APPENDIX: THE DIELECTRIC SUSCEPTIBILITY
A General *Dressed State* Formulation

Suppose that the complete Hamiltonian of a coupled system is parsed into two components

$$\mathcal{H} = \mathcal{H}_o + \mathcal{H}_{ex} . \qquad [\text{ VA-1 }]$$

The component $\mathcal{H}_o$ includes the Hamiltonians for the unperturbed material system, the free radiation field and interactions of the material system with available *cavity modes*. The component $\mathcal{H}_{ex}$ is the Hamiltonian for the interactions which couple the material system to **externally excited modes**. As the first step in finding a fully quantal expression for the dielectric susceptibility, let us expand the state vector in the Schrödinger picture in terms of, presumably, known eigenkets of $\mathcal{H}_o$ -- *viz*. the ***dressed states*** of the unperturbed system --

$$\left|\psi(t)\right\rangle = \sum_s C_s(t) \exp(-i\,\lambda_s\, t)\,|s\rangle . \qquad [\text{ VA-2 }]$$

Following a now familiar track, we can use the Schrödinger equation of motion -- *i.e*.

$$i\,\hbar \frac{d}{dt}\left|\psi(t)\right\rangle = \left[\mathcal{H}_o + \mathcal{H}_{ex}\right]\left|\psi(t)\right\rangle \qquad [\text{ VA-3 }]$$

to obtain

$$\begin{aligned} i\,\hbar \dot{C}_r(t) &= \sum_q C_q(t) \exp\left[-i\left(\lambda_q - \lambda_r\right) t\right] \left\langle r\middle|\mathcal{H}_{ex}\middle|q\right\rangle \\ -i\,\hbar \dot{C}_r^*(t) &= \sum_q C_q^*(t) \exp\left[+i\left(\lambda_q - \lambda_r\right) t\right] \left\langle q\middle|\mathcal{H}_{ex}\middle|r\right\rangle \end{aligned} . \qquad [\text{ VA-4 }]$$

In turn, we obtain the following expansion for the time dependent expectation value of induced material system dipole moment:

$$\begin{aligned}\langle \vec{\mathbf{p}}(t) \rangle &= -\langle \psi(t) |\, e\vec{\mathcal{D}}\, | \psi(t) \rangle \\ &= -\sum_r C_r(t) \exp[\, i\lambda_r\, t\,]\; \langle r |\, e\vec{\mathcal{D}} \sum_s C_s(t) \exp[-i\lambda_s\, t\,]\; | s \rangle \\ &= -\sum_r \sum_s C_r(t) C_s(t) \exp[\, i(\lambda_r - \lambda_s)\, t\,]\; e\langle r |\vec{\mathcal{D}}| s\rangle \end{aligned} \qquad \text{[VA-5]}$$

Differentiating this expression with respect to time and using Equation [VA-4] we obtain

$$\begin{aligned}\langle \dot{\vec{\mathbf{p}}}(t) \rangle = &-\sum_r \sum_s \left\{ i \sum_q C_q(t)\, C_s(t) \exp[\, i(\lambda_q - \lambda_s)\, t\,]\; \langle q|\mathcal{H}_{ex}|r\rangle \right\} \frac{e}{\hbar} \langle r|\vec{\mathcal{D}}|s\rangle \\ &+ \sum_r \sum_s \left\{ i \sum_q C_r(t)\, C_q(t) \exp[-i(\lambda_q - \lambda_r)\, t\,]\; \langle s|\mathcal{H}_{ex}|q\rangle \right\} \frac{e}{\hbar} \langle r|\vec{\mathcal{D}}|s\rangle \\ &- \sum_r \sum_s C_r(t) C_s(t)\; i(\lambda_r - \lambda_s) \exp[\, i(\lambda_r - \lambda_s)\, t\,]\; e\langle r|\vec{\mathcal{D}}|s\rangle \end{aligned} \qquad \text{[VA-6a]}$$

Regrouping, we see that this expression can be written

$$\begin{aligned}\langle \dot{\vec{\mathbf{p}}}(t) \rangle = &-\sum_r \sum_s \left\{ \frac{i\,e}{\hbar} \sum_q C_q(t)\, C_s(t) \exp[\, i(\lambda_q - \lambda_s)\, t\,]\; \left[\langle q|\mathcal{H}_{ex}|r\rangle\langle r|\vec{\mathcal{D}}|s\rangle - \langle r|\mathcal{H}_{ex}|s\rangle\langle q|\vec{\mathcal{D}}|r\rangle \right] \right\} \\ &- \sum_r \sum_s C_r(t) C_s(t)\; i(\lambda_r - \lambda_s) \exp[\, i(\lambda_r - \lambda_s)\, t\,]\; e\langle r|\vec{\mathcal{D}}|s\rangle \end{aligned} \qquad \text{[VA-6b]}$$

Since $\mathcal{H}_{ex}$ is proportional to $\vec{\mathcal{D}}$, we see that, ***like magic***, the first term vanishes!!!

