Macroscopic QED in Finite Particles

Jacob A Busche

27th February 2024

1 Introduction

1.1 Definitions of the coordinate fields

The canonical Lagrangian density of quantum electrodynamics is given by[1]

$$\begin{aligned}\mathscr{L}_{\text{QED}}\left[\mathbf{x}_1,\ldots,\mathbf{x}_N,\dot{\mathbf{x}}_1,\ldots,\dot{\mathbf{x}}_N;\mathbf{A},\dot{\mathbf{A}}\right] &= \sum_{i=1}^{N}\frac{1}{2}m_i\dot{\mathbf{x}}_i^2(t)\delta[\mathbf{r}-\mathbf{x}_i(t)] + \frac{1}{8\pi}\left[\mathbf{E}^2(\mathbf{r},t)-\mathbf{B}^2(\mathbf{r},t)\right] \\ &+ \sum_{i=1}^{N}\frac{e_i}{c}\dot{\mathbf{x}}_i(t)\delta[\mathbf{r}-\mathbf{x}_i(t)]\cdot\mathbf{A}(\mathbf{r},t) - \sum_{i=1}^{N}e_i\delta[\mathbf{r}-\mathbf{x}_i(t)]\Phi(\mathbf{r},t),\end{aligned} \tag{1}$$

and is appropriate for use in systems showcasing well-separated point-like charges interacting with electromagnetic field. However, in general, the charges that make up a material are not confined to small regions of space but are instead spread over larger volumes. In this sense, the usual charge densities $e_i\delta[\mathbf{r}-\mathbf{x}_i(t)]$ present in $\mathscr{L}_{\text{QED}}$ are a bad approximation and must be replaced by a more general description.

In our model, we assume that four separate charge distributions $\rho_\phi(\mathbf{r},t)$ exist within the volume $\mathbb{V}$ occupied by the material of our system, where $\phi = \{m,b,r,f\}$ denotes the "family" the charges belong to. The first is the distribution of the mobile ($\phi = m$) charges, which are assumed to be responsible for the material's interactions with the electromagnetic field. For brevity, we will omit the subscripts for any quantities pertaining to the mobile charges. For example, if we assert that the mobile charges form a continuous fluid with a velocity at each point in spacetime given by the field $\dot{\mathbf{Q}}(\mathbf{r},t)$, a bound current density $\mathbf{J}(\mathbf{r})$ arises. Given by

$$\begin{aligned}\mathbf{J}(\mathbf{r},t) &= \sum_i \mathbf{j}_i(\mathbf{r},t) \\ &= e\eta\Theta(\mathbf{r}\in\mathbb{V})\dot{\mathbf{Q}}(\mathbf{r},t),\end{aligned} \tag{2}$$

this current captures the contributions of the microscopic currents $\mathbf{j}_i(\mathbf{r},t)$ of each mobile particle i and can be calculated through atomistic models. Here e is the elementary charge and is used as a characteristic charge scale, while η is a characteristic number density. In general, we will assume $\mathbb{V}$ is a finite domain, such that $\dot{\mathbf{Q}}(\mathbf{r},t)$ can be mathematically defined over all of $\mathbb{R}^3$ as a periodic function in whichever sense best fits the system geometry. Thus, $\Theta(\mathbf{r}\in\mathbb{V})\dot{\mathbf{Q}}(\mathbf{r},t)$ represents the first "unit-cell" of $\dot{\mathbf{Q}}(\mathbf{r},t)$ and is the physical quantity of interest.

The related continuity equation states that

$$\begin{aligned}\frac{\partial\rho(\mathbf{r},t)}{\partial t} &= -\nabla\cdot\mathbf{J}(\mathbf{r},t) \\ &= -e\eta\left[\Theta(\mathbf{r}\in\mathbb{V})\nabla\cdot\dot{\mathbf{Q}}(\mathbf{r},t) - \delta(\mathbf{r}\in\partial\mathbb{V})\hat{\mathbf{n}}(\mathbf{r})\cdot\dot{\mathbf{Q}}(\mathbf{r},t)\right],\end{aligned} \tag{3}$$

wherein $\rho(\mathbf{r},t)$ is the mobile charge density and $\hat{\mathbf{n}}(\mathbf{r})$ is the outward-facing surface-normal unit vector.

The connection between the velocity field and the mobile charge coordinate field is now straightforward:

$$\mathbf{Q}(\mathbf{r},t) = \int_{-\infty}^{t}\dot{\mathbf{Q}}(\mathbf{r},t')\,\mathrm{d}t', \tag{4}$$

where the system is assumed to be at rest at $t \to -\infty$ such that $\lim_{t\to-\infty} \mathbf{Q}(\mathbf{r},t) = 0$. In parallel fashion, the mobile charge polarization field $\mathbf{P}(\mathbf{r},t)$ is related to the current density via

$$\begin{aligned} \mathbf{P}(\mathbf{r},t) &= \int\limits_{-\infty}^{t} \mathbf{J}(\mathbf{r},t')\,\mathrm{d}t' \\ &= e\eta\Theta(\mathbf{r}\in\mathbb{V})\mathbf{Q}(\mathbf{r},t), \end{aligned} \tag{5}$$

such that

$$\begin{aligned} \nabla\cdot\mathbf{P}(\mathbf{r},t) &= -\rho(\mathbf{r},t) \\ &= e\eta\left[\Theta(\mathbf{r}\in\mathbb{V})\nabla\cdot\mathbf{Q}(\mathbf{r},t) - \delta(\mathbf{r}\in\partial\mathbb{V})\hat{\mathbf{n}}(\mathbf{r})\cdot\mathbf{Q}(\mathbf{r},t)\right] - \bar{\rho}(\mathbf{r}). \end{aligned} \tag{6}$$

Here, $\bar{\rho}(\mathbf{r})$ is the equilibrium mobile charge density, defined by the limit $\lim_{t\to-\infty}\rho(\mathbf{r},t) = \bar{\rho}(\mathbf{r})$.

The background ($\phi = b$) charge density is assumed to be static, such that $\rho_b(\mathbf{r},t) = \rho_b(\mathbf{r})$. We assume it has an equal number density to that of the mobile charges and an equal and opposite characteristic charge, such that $\rho_b(\mathbf{r}) = -\bar{\rho}(\mathbf{r})$.

The reservoir charges ($\phi = r$) are treated in the same manner, with the exception that they are assumed to form a globally neutral system comprised of many independent densities $\rho_\nu(\mathbf{r},t)$. Each reservoir charge density is given a characteristic number density η_ν such that reservoir currents $\mathbf{J}_\nu(\mathbf{r},t) = e\eta_\nu\Theta(\mathbf{r}\in\mathbb{V})\dot{\mathbf{Q}}_\nu(\mathbf{r},t)$ can be defined via the coordinate fields $\mathbf{Q}_\nu(\mathbf{r},t)$.

Finally, the free charges ($\phi = f$) are left as the general charge distribution $\rho_f(\mathbf{r},t) = \sum_{i=1}^{N_f} e_{fi}\delta[\mathbf{r}-\mathbf{x}_{fi}(t)]$ with associated charges e_{fi} and coordinates $\mathbf{x}_{fi}(t)$ such that a more specific driving charge source can be specified later.

1.2 The First-Principles Lagrangian

The Lagrangian density of our system can now be clearly defined. In total,

$$\begin{aligned} &\mathscr{L}\left[\{\mathbf{x}_{fi}(t)\},\{\dot{\mathbf{x}}_{fi}(t)\};\mathbf{Q}(\mathbf{r},t),\dot{\mathbf{Q}}(\mathbf{r},t);\{\mathbf{Q}_\nu(\mathbf{r},t)\},\{\dot{\mathbf{Q}}_\nu(\mathbf{r},t)\};\mathbf{A}_f(\mathbf{r},t),\dot{\mathbf{A}}_f(\mathbf{r},t);\mathbf{A}(\mathbf{r},t),\dot{\mathbf{A}}(\mathbf{r},t)\right] \\ &\quad = \mathscr{L}_f + \mathscr{L}_m + \mathscr{L}_r + \mathscr{L}_{\text{int}} + \mathscr{L}_{\text{bind}} + \mathscr{L}_{\text{EM}} \end{aligned} \tag{7}$$

where

$$\begin{aligned} \mathscr{L}_f = \sum_{i=1}^{N_f}\frac{1}{2}m_{fi}\dot{\mathbf{x}}_{fi}^2(t)\delta[\mathbf{r}-\mathbf{x}_{fi}(t)] + \frac{1}{8\pi}\left[-\nabla\Phi_f(\mathbf{r},t) - \frac{1}{c}\dot{\mathbf{A}}(\mathbf{r},t)\right]^2 - \left[\nabla\times\mathbf{A}_f(\mathbf{r},t)\right]^2 \\ - \rho_f(\mathbf{r},t)\Phi_f(\mathbf{r},t) + \frac{1}{c}\mathbf{J}_f(\mathbf{r},t)\cdot\mathbf{A}_f(\mathbf{r},t). \end{aligned} \tag{8}$$

describes the energy of the free charges and fields. The free charges have masses m_{fi}, coordinates $\mathbf{x}_{fi}(t)$, and velocities $\dot{\mathbf{x}}_{fi}(t)$, while the fields they set up are defined by the scalar and vector potentials $\Phi_f(\mathbf{r},t)$ and $\mathbf{A}_f(\mathbf{r},t)$, respectively. Further,

$$\mathscr{L}_m = \frac{\mu}{2}\left[\dot{\mathbf{Q}}^2(\mathbf{r},t) - \Omega^2\mathbf{Q}^2(\mathbf{r},t)\right]\Theta(\mathbf{r}\in\mathbb{V}) \tag{9}$$

describes the kinetic and harmonic potential energies experienced by the mobile material charges with characteristic mass-densities μ, coordinate fields $\mathbf{Q}(\mathbf{r},t)$, velocity fields $\dot{\mathbf{Q}}(\mathbf{r},t)$, and natural frequencies ω_0, and

$$\mathscr{L}_r = \int\limits_0^\infty \frac{\mu}{2}\left[\dot{\mathbf{Q}}_\nu^2(\mathbf{r},t) - \nu^2\mathbf{Q}_\nu^2(\mathbf{r},t)\right]\Theta(\mathbf{r}\in\mathbb{V})\,\mathrm{d}\nu \tag{10}$$

describes the kinetic and potential energies of reservoir charges that build in the system's damping and have mass-densities μ, coordinate fields $\mathbf{Q}_\nu(\mathbf{r},t)$, velocity fields $\dot{\mathbf{Q}}_\nu(\mathbf{r},t)$, and natural frequencies ν. These forms for the mobile charge and reservoir Lagrangian densities impose the restriction on our model that the natural frequencies of each coordinate field are spatially isotropic, but are otherwise general. Additionally,

$$\begin{aligned}\mathscr{L}_{\mathrm{int}} &= e\eta\nabla\cdot\{\Theta(\mathbf{r}\in\mathbb{V})\mathbf{Q}(\mathbf{r},t)\}\left[\Phi(\mathbf{r},t)+\Phi_f(\mathbf{r},t)\right]+\frac{1}{c}e\eta\Theta(\mathbf{r}\in\mathbb{V})\dot{\mathbf{Q}}(\mathbf{r},t)\cdot\left[\mathbf{A}(\mathbf{r},t)+\mathbf{A}_f(\mathbf{r},t)\right]\\ &\quad-\eta\int_0^\infty v(\nu)\mathbf{Q}(\mathbf{r},t)\cdot\dot{\mathbf{Q}}_\nu(\mathbf{r},t)\Theta(\mathbf{r}\in\mathbb{V})\,\mathrm{d}\nu\end{aligned} \tag{11}$$

describes the interactions between the charges and fields of the system. The scalar and vector potentials $\Phi(\mathbf{r},t)$ and $\mathbf{A}(\mathbf{r},t)$ are set up by the mobile charges and also drive them, as do the free potentials. We also introduce coupling to the reservoir here, with $v(\nu)$ a characteristic coupling strength between the mobile charges and reservoir. Coupling between the free charges and the mobile charge potentials is assumed to be negligible, and coupling between the free charges and free fields is contained above in $\mathscr{L}_f$. Finally,

$$\mathscr{L}_{\mathrm{bind}} = -\sum_{\alpha,\beta=m,b,r}\left[u^{\infty}_{\alpha\beta}(\mathbf{r})+u^{\infty}_{\beta\alpha}(\mathbf{r})\right]-\sum_{\alpha=m,b,r,f}u^{\mathrm{self}}_{\alpha} \tag{12}$$

describes the binding potentials $u^{\infty}_{\alpha\beta}(\mathbf{r})$ and self-energies $u^{\mathrm{self}}_{\alpha}$ experienced by each charge, and

$$\mathscr{L}_{\mathrm{EM}} = \frac{1}{8\pi}\left[\left[\nabla\Phi(\mathbf{r},t)\right]^2+\frac{1}{c^2}\dot{\mathbf{A}}^2(\mathbf{r},t)-\left[\nabla\times\mathbf{A}(\mathbf{r},t)\right]^2+\frac{2}{c}\nabla\Phi(\mathbf{r},t)\cdot\dot{\mathbf{A}}(\mathbf{r},t)\right] \tag{13}$$

describes the energy bound in the electromagnetic fields set up by the mobile charges.

2 Construction of a Mechanical Picture for MQED

From this point, we will simplify the mobile-charge equilibrium density to a constant with a sharp cutoff at a spherical boundary $r=a$, such that $\Theta(\mathbf{r}\in\mathbb{V})=\Theta(a-r)$. Our goal is then to map the dynamics of this system onto a set of coupled oscillators such that the standard tricks of quantization function normally. We can do this via the expansions of the vector potential, mobile charge field, and reservoir field into sets of normal modes and analyzing the time-dependence of those modes' amplitudes. These expansions are given by

$$\begin{aligned}\mathbf{A}(\mathbf{r},t) &= \sum_{\boldsymbol{\alpha}}\int_0^\infty \frac{e\omega^{\frac{3}{2}}}{c^2}\mathscr{A}_{\boldsymbol{\alpha}}(\omega,t)\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)\,\mathrm{d}\omega,\\ \mathbf{Q}(\mathbf{r},t) &= \sum_{\boldsymbol{\beta}}\sum_n \mathscr{B}_{\boldsymbol{\beta}n}(t)\mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k_{\boldsymbol{\beta}n}),\\ \mathbf{Q}_\nu(\mathbf{r},t) &= \sum_{\boldsymbol{\gamma}}\sum_n \mathscr{C}_{\boldsymbol{\gamma}n}(\nu,t)\mathbf{X}_{\boldsymbol{\gamma}}(\mathbf{r},k_{\boldsymbol{\gamma}n}).\end{aligned} \tag{14}$$

The mode functions $\mathbf{X}_{\boldsymbol{\alpha}}$ are chosen to match the symmetry of our problem (details in Appendix B) such that the definition of the vector potential can be extrapolated from any introductory quantum optics text. Specifically, the vector potential is, in the Coulomb gauge, comprised of the excitation of a series of photon modes[1], each of which has angular indices $\boldsymbol{\alpha}=(T',p',\ell',m')$ and a radial index $k=\omega/c$ that determine its wave pattern in space. The 'type' index T of the vector potential can take the values M and E, in which cases the mode functions are transverse vector spherical harmonics of magnetic and electric type, respectively. The other three indices are the parity p, spherical harmonic order ℓ, and spherical harmonic degree m that determine the mode's angular symmetries. In these notes, we will generally used primed index labels for the vector potential modes and related quantities.

The expansions for the matter and bath fields are similar, with angular indices given by $\boldsymbol{\beta}=(T,p,\ell,m)$ and $\boldsymbol{\gamma}=(T'',p'',\ell'',m'')$, respectively. However, their definitions involve sums over discrete wavenumbers $k_{\boldsymbol{\beta}n}$ and $k_{\boldsymbol{\gamma}n}$, rather than the continuously-varying k, and have an additional allowed value of the type index,

$T = L$ (we will assume from here that $\mathcal{A}_{Lp\ell m}(\omega, t) = 0$). The additional allowed mode functions are the longitudinal vector spherical harmonics that describe longitudinal matter waves

The discrete wavenumber behavior is generated by the fact that these mode functions are being used to reconstruct periodic fields with a finite unit cell. The proof is complicated[2] and will not be shown here, but relies on the as afact that the vector spherical harmonics $\mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r}, k)$ are vector eigenfunctions of the Sturm-Liouville operator $\nabla^2 + k^2$.

A consequence of this proof is that, in order to form a complete set on a finite region (in this case, the interior of the sphere), one of the spherical vector harmonics' components, the derivative of one of their components, or a linear combination of the two must be equal to zero at the boundary $r = a$. Within this restriction, we have nearly complete freedom to define which discrete set of wavenumbers $k_{\boldsymbol{\beta}n}$ we use in our expansion.

2.1 Satisfying Boundary Conditions

The main caveat to this last point is that our expansion must produce fields that satisfy Maxwell's boundary conditions for nonmagnetic media. We will first assume that the free fields are smooth everywhere except near $\mathbf{x}_{fi}(t)$. Assuming that $\mathbf{x}_{fi}(t)$ are well-separated from our system, the free fields are therefore well-behaved near $\mathbb{V}$ and cannot contribute any nonzero field differences across the boundary $\partial\mathbb{V}$. Therefore, the boundary conditions

$$
\begin{aligned}
\hat{\mathbf{r}} \times \Big[\mathbf{E}^{(+)}(\mathbf{r}, t) - \mathbf{E}^{(-)}(\mathbf{r}, t)\Big]_{r\to a} &= 0, \\
\hat{\mathbf{r}} \cdot \Big[\mathbf{E}^{(+)}(\mathbf{r}, t) + 4\pi\mathbf{P}^{(+)}(\mathbf{r}, t) - \mathbf{E}^{(-)}(\mathbf{r}, t) - 4\pi\mathbf{P}^{(-)}(\mathbf{r}, t)\Big]_{r\to a} &= 0, \\
\hat{\mathbf{r}} \times \Big[\mathbf{B}^{(+)}(\mathbf{r}, t) - \mathbf{B}^{(-)}(\mathbf{r}, t)\Big]_{r\to a} &= 0, \\
\hat{\mathbf{r}} \cdot \Big[\mathbf{B}^{(+)}(\mathbf{r}, t) - \mathbf{B}^{(-)}(\mathbf{r}, t)\Big]_{r\to a} &= 0
\end{aligned} \tag{15}
$$

only concern the fields set up by the mobile charges. Here, vector and fields outside and inside the boundary are labeled with superscripts $(\pm)$, respectively.

Letting the fields be broken into their Helmholtz components such that $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{\perp}(\mathbf{r}, t) + \mathbf{E}^{\parallel}(\mathbf{r}, t)$ and $\mathbf{B}(\mathbf{r}, t) = \mathbf{B}^{\perp}(\mathbf{r}, t)$, we can first notice that the transverse components $\perp$ are entirely comprised of the photon field, i.e. $\mathbf{E}^{\perp}(\mathbf{r}, t) = -(1/c)\dot{\mathbf{A}}(\mathbf{r}, t)$ and $\mathbf{B}^{\perp}(\mathbf{r}, t) = \nabla \times \mathbf{A}(\mathbf{r}, t)$. The photon field is smooth everywhere, such that the transverse components of the fields cancel in each of the above equations. The latter two boundary conditions, involving only the (transverse) magnetic fields, are therefore satisfied.

