1.4: Summary - General Expressions for nth Order Nonlinearity

For an nth-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 nth order spectroscopy is at times referred to as (*n*+1)- wave mixing. The radiated nonlinear signal field is proportional to the nonlinear polarization:

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

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

Here the interactions of the light and matter are expressed in terms of a sequence of consecutive time intervals $\tau_1 \ldots \tau_n$ prior to observing 14 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

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