Hence,

$$\langle \dot{\vec{\mathbf{p}}}(t) \rangle = -\sum_r \sum_s C_r(t) C_s(t)\; i(\lambda_r - \lambda_s) \exp[\, i(\lambda_r - \lambda_s)\, t\,]\; e\langle r|\vec{\mathcal{D}}|s\rangle\;. \qquad \text{[VA-6c]}$$

Differentiating this expression with respect to time and, again, using Equation [VA-4] we obtain

$$\left\langle\, \ddot{\vec{\mathbf{p}}}(t)\,\right\rangle = -\sum_r\sum_s\sum_q C_q(t)C_s(t)\,\frac{e}{\hbar}\exp\left[\,i\left(\lambda_q-\lambda_s\right)\,t\right]$$
$$\times\left[\left(\lambda_r-\lambda_s\right)\langle q|\mathcal{H}_{ex}|r\rangle\langle r|\vec{\mathcal{D}}\,|s\rangle+\left(\lambda_r-\lambda_q\right)\langle q|\vec{\mathcal{D}}\,|r\rangle\langle r|\mathcal{H}_{ex}|s\rangle\right]\,. \qquad \text{[VA-7]}$$
$$-\sum_r\sum_s C_r(t)C_s(t)\left(\lambda_r-\lambda_s\right)^2\exp\left[\,i\left(\lambda_r-\lambda_s\right)t\right]\;e\langle r|\vec{\mathcal{D}}\,|s\rangle$$

Our task is to now to attempt an interpretation this very nasty expression. To that end, we make use of Equation [V-33] to write Equation [VA-6c] as

$$\left\langle\,\vec{\mathbf{p}}(t)\,\right\rangle = -\sum_r\sum_s C_r(t)C_s(t)\,\exp\left[\,i\left(\lambda_r-\lambda_s\right)t\right]$$
$$\times\, e\,\langle r|\sum_t\left\{\langle a|\vec{\mathcal{D}}\,|b\rangle\,\mathrm{U}_{t1}\,\mu_t^\dagger+\langle b|\vec{\mathcal{D}}\,|a\rangle\,\mathrm{U}_{t1}\,\mu_t\right\}|s\rangle\,. \qquad \text{[VA-8a]}$$

Using the properties of the $\mu_t^\dagger$ and μ_t operators (*viz.* $|s\rangle=\mu_s^\dagger|g\rangle$ and $|g\rangle=\mu_s|s\rangle$), this expression reduces to

$$\left\langle\,\vec{\mathbf{p}}(t)\,\right\rangle = -\sum_r\sum_s C_r(t)C_s(t)\,\exp\left[\,i\left(\lambda_r-\lambda_s\right)t\right]\;e\left\{\langle a|\vec{\mathcal{D}}\,|b\rangle\,\mathrm{U}_{r1}\,\delta_{s\,g}+\langle b|\vec{\mathcal{D}}\,|a\rangle\,\mathrm{U}_{s1}\,\delta_{r\,g}\right\} \qquad \text{[VA-8b]}$$

which may interpreted as a sum of a series of dipole moment components -- *viz.*

$$\left\langle\,\vec{\mathbf{p}}(t)\,\right\rangle = \sum_s\,\vec{\mathbf{p}}_s(t) = -\sum_s\left\{C_s(t)C_g(t)\,\exp\left[\,i\,\lambda_s\,t\right]\;e\langle a|\vec{\mathcal{D}}\,|b\rangle\,\mathrm{U}_{s1}+c.c.\right\} \qquad \text{[VA-8c]}$$

Given this interpretation, we return to Equation [VA-7] and use Equation [V-6] to obtain

$$\left\langle\,\ddot{\vec{\mathbf{p}}}(t)\,\right\rangle = +\sum_r\sum_s\sum_q C_q(t)C_s(t)\,\frac{e^2}{\hbar}\left[2\lambda_r-\lambda_s-\lambda_q\right]\times\exp\left[\,i\left(\lambda_q-\lambda_s\right)t\right]\;\vec{\mathbf{E}}_{\mathrm{T}}(0)\;\langle q|\vec{\mathcal{D}}\,|r\rangle\langle r|\vec{\mathcal{D}}\,|s\rangle$$
$$+\sum_r\sum_s C_r(t)C_s(t)\left(\lambda_r-\lambda_s\right)^2\exp\left[\,i\left(\lambda_r-\lambda_s\right)t\right]\;e\langle r|\vec{\mathcal{D}}\,|s\rangle \qquad .\text{[VA-9]}$$