We are then left with the task of showing that $-\nabla\Phi(\mathbf{r}, t)$ satisfies the simplified boundary conditions

$$
\begin{aligned}
\hat{\mathbf{r}} \times \Big[-\nabla\Phi^{(+)}(\mathbf{r}, t) + \nabla\Phi^{(-)}(\mathbf{r}, t)\Big] &= 0, \\
\hat{\mathbf{r}} \cdot \Big[-\nabla\Phi^{(+)}(\mathbf{r}, t) + \nabla\Phi^{(-)}(\mathbf{r}, t)\Big] &= \hat{\mathbf{r}} \cdot 4\pi\mathbf{P}(\mathbf{r}, t),
\end{aligned} \tag{16}
$$

wherein we have noted that $\mathbf{P}^{(+)}(\mathbf{r}, t) = 0$ such that $\mathbf{P}^{(-)}(\mathbf{r}, t) = \mathbf{P}(\mathbf{r}, t)$. To do so, we can begin by expanding the mobile-charge scalar potential as

$$
\begin{aligned}
\Phi(\mathbf{r},t) &= -\int \frac{\nabla'\cdot\mathbf{P}(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|}\,\mathrm{d}^3\mathbf{r}' \\
&= e\eta a^2 \sum_{p\ell mn} \frac{4\pi(1-\delta_{p1}\delta_{m0})}{\sqrt{\ell(\ell+1)}\sqrt{2\ell+1}} \left(f^{>}_{p\ell m}(\mathbf{r}) a^{\ell}\Theta(r-a) + f^{<}_{p\ell m}(\mathbf{r})\frac{1}{a^{\ell+1}}\Theta(a-r)\right) \\
&\quad \times \left(\mathcal{B}_{Ep\ell mn}(t)\frac{\ell(\ell+1)}{k_{E\ell n}a} j_\ell(k_{E\ell n}a) + \mathcal{B}_{Lp\ell mn}(t)\frac{\sqrt{\ell(\ell+1)}}{k_{L\ell n}}\frac{\partial j_\ell(k_{L\ell n}a)}{\partial a}\right) \\
&\quad + e\eta \sum_{p\ell mn} 4\pi(1-\delta_{p1}\delta_{m0}) \frac{k_{L\ell n}}{\sqrt{2\ell+1}}\mathcal{B}_{Lp\ell mn}(t) \left(f^{>}_{p\ell m}(\mathbf{r})\frac{r^{\ell+2}}{k_{L\ell n}} j_{\ell+1}(k_{L\ell n}r)\Theta(a-r) \right. \\
&\qquad + f^{>}_{p\ell m}(\mathbf{r})\frac{a^{\ell+2}}{k_{L\ell n}} j_{\ell+1}(k_{L\ell n}a)\Theta(r-a) + f^{<}_{p\ell m}(\mathbf{r})\frac{1}{k_{L\ell n}r^{\ell-1}} j_{\ell-1}(k_{L\ell n}r)\Theta(a-r) \\
&\qquad \left. - f^{<}_{p\ell m}(\mathbf{r})\frac{1}{k_{L\ell n}a^{\ell-1}} j_{\ell-1}(k_{L\ell n}a)\Theta(a-r)\right).
\end{aligned}
\tag{17}
$$

Noting that

$$
\begin{aligned}
\nabla\left\{ f^{>}_{p\ell m}(\mathbf{r})a^{\ell}\Theta(r-a) + \frac{f^{<}_{p\ell m}(\mathbf{r})}{a^{\ell+1}}\Theta(a-r)\right\} &= \nabla f^{>}_{p\ell m}(\mathbf{r})a^{\ell}\Theta(r-a) + \frac{\nabla f^{<}_{p\ell m}(\mathbf{r})}{a^{\ell+1}}\Theta(a-r) \\
&= \frac{1}{a^2}\mathbf{Z}_{p\ell m}(\mathbf{r};a),
\end{aligned}
\tag{18}
$$

with the vector harmonics $\mathbf{Z}_{p\ell m}(\mathbf{r};a)$ defined implicitly for convenience, we can see that

$$
\begin{aligned}
-\nabla\Phi(\mathbf{r},t) &= -e\eta \sum_{p\ell mn} \frac{4\pi(1-\delta_{p1}\delta_{m0})}{\sqrt{\ell(\ell+1)}\sqrt{2\ell+1}} \mathbf{Z}_{p\ell m}(\mathbf{r};a) \left(\mathcal{B}_{Ep\ell mn}(t)\frac{\ell(\ell+1)}{k_{E\ell n}a} j_\ell(k_{E\ell n}a) \right. \\
&\qquad \left. + \mathcal{B}_{Lp\ell mn}(t)\frac{\sqrt{\ell(\ell+1)}}{k_{L\ell n}}\frac{\partial j_\ell(k_{L\ell n}a)}{\partial a}\right) - e\eta\sum_{p\ell mn} 4\pi(1-\delta_{p1}\delta_{m0})\frac{1}{\sqrt{2\ell+1}}\mathcal{B}_{Lp\ell mn}(t) \\
&\qquad \times \left(\nabla f^{>}_{p\ell m}(\mathbf{r})\Theta(a-r) r^{\ell+2} j_{\ell+1}(k_{L\ell n}r) + \nabla f^{<}_{p\ell m}(\mathbf{r})\Theta(a-r)\frac{j_{\ell-1}(k_{L\ell n}r)}{r^{\ell-1}} \right. \\
&\qquad + \nabla f^{>}_{p\ell m}(\mathbf{r})\Theta(r-a) a^{\ell+2} j_{\ell+1}(k_{L\ell n}a) - \nabla f^{<}_{p\ell m}(\mathbf{r})\Theta(a-r)\frac{j_{\ell-1}(k_{L\ell n}a)}{a^{\ell-1}} \\
&\qquad + f^{>}_{p\ell m}(\mathbf{r})\Theta(a-r)(\ell+2) r^{\ell+1} j_{\ell+1}(k_{L\ell n}r)\hat{\mathbf{r}} + (-\ell+1) f^{<}_{p\ell m}(\mathbf{r})\Theta(a-r)\frac{j_{\ell-1}(k_{L\ell n}r)}{r^{\ell}}\hat{\mathbf{r}} \\
&\qquad \left. + f^{>}_{p\ell m}(\mathbf{r})\Theta(a-r) r^{\ell+2}\frac{\partial j_{\ell+1}(k_{L\ell n}r)}{\partial r}\hat{\mathbf{r}} + f^{<}_{p\ell m}(\mathbf{r})\Theta(a-r)\frac{1}{r^{\ell-1}}\frac{\partial j_{\ell-1}(k_{L\ell n}r)}{\partial r}\hat{\mathbf{r}}\right).
\end{aligned}
\tag{19}
$$

Therefore, noting that

$$
\begin{aligned}
\hat{\mathbf{r}}\times\nabla f^{<}_{p\ell m}(\mathbf{r}) &= \sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}} r^{\ell-1}\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta} S_p(m\phi)\hat{\boldsymbol{\phi}} + \frac{(-1)^p m}{\sin\theta} P_{\ell m}(\cos\theta) S_{p+1}(m\phi)\hat{\boldsymbol{\theta}}\right), \\
\hat{\mathbf{r}}\times\nabla f^{>}_{p\ell m}(\mathbf{r}) &= \sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}} \frac{1}{r^{\ell+2}}\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta} S_p(m\phi)\hat{\boldsymbol{\phi}} + \frac{(-1)^p m}{\sin\theta} P_{\ell m}(\cos\theta) S_{p+1}(m\phi)\hat{\boldsymbol{\theta}}\right), \\
\hat{\mathbf{r}}\cdot\nabla f^{<}_{p\ell m}(\mathbf{r}) &= \sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}} \ell r^{\ell-1} P_{\ell m}(\cos\theta) S_p(m\phi), \\
\hat{\mathbf{r}}\cdot\nabla f^{>}_{p\ell m}(\mathbf{r}) &= \sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}} \frac{(-\ell-1)}{r^{\ell+2}} P_{\ell m}(\cos\theta) S_p(m\phi),
\end{aligned}
\tag{20}
$$

we can say

$$
\begin{aligned}
-\hat{\mathbf{r}}\times\nabla\Phi(\mathbf{r},t) = &-e\eta\sum_{p\ell mn}\frac{4\pi(1-\delta_{p1}\delta_{m0})}{\sqrt{\ell(\ell+1)}\sqrt{2\ell+1}}\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}\left(\frac{a^{\ell+2}}{r^{\ell+2}}\Theta(r-a)+\frac{r^{\ell-1}}{a^{\ell-1}}\Theta(a-r)\right)\\
&\times\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\phi}}+\frac{(-1)^p m}{\sin\theta}P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\theta}}\right)\\
&\times\left(\mathcal{B}_{Ep\ell mn}(t)\frac{\ell(\ell+1)}{k_{Ep\ell mn}a}j_\ell(k_{E\ell n}a)+\mathcal{B}_{Lp\ell mn}(t)\frac{\sqrt{\ell(\ell+1)}}{k_{L\ell n}}\frac{\partial j_\ell(k_{L\ell n}a)}{\partial a}\right)\\
&-e\eta\sum_{p\ell mn}4\pi(1-\delta_{p1}\delta_{m0})\frac{1}{\sqrt{2\ell+1}}\mathcal{B}_{Lp\ell mn}(t)\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}\\
&\times\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\phi}}+\frac{(-1)^p m}{\sin\theta}P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\theta}}\right)\\
&\times\Bigg[j_{\ell+1}(k_{L\ell n}r)\Theta(a-r)+j_{\ell-1}(k_{L\ell n}r)\Theta(a-r)+\frac{a^{\ell+2}}{r^{\ell+2}}j_{\ell+1}(k_{L\ell n}a)\Theta(r-a)\\
&\quad-\frac{r^{\ell-1}}{a^{\ell-1}}j_{\ell-1}(k_{L\ell n}a)\Theta(a-r)\Bigg].
\end{aligned}
\tag{21}
$$

Due to the symmetric structure of the r-dependent factor of each term of the first sum above, the first sum approaches the same value as $r\to a$ from inside or outside the boundary. The r-dependent factors of the second sum are, after some cancellation, similarly identical on either side of the boundary as $r\to a$. Therefore,

$$
-\hat{\mathbf{r}}\times\left(\lim_{r\to a^+}\nabla\Phi(\mathbf{r},t)+\lim_{r\to a^-}\nabla\Phi(\mathbf{r},t)\right)=0 \tag{22}
$$

such that the surface-tangential components of the longitudinal electric field obey Maxwell's boundary conditions.

The component of $-\nabla\Phi(\mathbf{r},t)$ normal to the surface is

$$
\begin{aligned}
-\hat{\mathbf{r}}\cdot\nabla\Phi(\mathbf{r},t) = &-en\sum_{p\ell mn}\frac{4\pi(1-\delta_{p1}\delta_{m0})}{\sqrt{\ell(\ell+1)}\sqrt{2\ell+1}}\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}P_{\ell m}(\cos\theta)S_p(m\phi)\\
&\times\left(-(\ell+1)\frac{a^{\ell+2}}{r^{\ell+2}}\Theta(r-a)+\ell\frac{r^{\ell-1}}{a^{\ell-1}}\Theta(a-r)\right)\\
&\times\left(\mathcal{B}_{Ep\ell mn}(t)\frac{\ell(\ell+1)}{k_{Ep\ell mn}a}j_\ell(k_{E\ell n}a)+\mathcal{B}_{Lp\ell mn}(t)\frac{\sqrt{\ell(\ell+1)}}{k_{L\ell n}}\frac{\partial j_\ell(k_{L\ell n}a)}{\partial a}\right)\\
&-en\sum_{p\ell mn}4\pi(1-\delta_{p1}\delta_{m0})\frac{1}{\sqrt{2\ell+1}}\mathcal{B}_{Lp\ell mn}(t)\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}P_{\ell m}(\cos\theta)S_p(m\phi)\\
&\times\Bigg[-(\ell+1)j_{\ell+1}(k_{L\ell n}r)\Theta(a-r)+\ell j_{\ell-1}(k_{L\ell n}r)\Theta(a-r)\\
&-(\ell+1)\frac{a^{\ell+2}}{r^{\ell+2}}j_{\ell+1}(k_{L\ell n}a)\Theta(r-a)-\ell\frac{r^{\ell-1}}{a^{\ell-1}}j_{\ell-1}(k_{L\ell n}a)\Theta(a-r)\\
&+(\ell+2)j_{\ell+1}(k_{L\ell n}r)\Theta(a-r)+(-\ell+1)j_{\ell-1}(k_{L\ell n}r)\Theta(a-r)\\
&+r\frac{\partial j_{\ell+1}(k_{L\ell n}r)}{\partial r}\Theta(a-r)+r\frac{\partial j_{\ell-1}(k_{L\ell n}r)}{\partial r}\Theta(a-r)\Bigg].
\end{aligned}
\tag{23}
$$

Analyzing this term takes a little more work. Taking limits on either side of the boundary, one finds

$$\begin{aligned}
-\lim_{r\to a^+}\hat{\mathbf{r}}\cdot\nabla\Phi(\mathbf{r},t) = e\eta\sum_{p\ell mn}&\frac{4\pi(1-\delta_{p1}\delta_{m0})}{\sqrt{\ell(\ell+1)}\sqrt{2\ell+1}}\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}P_{\ell m}(\cos\theta)S_p(m\phi)(\ell+1)\\
&\times\left(\mathcal{B}_{Ep\ell mn}(t)\frac{\ell(\ell+1)}{k_{Ep\ell mn}a}j_\ell(k_{E\ell n}a)+\mathcal{B}_{Lp\ell mn}(t)\frac{\sqrt{\ell(\ell+1)}}{k_{L\ell n}}\frac{\partial j_\ell(k_{L\ell n}a)}{\partial a}\right)\\
&+e\eta\sum_{p\ell mn}4\pi(1-\delta_{p1}\delta_{m0})\frac{1}{\sqrt{2\ell+1}}\mathcal{B}_{Lp\ell mn}(t)\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}\\
&\quad\times P_{\ell m}(\cos\theta)S_p(m\phi)(\ell+1)j_{\ell+1}(k_{L\ell n}a)
\end{aligned}\tag{24}$$

and

$$\begin{aligned}
-\lim_{r\to a^-}\hat{\mathbf{r}}\cdot\nabla\Phi(\mathbf{r},t) = -e\eta\sum_{p\ell mn}&\frac{4\pi(1-\delta_{p1}\delta_{m0})}{\sqrt{\ell(\ell+1)}\sqrt{2\ell+1}}\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}P_{\ell m}(\cos\theta)S_p(m\phi)\ell\\
&\times\left(\mathcal{B}_{Ep\ell mn}(t)\frac{\ell(\ell+1)}{k_{Ep\ell mn}a}j_\ell(k_{E\ell n}a)+\mathcal{B}_{Lp\ell mn}(t)\frac{\sqrt{\ell(\ell+1)}}{k_{L\ell n}}\frac{\partial j_\ell(k_{L\ell n}a)}{\partial a}\right)\\
&+e\eta\sum_{p\ell mn}4\pi(1-\delta_{p1}\delta_{m0})\frac{1}{\sqrt{2\ell+1}}\mathcal{B}_{Lp\ell mn}(t)\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}\\
&\quad\times P_{\ell m}(\cos\theta)S_p(m\phi)(\ell+1)j_{\ell+1}(k_{L\ell n}a),
\end{aligned}\tag{25}$$

wherein we have used the recurrence relations

$$\begin{aligned}
\frac{\partial j_{\ell+1}(x)}{\partial x} &= j_\ell(x)-\frac{\ell+2}{x}j_{\ell+1}(x),\\
\frac{\partial j_{\ell-1}(x)}{\partial x} &= -j_\ell(x)+\frac{\ell-1}{x}j_{\ell-1}(x).
\end{aligned}\tag{26}$$

Therefore, using the identities

$$\begin{aligned}
\hat{\mathbf{r}}\cdot\mathbf{N}_{p\ell m}(\mathbf{r},k) &= \sqrt{(2-\delta_{m0})\frac{2\ell+1}{\ell(\ell+1)}\frac{(\ell-m)!}{(\ell+m)!}}\frac{\ell(\ell+1)}{kr}j_\ell(kr)P_{\ell m}(\cos\theta)S_p(m\phi),\\
\hat{\mathbf{r}}\cdot\mathbf{L}_{p\ell m}(\mathbf{r},k) &= \sqrt{(2-\delta_{m0})\frac{2\ell+1}{\ell(\ell+1)}\frac{(\ell-m)!}{(\ell+m)!}}\sqrt{\ell(\ell+1)}\frac{1}{k}\frac{\partial j_\ell(kr)}{\partial r}P_{\ell m}(\cos\theta)S_p(m\phi),
\end{aligned}\tag{27}$$

and $(1-\delta_{p1}\delta_{m0})S_p(m\phi)=S_p(m\phi)$, we can see that

$$= 4\pi\hat{\mathbf{r}}\cdot\mathbf{P}(\mathbf{r},t)|_{r\to a}\tag{28}$$

The surface normal components of the longitudinal electric field therefore also obey Maxwell's boundary conditions. Therefore, there are no restrictions on the wavenumbers $k_{\boldsymbol{\beta}n}$ other than those set by the requirements of Sturm-Liouville expansions.

2.2 Expansion of the Lagrangian

For simplicity, then, we will choose the wavenumbers $k_{\boldsymbol{\beta}n}=(\delta_{TM}+\delta_{TE})z_{\ell n}/a+\delta_{TL}w_{\ell n}/a$, where $z_{\ell n}$ are the roots of the spherical Bessel function $j_\ell(x)$ and $w_{\ell n}$ are the roots of the derivative $\partial j_\ell(x)/\partial x$, with $n=1,2,\ldots$ identifying the first, second, etc. root above zero. These wavenumbers guarantee that the radial components of each vector harmonic are zero at the boundary, and they simplify many of our calculations while providing a complete basis for expansion.

Explicitly, we can define the system Lagrangian as $L = \int \mathscr{L}\, \mathrm{d}^3\mathbf{r}$. Using the identities of Appendix B, we can say

$$\begin{aligned} L_m &= \frac{\mu}{2} \int\limits_{r<a} \left(\dot{\mathbf{Q}}^2(\mathbf{r},t) - \Omega^2 \mathbf{Q}^2(\mathbf{r},t)\right) \mathrm{d}^3\mathbf{r} \\ &= \sum_{\boldsymbol{\beta} n} \frac{m_{\boldsymbol{\beta} n}}{2} \left(\dot{\mathscr{B}}^2_{\boldsymbol{\beta} n}(t) - \Omega^2 \mathscr{B}^2_{\boldsymbol{\beta} n}(t)\right) \end{aligned} \tag{29}$$

and

$$\begin{aligned} L_r &= \int\limits_{r<a} \int\limits_0^\infty \frac{\mu}{2} \left(\dot{\mathbf{Q}}^2_\nu(\mathbf{r},t) - \nu^2 \mathbf{Q}^2_\nu(\mathbf{r},t)\right) \mathrm{d}\nu\, \mathrm{d}^3\mathbf{r} \\ &= \sum_{\boldsymbol{\gamma} n} \int\limits_0^\infty \frac{m_{\boldsymbol{\gamma} n}}{2} \left(\dot{\mathscr{C}}^2_{\boldsymbol{\gamma} n}(\nu,t) - \nu^2 \mathscr{C}^2_{\boldsymbol{\gamma} n}(\nu,t)\right) \mathrm{d}\nu \end{aligned} \tag{30}$$

where

$$m_{\boldsymbol{\beta} n} = 2\pi a^3 \mu \left[(\delta_{TM} + \delta_{TE}) j^2_{\ell+1}(z_{\ell n}) + \delta_{TL}\left(1 - \frac{\ell(\ell+1)}{w^2_{\ell n}}\right) j^2_\ell(w_{\ell n})\right] \tag{31}$$

is the effective mass of the $(\boldsymbol{\beta}, n)^{\text{th}}$ mode.

Further, after inserting our specified values of the roots $k_{\boldsymbol{\beta} n}$ into the matter field such that it simplifies to

$$\begin{aligned} -\nabla\Phi(\mathbf{r},t) = -e\eta \sum_{p\ell mn} \frac{4\pi}{\sqrt{2\ell+1}} \mathscr{B}_{Lp\ell mn}(t) \Bigg[&\sqrt{2\ell+1}\,\mathbf{L}_{p\ell m}\left(\mathbf{r}, \frac{w_{\ell n}}{a}\right) \Theta(a-r) \\ &+ a^{\ell+2} j_{\ell+1}(w_{\ell n}) \nabla f^>_{p\ell m}(\mathbf{r}) \Theta(r-a) - \frac{j_{\ell-1}(w_{\ell n})}{a^{\ell-1}} \nabla f^<_{p\ell m}(\mathbf{r}) \Theta(a-r) \Bigg], \end{aligned} \tag{32}$$

we can see that

$$\begin{aligned} &\int \nabla\Phi(\mathbf{r},t) \cdot \nabla\Phi(\mathbf{r},t)\, \mathrm{d}^3\mathbf{r} \\ &\quad = -8\pi \sum_{\boldsymbol{\beta}} \sum_{nn'} g^{(1)}_{\boldsymbol{\beta} nn'} \mathscr{B}_{Lp\ell mn}(t) \mathscr{B}_{Lp\ell mn'}(t), \end{aligned} \tag{33}$$

where

$$g^{(1)}_{\boldsymbol{\beta} nn'} = -\delta_{TL} 4\pi^2 e^2 \eta^2 a^3 \left[j^2_\ell(w_{\ell n}) \left(1 - \frac{\ell(\ell+1)}{w^2_{\ell n}}\right) \delta_{nn'} - \frac{2\ell(\ell+1)}{2\ell+1} \frac{j_\ell(w_{\ell n}) j_\ell(w_{\ell n'})}{w_{\ell n} w_{\ell n'}} \right] \tag{34}$$

is a characteristic coupling strength between the mobile charge modes arising from the energy bound in their electric fields. We can then say

$$\begin{aligned} L_{\text{EM}} &= \frac{1}{8\pi} \int \left(\nabla\Phi(\mathbf{r},t) \cdot \nabla\Phi(\mathbf{r},t) + \frac{1}{c^2} \dot{\mathbf{A}}^2(\mathbf{r},t) - [\nabla \times \mathbf{A}(\mathbf{r},t)]^2 \right) \mathrm{d}^3\mathbf{r} \\ &= -\sum_{\boldsymbol{\beta}} \sum_{nn'} g^{(1)}_{\boldsymbol{\beta} nn'} \mathscr{B}_{\boldsymbol{\beta} n}(t) \mathscr{B}_{\boldsymbol{\beta} n'}(t) + \sum_{\boldsymbol{\alpha}} \int\limits_0^\infty \frac{\pi}{4} \frac{e^2 \omega}{c^3} \left[\dot{\mathscr{A}}^2_{\boldsymbol{\alpha}}(\omega,t) - \omega^2 \mathscr{A}^2_{\boldsymbol{\alpha}}(\omega,t) \right] \mathrm{d}\omega \end{aligned} \tag{35}$$

