

Nonlinear Optical Engineering

Nonlinear Susceptibility (2)

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Nonlinear Polarisation

Constitutive relations:

$$\mathbf{D} = \epsilon \mathbf{E} = \epsilon_0 \mathbf{E} + \mathbf{P},$$

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}$$

Origin of the nonlinear response:

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E} = \epsilon_0 \left(\chi^{(1)} \mathbf{E} + \chi^{(2)} \mathbf{E} \mathbf{E} + \chi^{(3)} \mathbf{E} \mathbf{E} \mathbf{E} + \dots \right)$$

← Anharmonic motion of bound electrons

Note: Non-zero $\chi^{(2)}$ only for media that lack an inversion symmetry (centrosymmetry)

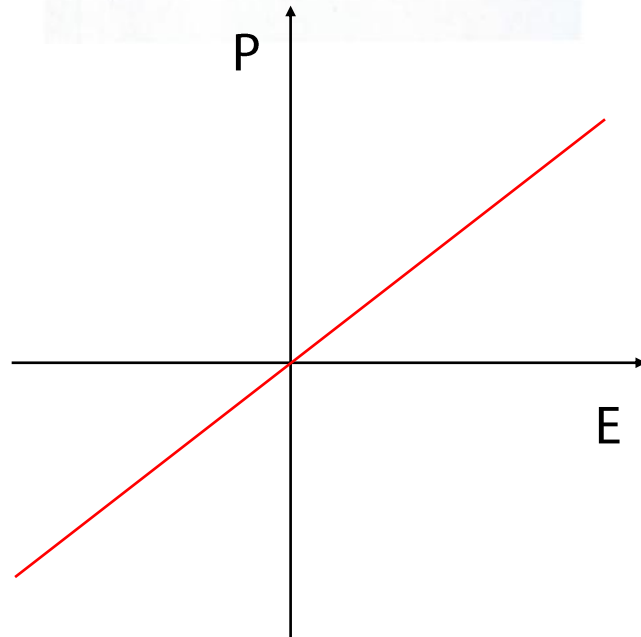
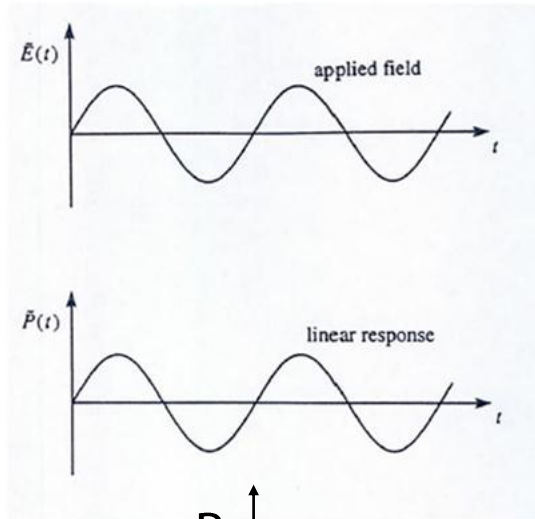
Second-order and third-order nonlinear susceptibilities:

$$\rightarrow \chi^{(2)} \equiv d_{ijk}$$

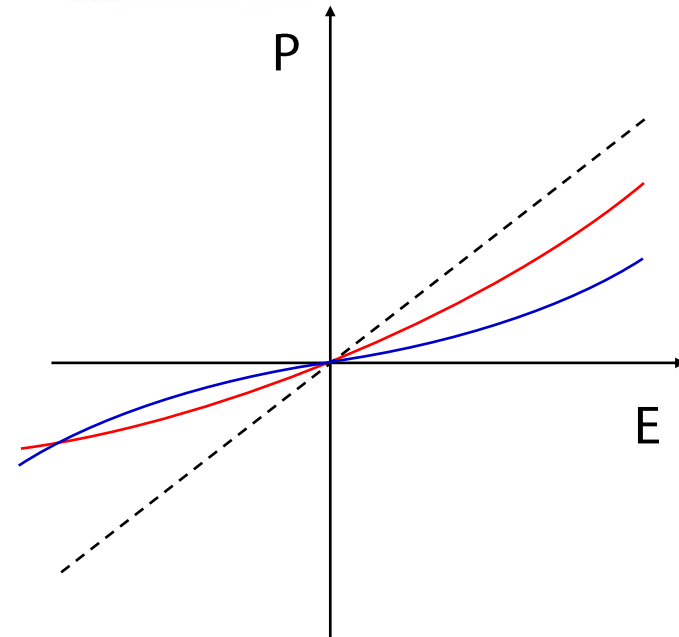
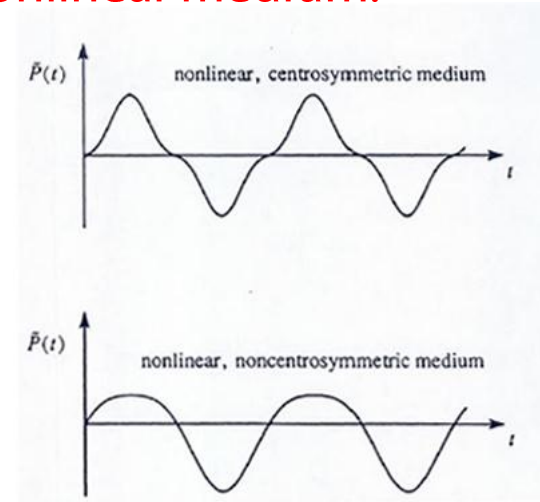
$$\rightarrow \chi^{(3)} \equiv \chi_{ijkl}$$

Linear and Nonlinear Atomic Response

Linear medium:



Nonlinear medium:



Tensor Notation for the 3rd-Order Susceptibility

How many elements for χ_{ijkl} ? $\rightarrow 81$ elements

For isotropic media: $\rightarrow 21$ nonzero elements

$$\chi_{1111} = \chi_{2222} = \chi_{3333},$$

$$\chi_{1122} = \chi_{1133} = \chi_{2211} = \chi_{2233} = \chi_{3311} = \chi_{3322},$$

$$\chi_{1212} = \chi_{1313} = \chi_{2323} = \chi_{2121} = \chi_{3131} = \chi_{3232},$$

$$\chi_{1221} = \chi_{1331} = \chi_{2112} = \chi_{2332} = \chi_{3113} = \chi_{3223}.$$

\leftarrow Centrosymmetry

In addition:

$$\chi_{1111} = \chi_{1122} + \chi_{1212} + \chi_{1221} \quad \rightarrow \text{Why?}$$

\rightarrow In the compact form:

$$\chi_{ijkl} = \chi_{1122} \delta_{ij} \delta_{kl} + \chi_{1212} \delta_{ik} \delta_{jl} + \chi_{1221} \delta_{il} \delta_{jk}$$

Tensor Notation for the 2nd-Order Susceptibility

How many elements for d_{ijk} ? $\rightarrow 27$ elements

For cubic media: $\rightarrow 6$ nonzero elements

$$d_{123} = d_{132} = d_{213} = d_{231} = d_{312} = d_{321}$$

\rightarrow Why?