Again using Equation [V-33] and the properties of the $\mu_t^\dagger$ and μ_t operators, we see that

$$
\begin{aligned}
\langle q|\vec{\mathcal{D}}|r\rangle\langle r|\vec{\mathcal{D}}|s\rangle &= \langle q|\sum_x\Big\{\langle a|\vec{\mathcal{D}}|b\rangle\,\mathrm{U}_{x1}\,\mu_x^\dagger+\langle b|\vec{\mathcal{D}}|a\rangle\,\mathrm{U}_{x1}\,\mu_x\Big\}|r\rangle \\
&\quad\times\langle r|\sum_y\Big\{\langle a|\vec{\mathcal{D}}|b\rangle\,\mathrm{U}_{y1}\,\mu_y^\dagger+\langle b|\vec{\mathcal{D}}|a\rangle\,\mathrm{U}_{y1}\,\mu_y\Big\}|s\rangle \\
&= \sum_x\sum_y\Big\{\langle a|\vec{\mathcal{D}}|b\rangle^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x^\dagger|r\rangle\langle r|\mu_y^\dagger|s\rangle \\
&\quad+\big|\langle a|\vec{\mathcal{D}}|b\rangle\big|^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x^\dagger|r\rangle\langle r|\mu_y|s\rangle \qquad \text{[VA-10a]} \\
&\quad+\langle b|\vec{\mathcal{D}}|a\rangle^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x|r\rangle\langle r|\mu_y|s\rangle \\
&\quad+\big|\langle b|\vec{\mathcal{D}}|a\rangle\big|^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x|r\rangle\langle r|\mu_y^\dagger|s\rangle\Big\}
\end{aligned}
$$

which reduces to

$$
\langle q|\vec{\mathcal{D}}|r\rangle\langle r|\vec{\mathcal{D}}|s\rangle = \langle a|\vec{\mathcal{D}}|b\rangle\langle b|\vec{\mathcal{D}}|a\rangle\,\mathrm{U}_{q1}\,\mathrm{U}_{s1}\,\delta_{r\,g}+\langle b|\vec{\mathcal{D}}|a\rangle\langle a|\vec{\mathcal{D}}|b\rangle\,|\mathrm{U}_{r1}|^2\,\delta_{q\,g}\,\delta_{s\,g} \qquad \text{[VA-10b]}
$$

Substituting this expression and the expression in Equation [V-33] into Equation [VA-9] and, again, using the properties of the $\mu_t^\dagger$ and μ_t operators it relatively straightforward to obtain

$$
\begin{aligned}
\Big\langle\,\ddot{\vec{\mathbf{p}}}(t)\,\Big\rangle+\sum_s\lambda_s^2\,\vec{\mathbf{p}}_s(t) &= -\frac{e^2}{\hbar}\sum_q\sum_s\big[\lambda_s+\lambda_q\big]\,C_q(t)C_s(t)\,\mathrm{U}_{q1}\,\mathrm{U}_{s1} \\
&\qquad\times\exp\Big[\,i\,\big(\lambda_q-\lambda_s\big)\,t\Big]\,\vec{\mathbf{E}}_{\mathrm{T}}(0)\,\langle a|\vec{\mathcal{D}}|b\rangle\langle b|\vec{\mathcal{D}}|a\rangle\,. \qquad \text{[VA-11a]} \\
&\quad+\frac{e^2}{\hbar}\sum_r 2\lambda_r\,\big|C_g(t)\big|^2\,\big|\mathrm{U}_{r1}\big|^2\,\vec{\mathbf{E}}_{\mathrm{T}}(0)\,\langle b|\vec{\mathcal{D}}|a\rangle\langle a|\vec{\mathcal{D}}|b\rangle
\end{aligned}
$$

Thus, in a kind of ***rotating field approximation***, we get a set of driven harmonic oscillator equations of the form

$$\ddot{\vec{\mathbf{p}}}_s(t) + \lambda_s^2\, \vec{\mathbf{p}}_s(t) \approx \frac{e^2}{\hbar}\, \vec{\mathbf{E}}_T(0)\, \left|\langle a|\,\vec{\mathcal{D}}\,|b\rangle\right|^2\, 2\lambda_s \left[\left|C_g(t)\right|^2 - \left|C_s(t)\right|^2 \right] \left|\mathrm{U}_{s1}\right|^2 . \qquad \text{[VA-11b]}$$

In the Weisskopf-Wigner approximation[1] -- *i.e.* $\left|C_g(t)\right|^2 \approx 1$ -- we can easily solve these equations and sum their results to obtain a ***standardized form*** for the frequency dependent of the **dressed** dielectric susceptibility of the system

$$\begin{aligned} \tilde{\chi}(\omega) &\approx \frac{N\, e^2}{\hbar\, \varepsilon_0} \left|\langle a|\,\vec{\mathcal{D}}\,|b\rangle\right|^2 \sum_s \frac{2\lambda_s}{\lambda_s^2 - \omega^2} \left|\mathrm{U}_{r1}\right|^2 \\ &= \frac{N\, e^2}{\hbar\, \varepsilon_o} \left|\langle a|\,\vec{\mathcal{D}}\,|b\rangle\right|^2 \sum_s \left|\mathrm{U}_{r1}\right|^2 \left[\frac{1}{\lambda_s - \omega} + \frac{1}{\lambda_s + \omega} \right] \end{aligned} \qquad \text{[VA-12]}$$

1 V. Weisskopf and E. Wigner, Z. Phys., **63**, 54 (1930).