The interaction Lagrangian can be handled similarly. We can use integration by parts and separate the scalar potential into its constituent parts to say

$$\begin{aligned} L_{\text{int}} = &-e\eta \int \Theta(a-r) \mathbf{Q}(\mathbf{r},t) \cdot [\nabla\Phi(\mathbf{r},t) + \nabla\Phi_f(\mathbf{r},t)]\ \mathrm{d}^3\mathbf{r} \\ &+ \frac{e\eta}{c} \int \Theta(a-r) \dot{\mathbf{Q}}(\mathbf{r},t) \cdot [\mathbf{A}(\mathbf{r},t) + \mathbf{A}_f(\mathbf{r},t)]\ \mathrm{d}^3\mathbf{r} \\ &- \int \int\limits_0^\infty \eta v(\nu) \mathbf{Q}(\mathbf{r},t) \cdot \dot{\mathbf{Q}}_\nu(\mathbf{r},t) \Theta(a-r)\, \mathrm{d}\nu\, \mathrm{d}^3\mathbf{r}. \end{aligned} \tag{36}$$

We can then simplify the terms detailing the interaction energies between the free fields and the mobile charges as

$$\begin{aligned}
-e\eta \int \Theta(a-r)\mathbf{Q}(\mathbf{r},t)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} &= -\sum_{\boldsymbol{\beta}n} F_{\boldsymbol{\beta}n}(t)\,\mathscr{B}_{\boldsymbol{\beta}n}(t),\\
\frac{e\eta}{c}\int \Theta(a-r)\dot{\mathbf{Q}}(\mathbf{r},t)\cdot\mathbf{A}_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} &= \sum_{\boldsymbol{\beta}n} G_{\boldsymbol{\beta}n}(t)\,\dot{\mathscr{B}}_{\boldsymbol{\beta}n}(t),
\end{aligned}\tag{37}$$

with

$$\begin{aligned}
F_{\boldsymbol{\beta}n}(t) &= e\eta \int\limits_{r<a} \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k_{\boldsymbol{\beta}n})\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r},\\
G_{\boldsymbol{\beta}n}(t) &= \frac{e\eta}{c}\int\limits_{r<a} \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k_{\boldsymbol{\beta}n})\cdot\mathbf{A}_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}.
\end{aligned}\tag{38}$$

The remaining terms describe the interactions between the mobile charge, transverse electromagnetic field, and reservoir modes. Analyzing these terms in order of appearance, we find that

$$-e\eta \int\limits_{r<a} \mathbf{Q}(\mathbf{r},t)\cdot\nabla\Phi(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} = -\sum_{\boldsymbol{\beta}}\sum_{nn'} g^{(2)}_{\boldsymbol{\beta}nn'}\mathscr{B}_{\boldsymbol{\beta}n}(t)\mathscr{B}_{\boldsymbol{\beta}n'}(t),\tag{39}$$

and

$$\frac{e\eta}{c}\int\limits_{r<a} \dot{\mathbf{Q}}(\mathbf{r},t)\cdot\mathbf{A}(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} = \sum_{\boldsymbol{\alpha\beta}}\sum_{n}\int\limits_0^\infty \lambda_{\boldsymbol{\alpha\beta}n}(\omega)\mathscr{A}_{\boldsymbol{\alpha}}(\omega,t)\dot{\mathscr{B}}_{\boldsymbol{\beta}n}(t)\,\mathrm{d}\omega,\tag{40}$$

and

$$-\eta\int\limits_0^\infty\int\limits_{r<a} v(\nu)\mathbf{Q}(\mathbf{r},t)\cdot\dot{\mathbf{Q}}_\nu(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}\nu = -\sum_{\boldsymbol{\beta}}\sum_{n}\int\limits_0^\infty v_{\boldsymbol{\beta}n}(\nu)\mathscr{B}_{\boldsymbol{\beta}n}(t)\dot{\mathscr{C}}_{\boldsymbol{\beta}n}(\nu,t),\tag{41}$$

where

$$g^{(2)}_{\boldsymbol{\beta}nn'} = \delta_{TL}16\pi^2e^2a^3\eta^2\left[\delta_{nn'}\frac{j_\ell^2(w_{\ell n})}{2}\left(1-\frac{\ell(\ell+1)}{w_{\ell n}^2}\right)-\frac{\ell(\ell+1)}{2\ell+1}\frac{j_\ell(w_{\ell n})j_\ell(w_{\ell n'})}{w_{\ell n}w_{\ell n'}}\right].\tag{42}$$

and

$$\lambda_{\boldsymbol{\alpha\beta}n}(\omega) = \frac{4\pi e^2\omega^{\frac{3}{2}}\eta}{c^3}\left[\delta_{T_\alpha T}R^{\lessgtr}_{T\ell}\left(\frac{z_{\ell n}}{a},\frac{\omega}{c};0,a\right)+\delta_{TL}\delta_{T_\alpha E}\sqrt{\ell(\ell+1)}\frac{a^2c}{w_{\ell n}\omega}j_\ell(w_{\ell n})j_\ell\left(\frac{\omega a}{c}\right)\right]\delta_{pp_\alpha}\delta_{\ell\ell_\alpha}\delta_{mm_\alpha}\tag{43}$$

and

$$v_{\boldsymbol{\beta}n}(\nu) = -2\pi a^3\eta v(\nu)\left[(\delta_{TM}+\delta_{TE})j^2_{\ell+1}(z_{\ell n})+\delta_{TL}\left(1-\frac{\ell(\ell+1)}{w_{\ell n}^2}\right)j_\ell^2(w_{\ell n})\right].\tag{44}$$

Altogether, then, we can say

$$\begin{aligned}
L_{\text{int}} = &-\sum_{\boldsymbol{\beta}}\sum_{n} F^{(2)}_{\boldsymbol{\beta}n}(t)\mathscr{B}_{\boldsymbol{\beta}n}(t)+\sum_{\boldsymbol{\beta}n} G_{\boldsymbol{\beta}n}(t)\dot{\mathscr{B}}_{\boldsymbol{\beta}n}(t)-\sum_{\boldsymbol{\beta}nn'} g^{(2)}_{\boldsymbol{\beta}nn'}\mathscr{B}_{\boldsymbol{\beta}n}(t)\mathscr{B}_{\boldsymbol{\beta}n'}(t)\\
&+\sum_{\boldsymbol{\alpha\beta}n}\int\limits_0^\infty \lambda_{\boldsymbol{\alpha\beta}n}(\omega)\mathscr{A}_{\boldsymbol{\alpha}}(\omega,t)\dot{\mathscr{B}}_{\boldsymbol{\beta}n}(t)\,\mathrm{d}\omega-\sum_{\boldsymbol{\beta}}\sum_{n}\int\limits_0^\infty v_{\boldsymbol{\beta}n}(\nu)\mathscr{B}_{\boldsymbol{\beta}n}\dot{\mathscr{C}}_{\boldsymbol{\beta}n}(\nu,t)\,\mathrm{d}\nu
\end{aligned}\tag{45}$$

such that

$$
\begin{aligned}
L = L_f + L_{\text{bind}} &+ \sum_{\boldsymbol{\beta}n} \frac{m_{\boldsymbol{\beta}n}}{2} \left(\dot{\mathcal{B}}^2_{\boldsymbol{\beta}n}(t) - \Omega^2 \mathcal{B}^2_{\boldsymbol{\beta}n}(t) \right) - \sum_{\boldsymbol{\beta}} \sum_{nn'} g_{\boldsymbol{\beta}nn'} \mathcal{B}_{\boldsymbol{\beta}n}(t) \mathcal{B}_{\boldsymbol{\beta}n'}(t) \\
&+ \sum_{\boldsymbol{\gamma}n} \int_0^\infty \frac{m_{\boldsymbol{\gamma}n}}{2} \left(\dot{\mathcal{C}}^2_{\boldsymbol{\gamma}n}(\nu,t) - \nu^2 \mathcal{C}^2_{\boldsymbol{\gamma}n}(t) \right) \mathrm{d}\nu - \sum_{\boldsymbol{\beta}n} \int_0^\infty v_{\boldsymbol{\beta}n}(\nu) \mathcal{B}_{\boldsymbol{\beta}n}(t) \dot{\mathcal{C}}_{\boldsymbol{\beta}n}(\nu,t) \, \mathrm{d}\nu \\
&+ \sum_{\boldsymbol{\alpha}} \int_0^\infty \frac{\pi e^2 \omega}{4c^3} \left[\dot{\mathcal{A}}^2_{\boldsymbol{\alpha}}(\omega,t) - \omega^2 \mathcal{A}^2_{\boldsymbol{\alpha}}(\omega,t) \right] \mathrm{d}\omega + \sum_{\boldsymbol{\alpha\beta}n} \int_0^\infty \lambda_{\boldsymbol{\alpha\beta}n}(\omega) \mathcal{A}_{\boldsymbol{\alpha}}(\omega,t) \dot{\mathcal{B}}_{\boldsymbol{\beta}n}(t) \, \mathrm{d}\omega \\
&- \sum_{\boldsymbol{\beta}n} F_{\boldsymbol{\beta}n}(t) \mathcal{B}_{\boldsymbol{\beta}n}(t) + \sum_{\boldsymbol{\beta}n} G_{\boldsymbol{\beta}n}(t) \dot{\mathcal{B}}_{\boldsymbol{\beta}n}(t)
\end{aligned}
\tag{46}
$$

where

$$
\begin{aligned}
g_{\boldsymbol{\beta}nn'} &= g^{(1)}_{\boldsymbol{\beta}nn'} + g^{(2)}_{\boldsymbol{\beta}nn'} \\
&= 4\pi^2 \delta_{TL} \delta_{nn'} e^2 a^3 \eta^2 j^2_\ell(w_{\ell n}) \left(1 - \frac{\ell(\ell+1)}{w^2_{\ell n}} \right) - 8\pi^2 \delta_{TL} e^2 \eta^2 a^3 \frac{\ell(\ell+1)}{2\ell+1} \frac{j_\ell(w_{\ell n}) j_\ell(w_{\ell n'})}{w_{\ell n} w_{\ell n'}}
\end{aligned}
\tag{47}
$$

2.3 Quantization with Classical External Fields

In order to quantize our model, we need to define the system's Hamiltonian. It will be advantageous to first define a new Lagrangian

$$
\begin{aligned}
L' &= L - \frac{\mathrm{d}}{\mathrm{d}t} \left\{ \sum_{\boldsymbol{\alpha\beta}n} \int_0^\infty \lambda_{\boldsymbol{\alpha\beta}n}(\omega) \mathcal{A}_{\boldsymbol{\alpha}}(\omega,t) \mathcal{B}_{\boldsymbol{\beta}n}(t) \, \mathrm{d}\omega \right\} - \frac{\mathrm{d}}{\mathrm{d}t} \left\{ \sum_{\boldsymbol{\beta}n} G_{\boldsymbol{\beta}n}(t) \mathcal{B}_{\boldsymbol{\beta}n}(t) \right\} \\
&= L_f + L_{\text{bind}} + \sum_{\boldsymbol{\beta}n} \frac{m_{\boldsymbol{\beta}n}}{2} \left(\dot{\mathcal{B}}^2_{\boldsymbol{\beta}n}(t) - \Omega^2 \mathcal{B}^2_{\boldsymbol{\beta}n}(t) \right) - \sum_{\boldsymbol{\beta}} \sum_{nn'} g_{\boldsymbol{\beta}nn'} \mathcal{B}_{\boldsymbol{\beta}n}(t) \mathcal{B}_{\boldsymbol{\beta}n'}(t) \\
&\quad + \sum_{\boldsymbol{\gamma}n} \int_0^\infty \frac{m_{\boldsymbol{\gamma}n}}{2} \left(\dot{\mathcal{C}}^2_{\boldsymbol{\gamma}n}(\nu,t) - \nu^2 \mathcal{C}^2_{\boldsymbol{\gamma}n}(t) \right) \mathrm{d}\nu - \sum_{\boldsymbol{\beta}n} \int_0^\infty v_{\boldsymbol{\beta}n}(\nu) \mathcal{B}_{\boldsymbol{\beta}n}(t) \dot{\mathcal{C}}_{\boldsymbol{\beta}n}(\nu,t) \, \mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\alpha}} \int_0^\infty \frac{\pi e^2 \omega}{4c^3} \left[\dot{\mathcal{A}}^2_{\boldsymbol{\alpha}}(\omega,t) - \omega^2 \mathcal{A}^2_{\boldsymbol{\alpha}}(\omega,t) \right] \mathrm{d}\omega - \sum_{\boldsymbol{\alpha\beta}n} \int_0^\infty \lambda_{\boldsymbol{\alpha\beta}n}(\omega) \dot{\mathcal{A}}_{\boldsymbol{\alpha}}(\omega,t) \mathcal{B}_{\boldsymbol{\beta}n}(t) \, \mathrm{d}\omega \\
&\quad - \sum_{\boldsymbol{\beta}n} F_{\boldsymbol{\beta}n}(t) \mathcal{B}_{\boldsymbol{\beta}n}(t) - \sum_{\boldsymbol{\beta}n} \dot{G}_{\boldsymbol{\beta}n}(t) \mathcal{B}_{\boldsymbol{\beta}n}(t)
\end{aligned}
\tag{48}
$$

that describes the same motion as L. We can then define the canonical momenta of our coordinates as

$$
\begin{aligned}
\mathcal{P}_{\boldsymbol{\beta}n}(t) &= \frac{\partial L'}{\partial \dot{\mathcal{B}}_{\boldsymbol{\beta}n}(t)} = m_{\boldsymbol{\beta}n} \dot{\mathcal{B}}_{\boldsymbol{\beta}n}(t), \\
\mathcal{Q}_{\boldsymbol{\gamma}n}(\nu,t) &= \frac{\partial L'}{\partial \dot{\mathcal{C}}_{\boldsymbol{\gamma}n}(\nu,t)} = m_{\boldsymbol{\gamma}n} \dot{\mathcal{C}}_{\boldsymbol{\gamma}n}(\nu,t) - v_{\boldsymbol{\gamma}n}(\nu) \mathcal{B}_{\boldsymbol{\gamma}n}(t), \\
\mathit{\Pi}_{\boldsymbol{\alpha}}(\omega,t) &= \frac{\partial L'}{\partial \dot{\mathcal{A}}_{\boldsymbol{\alpha}}(\omega,t)} = \frac{\pi e^2 \omega}{2c^3} \dot{\mathcal{A}}_{\boldsymbol{\alpha}}(\omega,t) - \sum_{\boldsymbol{\beta}n} \lambda_{\boldsymbol{\alpha\beta}n}(\omega) \mathcal{B}_{\boldsymbol{\beta}n}(t).
\end{aligned}
\tag{49}
$$

Calculation of the Hamiltonian is then straightforward:

$$
\begin{aligned}
H &= \sum_{\boldsymbol{\alpha}} \int_0^\infty \dot{\mathcal{A}}_{\boldsymbol{\alpha}}(\omega,t) \mathit{\Pi}_{\boldsymbol{\alpha}}(\omega,t)\, \mathrm{d}\omega + \sum_{\boldsymbol{\beta} n} \dot{\mathcal{B}}_{\boldsymbol{\beta} n}(t) \mathcal{P}_{\boldsymbol{\beta} n}(t) + \sum_{\boldsymbol{\gamma} n} \int_0^\infty \dot{\mathcal{C}}_{\boldsymbol{\gamma} n}(\nu,t) \mathcal{Q}_{\boldsymbol{\gamma} n}(\nu,t)\, \mathrm{d}\nu - L' \\
&= \sum_{\boldsymbol{\alpha}} \int_0^\infty \left(\frac{c^3}{\pi e^2 \omega} \mathit{\Pi}^2_{\boldsymbol{\alpha}}(\omega,t) + \frac{\pi e^2 \omega}{4c^3} \omega^2 \mathcal{A}^2_{\boldsymbol{\alpha}}(\omega,t) \right) \mathrm{d}\omega + \sum_{\boldsymbol{\beta} n} \left(\frac{\mathcal{P}^2_{\boldsymbol{\beta}}(t)}{2 m_{\boldsymbol{\beta} n}} + \frac{1}{2} m_{\boldsymbol{\beta} n} \tilde{\Omega}^2 \mathcal{B}^2_{\boldsymbol{\beta} n}(t) \right) \\
&\quad + \sum_{\boldsymbol{\gamma} n} \int_0^\infty \left(\frac{\mathcal{Q}^2_{\boldsymbol{\gamma} n}(\nu,t)}{2 m_{\boldsymbol{\gamma} n}} + \frac{1}{2} m_{\boldsymbol{\gamma} n} \nu^2 \mathcal{C}^2_{\boldsymbol{\gamma} n}(\nu,t) \right) \mathrm{d}\nu + \sum_{\boldsymbol{\beta}\boldsymbol{\beta}'} \sum_{nn'} \tilde{g}_{\boldsymbol{\beta}\boldsymbol{\beta}' nn'} \mathcal{B}_{\boldsymbol{\beta} n}(t) \mathcal{B}_{\boldsymbol{\beta}' n'}(t) \\
&\quad + \sum_{\boldsymbol{\alpha}\boldsymbol{\beta} n} \int_0^\infty \frac{2c^3}{\pi e^2 \omega} \lambda_{\boldsymbol{\alpha}\boldsymbol{\beta} n}(\omega) \mathit{\Pi}_{\boldsymbol{\alpha}}(\omega,t) \mathcal{B}_{\boldsymbol{\beta} n}(t)\, \mathrm{d}\omega - \sum_{\boldsymbol{\beta} n} \int_0^\infty \frac{\eta v(\nu)}{\mu} \mathcal{B}_{\boldsymbol{\beta} n}(t) \mathcal{Q}_{\boldsymbol{\beta} n}(\nu,t)\, \mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta} n} \left[F_{\boldsymbol{\beta} n}(t) + \dot{G}_{\boldsymbol{\beta} n}(t) \right] \mathcal{B}_{\boldsymbol{\beta} n}(t),
\end{aligned}
\tag{50}
$$

where

$$
\tilde{\Omega} = \left[\Omega^2 + \int_0^\infty \frac{2 v^2_{\boldsymbol{\beta} n}(\nu)}{m^2_{\boldsymbol{\beta} n}}\, \mathrm{d}\nu \right]^{\frac{1}{2}} = \left[\Omega^2 + \frac{2\eta^2}{\mu^2} \int_0^\infty v^2(\nu)\, \mathrm{d}\nu \right]^{\frac{1}{2}}
\tag{51}
$$

and

$$
\tilde{g}_{\boldsymbol{\beta}\boldsymbol{\beta}' nn'} = g_{\boldsymbol{\beta} nn'} \delta_{TL} \delta_{\boldsymbol{\beta}\boldsymbol{\beta}'} + \sum_{\boldsymbol{\alpha}} \int_0^\infty \frac{c^3}{\pi e^2 \omega} \lambda_{\boldsymbol{\alpha}\boldsymbol{\beta} n}(\omega) \lambda_{\boldsymbol{\alpha}\boldsymbol{\beta}' n'}(\omega)\, \mathrm{d}\omega .
\tag{52}
$$

Quantization is trivial here, with each coordinate and momentum replaced by an operator as

$$
\begin{aligned}
\mathcal{A}_{\boldsymbol{\alpha}}(\omega,t) &\to \hat{\mathcal{A}}_{\boldsymbol{\alpha}}(\omega,t), \\
\mathit{\Pi}_{\boldsymbol{\alpha}}(\omega,t) &\to \hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}(\omega,t), \\
\mathcal{B}_{\boldsymbol{\beta} n}(t) &\to \hat{\mathcal{B}}_{\boldsymbol{\beta} n}(t), \\
\mathcal{P}_{\boldsymbol{\beta} n}(t) &\to \hat{\mathcal{P}}_{\boldsymbol{\beta} n}(t), \\
\mathcal{C}_{\boldsymbol{\gamma} n}(\nu,t) &\to \hat{\mathcal{C}}_{\boldsymbol{\gamma} n}(\nu,t), \\
\mathcal{Q}_{\boldsymbol{\gamma} n}(\nu,t) &\to \hat{\mathcal{Q}}_{\boldsymbol{\gamma} n}(\nu,t).
\end{aligned}
\tag{53}
$$

Further assuming that each coordinate represents an excitation of a bosonic field, we can define equal-time commutation relations as

$$
\begin{aligned}
\left[\hat{\mathcal{A}}_{\boldsymbol{\alpha}}(\omega,t), \hat{\mathit{\Pi}}_{\boldsymbol{\alpha}'}(\omega',t) \right] &= \mathrm{i}\hbar \delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'} \delta(\omega-\omega'), \\
\left[\hat{\mathcal{B}}_{\boldsymbol{\beta} n}(t), \hat{\mathcal{P}}_{\boldsymbol{\beta}' n'}(t) \right] &= \mathrm{i}\hbar \delta_{\boldsymbol{\beta}\boldsymbol{\beta}'} \delta_{nn'}, \\
\left[\hat{\mathcal{C}}_{\boldsymbol{\gamma} n}(\nu,t), \hat{\mathcal{Q}}_{\boldsymbol{\gamma}' n'}(\nu',t) \right] &= \mathrm{i}\hbar \delta_{\boldsymbol{\gamma}\boldsymbol{\gamma}'} \delta_{nn'} \delta(\nu-\nu').
\end{aligned}
\tag{54}
$$

