5.3: The Density Matrix in the Interaction Picture

For the case in which we wish to describe a material Hamiltonian H_0 under the influence of an external potential $V(t)$,

$$H(t) = H_0 + V(t) \tag{5.3.1}$$

we can also formulate the density operator in the interaction picture, ρ_I. From our original definition of the interaction picture wavefunctions

$$|\psi_I\rangle = U_0^\dagger |\psi_S\rangle \tag{5.3.2}$$

We obtain ρ_I as

$$\rho_I = U_0^\dagger \rho_S U_0 \tag{5.3.3}$$

Similar to the discussion of the density operator in the Schrödinger equation, above, the equation of motion in the interaction picture is

$$\frac{\partial \rho_I}{\partial t} = -\frac{i}{\hbar}[V_I(t), \rho_I(t)] \tag{5.3.4}$$

where, as before, $V_I = U_0^\dagger V U_0$.

Equation 5.3.4 can be integrated to obtain

$$\rho_I(t) = \rho_I(t_0) - \frac{i}{\hbar}\int_{t_0}^{t} dt' \left[V_I(t'), \rho_I(t')\right] \tag{5.3.5}$$

Repeated substitution of $\rho_I(t)$ into itself in this expression gives a perturbation series expansion

$$\begin{aligned}
\rho_I(t) &= \rho_0 - \frac{i}{\hbar}\int_{t_0}^{t} dt_2 \left[V_I(t_1), \rho_0\right] && (5.3.6)\\
&+ \left(-\frac{i}{\hbar}\right)\int_{t_0}^{t} dt_2 \int_{t_0}^{t_2} dt_1 \left[V_I(t_2), \left[V_I(t_1), \rho_0\right]\right] + \cdots && (5.3.7)\\
&+ \left(-\frac{i}{\hbar}\right)^n \int_{t_0}^{t} dt_n \int_{t_0}^{t_n} dt_{n-1} \cdots \int_{t_0}^{t_2} dt_1 \left[V_I(t_n), \left[V_I(t_{n-1}), \cdots \left[V_I(t_1), \rho_0\right]\right]\right] && (5.3.8)\\
&= \rho^{(0)} + \rho^{(1)} + \rho^{(2)} + \cdots + \rho^{(n)} + \cdots && (5.3.9)
\end{aligned}$$

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Here $\rho_0 = \rho(t_0)$ and $\rho^{(n)}$ is the n$^{\text{th}}$-order expansion of the density matrix. This perturbative expansion will play an important role later in the description of nonlinear spectroscopy. An n$^{\text{th}}$ order expansion term will be proportional to the observed polarization in an n$^{\text{th}}$-order nonlinear spectroscopy, and the commutators observed in Equation 5.3.8 are closely related to nonlinear response functions. Equation 5.3.8 can also be expressed as

$$\rho_I(t) = U_0 \rho_I(0) U_0^\dagger \tag{5.3.10}$$

This is the solution to the Liouville equation in the interaction picture.