

Nonlinear polarization for an isotropic chi-3 material

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May 6, 2018

The most general expression for the nonlinear polarization density of a chi-3 material is

$$P_i(t) = \frac{\varepsilon_0}{(2\pi)^3} \sum_{jkl} \int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 \int_{-\infty}^{\infty} d\omega_3 \chi_{ijkl}^{(3)}(\omega_1 + \omega_2 + \omega_3; \omega_1, \omega_2, \omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3) e^{i(\omega_1 + \omega_2 + \omega_3)t} \quad (1)$$

If the material is dispersionless, $\chi^{(3)}$ is frequency independent. Then, Eq. 1 becomes

$$\begin{aligned} P_i(t) &= \frac{\varepsilon_0}{(2\pi)^3} \sum_{jkl} \chi_{ijkl}^{(3)} \int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 \int_{-\infty}^{\infty} d\omega_3 E_j(\omega_1) E_k(\omega_2) E_l(\omega_3) e^{i(\omega_1 + \omega_2 + \omega_3)t} \\ &= \varepsilon_0 \sum_{jkl} \chi_{ijkl}^{(3)} E_j(t) E_k(t) E_l(t) \end{aligned} \quad (2)$$

The most general form for an isotropic fourth order tensor is

$$\chi_{ijkl}^{(3)} = \alpha \delta_{ij} \delta_{kl} + \beta \delta_{ik} \delta_{jl} + \gamma \delta_{il} \delta_{jk} \quad (3)$$

Inserting Eq. 3 into Eq. 2 yields

$$\begin{aligned} P_i(t) &= \varepsilon_0 \sum_{jkl} (\alpha \delta_{ij} \delta_{kl} + \beta \delta_{ik} \delta_{jl} + \gamma \delta_{il} \delta_{jk}) E_j(t) E_k(t) E_l(t) \\ &= \varepsilon_0 \left[\alpha E_i(t) \sum_k E_k^2(t) + \beta E_i(t) \sum_j E_j^2(t) + \gamma E_i(t) \sum_j E_j^2(t) \right] \\ &= \varepsilon_0 \chi^{(3)} E_i(t) [\vec{E}(t)]^2 \end{aligned} \quad (4)$$

where $\chi^{(3)} = \alpha + \beta + \gamma$. Eq. 4 is written in vectorial form as

$$\vec{P}(t) = \varepsilon_0 \chi^{(3)} [\vec{E}(t)]^2 \vec{E}(t) \quad (5)$$