Before moving onto the usual ladder operator description of the system, it is convenient to analyze the parts of the above Hamiltonians that depend only on the coordinates and momenta of the matter resonances. The characteristic coupling constants of these terms are the constants $\tilde{g}_{\boldsymbol{\beta}\boldsymbol{\beta}' nn'}$, which take the relatively simple forms

$$
\tilde{g}_{\boldsymbol{\beta}\boldsymbol{\beta}' nn'} = \delta_{\boldsymbol{\beta}\boldsymbol{\beta}'} \delta_{nn'} 4\pi^2 e^2 a^3 \eta^2 \left([\delta_{TM} + \delta_{TE}] j^2_{\ell-1}(z_{\ell n}) + \delta_{TL} j^2_{\ell}(w_{\ell n}) \left[1 - \frac{\ell(\ell+1)}{w^2_{\ell n}} \right] \right)
\tag{55}
$$

as can be seen be combining the results for $g_{\boldsymbol{\beta} nn'}$ above with the derivations of Appendix G. Importantly, these coupling constants do not allow for energy to pass between any two modes of different indices, such that they only serve to modify the resonance frequencies of each mode via a "self-coupling" interaction. Explicitly, we then have

$$
\begin{aligned}
\hat{H} = &\sum_{\boldsymbol{\alpha}} \int_0^\infty \left(\frac{\hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}^2(\omega,t)}{2m_0(\omega)} + \frac{1}{2} m_0(\omega)\omega^2 \hat{\mathscr{A}}_{\boldsymbol{\alpha}}^2(\omega,t) \right) \mathrm{d}\omega + \sum_{\boldsymbol{\beta} n} \left(\frac{\hat{\mathscr{P}}_{\boldsymbol{\beta}}^2(t)}{2m_{\boldsymbol{\beta} n}} + \frac{1}{2} m_{\boldsymbol{\beta} n} \bar{\Omega}^2 \hat{\mathscr{B}}_{\boldsymbol{\beta} n}^2(t) \right) \\
&+ \sum_{\boldsymbol{\gamma} n} \int_0^\infty \left(\frac{\hat{\mathscr{Q}}_{\boldsymbol{\gamma} n}^2(\nu,t)}{2m_{\boldsymbol{\gamma} n}} + \frac{1}{2} m_{\boldsymbol{\gamma} n} \nu^2 \hat{C}_{\boldsymbol{\gamma} n}^2(\nu,t) \right) \mathrm{d}\nu + \sum_{\boldsymbol{\alpha\beta} n} \int_0^\infty \frac{\lambda_{\boldsymbol{\alpha\beta} n}(\omega)}{m_0(\omega)} \hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}(\omega,t) \hat{\mathscr{B}}_{\boldsymbol{\beta} n}(t)\, \mathrm{d}\omega \\
&- \sum_{\boldsymbol{\beta} n} \int_0^\infty \frac{\eta v(\nu)}{\mu} \hat{\mathscr{B}}_{\boldsymbol{\beta} n}(t) \hat{\mathscr{Q}}_{\boldsymbol{\beta} n}(\nu,t)\, \mathrm{d}\nu + \sum_{\boldsymbol{\beta} n} \left[F_{\boldsymbol{\beta} n}(t) + \dot{G}_{\boldsymbol{\beta} n}(t) \right] \hat{\mathscr{B}}_{\boldsymbol{\beta} n}(t),
\end{aligned}
\tag{56}
$$

where

$$
\begin{aligned}
\bar{\Omega} &= \left[\tilde{\Omega}^2 + \frac{2\tilde{g}_{\boldsymbol{\beta\beta} nn}}{m_{\boldsymbol{\beta} n}} \right]^{\frac{1}{2}} \\
&= \left[\Omega^2 + \frac{2\eta^2}{\mu^2} \int_0^\infty v^2(\nu)\, \mathrm{d}\nu + \frac{4\pi e^2 \eta^2}{\mu} \right]^{\frac{1}{2}}
\end{aligned}
\tag{57}
$$

are the natural frequencies of each oscillator raised by the self-coupling and $m_0(\omega) = \pi e^3 \omega / 2c^3$ is the photon effective mass.

From here, a transition into a ladder-operator picture is straightforward. Using the phase conventions of Huttner and Barnett [3] for "bath" operators with continuous indices, we find

$$
\begin{aligned}
\hat{a}_{\boldsymbol{\alpha}}(\omega,t) &= \sqrt{\frac{m_0(\omega)}{2\hbar\omega}} \left[-\mathrm{i}\omega \hat{\mathscr{A}}_{\boldsymbol{\alpha}}(\omega,t) + \frac{1}{m_0(\omega)} \hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}(\omega,t) \right], \\
\hat{b}_{\boldsymbol{\beta} n}(t) &= \sqrt{\frac{m_{\boldsymbol{\beta} n}}{2\hbar\bar{\Omega}}} \left[\bar{\Omega} \hat{\bar{\mathscr{B}}}_{\boldsymbol{\beta} n}(t) + \frac{\mathrm{i}}{m_{\boldsymbol{\beta} n}} \hat{\bar{\mathscr{P}}}_{\boldsymbol{\beta} n}(t) \right], \\
\hat{c}_{\boldsymbol{\gamma} n}(\nu,t) &= \sqrt{\frac{m_{\boldsymbol{\gamma} n}}{2\hbar\nu}} \left[-\mathrm{i}\nu \hat{C}_{\boldsymbol{\gamma} n}(\nu,t) + \frac{1}{m_{\boldsymbol{\gamma} n}} \hat{\mathscr{Q}}_{\boldsymbol{\gamma} n}(\nu,t) \right],
\end{aligned}
\tag{58}
$$

such that

$$
\begin{aligned}
\left[\hat{a}_{\boldsymbol{\alpha}}(\omega,t), \hat{a}_{\boldsymbol{\alpha}'}^\dagger(\omega',t) \right] &= \delta_{\boldsymbol{\alpha\alpha}'} \delta(\omega - \omega'), \\
\left[\hat{b}_{\boldsymbol{\beta} n}(t), \hat{b}_{\boldsymbol{\beta}' n'}^\dagger(t) \right] &= \delta_{\boldsymbol{\beta\beta}'} \delta_{nn'}, \\
\left[\hat{c}_{\boldsymbol{\gamma} n}(\nu,t), \hat{c}_{\boldsymbol{\gamma}' n'}^\dagger(\nu',t) \right] &= \delta_{\boldsymbol{\gamma\gamma}'} \delta_{nn'} \delta(\nu - \nu'),
\end{aligned}
\tag{59}
$$

and

$$
\begin{aligned}
\left[\hat{a}_{\boldsymbol{\alpha}}(\omega,t), \hat{a}_{\boldsymbol{\alpha}'}(\omega',t) \right] &= \left[\hat{a}_{\boldsymbol{\alpha}}^\dagger(\omega,t), \hat{a}_{\boldsymbol{\alpha}'}^\dagger(\omega',t) \right] = 0, \\
\left[\hat{b}_{\boldsymbol{\beta} n}(t), \hat{b}_{\boldsymbol{\beta}' n'}(t) \right] &= \left[\hat{b}_{\boldsymbol{\beta} n}^\dagger(t), \hat{b}_{\boldsymbol{\beta}' n'}^\dagger(t) \right] = 0, \\
\left[\hat{c}_{\boldsymbol{\gamma} n}(\nu,t), \hat{c}_{\boldsymbol{\gamma}' n'}(\nu',t) \right] &= \left[\hat{c}_{\boldsymbol{\gamma} n}^\dagger(\nu,t), \hat{c}_{\boldsymbol{\gamma}' n'}^\dagger(\nu',t) \right] = 0.
\end{aligned}
\tag{60}
$$

The coordinate and momentum operators can then be reconstructed as

$$
\begin{aligned}
\hat{\mathscr{A}}_{\boldsymbol{\alpha}}(\omega,t) &= \mathrm{i}\sqrt{\frac{\hbar}{2\omega m_0(\omega)}}\left[\hat{a}_{\boldsymbol{\alpha}}(\omega,t)-\hat{a}^{\dagger}_{\boldsymbol{\alpha}}(\omega,t)\right],\\
\hat{\Pi}_{\boldsymbol{\alpha}}(\omega,t) &= \sqrt{\frac{m_0(\omega)\hbar\omega}{2}}\left[\hat{a}_{\boldsymbol{\alpha}}(\omega,t)+\hat{a}^{\dagger}_{\boldsymbol{\alpha}}(\omega,t)\right],\\
\hat{\bar{\mathscr{B}}}_{\boldsymbol{\beta}n}(t) &= \sqrt{\frac{\hbar}{2\bar{\Omega}m_{\boldsymbol{\beta}n}}}\left[\hat{b}_{\boldsymbol{\beta}n}(t)+\hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t)\right],\\
\hat{\bar{\mathscr{P}}}_{\boldsymbol{\beta}n}(t) &= -\mathrm{i}\sqrt{\frac{m_{\boldsymbol{\beta}n}\hbar\bar{\Omega}}{2}}\left[\hat{b}_{\boldsymbol{\beta}n}(t)-\hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t)\right],\\
\hat{\mathscr{C}}_{\boldsymbol{\gamma}n}(\nu,t) &= \mathrm{i}\sqrt{\frac{\hbar}{2\nu m_{\boldsymbol{\gamma}n}}}\left[\hat{c}_{\boldsymbol{\gamma}n}(\nu,t)-\hat{c}^{\dagger}_{\boldsymbol{\gamma}n}(\nu,t)\right],\\
\hat{\mathscr{Q}}_{\boldsymbol{\gamma}n}(\nu,t) &= \sqrt{\frac{m_{\boldsymbol{\gamma}n}\hbar\nu}{2}}\left[\hat{c}_{\boldsymbol{\gamma}n}(\nu,t)+\hat{c}^{\dagger}_{\boldsymbol{\gamma}n}(\nu,t)\right],
\end{aligned}
\tag{61}
$$

providing, after substitution, a Hamiltonian

$$
\begin{aligned}
\hat{H} = {} & \sum_{\boldsymbol{\alpha}}\int_0^\infty \hbar\omega\hat{a}^{\dagger}_{\boldsymbol{\alpha}}(\omega,t)\hat{a}_{\boldsymbol{\alpha}}(\omega,t)\,\mathrm{d}\omega + \sum_{\boldsymbol{\beta}n}\hbar\bar{\Omega}\hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t)\hat{b}_{\boldsymbol{\beta}n}(t) + \sum_{\boldsymbol{\gamma}n}\int_0^\infty \hbar\nu\hat{c}^{\dagger}_{\boldsymbol{\gamma}n}(\nu,t)\hat{c}_{\boldsymbol{\gamma}n}\,\mathrm{d}\nu(\nu,t)\\
& + \sum_{\boldsymbol{\alpha\beta}n}\int_0^\infty \hbar\Lambda_{\boldsymbol{\alpha\beta}n}(\nu)\left[\hat{a}_{\boldsymbol{\alpha}}(\omega,t)+\hat{a}^{\dagger}_{\boldsymbol{\alpha}}(\omega,t)\right]\left[\hat{b}_{\boldsymbol{\beta}n}(t)+\hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t)\right]\mathrm{d}\omega\\
& + \sum_{\boldsymbol{\beta}n}\int_0^\infty \hbar V(\nu)\left[\hat{b}_{\boldsymbol{\beta}n}(t)+\hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t)\right]\left[\hat{c}_{\boldsymbol{\beta}n}(\nu,t)+\hat{c}^{\dagger}_{\boldsymbol{\beta}n}(\nu,t)\right]\mathrm{d}\nu\\
& + \sum_{\boldsymbol{\beta}n}\hbar J_{\boldsymbol{\beta}n}(t)\left[\hat{b}_{\boldsymbol{\beta}n}(t)+\hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t)\right],
\end{aligned}
\tag{62}
$$

wherein we have exploited the identities

$$
\begin{aligned}
\hat{a}_{\boldsymbol{\alpha}}(\omega,t)\hat{a}^{\dagger}_{\boldsymbol{\alpha}}(\omega,t) &= \hat{a}^{\dagger}_{\boldsymbol{\alpha}}(\omega,t)\hat{a}_{\boldsymbol{\alpha}}(\omega,t)+1,\\
\hat{b}_{\boldsymbol{\beta}n}(t)\hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t) &= \hat{b}^{\dagger}_{\boldsymbol{\beta}n}(t)\hat{b}_{\boldsymbol{\beta}n}(t)+1,\\
\hat{c}_{\boldsymbol{\gamma}n}(\nu,t)\hat{c}^{\dagger}_{\boldsymbol{\gamma}n}(\nu,t) &= \hat{c}^{\dagger}_{\boldsymbol{\gamma}n}(\nu,t)\hat{c}_{\boldsymbol{\gamma}n}(\nu,t)+1,
\end{aligned}
\tag{63}
$$

and dropped all constant terms. Further, we have defined new coupling constants

$$
\begin{aligned}
\Lambda_{\boldsymbol{\alpha\beta}n}(\omega) &= \lambda_{\boldsymbol{\alpha\beta}n}\sqrt{\frac{\omega}{4m_0(\omega)m_{\boldsymbol{\beta}n}\bar{\Omega}}},\\
V(\nu) &= \frac{\eta v(\nu)}{\mu}\sqrt{\frac{\nu}{4\bar{\Omega}}},\\
J_{\boldsymbol{\beta}n}(t) &= \sqrt{\frac{\hbar}{2\bar{\Omega}m_{\boldsymbol{\beta}n}}}\left[F_{\boldsymbol{\beta}n}(t)+\dot{G}_{\boldsymbol{\beta}n}(t)\right].
\end{aligned}
\tag{64}
$$

A Unit Conventions

The chosen dimensions and units for each important quantity are shown below.

Symbol Name	Dimensions	Units (cgs Gaussian)
$\mathscr{L}$	$\frac{[U]}{[L]^3} = \frac{[M]}{[L][T^2]}$	$\frac{\text{erg}}{\text{cm}^3}$
$L,\ H$	$[U]$	erg
$\mathscr{A}_{\boldsymbol{\alpha}}(\omega, t)$	$[L][T]^{\frac{1}{2}}$	$\text{cm} \cdot \text{s}^{\frac{1}{2}}$
$\mathscr{B}_{\boldsymbol{\beta} n}(t)$	$[L]$	cm
$\mathscr{C}_{\boldsymbol{\gamma} n}(\nu, t)$	$[L][T]^{\frac{1}{2}}$	$\text{cm} \cdot \text{s}^{\frac{1}{2}}$
$\mathit{\Pi}_{\boldsymbol{\alpha}}(\omega, t)$	$\frac{[L][M]}{[T]^{\frac{1}{2}}}$	$\frac{\text{cm}\cdot\text{g}}{\text{s}^{\frac{1}{2}}}$
$\mathscr{P}_{\boldsymbol{\beta} n}(t)$	$\frac{[M][L]}{[T]}$	$\frac{\text{cm}\cdot\text{cm}}{\text{s}}$
$\mathscr{Q}_{\boldsymbol{\gamma} n}(\nu, t)$	$\frac{[L][M]}{[T]^{\frac{1}{2}}}$	$\frac{\text{cm}\cdot\text{g}}{\text{s}^{\frac{1}{2}}}$
$g_{\boldsymbol{\beta} nn},\ \tilde{g}_{\boldsymbol{\beta}\boldsymbol{\beta}' nn'}$	$\frac{[M]}{[T]^2}$	$\frac{\text{g}}{\text{s}^2}$
$\lambda_{\boldsymbol{\alpha}\boldsymbol{\beta} n}(\omega)$	$\frac{[M]}{[T]^{\frac{1}{2}}}$	$\frac{\text{g}}{\text{s}^{\frac{1}{2}}}$
$v(\nu),\ v_{\boldsymbol{\beta}\nu}(\nu)$	$\frac{[M]}{[T]^{\frac{1}{2}}}$	$\frac{\text{g}}{\text{s}^{\frac{1}{2}}}$
$\hat{a}_{\boldsymbol{\alpha}}(\omega, t)$	$[T]^{\frac{1}{2}}$	$\text{s}^{\frac{1}{2}}$
$\hat{b}_{\boldsymbol{\beta} n}(t)$	1	1
$\hat{c}_{\boldsymbol{\gamma} n}(\nu, t)$	$[T]^{\frac{1}{2}}$	$\text{s}^{\frac{1}{2}}$
$\sigma_{\boldsymbol{\beta}\boldsymbol{\beta} nn'}$	$\frac{1}{[T]}$	s^{-1}
$\Lambda_{\boldsymbol{\alpha}\boldsymbol{\beta} n}(\omega),\ \tilde{\Lambda}_{\boldsymbol{\alpha}\boldsymbol{\beta} n}(\omega)$	$\frac{1}{[T]^{\frac{1}{2}}}$	$\text{s}^{-\frac{1}{2}}$
$V_{\boldsymbol{\beta} n}(\nu),\ \tilde{V}_{\boldsymbol{\beta} n}(\nu)$	$\frac{1}{[T]^{\frac{1}{2}}}$	$\text{s}^{-\frac{1}{2}}$

B Harmonic Definitions

The dyadic Green's function of a sphere is composed of transverse vector spherical harmonics

$$\begin{aligned}
\mathbf{M}_{p\ell m}(\mathbf{r}, k) &= \sqrt{K_{\ell m}} \left[\frac{(-1)^{p+1} m}{\sin\theta} j_\ell(kr) P_{\ell m}(\cos\theta) S_{p+1}(m\phi) \hat{\boldsymbol{\theta}} - j_\ell(kr) \frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta} S_p(m\phi) \hat{\boldsymbol{\phi}} \right], \\
\mathbf{N}_{p\ell m}(\mathbf{r}, k) &= \sqrt{K_{\ell m}} \left[\frac{\ell(\ell+1)}{kr} j_\ell(kr) P_{\ell m}(\cos\theta) S_p(m\phi) \hat{\mathbf{r}} \right. \\
&\quad \left. + \frac{1}{kr} \frac{\partial\{r j_\ell(kr)\}}{\partial r} \left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta} S_p(m\phi) \hat{\boldsymbol{\theta}} + \frac{(-1)^{p+1} m}{\sin\theta} P_{\ell m}(\cos\theta) S_{p+1}(m\phi) \hat{\boldsymbol{\phi}} \right) \right],
\end{aligned} \tag{B.1}$$

which each have zero divergence, i.e. $\nabla \cdot \mathbf{M}_{p\ell m}(\mathbf{r}, k) = \nabla \cdot \mathbf{N}_{p\ell m}(\mathbf{r}, k) = 0$, and longitudinal vector spherical harmonics

$$\begin{aligned}\mathbf{L}_{p\ell m}(\mathbf{r}, k) &= \frac{1}{k}\nabla\left\{\sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}j_\ell(kr)P_{\ell m}(\cos\theta)S_p(m\phi)\right\} \\ &= \sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}\left[\frac{1}{k}\frac{\partial\{j_\ell(kr)\}}{\partial r}P_{\ell m}(\cos\theta)S_p(m\phi)\hat{\mathbf{r}} + \frac{1}{kr}j_\ell(kr)\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\theta}}\right. \\ &\left. + \frac{(-1)^{p+1}m}{kr\sin\theta}j_\ell(kr)P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\phi}}\right]\end{aligned} \tag{B.2}$$

which obey $\nabla \times \mathbf{L}_{p\ell m}(\mathbf{r}, k) = 0$. Here $j_\ell(x)$ are spherical Bessel functions with $\ell = 1, 2, 3, \ldots, \infty$; $P_{\ell m}(x)$ are associated Legendre polynomials with the same range in ℓ; $m = 0, 1, 2, \ldots, \ell$; and

$$S_p(mx) = \begin{cases}\cos(mx), & p \text{ even,} \\ \sin(mx), & p \text{ odd}\end{cases} \tag{B.3}$$

and the same range in m. Due to the binary nature of p, we will in general need to use special notation to assert that $S_p(x) = S_{p+2}(x) = S_{p+4}(x) = \cdots$. However, to save space, we will use the convention that any nonzero even value of p is automatically replaced by 0 and any non-one odd value of p is automatically replaced by 1 in what follows. Further, the prefactors $K_{\ell m}$, which are set to 1 in most texts, are set to

$$K_{\ell m} = (2 - \delta_{m0})\frac{2\ell+1}{\ell(\ell+1)}\frac{(\ell-m)!}{(\ell+m)!} \tag{B.4}$$

in an effort to regularize the transverse harmonics, i.e. ensure that their maximum magnitudes in $\mathbf{r}$ for fixed k are similar for all index combinations T, p, ℓ, m. The prefactors of the Laplacian harmonics are defined similarly, but with an added factor of δ_{TE}. Here, T is a fourth "type" index, similar to the parity (p), order (ℓ), and degree (m) indices, that takes values of M (magnetic type) or E (electric type) to describe whether the characteristic field profiles of each mode resemble those of magnetic or electric multipoles. Only the electric longitudinal modes $\mathbf{L}_{p\ell m}(\mathbf{r})$ are nonzero such that we won't bother to invent a second symbol for magnetic longitudinal modes, but the transverse harmonics clearly come in two different types: magnetic modes $\mathbf{M}_{p\ell m}(\mathbf{r}, k)$ ($T = M$) and electric modes $\mathbf{N}_{p\ell m}(\mathbf{r}, k)$ ($T = E$).

