

$\chi_i(t < 0) = 0$. The Fourier transform of Eq. 3 for a purely ac field $E(t) = E \cos(\omega t)$ gives:

$$\begin{aligned} \frac{P(\omega')}{\epsilon_0} = & \frac{E}{2} \left[\chi_1(\omega) + \frac{3E^2}{4} \chi_3(-\omega, \omega, \omega) + \dots \right] \delta(\omega' - \omega) \\ & + \frac{E}{2} \left[\chi_1(-\omega) + \frac{3E^2}{4} \chi_3(\omega, -\omega, -\omega) + \dots \right] \delta(\omega' + \omega) \\ & + \frac{E^3}{8} \chi_3(\omega, \omega, \omega) \delta(\omega' - 3\omega) \\ & + \frac{E^3}{8} \chi_3(-\omega, -\omega, -\omega) \delta(\omega' + 3\omega) + \dots, \end{aligned} \quad (4)$$

where the polarization P and the susceptibilities χ_i are now taken in the frequency domain and the dots indicate again infinite sums involving higher order terms. The response $P(t)$ to $E(t) = E \cos(\omega t)$ can thus be written

$$P(t)/\epsilon_0 = \text{Re} [(E\chi_1(\omega) + 3/4E^3\chi_3(\omega) + \dots)e^{-i\omega t}] + \text{Re} [1/4E^3\chi_3(\omega)e^{-i3\omega t} + \dots] + \dots \quad (5)$$

To obtain Eq. (5), we have used the fact that because χ_1 and χ_3 are real in the time domain, their Fourier transform verify $\chi_1^*(\omega) = \chi_1(-\omega)$ and $\chi_3^*(\omega_1, \omega_2, \omega_3) = \chi_3(-\omega_1, -\omega_2, -\omega_3)$ (the star denotes the complex conjugate), and the invariance of χ_3 by permutation of its arguments. For simplicity, we write $\chi_3(\omega) = \chi_3(\omega, \omega, \omega)$ and $\chi_3(\omega) = \chi_3(-\omega, \omega, \omega)$. Eq. (5) can be written

$$\begin{aligned} P(t)/\epsilon_0 = & E(\chi_1' \cos \omega t + \chi_1'' \sin \omega t) + 3/4E^3(\chi_3' \cos \omega t + \chi_3'' \sin \omega t) + \dots \\ & + 1/4E^3(\chi_3' \cos 3\omega t + \chi_3'' \sin 3\omega t) + \dots, \end{aligned} \quad (6)$$

where the susceptibilities χ_i are given as a function of their real and imaginary parts χ_i' and χ_i'' . For practical applications, the moduli and arguments $|\chi_i|$ and δ_i are rather used:

$$\begin{aligned} P(t)/\epsilon_0 = & E |\chi_1| \cos(\omega t - \delta_1) + 3/4E^3 |\chi_3| \cos(\omega t - \delta_3) + \\ & + \dots + 1/4E^3 |\chi_3| \cos(3\omega t - \delta_3) + \dots \end{aligned} \quad (7)$$

We see in the rhs in Eqs 5-7 that the nonlinear susceptibility of interest, namely χ_3