2.6: Frequency Domain Representation(1)

A Fourier-Laplace transform of $P^{(3)}(t)$ with respect to the time intervals allows us to obtain an expression for the third order nonlinear susceptibility, $\chi^{(3)}\left(\omega_1, \omega_2, \omega_3\right)$:

$$P^{(3)}\left(\omega_{sig}\right) = \chi^{(3)}\left(\omega_{sig}; \omega_1, \omega_2, \omega_3\right) \bar{E}_1 \bar{E}_2 \bar{E}_3 \tag{2.6.1}$$

$$\chi^{(n)} = \int_0^{\infty} d\tau_n e^{i\Omega_n \tau_n} \cdots \int_0^{\infty} d\tau_1 e^{i\Omega_1 \tau_1} R^{(n)}\left(\tau_1, \tau_2, \ldots \tau_n\right) \tag{2.6.2}$$

Here the Fourier transform conjugate variables Ω_m to the time-interval τ_m are the sum over all frequencies for the incident field interactions up to the period for which you are evolving:

$$\Omega_m = \sum_{i=1}^{m} \omega_i \tag{2.6.3}$$

For instance, the conjugate variable for the third time-interval of a $k_1 - k_2 + k_3$ experiment is the sum over the three preceding incident frequencies $\Omega_3 = \omega_1 - \omega_2 + \omega_3$.

In general, \chi^{(3)} is a sum over many correlation functions and includes a sum over states:

$$\chi^{(3)}\left(\omega_1, \omega_2, \omega_3\right) = \frac{1}{6}\left(\frac{i}{\hbar}\right)^3 \sum_{abcd} p_a \sum_{\alpha=1}^{4} \left[\chi_\alpha - \chi_\alpha^*\right] \tag{2.6.4}$$

Here *a* is the initial state and the sum is over all possible intermediate states. Also, to describe frequency domain experiments, we have to permute over all possible time orderings. Most generally, the eight terms in $R^{(3)}$ lead to 48 terms for $\chi^{(3)}$, as a result of the 3!=6 permutations of the time-ordering of the input fields.[2]

Given a set of diagrams, we can write the nonlinear susceptibility directly as follows:

1) Read off products of light-matter interaction factors.

2) Multiply by resonance denominator terms that describe the propagation under H_0. In the frequency domain, if we apply eq. (3.6.2) to response functions that use phenomenological time-propagators of the form eq. (3.2.1), we obtain

$$\hat{G}(\tau_m)\rho_{ab} \implies \frac{1}{\left(\Omega_m - \omega_{ba}\right) - i\Gamma_{ba}} \tag{2.6.5}$$

Ω_m is defined in eq. (3.6.3).

3) As for the time domain, multiply by a factor of $(-1)^n$ for *n bra* side interactions.

4) The radiated signal will have frequency $\omega_{sig} = \sum_i \omega_i$ and wavevector $\bar{k}_{sig} = \sum_i \bar{k}_i$.

As an example, consider the term for R_2 applied to a two-level system that we wrote in the time domain in eq. (3.5.2)

$$\begin{aligned} \chi_2 &= \left|\mu_{ba}\right|^4 \frac{(-1)}{\omega_{ab} - (-\omega_1) - i\Gamma_{ab}} \cdot \frac{1}{\omega_{\cancel{bb}} - (\omega_2 - \omega_1) - i\Gamma_{bb}} \cdot \frac{(-1)}{\omega_{ba} - (\omega_3 + \omega_2 - \omega_1) - i\Gamma_{ba}} \\ &= \left|\mu_{ba}\right|^4 \frac{1}{\omega_1 - \omega_{ba} - i\Gamma_{ba}} \cdot \frac{1}{-(\omega_2 - \omega_1) - i\Gamma_{bb}} \cdot \frac{1}{-(\omega_3 + \omega_2 - \omega_1 - \omega_{ba}) - i\Gamma_{ba}} \end{aligned}$$

The terms are written from a diagram with each interaction and propagation adding a resonant denominator term (here reading left to right). The full frequency domain response is a sum over multiple terms like these.

1. Prior, Y. A complete expression for the third order susceptibility $\chi^{(3)}$-perturbative and diagramatic approaches. *IEEE J. Quantum Electron*. **QE-20**, 37 (1984).

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2. Bloembergen, N., Lotem, H. & Lynch, R. T. Lineshapes in coherent resonant Raman scattering. *Indian J. Pure Appl. Phys.* **16**, 151 (1978).