Both the transverse and Laplacian modes also come with a fifth modifier, the "region" index, taking values of $<$ for *interior* harmonics and $>$ for *exterior* harmonics, although we have neglected append our definitions with yet another symbol. The interior harmonics, listed above as $\mathbf{L}$, $\mathbf{M}$, and $\mathbf{N}$, are generally used for $\mathbf{r}$ confined within some closed spherical surface such that $r < \infty$ always and $r = 0$ somwhere within the region. To describe fields in regions that do not include the origin, one can find the second set of solutions to the second-order wave equation PDE, which are the so-called *exterior* vector spherical harmonics $\boldsymbol{\mathcal{L}}_{p\ell m}(\mathbf{r})$, $\boldsymbol{\mathcal{M}}_{p\ell m}(\mathbf{r}, k)$, and $\boldsymbol{\mathcal{N}}_{p\ell m}(\mathbf{r}, k)$. These harmonics are defined with the simple substitutions $j_\ell(kr) \to h_\ell(kr)$ and are explicitly given by

$$\begin{aligned}\boldsymbol{\mathcal{L}}_{p\ell m}(\mathbf{r}, k) &= \frac{1}{k}\nabla\left\{\sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}h_\ell(kr)P_{\ell m}(\cos\theta)S_p(m\phi)\right\}, \\ &= \sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}\left[\frac{1}{k}\frac{\partial\{h_\ell(kr)\}}{\partial r}P_{\ell m}(\cos\theta)S_p(m\phi)\hat{\mathbf{r}} + \frac{1}{kr}h_\ell(kr)\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\theta}}\right. \\ &\left. + \frac{(-1)^{p+1}m}{kr\sin\theta}h_\ell(kr)P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\phi}}\right], \\ \boldsymbol{\mathcal{M}}_{p\ell m}(\mathbf{r}, k) &= \sqrt{K_{\ell m}}\left[\frac{(-1)^{p+1}m}{\sin\theta}h_\ell(kr)P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\theta}} - h_\ell(kr)\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\phi}}\right], \\ \boldsymbol{\mathcal{N}}_{p\ell m}(\mathbf{r}, k) &= \sqrt{K_{\ell m}}\left[\frac{\ell(\ell+1)}{kr}h_\ell(kr)P_{\ell m}(\cos\theta)S_p(m\phi)\hat{\mathbf{r}}\right. \\ &\left. + \frac{1}{kr}\frac{\partial\{rh_\ell(kr)\}}{\partial r}\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\theta}} + \frac{(-1)^{p+1}m}{\sin\theta}P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\phi}}\right)\right].\end{aligned} \tag{B.5}$$

The functions $h_\ell(kr) = j_\ell(kr) + \mathrm{i}y_\ell(kr)$ are the spherical Hankel functions of the first kind and involve the spherical Bessel functions of both the first kind ($j_\ell[kr]$) and the second kind, $y_\ell(kr)$, the latter of which are irregular at $r \to 0$, justifying their exclusion from use in all regions that include the point $r = 0$.

We would like the notation for the harmonics to be as succinct as possible. Therefore, for shorthand, we can define

$$\begin{aligned} \mathbf{X}_{Tp\ell m}(\mathbf{r},k) &= \begin{cases} \mathbf{L}_{p\ell m}(\mathbf{r},k), & T = L, \\ \mathbf{M}_{p\ell m}(\mathbf{r},k), & T = M, \\ \mathbf{N}_{p\ell m}(\mathbf{r},k), & T = E; \end{cases} \\ \boldsymbol{\mathcal{X}}_{Tp\ell m}(\mathbf{r},k) &= \begin{cases} \boldsymbol{\mathcal{L}}_{p\ell m}(\mathbf{r},k), & T = L, \\ \boldsymbol{\mathcal{M}}_{p\ell m}(\mathbf{r},k), & T = M, \\ \boldsymbol{\mathcal{N}}_{p\ell m}(\mathbf{r},k), & T = E. \end{cases} \end{aligned} \tag{B.6}$$

Here, we have finally introduced the fourth "type" index to the harmonic notation, with L, M, and E denoting the Longitudinal, transverse Magnetic, and transverse Electric harmonics, respectively. We can condense the notation further by writing the string of four indices as $\boldsymbol{\alpha} = (T, p, \ell, m)$, such that our harmonics can now be written as $\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)$ and $\boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}(\mathbf{r},k)$.

The symmetries of the vector spherical harmonics are such that

$$\begin{aligned} \mathbf{L}_{p\ell m}(\mathbf{r},k) &= (-1)^{\ell+1}\mathbf{L}_{p\ell m}(\mathbf{r},-k), \\ \mathbf{M}^*_{p\ell m}(\mathbf{r},k) &= (-1)^{\ell}\mathbf{M}_{p\ell m}(\mathbf{r},-k), \\ \mathbf{N}^*_{p\ell m}(\mathbf{r},k) &= (-1)^{\ell+1}\mathbf{N}_{p\ell m}(\mathbf{r},-k), \end{aligned} \tag{B.7}$$

with identical relations for the respective exterior harmonics. Further, the transverse harmonics obey the duality

$$\begin{aligned} \nabla \times \mathbf{M}_{p\ell m}(\mathbf{r},k) &= k\mathbf{N}_{p\ell m}(\mathbf{r},k), \\ \nabla \times \mathbf{N}_{p\ell m}(\mathbf{r},k) &= k\mathbf{M}_{p\ell m}(\mathbf{r},k), \end{aligned} \tag{B.8}$$

and similar for the exterior transverse harmonics, such that a second set of harmonics can be defined as

$$\begin{aligned} \mathbf{Y}_{Lp\ell m}(\mathbf{r},t) &= 0, \\ \mathbf{Y}_{Mp\ell m}(\mathbf{r},k) &= \mathbf{N}_{p\ell m}(\mathbf{r},t), \\ \mathbf{Y}_{Ep\ell m}(\mathbf{r},k) &= \mathbf{M}_{p\ell m}(\mathbf{r},t). \end{aligned} \tag{B.9}$$

More succinctly,

$$\begin{aligned} \mathbf{Y}_{\boldsymbol{\alpha}}(\mathbf{r},k) &= \frac{1}{k}\nabla \times \mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k), \\ \boldsymbol{\mathcal{Y}}_{\boldsymbol{\alpha}}(\mathbf{r},k) &= \frac{1}{k}\nabla \times \boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}(\mathbf{r},k), \end{aligned} \tag{B.10}$$

where $\boldsymbol{\mathcal{Y}}(\mathbf{r},k)$ are the corresponding exterior harmonics to $\mathbf{Y}_{\boldsymbol{\alpha}}(\mathbf{r},k)$.

Due to their regularity at all points $\mathbf{r}$, the interior harmonics obey the orthogonality condition

$$\int \mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k) \cdot \mathbf{X}_{\boldsymbol{\alpha}'}(\mathbf{r},k')\,\mathrm{d}^3\mathbf{r} = \frac{2\pi^2}{k^2}\delta(k-k')(1-\delta_{p1}\delta_{m0})\delta_{TT'}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'} \tag{B.11}$$

for $k, k' > 0$ and integration across the entire universe. The transverse magnetic harmonics also satisfy convenient orthogonality relations when integrated over finite regions in r, with

$$\begin{aligned} \int_0^{2\pi}\int_0^{\pi}\int_a^{b} \mathbf{L}_{p\ell m}(\mathbf{r},k) \cdot \mathbf{M}_{p'\ell'm'}(\mathbf{r},k')r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi &= 0, \\ \int_0^{2\pi}\int_0^{\pi}\int_a^{b} \mathbf{N}_{p\ell m}(\mathbf{r},k) \cdot \mathbf{M}_{p'\ell'm'}(\mathbf{r},k')r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi &= 0. \end{aligned} \tag{B.12}$$

However, finite spherical integrals of the longitudinal and transverse electric harmonics produce

$$\int_0^{2\pi}\int_0^{\pi}\int_a^{b} \mathbf{L}_{p\ell m}(\mathbf{r},k)\cdot\mathbf{N}_{p'\ell'm'}(\mathbf{r},k')r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = 4\pi(1-\delta_{p1}\delta_{m0})\sqrt{\ell(\ell+1)}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'} \times\frac{1}{kk'}\left[bj_\ell(kb)j_\ell(k'b)-aj_\ell(ka)j_\ell(k'a)\right], \tag{B.13}$$

which can be seen to produce zero in the limits $a\to 0$, $b\to\infty$ as $\lim_{r\to 0} rj_\ell(kr)j_\ell(k'r) = \lim_{r\to\infty} rj_\ell(kr)j_\ell(k'r) = 0$ for all real, positive k and k' if ℓ is an integer greater than 0. Finally, the harmonics of the same type are normalized such that integrals over finite spherical regions produce

$$\int_0^{2\pi}\int_0^{\pi}\int_a^{b} \mathbf{X}_{Tp\ell m}(\mathbf{r},k)\cdot\mathbf{X}_{Tp'\ell'm'}(\mathbf{r},k')r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = 4\pi(1-\delta_{p1}\delta_{m0})\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}R^{\ll}_{T\ell}(k,k';a,b), \tag{B.14}$$

where $R^{\ll}_{T\ell}(k,k';a,b)$ are radial integral functions defined by

$$\begin{aligned}
R^{\ll}_{M\ell}(k,k';a,b) &= \int_a^b j_\ell(kr)j_\ell(k'r)r^2\,\mathrm{d}r \\
&= \left.\frac{r^2}{k^2-k'^2}\left[k'j_{\ell-1}(k'r)j_\ell(kr)-kj_{\ell-1}(kr)j_\ell(k'r)\right]\right|_a^b, \\
R^{\ll}_{E\ell}(k,k';a,b) &= \frac{\ell+1}{2\ell+1}R^{\ll}_{M,\ell-1}(k,k';a,b)+\frac{\ell}{2\ell+1}R^{\ll}_{M,\ell+1}(k,k';a,b), \\
R^{\ll}_{L\ell}(k,k';a,b) &= \frac{\ell+1}{2\ell+1}R^{\ll}_{M,\ell+1}(k,k';a,b)+\frac{\ell}{2\ell+1}R^{\ll}_{M,\ell-1}(k,k';a,b).
\end{aligned} \tag{B.15}$$

The superscript $\ll$ indicates that the integral involves two spherical Bessel functions of the first kind and thus arises from the integration of two interior harmonics.

An important case of this integral occurs when $b\to a$ and $a\to 0$, i.e. during integration over a finite sphere. In this case, the characteristic wavenumbers are often defined such that $k=z_{\ell n}/a$ and $k'=z_{\ell n'}/a$, where $z_{\ell n}$ is the n^{th} unitless root of $j_\ell(x)$ with $n=1,2,\ldots$, or as $k=w_{\ell n}/a$ and $k'=w_{\ell n'}/a$, where $w_{\ell n}$ is the n^{th} unitless root of $\partial j_\ell(x)/\partial x$ with $n=1,2,\ldots$. With these values of k and k' fixed, we can use the identity $j_{\ell-1}(w_{\ell n})=(\ell+1)j_\ell(w_{\ell n})/w_{\ell n}$ to see that $R^{\ll}_{M\ell}(z_{\ell n}/a,z_{\ell n'}/a;0,a)=R^{\ll}_{M\ell}(w_{\ell n}/a,w_{\ell n'}/a;0,a)=0$ for $n\neq n'$. Further, with

$$\lim_{k'\to k} R^{\ll}_{M\ell}(k,k';0,a)=\frac{a^3}{2}\left[j_\ell^2(ka)-j_{\ell-1}(ka)j_{\ell+1}(ka)\right] \tag{B.16}$$

by L'Hopital's rule, we can use the identity $j_{\ell-1}(z_{n\ell})=-j_{\ell+1}(z_{n\ell})$ to say

$$R^{\ll}_{M\ell}\left(\frac{z_{\ell n}}{a},\frac{z_{\ell n'}}{a};0,a\right)=R^{\ll}_{M,\ell-1}\left(\frac{z_{\ell n}}{a},\frac{z_{\ell n'}}{a};0,a\right)=R^{\ll}_{M,\ell+1}\left(\frac{z_{\ell n}}{a},\frac{z_{\ell n'}}{a};0,a\right)=\delta_{nn'}\frac{a^3}{2}j^2_{\ell+1}(z_{\ell n}). \tag{B.17}$$

Further, we can use the identities $j_{\ell+1}(w_{\ell n})=\ell j_\ell(w_{\ell n})/w_{\ell n}$, $j_{\ell-2}(x)=(2\ell-1)j_{\ell-1}(x)/x-j_\ell(x)$, and $j_{\ell+2}(x)=(2\ell+3)j_{\ell+1}(x)/x-j_\ell(x)$ to say

$$\begin{aligned}
R^{\ll}_{M\ell}\left(\frac{w_{\ell n}}{a},\frac{w_{\ell n'}}{a};0,a\right) &= \delta_{nn'}\frac{a^3}{2}j_\ell^2(w_{\ell n})\left(1-\frac{\ell(\ell+1)}{w^2_{\ell n}}\right), \\
R^{\ll}_{M,\ell-1}\left(\frac{w_{\ell n}}{a},\frac{w_{\ell n'}}{a};0,a\right) &= \delta_{nn'}\frac{a^3}{2}j_\ell^2(w_{\ell n})\left(1-\frac{\ell^2-\ell-2}{w^2_{\ell n}}\right), \\
R^{\ll}_{M,\ell+1}\left(\frac{w_{\ell n}}{a},\frac{w_{\ell n'}}{a};0,a\right) &= \delta_{nn'}\frac{a^3}{2}j_\ell^2(w_{\ell n})\left(1-\frac{\ell(\ell+3)}{w^2_{\ell n}}\right).
\end{aligned} \tag{B.18}$$

These expressions can be used to construct the useful identities

$$\begin{aligned} R_{E\ell}^{\lll}\left(\frac{z_{\ell n}}{a}, \frac{z_{\ell n'}}{a}; 0, a\right) &= \delta_{nn'} \frac{a^3}{2} j_{\ell+1}^2(z_{\ell n}), \\ R_{L\ell}^{\lll}\left(\frac{w_{\ell n}}{a}, \frac{w_{\ell n'}}{a}; 0, a\right) &= \delta_{nn'} \frac{a^3}{2} \left(1 - \frac{\ell(\ell+1)}{w_{\ell n}^2}\right) j_\ell^2(w_{\ell n}). \end{aligned} \tag{B.19}$$

Therefore, letting $k_{Mp\ell mn} = k_{Ep\ell mn} = z_{\ell n}/a$ and $k_{Lp\ell mn} = w_{\ell n}/a$, we have

$$\begin{aligned} \int_0^{2\pi}\int_0^{\pi}\int_0^{a} \mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r}, k_{\boldsymbol{\alpha} n}) \cdot \mathbf{X}_{\boldsymbol{\alpha}'}(\mathbf{r}, k_{\boldsymbol{\alpha}' n'}) r^2 \sin\theta \, \mathrm{d}r \, \mathrm{d}\theta \, \mathrm{d}\phi &= 2\pi a^3 (1 - \delta_{p1}\delta_{m0}) \delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'} \delta_{nn'} \\ &\times \left[(\delta_{TM} + \delta_{TE}) j_{\ell+1}^2(z_{\ell n}) + \delta_{TL} \left(1 - \frac{\ell(\ell+1)}{w_{\ell n}^2}\right) j_\ell^2(w_{\ell n}) \right]. \end{aligned} \tag{B.20}$$

In related fashion, using the identity

$$\nabla \cdot \mathbf{L}_{p\ell m}(\mathbf{r}, k) = -k\sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)} j_\ell(kr) P_{\ell m}(\cos\theta) S_p(m\phi), \tag{B.21}$$

we have

$$\begin{aligned} &\int_0^{2\pi}\int_0^{\pi}\int_0^{a} \left[\nabla \cdot \mathbf{L}_{p\ell m}(\mathbf{r}, k_{L\ell n})\right] \left[\nabla \cdot \mathbf{L}_{p'\ell' m'}(\mathbf{r}, k_{L\ell' n'})\right] r^2 \sin\theta \, \mathrm{d}r \, \mathrm{d}\theta \, \mathrm{d}\phi \\ &= 2\pi a^3 (1 - \delta_{p1}\delta_{m0}) \delta_{pp'} \delta_{\ell\ell'} \delta_{mm'} \delta_{nn'} k_{L\ell n}^2 j_\ell^2(k_{L\ell n} a) \left(1 - \frac{\ell(\ell+1)}{(k_{L\ell n} a)^2}\right). \end{aligned} \tag{B.22}$$

In addition to our vector spherical harmonics, the expansion of the Coulomb Green's function

$$1/|\mathbf{r} - \mathbf{r}'| = \sum_{p\ell m} [f_{p\ell m}^{>}(\mathbf{r}) f_{p\ell m}^{<}(\mathbf{r}') \Theta(r - r') + f_{p\ell m}^{<}(\mathbf{r}) f_{p\ell m}^{>}(\mathbf{r}') \Theta(r' - r)] \tag{B.23}$$

into *scalar* spherical harmonics

$$f_{p\ell m}^{i}(\mathbf{r}) = \sqrt{K_{\ell m}} \sqrt{\frac{\ell(\ell+1)}{2\ell+1}} P_{\ell m}(\cos\theta) S_p(m\phi) \times \begin{cases} r^\ell, & i = <, \\ \dfrac{1}{r^{\ell+1}}, & i = > \end{cases} \tag{B.24}$$

motivates the calculation of a further set of integral identities. In particular, the radial integrals

$$\begin{aligned} \int_a^b j_\ell(kr) r^{\ell+2} \, \mathrm{d}r &= \frac{b^{\ell+2}}{k} j_{\ell+1}(kb) - \frac{a^{\ell+2}}{k} j_{\ell+1}(ka), \\ \int_a^b j_\ell(kr) r^{-\ell+1} \, \mathrm{d}r &= \frac{1}{k a^{\ell-1}} j_{\ell-1}(ka) - \frac{1}{k b^{\ell-1}} j_{\ell-1}(kb). \end{aligned} \tag{B.25}$$

are useful to define, as are the volume integrals

$$\int_{r<a} \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r}, k_{\boldsymbol{\beta} n}) \cdot \nabla f_{p'\ell' m'}^{<}(\mathbf{r}) \, \mathrm{d}^3\mathbf{r} = 4\pi \delta_{TL} (1 - \delta_{p1}\delta_{m0}) \delta_{pp'} \delta_{\ell\ell'} \delta_{mm'} \frac{a^{\ell+2}}{\sqrt{2\ell+1}} j_{\ell+1}(w_{\ell n}). \tag{B.26}$$

and

$$\int_0^{2\pi}\int_0^{\pi}\int_a^{b} \nabla f^{<}_{p\ell m}(\mathbf{r})\cdot\nabla f^{<}_{p'\ell'm'}(\mathbf{r}) r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = 4\pi\frac{\ell}{2\ell+1}\left(b^{2\ell+1}-a^{2\ell+1}\right)\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'},$$
$$\int_0^{2\pi}\int_0^{\pi}\int_a^{b} \nabla f^{>}_{p\ell m}(\mathbf{r})\cdot\nabla f^{>}_{p'\ell'm'}(\mathbf{r}) r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = -4\pi\frac{\ell+1}{2\ell+1}\left(b^{-2\ell-1}-a^{-2\ell-1}\right)\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}, \tag{B.27}$$
$$\int_0^{2\pi}\int_0^{\pi}\int_a^{b} \nabla f^{<}_{p\ell m}(\mathbf{r})\cdot\nabla f^{>}_{p'\ell'm'}(\mathbf{r}) r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = 0.$$

B.1 Useful Orthogonality Relations

A list of the useful and simple orthogonality relations useful to our calculations begins with the angular integral

$$\int_0^{2\pi} S_p(m\phi)S_{p'}(m'\phi)\,\mathrm{d}\phi = \pi(1+\delta_{p0}\delta_{m0}-\delta_{p1}\delta_{m0})\delta_{pp'}\delta_{mm'}, \tag{B.28}$$

wherein the convention is used that the replacements $p+2n\to 0$ for even p and $p+2n\to 1$ for odd p are taken automatically (here $n=0,1,2,\ldots$ is a nonnegative integer). Similarly, $p+2n+1\to 1$ for even p and $p+2n+1\to 0$ for odd p are implied.

