial\vec{E}}{\partial t} + \mu\varepsilon\frac{\partial^2\vec{E}}{\partial t^2} + \mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

- take one component of linear polarization
- propagate plane wave along z-axis

$$E_1(z,t) = E_1(z)\exp(i\omega_1 t - ik_1 z)$$
$$E_2(z,t) = E_2(z)\exp(i\omega_2 t - ik_2 z)$$

Producing a non-linear polarization at sum.

$$P_{NL}(z,t) = dE_1(z)E_2(z)$$
$$\times\exp[i(\omega_1+\omega_2)t - i(k_1+k_2)z]$$

A new field is created at $\omega_3 = \omega_1 + \omega_2$

$$E_3(z,t) = E_3(z)\exp(i\omega_3 t - ik_3 z)$$

All this is subsituted into Maxwell's equation

and $\nabla^2 E_3(z,t) = \frac{d^2}{dz^2}E_3(z)$

Coupled Wave Equations - 2

Again

$$\nabla^2\vec{E} - \mu\sigma\frac{\partial\vec{E}}{\partial t} - \mu\varepsilon\frac{\partial^2\vec{E}}{\partial t^2} = \mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

Substitute left side:

$$\frac{d^2}{dz^2}E_3(z,t) - \mu\sigma\frac{d}{dt}E_3(z,t) - \mu\varepsilon\frac{d^2}{dt^2}E_3(z,t) =$$

$$\frac{d^2}{dz^2}E_3(z,t) + 2ik_3\frac{d}{dz}E_3(z,t) - k_3^2E_3(z,t)$$

$$+ i\omega_3\mu\sigma E_3(z,t) + \mu\varepsilon\omega_3^2E_3(z,t)$$

Slowly varying amplitude approximation

$$\left|\frac{d^2}{dz^2}E_3(z,t)\right| << \left|2ik_3\frac{d}{dz}E_3(z,t)\right|$$

Variation of the amplitude of the distance of a wavelength is small

$$\frac{d^2}{dz^2}E_3(z,t) + 2ik_3\frac{d}{dz}E_3(z,t) - k_3^2E_3(z,t)$$

$$+ i\omega_3\mu\sigma E_3(z,t) + \mu\varepsilon\omega_3^2E_3(z,t)$$

For plane waves in a medium;

$$\mu\varepsilon\omega_3^2 - k_3^2 = \frac{\omega_3^2}{c^2} - k_3^2 = 0$$

So left side of wave equation;

$$2ik_3\frac{d}{dz}E_3(z,t) + i\omega_3\mu\sigma E_3(z,t)$$

Right side of wave equation

$$\mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL} =$$

$$\mu\frac{d^2}{dt^2}dE_1(z)E_2(z)\exp[i(\omega_1+\omega_2)t - i(k_1+k_2)z] =$$

$$-\mu(\omega_1+\omega_2)^2 dE_1(z)E_2(z)\exp[i(\omega_1+\omega_2)t - i(k_1+k_2)z]$$

Coupled Wave Equations - 3

Equate left and right side and use:

$$\omega_3 = ck_3 \qquad \omega_3 = \omega_1 + \omega_2$$

Then:

$$\frac{d}{dz}E_3(z) = -\frac{\sigma}{2}\sqrt{\frac{\mu}{\varepsilon_3}}E_3(z) - \frac{i\omega_3}{2}\sqrt{\frac{\mu}{\varepsilon_3}}dE_1(z)E_2(z)\exp\left[-i(k_1 + k_2 - k_3)z\right]$$

This is a coupled-wave equation.
Also reverse processes occur: $\omega_3 - \omega_2 \rightarrow \omega_1$

Leading to other coupled equations

$$\frac{d}{dz}E_1(z) = -\frac{\sigma}{2}\sqrt{\frac{\mu}{\varepsilon_1}}E_1(z) - \frac{i\omega_1}{2}\sqrt{\frac{\mu}{\varepsilon_1}}dE_3(z)E_2(z)^*\exp\left[-i(k_3 - k_2 - k_1)z\right]$$

$$\frac{d}{dz}E_2(z)^* = -\frac{\sigma}{2}\sqrt{\frac{\mu}{\varepsilon_2}}E_2(z)^* + \frac{i\omega_2}{2}\sqrt{\frac{\mu}{\varepsilon_2}}dE_1(z)E_3(z)^*\exp\left[-i(k_1 + k_2 - k_3)z\right]$$

Three differential equations describe the couplings of the fields

Note that we used cancellation of the frequency terms via:

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

But this does not hold for the spatial phase factors, because:

$$\omega_i = \frac{k_i}{\sqrt{\mu\varepsilon(\omega_i)}} = \frac{ck_i}{n(\omega_i)}$$

Hence:

$$k_1 + k_2 - k_3 \neq 0$$

There is a phase-mismatch because of dispersion in the medium.

Define the wave vector mismatch:

$$\Delta\vec{k} = \vec{k}_3 - \vec{k}_1 - \vec{k}_2$$

This relation pertains to plane waves;
Later we will use focused beams.