

1.4: Summary - General Expressions for nth Order Nonlinearity

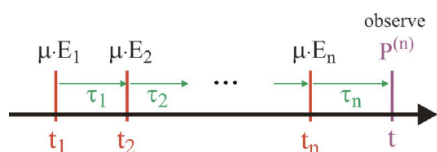
For an n^{th} -order nonlinear signal, there are n interactions with the incident electric field or fields that give rise to the radiated signal. Counting the radiated signal there are $n+1$ fields involved ($n+1$ light-matter interactions), so that n^{th} order spectroscopy is at times referred to as $(n+1)$ - wave mixing. The radiated nonlinear signal field is proportional to the nonlinear polarization:

$$P^{(n)}(t) = \int_0^\infty d\tau_n \cdots \int_0^\infty d\tau_1 R^{(n)}(\tau_1, \tau_2, \dots, \tau_n) \bar{E}_1(t - \tau_n - \cdots - \tau_1) \cdots \bar{E}_n(t - \tau_n) \quad (1.4.1)$$

$$R^{(n)}(\tau_1, \tau_2, \dots, \tau_n) = \left(\frac{i}{\hbar}\right)^n \theta(\tau_1) \theta(\tau_2) \cdots \theta(\tau_n) \\ \times \text{Tr}\{[\dots [\mu_I(\tau_n + \tau_{n-1} + \dots + \tau_1), \mu_I(\tau_{n-1} + \tau_n + \dots + \tau_1)], \dots] \mu_I(0) \rho_{eq}\}$$

Here the interactions of the light and matter are expressed in terms of a sequence of consecutive time intervals $\tau_1 \dots \tau_n$ prior to observing the system. For delta-function interactions, $\bar{E}_i(t - t_0) = |\bar{E}_i| \delta(t - t_0)$, the polarization and response function are directly proportional

$$P^{(n)}(t) = R^{(n)}(\tau_1, \tau_2, \dots, \tau_{n-1}, t) |\bar{E}_1| \cdots |\bar{E}_n| \quad (1.4.2)$$



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