Two more angular integrals, this time in θ, are of use:

$$\int_0^{\pi} P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\sin\theta\,\mathrm{d}\theta = \frac{2}{(2\ell+1)}\frac{(\ell+m)!}{(\ell-m)!}\delta_{\ell\ell'} \tag{B.29}$$

and

$$\begin{aligned}\int_0^{\pi}\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta}+m^2\frac{P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)}{\sin^2\theta}\right)\sin\theta\,\mathrm{d}\theta \\ = \int_0^{\pi}\ell(\ell+1)P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\sin\theta\,\mathrm{d}\theta \\ = \frac{2\ell(\ell+1)}{(2\ell+1)}\frac{(\ell+m)!}{(\ell-m)!}\delta_{\ell\ell'},\end{aligned} \tag{B.30}$$

for $\ell>0$, $0\le m\le\ell$. The second identity can be derived using the identity

$$\begin{aligned}&\frac{\partial}{\partial\theta}\left\{\sin\theta\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta}P_{\ell m}(\cos\theta)+\sin\theta\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}P_{\ell' m}(\cos\theta)\right\}\\ &=2\sin\theta\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta}+m^2\frac{P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)}{\sin^2\theta}\right)-2\ell(\ell+1)\sin\theta P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta),\end{aligned} \tag{B.31}$$

which can in turn be derived from the generalized Legendre equation defining the functions $P_{\ell m}(\cos\theta)$,

$$\frac{\partial^2}{\partial\theta^2}P_{\ell m}(\cos\theta)+\frac{\cos\theta}{\sin\theta}\frac{\partial}{\partial\theta}P_{\ell m}(\cos\theta)+\left(\ell(\ell+1)-\frac{m^2}{\sin^2\theta}\right)P_{\ell m}(\cos\theta)=0. \tag{B.32}$$

Further,

$$
\begin{aligned}
\int_0^\pi \left[\frac{P_{\ell m}(\cos\theta)}{\sin\theta}\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta} + \frac{P_{\ell' m}(\cos\theta)}{\sin\theta}\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}\right]\sin\theta\,\mathrm{d}\theta &= \int_0^\pi \frac{\partial}{\partial\theta}\left\{P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\right\}\mathrm{d}\theta \\
&= P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\big|_0^\pi \\
&= 2\delta_{m0}\delta_{\mathrm{par}(\ell),1},
\end{aligned}
\tag{B.33}
$$

where

$$
\mathrm{par}(n) = \begin{cases} 0, & |n| \text{ even}, \\ 1, & |n| \text{ odd} \end{cases}
\tag{B.34}
$$

is the parity function that returns 0 for even integers and 1 for odd integers.

B.2 Useful recursion relations

It is sometimes useful to know that

$$
\begin{aligned}
z_\ell(x) &= \frac{x}{2\ell+1}\left[z_{\ell-1}(x) + z_{\ell+1}(x)\right], \\
\frac{\partial z_\ell(x)}{\partial x} &= z_{\ell-1}(x) - \frac{\ell+1}{x}z_\ell(x), \\
\frac{\partial z_\ell(x)}{\partial x} &= -z_{\ell+1}(x) + \frac{\ell}{x}z_\ell(x),
\end{aligned}
\tag{B.35}
$$

and

$$
\frac{\partial\{x z_\ell(x)\}}{\partial x} = \frac{x}{2\ell+1}\left[(\ell+1)z_{\ell-1}(x) - \ell z_{\ell+1}(x)\right]
\tag{B.36}
$$

where $z_\ell(x)$ is any spherical Bessel or Hankel function. These identities can be combined to give

$$
\begin{aligned}
\ell(\ell+1)\frac{w_\ell(x_1)z_\ell(x_2)}{x_1x_2} &+ \frac{1}{x_1x_2}\frac{\partial\{x_1w_\ell(x_1)\}}{\partial x_1}\frac{\partial\{x_2z_\ell(x_2)\}}{\partial x_2} \\
&= \frac{1}{2\ell+1}\left[(\ell+1)w_{\ell-1}(x_1)z_{\ell-1}(x_2) + \ell w_{\ell+1}(x_1)z_{\ell+1}(x_2)\right]
\end{aligned}
\tag{B.37}
$$

and

$$
\frac{\partial z_\ell(x_1)}{\partial x_1}\frac{\partial w_\ell(x_2)}{\partial x_2} = \frac{\ell+1}{2\ell+1}z_{\ell+1}(x_1)w_{\ell+1}(x_2) + \frac{\ell}{2\ell+1}z_{\ell-1}(x_1)w_{\ell-1}(x_2) - \frac{\ell(\ell+1)}{x_1x_2}z_\ell(x_1)w_\ell(x_2)
\tag{B.38}
$$

where w_ℓ and z_ℓ are any two spherical Bessel or Hankel functions.

C Fano Diagonalization of an Oscillator Coupled to a Bath

Beginning with a Hamiltonian

$$
\hat{H} = \hat{H}_0 + \hat{H}_{\mathrm{int}}
\tag{C.1}
$$

where

$$
\begin{aligned}
\hat{H}_0 = \sum_{\boldsymbol{\alpha}}\int_0^\infty &\frac{1}{2}\hbar\Omega\left[\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\hat{a}_{\boldsymbol{\alpha}}(k,t) + \hat{a}_{\boldsymbol{\alpha}}(k,t)\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\right]\mathrm{d}k \\
&+ \sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty \frac{1}{2}\hbar\nu\left[\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu;t)\hat{b}_{\boldsymbol{\beta}}(k,\nu;t) + \hat{b}_{\boldsymbol{\beta}}(k,\nu;t)\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu;t)\right]\mathrm{d}\nu\,\mathrm{d}k
\end{aligned}
\tag{C.2}
$$

and

$$\hat{H}_{\text{int}} = \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar g(\nu) \left[\hat{a}_{\boldsymbol{\beta}}(k,t) + \hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)\right] \left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t) + \hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu;t)\right] \mathrm{d}\nu\, \mathrm{d}k, \tag{C.3}$$

we can define the boson operators $\hat{a}_{\boldsymbol{\alpha}}(k,t)$ and $\hat{b}_{\boldsymbol{\beta}}(k,\nu;t)$ by their commutation relations

$$\begin{aligned} \left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}^\dagger_{\boldsymbol{\alpha}'}(k',t)\right] &= \delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'}\delta(k-k'), \\ \left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t), \hat{b}^\dagger_{\boldsymbol{\beta}'}(k',\nu';t)\right] &= \delta_{\boldsymbol{\beta}\boldsymbol{\beta}'}\delta(k-k')\delta(\nu-\nu') \end{aligned} \tag{C.4}$$

and

$$\begin{aligned} \left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}_{\boldsymbol{\alpha}'}(k',t)\right] &= 0, \\ \left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t), \hat{b}_{\boldsymbol{\beta}'}(k',\nu';t)\right] &= 0, \\ \left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{b}_{\boldsymbol{\alpha}'}(k',\nu;t)\right] &= \left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{b}^\dagger_{\boldsymbol{\alpha}'}(k',\nu;t)\right] = 0. \end{aligned} \tag{C.5}$$

Note that commutation relations between the system's creation operators follow from the above via the Hermitian conjugate identity $[\hat{A},\hat{B}]^\dagger = -[\hat{A}^\dagger,\hat{B}^\dagger]$. Diagonalization of this system can be performed by defining new boson operators

$$\hat{B}_{\boldsymbol{\beta}}(k,\nu;t) = q(\nu)\hat{a}_{\boldsymbol{\beta}}(k,t) + s(\nu)\hat{a}^\dagger_{\boldsymbol{\beta}}(k,t) + \int_0^\infty \left[u(\nu,\nu')\hat{b}_{\boldsymbol{\beta}}(k,\nu';t) + v(\nu,\nu')\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\right] \mathrm{d}\nu' \tag{C.6}$$

that obey commutation relations

$$\begin{aligned} \left[\hat{B}_{\boldsymbol{\beta}}(k,\nu;t), \hat{B}^\dagger_{\boldsymbol{\beta}'}(k',\nu';t)\right] &= \delta_{\boldsymbol{\beta}\boldsymbol{\beta}'}\delta(k-k')\delta(\nu-\nu'), \\ \left[\hat{B}_{\boldsymbol{\beta}}(k,\nu;t), \hat{B}_{\boldsymbol{\beta}'}(k',\nu';t)\right] &= 0 \end{aligned} \tag{C.7}$$

and provide a new Hamiltonian

$$\hat{H} = \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\nu \hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu;t)\hat{B}_{\boldsymbol{\beta}}(k,\nu;t)\, \mathrm{d}\nu\, \mathrm{d}k. \tag{C.8}$$

The diagonalization process begins with the identification of the commutation relations

$$\begin{aligned} \left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{H}\right] = \sum_{\boldsymbol{\alpha}'} \int_0^\infty \frac{1}{2}\hbar\Omega \left(\left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}^\dagger_{\boldsymbol{\alpha}'}(k',t)\hat{a}_{\boldsymbol{\alpha}'}(k',t)\right] + \left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}_{\boldsymbol{\alpha}'}(k',t)\hat{a}^\dagger_{\boldsymbol{\alpha}'}(k',t)\right]\right) \mathrm{d}k' \\ + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar g(\nu) \left(\left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}_{\boldsymbol{\beta}}(k',t)\right] + \left[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}^\dagger_{\boldsymbol{\beta}}(k',t)\right]\right) \\ \times \left[\hat{b}_{\boldsymbol{\beta}}(k',\nu;t) + \hat{b}^\dagger_{\boldsymbol{\beta}}(k',\nu;t)\right] \mathrm{d}\nu\, \mathrm{d}k' \\ = \hbar\Omega\hat{a}_{\boldsymbol{\alpha}}(k,t) + \int_0^\infty \hbar g(\nu) \left[\hat{b}_{\boldsymbol{\alpha}}(k,\nu;t) + \hat{b}^\dagger_{\boldsymbol{\alpha}}(k,\nu;t)\right] \mathrm{d}\nu \end{aligned} \tag{C.9}$$

and

$$
\begin{aligned}
\left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t),\hat{H}\right] &= \sum_{\boldsymbol{\beta}'}\int_0^\infty\int_0^\infty \frac{1}{2}\hbar\nu'\left(\left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t),\hat{b}^\dagger_{\boldsymbol{\beta}'}(k',\nu';t)\hat{b}_{\boldsymbol{\beta}'}(k',\nu';t)\right]\right.\\
&\qquad\left.+\left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t),\hat{b}_{\boldsymbol{\beta}'}(k',\nu';t)\hat{b}^\dagger_{\boldsymbol{\beta}'}(k',\nu';t)\right]\right)\mathrm{d}\nu'\,\mathrm{d}k'\\
&\quad+\sum_{\boldsymbol{\beta}'}\int_0^\infty\int_0^\infty \hbar g(\nu')\left[\hat{a}_{\boldsymbol{\beta}'}(k',t)+\hat{a}^\dagger_{\boldsymbol{\beta}'}(k',t)\right]\\
&\qquad\times\left(\left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t),\hat{b}_{\boldsymbol{\beta}'}(k',\nu';t)\right]+\left[\hat{b}_{\boldsymbol{\beta}}(k,\nu;t),\hat{b}^\dagger_{\boldsymbol{\beta}'}(k',\nu';t)\right]\right)\mathrm{d}\nu'\,\mathrm{d}k'\\
&= \hbar\nu\hat{b}_{\boldsymbol{\beta}}(k,\nu;t)+\hbar g(\nu)\left[\hat{a}_{\boldsymbol{\beta}}(k,t)+\hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)\right].
\end{aligned}
\tag{C.10}
$$

With these and the identity $[\hat{A},\hat{B}]^\dagger = -[\hat{A}^\dagger,\hat{B}^\dagger]$, we can calculate the commutator of our hybridized annihilation operators with both the hybridized and unhybridized forms of the Hamtilonian. In detail,

$$
\begin{aligned}
\left[\hat{B}_{\boldsymbol{\beta}}(k,\nu;t),\hat{H}\right] &= \sum_{\boldsymbol{\beta}'}\int_0^\infty\int_0^\infty \hbar\nu'\left[\hat{B}_{\boldsymbol{\beta}}(k,\nu;t),\hat{B}^\dagger_{\boldsymbol{\beta}'}(k',\nu';t)\hat{B}_{\boldsymbol{\beta}'}(k',\nu';t)\right]\mathrm{d}\nu'\,\mathrm{d}k'\\
&= \hbar\nu\left(q(\nu)\hat{a}_{\boldsymbol{\beta}}(k,t)+s(\nu)\hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)+\int_0^\infty\left[u(\nu,\nu')\hat{b}_{\boldsymbol{\beta}}(k,\nu';t)+v(\nu,\nu')\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\right]\mathrm{d}\nu'\right).
\end{aligned}
\tag{C.11}
$$

and

$$
\begin{aligned}
\left[\hat{B}_{\boldsymbol{\beta}}(k,\nu;t),\hat{H}\right] &= \left[q(\nu)\hat{a}_{\boldsymbol{\beta}}(k,t)+s(\nu)\hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)+\int_0^\infty\left[u(\nu,\nu')\hat{b}_{\boldsymbol{\beta}}(k,\nu';t)+v(\nu,\nu')\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\right]\mathrm{d}\nu',\hat{H}\right]\\
&= q(\nu)\left(\hbar\Omega\hat{a}_{\boldsymbol{\alpha}}(k,t)+\int_0^\infty\hbar g(\nu')\left[\hat{b}_{\boldsymbol{\alpha}}(k,\nu';t)+\hat{b}^\dagger_{\boldsymbol{\alpha}}(k,\nu';t)\right]\mathrm{d}\nu'\right)\\
&\quad-s(\nu)\left(\hbar\Omega\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)+\int_0^\infty\hbar g(\nu')\left[\hat{b}_{\boldsymbol{\alpha}}(k,\nu';t)+\hat{b}^\dagger_{\boldsymbol{\alpha}}(k,\nu';t)\right]\mathrm{d}\nu'\right)\\
&\quad+\int_0^\infty u(\nu,\nu')\left(\hbar\nu'\hat{b}_{\boldsymbol{\beta}}(k,\nu';t)+\hbar g(\nu')\left[\hat{a}_{\boldsymbol{\beta}}(k,t)+\hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)\right]\right)\mathrm{d}\nu'\\
&\quad-\int_0^\infty v(\nu,\nu')\left(\hbar\nu'\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)+\hbar g(\nu')\left[\hat{a}_{\boldsymbol{\beta}}(k,t)+\hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)\right]\right)\mathrm{d}\nu'.
\end{aligned}
\tag{C.12}
$$

Noting that each operator is linearly independent from the others, we can see that Eqs. (C.11) and (C.12) form a system of four equations,

$$\begin{aligned}
\hbar\nu q(\nu) &= \hbar\Omega q(\nu) + \int_0^\infty \left[u(\nu,\nu') - v(\nu,\nu')\right]\hbar g(\nu')\mathrm{d}\nu', \\
\hbar\nu s(\nu) &= -\hbar\Omega s(\nu) + \int_0^\infty \left[u(\nu,\nu') - v(\nu,\nu')\right]\hbar g(\nu')\mathrm{d}\nu', \\
\hbar\nu u(\nu,\nu') &= \hbar g(\nu')[q(\nu) - s(\nu)] + \hbar\nu' u(\nu,\nu'), \\
\hbar\nu v(\nu,\nu') &= \hbar g(\nu')[q(\nu) - s(\nu)] - \hbar\nu' v(\nu,\nu').
\end{aligned} \tag{C.13}$$

From the first two equations of Eq. (C.13), we can see that

$$s(\nu) = \frac{\nu - \Omega}{\nu + \Omega} q(\nu). \tag{C.14}$$

This result can be plugged into the latter two lines to produce

$$\begin{aligned}
(\nu - \nu')u(\nu,\nu') &= \frac{2\Omega}{\nu + \Omega} g(\nu')q(\nu), \\
(\nu + \nu')v(\nu,\nu') &= \frac{2\Omega}{\nu + \Omega} g(\nu')q(\nu).
\end{aligned} \tag{C.15}$$

It will be useful later in our calculations to extend the definitions of our coefficients to negative bath frequencies, such that either equation above needs to be carefully handled where $\nu = \pm\nu'$, respectively. In these cases, the equations give no information about $u(\nu,\nu')$ or $v(\nu,\nu')$, such that we must use the formal solution $f(x) = PV\{1/x\} + C\delta(x)$ to the equation $xf(x) = 1$ in order to proceed. With PV indicating the Cauchy principal value and C a complex nonzero constant with respect to x, we can map this solution onto our current problem by letting ν be fixed such that

$$\begin{aligned}
u(\nu,\nu') &= \left[PV\left\{\frac{1}{\nu - \nu'}\right\} + [y(\nu) + r]\delta(\nu - \nu')\right] \frac{2\Omega}{\nu + \Omega} g(\nu')q(\nu), \\
v(\nu,\nu') &= \left[PV\left\{\frac{1}{\nu + \nu'}\right\} + [x(\nu) + p]\delta(\nu + \nu')\right] \frac{2\Omega}{\nu + \Omega} g(\nu')q(\nu).
\end{aligned} \tag{C.16}$$

Here, $x(\nu)$ and $y(\nu)$ are complex functions of ν and r and p are complex constants. Plugging these coefficients back into either of the first two lines of Eq. (C.13) produces the condition

$$y(\nu) + r = \frac{\nu^2 - \Omega^2}{2\Omega g^2(\nu)} - \frac{1}{g^2(\nu)} \int_0^\infty \left[PV\left\{\frac{1}{\nu - \nu'}\right\} - PV\left\{\frac{1}{\nu + \nu'}\right\}\right] g^2(\nu')\,\mathrm{d}\nu' \tag{C.17}$$

under the assumption that $g^2(-\nu) = -g^2(\nu)$. The simplest combination of constants that satisfies this condition is

$$\begin{aligned}
p &= 0, \\
x(\nu) &= 0, \\
y(\nu) + r &= \frac{\nu^2 - \Omega^2}{2\Omega g^2(\nu)} - \frac{1}{g^2(\nu)} \int_0^\infty \left[PV\left\{\frac{1}{\nu - \nu'}\right\} - PV\left\{\frac{1}{\nu + \nu'}\right\}\right] g^2(\nu')\,\mathrm{d}\nu' \\
&= \frac{\nu^2 - \Omega^2}{2\Omega g^2(\nu)} - \frac{1}{g^2(\nu)} \int_{-\infty}^\infty PV\left\{\frac{1}{\nu - \nu'}\right\} g^2(\nu')\,\mathrm{d}\nu',
\end{aligned} \tag{C.18}$$

such that we can now represent $u(\nu,\nu')$ and $v(\nu,\nu')$ as products of known quantities times $q(\nu)$.

We are now left with the task of calculating $q(\nu)$. To do so, we can use the commutation relation

$$\begin{aligned}\left[\hat{B}_{\boldsymbol{\beta}}(k,\nu;t),\hat{B}^{\dagger}_{\boldsymbol{\beta}'}(k',\nu';t)\right] &= \Bigg[\Bigg(q(\nu)\hat{a}_{\boldsymbol{\beta}}(k,t)+s(\nu)\hat{a}^{\dagger}_{\boldsymbol{\beta}}(k,t)+\int_0^\infty u(\nu,\omega)\hat{b}_{\boldsymbol{\beta}}(k,\omega;t)\,\mathrm{d}\omega \\ &\quad +\int_0^\infty v(\nu,\omega)\hat{b}^{\dagger}_{\boldsymbol{\beta}}(k,\omega;t)\,\mathrm{d}\omega\Bigg),\Bigg(q^*(\nu')\hat{a}^{\dagger}_{\boldsymbol{\beta}'}(k',\nu')+s^*(\nu')\hat{a}_{\boldsymbol{\beta}'}(k',\nu') \\ &\quad +\int_0^\infty\left[u^*(\nu',\omega')\hat{b}^{\dagger}_{\boldsymbol{\beta}'}(k',\omega';t)+v^*(\nu',\omega')\hat{b}_{\boldsymbol{\beta}'}(k',\omega';t)\right]\,\mathrm{d}\omega'\Bigg)\Bigg] \\ &= [q(\nu)q^*(\nu')-s(\nu)s^*(\nu')]\delta(k-k')\delta_{\boldsymbol{\beta}\boldsymbol{\beta}'} \\ &\quad +\delta(k-k')\delta_{\boldsymbol{\beta}\boldsymbol{\beta}'}\int_0^\infty[u(\nu,\omega)u^*(\nu',\omega)-v(\nu,\omega)v^*(\nu',\omega)]\,\mathrm{d}\omega\end{aligned} \tag{C.19}$$

Since we have demanded that

$$\left[\hat{B}_{\boldsymbol{\beta}}(k,\nu;t),\hat{B}^{\dagger}_{\boldsymbol{\beta}'}(k',\nu';t)\right]=\delta(k-k')\delta(\nu-\nu')\delta_{\boldsymbol{\beta}\boldsymbol{\beta}'}, \tag{C.20}$$

we can say

$$\begin{aligned}\delta(\nu-\nu') &= q(\nu)q^*(\nu')-s(\nu)s^*(\nu')+\int_0^\infty[u(\nu,\omega)u^*(\nu',\omega)-v(\nu,\omega)v^*(\nu',\omega)]\,\mathrm{d}\omega \\ &= q(\nu)q^*(\nu')\frac{4\Omega^2}{(\nu+\Omega)(\nu'+\Omega)}\Bigg(\frac{\nu+\nu'}{2\Omega}+g^2(\nu)[y(\nu)+r][y(\nu')+r]^*\delta(\nu-\nu') \\ &\quad +\int_{-\infty}^\infty PV\left\{\frac{1}{(\nu-\omega)(\nu'-\omega)}\right\}g^2(\omega)\,\mathrm{d}\omega \\ &\quad +\left[PV\left\{\frac{1}{\nu'-\nu}\right\}g^2(\nu)[y(\nu)+r]+PV\left\{\frac{1}{\nu-\nu'}\right\}g^2(\nu')[y(\nu')+r]^*\right]\Bigg)\end{aligned} \tag{C.21}$$

To simplify, we can note that

$$PV\left\{\frac{1}{x}\right\}=\lim_{\epsilon\to 0}\frac{x}{x^2+\epsilon^2} \tag{C.22}$$

such that $PV\{-1/x\}=-PV\{1/x\}$. Further, noting that

$$\begin{aligned}\lim_{\epsilon\to 0}\frac{\epsilon}{x^2+\epsilon^2}&=\pi\delta(x), \\ \lim_{\epsilon\to 0}\frac{x^2}{x^2+\epsilon^2}&=1,\end{aligned} \tag{C.23}$$

we can see that

$$\begin{aligned}&\left[PV\left\{\frac{1}{\nu'-\nu}\right\}g^2(\nu)[y(\nu)+r]+PV\left\{\frac{1}{\nu-\nu'}\right\}g^2(\nu')[y(\nu')+r]^*\right] \\ &=-\frac{\nu+\nu'}{2\Omega}-\int_{-\infty}^\infty PV\left\{\frac{1}{(\nu-\omega)(\nu'-\omega)}\right\}g^2(\omega)\,\mathrm{d}\omega+\pi^2\delta(\nu-\nu')g^2(\nu).\end{aligned} \tag{C.24}$$

Plugging this result back into Eq. (C.21), we find that

$$\delta(\nu-\nu')=q(\nu)q^*(\nu')\frac{4\Omega^2}{(\nu+\Omega)(\nu'+\Omega)}\left[g^2(\nu')[y(\nu')+r][y(\nu)+r]^*\delta(\nu-\nu')+\pi^2\delta(\nu-\nu')g^2(\nu)\right]. \tag{C.25}$$

This implies that

$$1 = |q(\nu)|^2(|y(\nu) + r|^2 + \pi^2)\frac{4\Omega^2 g^2(\nu)}{(\nu+\Omega)^2}. \tag{C.26}$$

The most advantageous value of r will turn out to be $r = 0$ such that

$$|y(\nu) + r|^2 + \pi^2 = |y(\nu) - \mathrm{i}\pi|^2 \tag{C.27}$$

and

$$q(\nu) = \frac{\nu + \Omega}{2\Omega g(\nu)[y(\nu) - \mathrm{i}\pi]}. \tag{C.28}$$

Further, with

$$PV\left\{\frac{1}{x}\right\} = \lim_{\epsilon\to 0}\frac{1}{x - \mathrm{i}\epsilon} - \mathrm{i}\pi\delta(x), \tag{C.29}$$

we can see that

$$\begin{aligned} y(\nu) - \mathrm{i}\pi &= \frac{\nu^2 - \Omega^2}{2\Omega g^2(\nu)} - \frac{1}{g^2(\nu)}\int_{-\infty}^{\infty}\left[\lim_{\epsilon\to 0}\frac{1}{\nu - \nu' - \mathrm{i}\epsilon} - \mathrm{i}\pi\delta(\nu - \nu')\right] g^2(\nu')\,\mathrm{d}\nu' - \mathrm{i}\pi \\ &= \frac{\nu^2 - \Omega^2}{2\Omega g^2(\nu)} - \frac{1}{g^2(\nu)}\int_{-\infty}^{\infty}\lim_{\epsilon\to 0}\frac{1}{\nu - \nu' - \mathrm{i}\epsilon} g^2(\nu')\,\mathrm{d}\nu'. \end{aligned} \tag{C.30}$$

We can then define a new function

$$z(\nu) = 1 + \frac{2}{\Omega}\int_{-\infty}^{\infty}\frac{g^2(\omega)}{\nu - \omega - \mathrm{i}\epsilon}\mathrm{d}\omega \tag{C.31}$$

such that

$$y(\nu) - \mathrm{i}\pi = \frac{\nu^2 - \Omega^2 z(\nu)}{2\Omega g^2(\nu)}, \tag{C.32}$$

which allows us to finally define our expansions coefficients:

$$\begin{aligned} q(\nu) &= \frac{(\nu + \Omega)g(\nu)}{\nu^2 - \Omega^2 z(\nu)}, \\ s(\nu) &= \frac{(\nu - \Omega)g(\nu)}{\nu^2 - \Omega^2 z(\nu)}, \\ u(\nu,\nu') &= \delta(\nu - \nu') + \frac{2\Omega g(\nu)}{\nu^2 - \Omega^2 z(\nu)}\frac{g(\nu')}{\nu - \nu' - \mathrm{i}\epsilon}, \\ v(\nu,\nu') &= \frac{2\Omega g(\nu)}{\nu^2 - \Omega^2 z(\nu)} PV\left\{\frac{g(\nu')}{\nu + \nu'}\right\}. \end{aligned} \tag{C.33}$$

Finally, we also need to be able to represent our unhybridized coordinates in terms of our hybridized ones. To do this, we can see that

$$\begin{aligned} &\int_0^{\infty}\left[q^*(\nu)\hat{B}_{\boldsymbol{\beta}}(k,\nu;t) - s(\nu)\hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu;t)\right]\mathrm{d}\nu = \hat{a}_{\boldsymbol{\beta}}(k,t)\int_0^{\infty}\left(|q(\nu)|^2 - |s(\nu)|^2\right)\mathrm{d}\nu \\ &\quad + \hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)\int_0^{\infty}[q^*(\nu)s(\nu) - s(\nu)q^*(\nu)]\,\mathrm{d}\nu + \int_0^{\infty}\int_0^{\infty}[q^*(\nu)u(\nu,\nu') - s(\nu)v^*(\nu,\nu')]\,\hat{b}_{\boldsymbol{\beta}}(k,\nu';t)\,\mathrm{d}\nu'\,\mathrm{d}\nu \\ &\quad + \int_0^{\infty}\int_0^{\infty}[q^*(\nu)v(\nu,\nu') - s(\nu)u^*(\nu,\nu')]\,\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\,\mathrm{d}\nu'\,\mathrm{d}\nu, \end{aligned} \tag{C.34}$$

wherein the terms under integration must go to zero independently for the linear combination of hybridized coordinates to return an expression solely dependent on the operator $\hat{a}_{\boldsymbol{\beta}}(k,t)$. We can first immediately see that $q^*(\nu)s(\nu) - s(\nu)q^*(\nu) = 0$. Second,

$$\begin{aligned}\int_0^\infty \left(|q(\nu)|^2 - |s(\nu)|^2\right) \mathrm{d}\nu &= \int_0^\infty \left[(\nu+\Omega)^2 - (\nu-\Omega)^2\right] \frac{g^2(\nu)}{[\nu^2 - \Omega^2 z(\nu)]^2} \\ &= \int_0^\infty \frac{4\nu\Omega g^2(\nu)}{|\nu^2 - \Omega^2 z(\nu)|^2}\,\mathrm{d}\nu.\end{aligned} \tag{C.35}$$

From here, we can note that integrals of the type in Eq. (C.35) are often done via contour integration using a closed contour comprised of the real line and a semicircle of infinite radius that lies in the complex plane. Before we can construct such a contour, however, it is useful to first analyze the pole structure of the function $\nu^2 - \Omega^2 z(\nu)$. Letting $\nu = a + \mathrm{i}b$, we can see that

$$\begin{aligned}(a+\mathrm{i}b)^2 - \Omega^2 z(a+\mathrm{i}b) &= a^2 + 2\mathrm{i}ab - b^2 - \Omega^2 - 2\Omega \int_{-\infty}^{\infty} \frac{g^2(\omega)}{a + \mathrm{i}b - \omega - \mathrm{i}\epsilon}\,\mathrm{d}\omega \\ &= a^2 - b^2 - \Omega^2 - 2\Omega \int_{-\infty}^{\infty} \frac{g^2(\omega)(a-\omega)}{(a-\omega)^2 + (b-\epsilon)^2}\,\mathrm{d}\omega \\ &\quad + \mathrm{i}(b-\epsilon)\left(2a + 2\Omega \int_{-\infty}^{\infty} \frac{g^2(\omega)}{(a-\omega)^2 + (b-\epsilon)^2}\,\mathrm{d}\omega\right).\end{aligned} \tag{C.36}$$

There are two cases to consider here:

- First, we can see that the imaginary part of $\nu^2 - \Omega^2 z(\nu)$ is zero when $b = \epsilon$. In this case, the function has a zero if there exists a value a such that

$$a^2 - \Omega^2 - \epsilon^2 - 2\Omega \int_{-\infty}^{\infty} \frac{g^2(\omega)}{a-\omega}\,\mathrm{d}\omega = 0. \tag{C.37}$$

  For functions $g^2(\omega)$ and frequencies a defined such that $g^2(a) \neq 0$ for any a, the integral above is divergent and $\nu^2 - \Omega^2 z(\nu)$ has no zeros with $b = \epsilon$. Otherwise, there can exist any number of zeros, depending on the functional form of $g^2(\omega)$. An important example of the latter case is that where $g^2(\omega)$ is an odd function such that $g^2(0) = 0$. Under this assumption, a candidate zero exists at $a = 0$, such that $a + \mathrm{i}b = \mathrm{i}\epsilon$ is a zero of $\nu^2 - \Omega^2 z(\nu)$ if

$$\frac{\Omega^2 + \epsilon^2}{\Omega} = \int_{-\infty}^{\infty} \frac{g^2(\omega)}{\omega}\,\mathrm{d}\omega. \tag{C.38}$$

- Second, in the case that $b \neq \epsilon$, we can see that the imaginary part of the RHS of Eq. (C.36) can be zero if the quantity inside the parentheses is zero. We will assume $b \neq \epsilon$ in the following logic. Because $g(\omega) \sim \omega\tilde{v}^2(\omega)$ and $\tilde{v}^2(\omega)$ is a nonnegative even function, the integrand of this term is always positive for positive ω and negative for negative ω. Moreover, because the area under $1/[(a-\omega)^2 + (b-\epsilon)^2]$ is larger on the same side of the origin as the sign of a, the integral is always positive for positive a and negative for negative a. The first term in the parentheses, $2a$, has the same signs in the same regions of the real line. Therefore, the imaginary part of $\nu^2 - \Omega^2 z(\nu)$ can only be zero when $a = \mathrm{Re}\{\nu\} = 0$, such that $\nu^2 - \Omega^2 z(\nu)$ can only have zeros along the imaginary axis. Further noting that $\int_{-\infty}^{\infty} g^2(\omega)/[\omega^2 + (b-\epsilon)^2]\,\mathrm{d}\omega \to 0$ as $\epsilon \to 0$ due to the odd and even parities of the numerator and denominator, respectively, we can see that, in the limit $a \to 0$, our function becomes

$$(\mathrm{i}b)^2 - \Omega^2 z(\mathrm{i}b) = -(b^2 + \Omega^2) + 2\Omega \int_{-\infty}^{\infty} \frac{\omega g^2(\omega)}{\omega^2 + (b-\epsilon)^2}\, \mathrm{d}\omega. \tag{C.39}$$

Because the first term on the RHS ($-[b^2 + \Omega^2]$) is strictly negative and the second is strictly positive, our function always has zeros at $b = \pm b_0$ except in the case where the two terms on the RHS never have equal magnitudes at any b. Because b^2 is strictly increasing with $|b|$ and $1/(\omega^2 + [b-\epsilon]^2)$ is strictly decreasing, the second term on the RHS can never be greater than the first for all b. Therefore, the only condition that guarantees the nonexistence of zeros is the condition that the second term on the RHS is always *less* than the first, i.e.

$$2\Omega \int_{-\infty}^{\infty} \frac{\omega g^2(\omega)}{\omega^2 + (b-\epsilon)^2}\, \mathrm{d}\omega < b^2 + \Omega^2 \tag{C.40}$$

for all b. We can guarantee this condition by guaranteeing it is satisfied when the LHS is maximized and the RHS is minimized, which occurs at $b = 0$. Therefore, we can guarantee that $\nu^2 - \Omega^2 z(\nu)$ has no zeros in the complex plane (other than $b = \epsilon$) only when

$$2\int_{-\infty}^{\infty} \frac{\omega g^2(\omega)}{\omega^2 + \epsilon^2}\, \mathrm{d}\omega = 4\int_0^{\infty} PV\left\{\frac{g^2(\omega)}{\omega}\right\} \mathrm{d}\omega < \Omega. \tag{C.41}$$

Assuming that the second condition on $g(\nu)$ is satisfied, using contour integration to simplify Eq. (C.35) becomes much simpler. In more detail, since a contour comprised of the real line and a great semicircle in the lower complex half-plane contains no poles, the residue theorem tells us that

$$\int_{-\infty}^{\infty} \frac{4\Omega\nu g^2(\nu)}{|\nu^2 - \Omega^2 z(\nu)|^2}\, \mathrm{d}\nu = -\int_{C_R^-} \frac{4\Omega\nu g^2(\nu)}{|\nu^2 - \Omega^2 z(\nu)|^2}\, \mathrm{d}\nu \tag{C.42}$$

with C_R^- symbolizing the great circle portion of the contour. For our hybridization scheme to be consistent, we require the integral of Eq. (C.35) to be equal to one. Conveniently, we can see that the method of partial fractions gives

$$\frac{\mathrm{i}}{[x-(a+\mathrm{i}b)][x-(a-\mathrm{i}b)]} = -\frac{1}{2b}\frac{1}{x-(a+\mathrm{i}b)} + \frac{\mathrm{i}}{2b}\frac{1}{x-(a-\mathrm{i}b)} \tag{C.43}$$

such that

$$\begin{aligned}\frac{1}{|\nu^2 - \Omega^2 z(\nu)|^2} &= \frac{1}{[\nu^2 - \Omega^2 z(\nu)][\nu^2 - \Omega^2 z^*(\nu)]} \\ &= -\frac{\mathrm{i}}{4\pi\Omega g^2(\nu)}\frac{1}{\nu^2 - \Omega^2 z(\nu)} + \frac{\mathrm{i}}{4\pi\Omega g^2(\nu)}\frac{1}{\nu^2 - \Omega^2 z^*(\nu)}.\end{aligned} \tag{C.44}$$

Therefore, using the fact that $z(-\nu) = z^*(\nu)$, as is clear from the expansion

$$\begin{aligned} z(\nu) &= 1 + \frac{2}{\Omega}\int_{-\infty}^{\infty} \frac{g^2(\omega)}{\nu - \omega - \mathrm{i}\epsilon}\mathrm{d}\omega \\ &= 1 + \frac{2}{\Omega}\left[\int_0^{\infty} \frac{g^2(\omega)}{\nu - \omega - \mathrm{i}\epsilon}\, \mathrm{d}\omega + \int_0^{\infty} \frac{g^2(\omega)}{-\nu - \omega + \mathrm{i}\epsilon}\, \mathrm{d}\omega\right],\end{aligned} \tag{C.45}$$

we can say

$$
\begin{aligned}
\int_0^\infty \frac{4\Omega\nu g^2(\nu)}{|\nu^2-\Omega^2 z(\nu)|^2}\,\mathrm{d}\nu &= -\frac{\mathrm{i}}{\pi}\int_0^\infty \left(\frac{\nu}{\nu^2-\Omega^2 z(\nu)} - \frac{\nu}{\nu^2-\Omega^2 z^*(\nu)}\right)\mathrm{d}\nu \\
&= \lim_{R\to\infty}\frac{\mathrm{i}}{\pi}\int_{-\pi}^{0}\frac{R\mathrm{e}^{\mathrm{i}\theta}}{R^2\mathrm{e}^{2\mathrm{i}\theta}-\Omega^2 z(R\mathrm{e}^{\mathrm{i}\theta})}R\mathrm{i}\mathrm{e}^{\mathrm{i}\theta}\,\mathrm{d}\theta \\
&= 1,
\end{aligned}
\tag{C.46}
$$

wherein we have noticed that $|z[R\exp(\mathrm{i}\theta)]| \ll R^2$ for large R. Therefore, as long as Eq. (C.41) is satisfied, we have

$$
\int_0^\infty \left(|q_\perp(\nu)|^2 - |s_\perp(\nu)|^2\right)\mathrm{d}\nu = 1. \tag{C.47}
$$

Using similar logic, we can see that

$$
\begin{aligned}
\int_0^\infty [q^*(\nu)u(\nu,\nu') - s(\nu)v^*(\nu,\nu')]\,\mathrm{d}\nu &= \int_0^\infty \left(\left[\delta(\nu-\nu') + \frac{2\Omega g(\nu)}{\nu^2-\Omega^2 z(\nu)}\frac{g(\nu')}{\nu-\nu'-\mathrm{i}\epsilon}\right]\frac{(\nu+\Omega)g(\nu)}{\nu^2-\Omega^2 z^*(\nu)}\right. \\
&\quad \left. -\frac{(\nu-\Omega)g(\nu)}{\nu^2-\Omega^2 z(\nu)}\frac{2\Omega g(\nu)}{\nu^2-\Omega^2 z^*(\nu)}PV\left\{\frac{g(\nu')}{\nu+\nu'}\right\}\right)\mathrm{d}\nu \\
&= \frac{(\nu'+\Omega)g(\nu')}{\nu'^2-\Omega^2 z^*(\nu')} - 0 - 0 + 0 - \frac{(\nu'+\Omega+\mathrm{i}\epsilon)g(\nu')}{(\nu'+\mathrm{i}\epsilon)^2-\Omega^2 z^*(\nu'+\mathrm{i}\epsilon)} \\
&= 0
\end{aligned}
\tag{C.48}
$$

and

$$
\begin{aligned}
\int_0^\infty [q^*(\nu)v(\nu,\nu') - s(\nu)u^*(\nu,\nu')]\,\mathrm{d}\nu &= \int_0^\infty \left(\frac{(\nu+\Omega)g(\nu)}{\nu^2-\Omega^2 z^*(\nu)}\frac{2\Omega g(\nu)}{\nu^2-\Omega^2 z(\nu)}PV\left\{\frac{g(\nu')}{\nu+\nu'}\right\}\right. \\
&\quad \left. -\frac{(\nu-\Omega)g(\nu)}{\nu^2-\Omega^2 z(\nu)}\left[\delta(\nu-\nu') + \frac{2\Omega g(\nu)}{\nu^2-\Omega^2 z^*(\nu)}\frac{g(\nu')}{\nu-\nu'+\mathrm{i}\epsilon}\right]\right)\mathrm{d}\nu \\
&= -\frac{(\nu'-\Omega)g(\nu')}{\nu'^2-\Omega^2 z^*(\nu')} + \int_0^\infty \frac{2\Omega g^2(\nu)g(\nu')}{|\nu^2-\Omega^2 z(\nu)|^2}\left(\frac{(\nu+\Omega)}{\nu+\nu'} - \frac{(\nu-\Omega)}{\nu-\nu'+\mathrm{i}\epsilon}\right)\mathrm{d}\nu \\
&= 0
\end{aligned}
\tag{C.49}
$$

such that

$$
\int_0^\infty \left[q^*(\nu)\hat{B}_{\boldsymbol{\beta}}(k,\nu;t) - s(\nu)\hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu;t)\right]\mathrm{d}\nu = \hat{a}_{\boldsymbol{\beta}}(k,t). \tag{C.50}
$$

We can repeat this process to find the representation of the bath operator $\hat{b}_{\boldsymbol{\beta}}(k,\nu;t)$ in terms of the hybrid operators. Beginning with

$$
\begin{aligned}
\int_0^\infty \Big[u^*(\nu',\nu)\hat{B}_{\boldsymbol{\beta}}(k,\nu';t) - v(\nu',\nu)\hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\Big]\,\mathrm{d}\nu' &= \hat{a}_{\boldsymbol{\beta}}(k,t)\int_0^\infty \left[q(\nu')u^*(\nu',\nu) - s^*(\nu')v(\nu',\nu)\right]\mathrm{d}\nu' \\
&+ \hat{a}^\dagger_{\boldsymbol{\beta}}(k,t)\int_0^\infty \left[s(\nu')u^*(\nu',\nu) - q^*(\nu')v(\nu',\nu)\right]\mathrm{d}\nu' \\
&+ \int_0^\infty\int_0^\infty \left[u^*(\nu',\nu)u(\nu',\omega) - v(\nu',\nu)v^*(\nu',\omega)\right]\hat{b}_{\boldsymbol{\beta}}(k,\omega;t)\,\mathrm{d}\nu'\,\mathrm{d}\omega \\
&+ \int_0^\infty\int_0^\infty \left[u^*(\nu',\nu)v(\nu',\omega) - v(\nu',\nu)u^*(\nu',\omega)\right]\hat{b}^\dagger_{\boldsymbol{\beta}}(k,\omega;t)\,\mathrm{d}\nu'\,\mathrm{d}\omega,
\end{aligned}
\tag{C.51}
$$

we can see that the integral prefactors of the first two terms on the RHS above are complex conjugates or reverses of integrals we already know to be zero and are thus themselves zero. The fourth term on the RHS is also zero, as is shown by

$$
\begin{aligned}
\int_0^\infty \left[u^*(\nu',\nu)v(\nu',\omega) - v(\nu',\nu)u^*(\nu',\omega)\right]\mathrm{d}\nu' &= \int_0^\infty \Bigg[\Bigg(\delta(\nu'-\nu) + \frac{2\Omega g(\nu')}{\nu'^2-\Omega^2 z^*(\nu')}\frac{g(\nu)}{\nu'-\nu+\mathrm{i}\epsilon}\Bigg) \\
&\quad\times \frac{2\Omega g(\nu')}{\nu'^2-\Omega^2 z(\nu')} PV\left\{\frac{g(\omega)}{\nu'+\omega}\right\} - \frac{2\Omega g(\nu')}{\nu'^2-\Omega^2 z(\nu')} PV\left\{\frac{g(\nu)}{\nu'+\nu}\right\}\Bigg(\delta(\nu'-\omega) \\
&\quad+ \frac{2\Omega g(\nu')}{\nu'^2-\Omega^2 z^*(\nu')}\frac{g(\omega)}{\nu'-\omega+\mathrm{i}\epsilon}\Bigg)\Bigg]\,\mathrm{d}\nu' \\
&= \frac{2\Omega g(\nu)}{\nu^2-\Omega^2 z(\nu)} PV\left\{\frac{g(\omega)}{\nu+\omega}\right\} + \int_0^\infty \frac{4\Omega^2 g^2(\nu')}{|\nu'^2-\Omega^2 z(\nu')|^2}\frac{g(\nu)}{\nu'-\nu+\mathrm{i}\epsilon} PV\left\{\frac{g(\omega)}{\nu'+\omega}\right\}\mathrm{d}\nu' \\
&\quad- \frac{2\Omega g(\omega)}{\omega^2-\Omega^2 z(\omega)} PV\left\{\frac{g(\nu)}{\omega+\nu}\right\} - \int_0^\infty \frac{4\Omega^2 g^2(\nu')}{|\nu'^2-\Omega^2 z(\nu')|^2}\frac{g(\omega)}{\nu'-\omega+\mathrm{i}\epsilon} PV\left\{\frac{g(\nu)}{\nu'+\nu}\right\}\mathrm{d}\nu' \\
&= 2\Omega g(\omega)g(\nu)\frac{\omega+\nu}{(\omega+\nu)^2+\epsilon^2}\left[\frac{1}{\nu^2-\omega^2 z(\nu)} - \frac{1}{\omega^2-\Omega^2 z(\omega)}\right] \\
&\quad+ 2\Omega g(\omega)g(\nu)\frac{1}{\omega+\nu}\left[\frac{1}{\omega^2-\Omega^2 z(\omega)} - \frac{1}{\nu^2-\Omega^2 z(\nu)}\right] \\
&= 0.
\end{aligned}
\tag{C.52}
$$

Finally, the integral prefactor of the third term gives

$$
\begin{aligned}
\int_0^\infty \left[u^*(\nu',\omega)u(\nu',\omega) - v(\nu',\nu)v^*(\nu',\omega)\right]\mathrm{d}\nu' = \int_0^\infty \Bigg[&\left(\delta(\nu'-\nu) + \frac{2\Omega g(\nu')}{\nu'^2 - \Omega^2 z^*(\nu')}\frac{g(\nu)}{\nu'-\nu+\mathrm{i}\epsilon}\right) \\
\times\left(\delta(\nu'-\omega) + \frac{2\Omega g(\nu')}{\nu'^2 - \Omega^2 z(\nu')}\frac{g(\omega)}{\nu'-\omega-\mathrm{i}\epsilon}\right) &- \frac{2\Omega g(\nu')}{\nu'^2-\Omega^2 z(\nu')} PV\left\{\frac{g(\omega)}{\nu'+\omega}\right\} \\
\times \frac{2\Omega g(\nu')}{\nu'^2-\Omega^2 z^*(\nu')} PV\left\{\frac{g(\nu)}{\nu'+\nu}\right\}\Bigg]\,\mathrm{d}\nu' & \\
= \delta(\nu-\omega) + \frac{2\Omega g(\omega)g(\nu)}{\nu-\omega-\mathrm{i}\epsilon}\left[\frac{1}{\nu^2-\Omega^2 z(\nu)} - \frac{1}{\omega^2-\Omega^2 z^*(\omega)}\right] & \\
+ \int_0^\infty \frac{4\Omega^2 g^2(\nu')}{|\nu'^2-\Omega^2 z(\nu')|^2}\frac{g(\nu)g(\omega)}{(\nu'-\nu+\mathrm{i}\epsilon)(\nu'-\omega-\mathrm{i}\epsilon)}\,\mathrm{d}\nu' & \\
+ \int_{-\infty}^0 \frac{4\Omega^2 g^2(\nu')}{|\nu'^2-\Omega^2 z(\nu')|^2}\frac{g(\nu)g(\omega)}{(\nu'-\nu+\mathrm{i}\epsilon)(\nu'-\omega-\mathrm{i}\epsilon)}\,\mathrm{d}\nu' & \\
= \delta(\nu-\omega) + \frac{2\Omega g(\omega)g(\nu)}{\nu-\omega-\mathrm{i}\epsilon}\left[\frac{1}{\nu^2-\Omega^2 z(\nu)} - \frac{1}{\omega^2-\Omega^2 z^*(\omega)}\right] & \\
+ \int_{-\infty}^\infty \frac{4\Omega^2 g^2(\nu')}{|\nu'^2-\Omega^2 z(\nu')|^2}\frac{g(\nu)g(\omega)}{(\nu'-\nu+\mathrm{i}\epsilon)(\nu'-\omega-\mathrm{i}\epsilon)}\,\mathrm{d}\nu' & \\
= \delta(\nu-\omega) + \frac{2\Omega g(\omega)g(\nu)}{\nu-\omega-\mathrm{i}\epsilon}\left[\frac{1}{\nu^2-\Omega^2 z(\nu)} - \frac{1}{\omega^2-\Omega^2 z^*(\omega)}\right] & \\
+ \int_{-\infty}^\infty 4\Omega^2 g^2(\nu')\left(-\frac{\mathrm{i}}{4\pi\Omega g^2(\nu')}\frac{1}{\nu'^2-\Omega^2 z(\nu')} + \frac{\mathrm{i}}{4\pi\Omega g^2(\nu')}\frac{1}{\nu'^2-\Omega^2 z^*(\nu')}\right) & \\
\times g(\nu)g(\omega)\left(\frac{1}{\nu-\omega-2\mathrm{i}\epsilon}\frac{1}{\nu'-\nu+\mathrm{i}\epsilon} - \frac{1}{\nu-\omega-2\mathrm{i}\epsilon}\frac{1}{\nu'-\omega-\mathrm{i}\epsilon}\right)\mathrm{d}\nu' & \\
= \delta(\nu-\omega) + \frac{2\Omega g(\omega)g(\nu)}{\nu-\omega-\mathrm{i}\epsilon}\left[\frac{1}{\nu^2-\Omega^2 z(\nu)} - \frac{1}{\omega^2-\Omega^2 z^*(\omega)}\right] & \\
- \frac{\mathrm{i}\Omega}{\pi} g(\nu)g(\omega)\frac{1}{\nu-\omega-2\mathrm{i}\epsilon}\int_{-\infty}^\infty \left(\frac{1}{\nu'^2-\Omega^2 z(\nu')} - \frac{1}{\nu'^2-\Omega^2 z^*(\nu')}\right)\left(\frac{1}{\nu'-\nu+\mathrm{i}\epsilon} - \frac{1}{\nu'-\omega-\mathrm{i}\epsilon}\right)\mathrm{d}\nu' & \\
= \delta(\nu-\omega) + \frac{2\Omega g(\omega)g(\nu)}{\nu-\omega-\mathrm{i}\epsilon}\left[\frac{1}{\nu^2-\Omega^2 z(\nu)} - \frac{1}{\omega^2-\Omega^2 z^*(\omega)}\right] & \\
- \frac{2\Omega g(\omega)g(\nu)}{\nu-\omega-2\mathrm{i}\epsilon}\left[\frac{1}{\nu^2-\Omega^2 z(\nu)} - \frac{1}{\omega^2-\Omega^2 z^*(\omega)}\right] & \\
= \delta(\nu-\omega) &
\end{aligned}
\tag{C.53}
$$

such that

$$
\begin{aligned}
\int_0^\infty \left[u^*(\nu',\nu)\hat{B}_{\boldsymbol{\beta}}(k,\nu';t) - v(\nu',\nu)\hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\right]\mathrm{d}\nu' &= \int_0^\infty \delta(\nu-\omega)\hat{b}_{\boldsymbol{\beta}}(k,\omega;t)\,\mathrm{d}\omega \\
&= \hat{b}_{\boldsymbol{\beta}}(k,\nu;t).
\end{aligned}
\tag{C.54}
$$

C.1 Hybridized Operator Expansion Coefficients

The first hybridization step from the main text produces expansion coefficients

$$\begin{aligned} q_{\perp,\parallel}(\nu) &= \frac{\nu+\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega_{\perp,\parallel}^2 z_{\perp,\parallel}(\nu)}, \\ s_{\perp,\parallel}(\nu) &= \frac{\nu-\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega_{\perp,\parallel}^2 z_{\perp,\parallel}(\nu)}, \\ u_{\perp,\parallel}(\nu,\nu') &= \delta(\nu-\nu') + \frac{\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega_{\perp,\parallel}^2 z_{\perp,\parallel}(\nu)}\frac{V_{\perp,\parallel}(\nu')}{\nu-\nu'-\mathrm{i}\epsilon}, \\ v_{\perp,\parallel}(\nu,\nu') &= \frac{\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega_{\perp,\parallel}^2 z_{\perp,\parallel}(\nu)} PV\left\{\frac{V_{\perp,\parallel}(\nu)}{\nu+\nu'}\right\}, \end{aligned} \tag{C.55}$$

with the substitutions $g(\nu)\to V_{\perp,\parallel}(\nu)/2$, $\Omega\to\Omega_{\perp,\parallel}$, and

$$z(\nu)\to z_{\perp,\parallel}(\nu) = 1+\frac{1}{2\Omega_{\perp,\parallel}}\int_{-\infty}^{\infty}\frac{V_{\perp,\parallel}^2(\omega)}{\nu-\omega-\mathrm{i}\epsilon}\,\mathrm{d}\omega. \tag{C.56}$$

The second step uses the substitutions $\Omega\to\tilde{k}c$,

$$\begin{aligned} g(\nu) &\to \frac{\mathrm{i}}{2}\Lambda(k)\left[q_\perp(\nu)+s_\perp(\nu)\right] \\ &= \frac{\mathrm{i}}{2}\Lambda(k)\left(\frac{\Omega_\perp V_\perp(\nu)}{\nu^2-\Omega_\perp^2 z_\perp(\nu)}\right), \end{aligned} \tag{C.57}$$

and

$$\begin{aligned} z(\nu)\to\tilde{z}(\nu) &= 1+\frac{2}{\tilde{k}c}\int_{-\infty}^{\infty}\frac{\left(\frac{\mathrm{i}}{2}\Lambda(k)[q_\perp(\omega)+s_\perp(\omega)]\right)^2}{\nu-\omega-\mathrm{i}\epsilon}\,\mathrm{d}\omega \\ &= 1-\frac{\Lambda^2(k)\Omega_\perp^2}{2\tilde{k}c}\int_{-\infty}^{\infty}\frac{V_\perp^2(\omega)}{[\omega^2-\Omega_\perp^2 z_\perp(\omega)]^2}\frac{1}{\nu-\omega-\mathrm{i}\epsilon}\,\mathrm{d}\omega \end{aligned} \tag{C.58}$$

to produce

$$\begin{aligned} \tilde{q}(k,\nu) &= \mathrm{i}\frac{(\nu+\tilde{k}c)\Lambda(k)}{2}\frac{\Omega_\perp V_\perp(\nu)}{\nu^2-\Omega_\perp^2 z_\perp(\nu)}\frac{1}{\nu^2-\tilde{k}^2c^2\tilde{z}(\nu)}, \\ \tilde{s}(k,\nu) &= \mathrm{i}\frac{(\nu-\tilde{k}c)\Lambda(k)}{2}\frac{\Omega_\perp V_\perp(\nu)}{\nu^2-\Omega_\perp^2 z_\perp(\nu)}\frac{1}{\nu^2-\tilde{k}^2c^2\tilde{z}(\nu)}, \\ \tilde{u}(k;\nu,\nu') &= \delta(\nu-\nu')+\frac{2\tilde{k}c}{\nu^2-\tilde{k}c\tilde{z}(\nu)}\frac{\mathrm{i}}{2}\Lambda(k)\frac{\Omega_\perp V_\perp(\nu)}{\nu^2-\Omega_\perp^2 z_\perp(\nu)}\frac{\mathrm{i}}{2}\Lambda(k)\frac{\Omega_\perp V_\perp(\nu')}{\nu'^2-\Omega_\perp^2 z_\perp(\nu')}\frac{1}{\nu-\nu'-\mathrm{i}\epsilon} \\ &= \delta(\nu-\nu')-\frac{\tilde{k}c\Lambda^2(k)\Omega_\perp^2}{2}\frac{V_\perp(\nu)}{\left[\nu^2-\tilde{k}^2c^2\tilde{z}(\nu)\right][\nu^2-\Omega_\perp^2 z_\perp(\nu)][\nu'^2-\Omega_\perp^2 z_\perp(\nu')]}\frac{V_\perp(\nu')}{\nu-\nu'-\mathrm{i}\epsilon}, \\ \tilde{v}(k;\nu,\nu') &= -\frac{\tilde{k}c\Lambda^2(k)\Omega_\perp^2}{2}\frac{V_\perp(\nu)}{\left[\nu^2-\tilde{k}^2c^2\tilde{z}(\nu)\right][\nu^2-\Omega_\perp^2 z_\perp(\nu)][\nu'^2-\Omega_\perp^2 z_\perp(\nu')]} PV\left\{\frac{V_\perp(\nu')}{\nu+\nu'}\right\}. \end{aligned} \tag{C.59}$$

D Fano Diagonalization of a Set of Oscillators Coupled to a Set of Baths

We begin with a Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} \tag{D.1}$$

where

$$\hat{H}_0 = \sum_{i=1}^{M} \hbar\Omega_i \hat{a}_i^\dagger(t)\hat{a}_i(t) + \sum_{\sigma=1}^{N} \int_0^\infty \hbar\nu \hat{b}_\sigma^\dagger(\nu,t)\hat{b}_\sigma(\nu,t)\,\mathrm{d}\nu \tag{D.2}$$

and

$$\hat{H}_{\text{int}} = \sum_{i,\sigma} \int_0^\infty \hbar g_{i\sigma}(\nu)\left[\hat{a}_i(t) + \hat{a}_i^\dagger(t)\right]\left[\hat{b}_\sigma(\nu,t) + \hat{b}_\sigma^\dagger(\nu,t)\right]\mathrm{d}\nu. \tag{D.3}$$

The M discrete boson operators $\hat{a}_i(t)$ and N sets of bath operators $\hat{b}_\sigma(\nu,t)$ have natural frequencies Ω_i and $\nu(\nu)$, respectively, and commutation relations

$$\begin{aligned}\left[\hat{a}_i(t), \hat{a}_j^\dagger(t)\right] &= \delta_{ij},\\ \left[\hat{b}_\sigma(\nu,t), \hat{b}_{\sigma'}^\dagger(\nu',t)\right] &= \delta_{\sigma\sigma'}\delta(\nu-\nu'),\end{aligned} \tag{D.4}$$

and

$$\begin{aligned}\left[\hat{a}_i(t), \hat{a}_j(t)\right] &= 0,\\ \left[\hat{b}_\sigma(\nu,t), \hat{b}_\sigma(\nu',t)\right] &= 0,\\ \left[\hat{a}_i(t), \hat{b}_\sigma(\nu,t)\right] &= \left[\hat{a}_i(t), \hat{b}_\sigma^\dagger(\nu,t)\right] = 0.\end{aligned} \tag{D.5}$$

The coupling strength between the i^{th} discrete state and the $(\sigma,\nu)^{\text{th}}$ bath operator is assumed to be real and is given by $\hbar g_{i\sigma}(\nu)$. As was shown in a simpler setup in Appendix C, diagonalization of this system can be performed by defining new hybridized boson operators

$$\hat{C}_n(\omega,t) = \sum_i \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega)\hat{a}_i(t) + s_{ni}(\omega)\hat{a}_i^\dagger(t)\right] + \sum_\sigma \int_0^\infty \left[u_{n\sigma}(\omega,\nu)\hat{b}_\sigma(\nu,t) + v_{n\sigma}(\omega,\nu)\hat{b}_\sigma^\dagger(\nu,t)\right]\mathrm{d}\nu \tag{D.6}$$

that provide a simple form for the Hamiltonian,

$$\hat{H} = \sum_n \int_0^\infty \hbar\omega \hat{C}_n^\dagger(\omega,t)\hat{C}_n(\omega,t)\,\mathrm{d}\omega, \tag{D.7}$$

and satisfy the commutation relations

$$\begin{aligned}\left[\hat{C}_n(\omega,t), \hat{C}_{n'}^\dagger(\omega',t)\right] &= \delta_{nn'}\delta(\omega-\omega'),\\ \left[\hat{C}_n(\omega,t), \hat{C}_{n'}(\omega',t)\right] &= 0.\end{aligned} \tag{D.8}$$

Further, using the algebra of the previous section, we can employ the commutators

$$\begin{aligned}\left[\hat{a}_i(t), \hat{H}\right] &= \hbar\Omega_i\hat{a}_i(t) + \sum_\sigma \int_0^\infty \hbar g_{i\sigma}(\nu)\left[\hat{b}_\sigma(\nu,t) + \hat{b}_\sigma^\dagger(\nu,t)\right]\mathrm{d}\nu,\\ \left[\hat{b}_\sigma(\nu,t), \hat{H}\right] &= \hbar\nu\hat{b}_\sigma(\nu,t) + \sum_i \hbar g_{i\sigma}(\nu)\left[\hat{a}_i(t) + \hat{a}_i^\dagger(t)\right],\end{aligned} \tag{D.9}$$

and

$$\begin{aligned}\left[\hat{a}_i^\dagger(t), \hat{H}\right] &= -\hbar\Omega_i \hat{a}_i^\dagger(t) - \sum_\sigma \int_0^\infty \hbar g_{i\sigma}(\nu) \left[\hat{b}_\sigma(\nu,t) + \hat{b}_\sigma^\dagger(\nu,t)\right] \mathrm{d}\nu, \\ \left[\hat{b}_\sigma^\dagger(\nu,t), \hat{H}\right] &= -\hbar\nu \hat{b}_\sigma^\dagger(\nu,t) - \sum_i \hbar g_{i\sigma}(\nu) \left[\hat{a}_i(t) + \hat{a}_i^\dagger(t)\right],\end{aligned} \tag{D.10}$$

via Eq. (D.1) as well as

$$\left[\hat{C}_n(\omega,t), \hat{H}\right] = \hbar\omega \hat{C}_n(\omega,t) \tag{D.11}$$

via Eq. (D.7). Alongside the definition of the hybridized operators (Eq. [D.6]), these identities can be to say

$$\begin{aligned}&\hbar\omega\left(\sum_i \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega)\hat{a}_i(t) + s_{ni}(\omega)\hat{a}_i^\dagger(t)\right] + \sum_\sigma \int_0^\infty \left[u_{n\sigma}(\omega,\nu)\hat{b}_\sigma(\nu,t) + v_{n\sigma}(\omega,\nu)\hat{b}_\sigma^\dagger(\nu,t)\right] \mathrm{d}\nu\right) \\ &= \sum_i \left(\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega)\left[\hat{a}_i(t), \hat{H}\right] + s_{ni}(\omega)\left[\hat{a}_i^\dagger(t), \hat{H}\right]\right) + \sum_\sigma \int_0^\infty \left(u_{n\sigma}(\omega,\nu)\left[\hat{b}_\sigma(\nu,t), \hat{H}\right]\right. \\ &\quad \left.+ v_{n\sigma}(\omega,\nu)\left[\hat{b}_\sigma^\dagger(\nu,t), \hat{H}\right]\right) \mathrm{d}\nu \\ &= \sum_i \left(\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega)\left[\hbar\Omega_i \hat{a}_i(t) + \sum_\sigma \int_0^\infty \hbar g_{i\sigma}(\nu)\left(\hat{b}_\sigma(\nu,t) + \hat{b}_\sigma^\dagger(\nu,t)\right)\mathrm{d}\nu\right]\right. \\ &\quad \left.+ s_{ni}(\omega)\left[-\hbar\Omega_i \hat{a}_i^\dagger(t) - \sum_\sigma \int_0^\infty \hbar g_{i\sigma}(\nu)\left(\hat{b}_\sigma(\nu,t) + \hat{b}_\sigma^\dagger(\nu,t)\right)\mathrm{d}\nu\right]\right) \\ &\quad + \sum_\sigma \int_0^\infty \left(u_{n\sigma}(\omega,\nu)\left[\hbar\nu \hat{b}_\sigma(\nu,t) + \sum_i \hbar g_{i\sigma}(\nu)\left(\hat{a}_i(t) + \hat{a}_i^\dagger(t)\right)\right]\right. \\ &\quad \left.+ v_{n\sigma}(\omega,\nu)\left[-\hbar\nu \hat{b}_\sigma^\dagger(\nu,t) - \sum_i \hbar g_{i\sigma}(\nu)\left(\hat{a}_i(t) + \hat{a}_i^\dagger(t)\right)\right]\right)\mathrm{d}\nu.\end{aligned} \tag{D.12}$$

This provides a set of equations, one for each independent operator, that serve to fix the values of each expansion coefficient $q_{ni}(\omega)$, $s_{ni}(\omega)$, $u_{n\sigma}(\omega,\nu)$, and $v_{n\sigma}(\omega,\nu)$. Explicitly,

$$\begin{aligned}(\omega - \Omega_i)\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega) &= \sum_\sigma \int_0^\infty \left[u_{n\sigma}(\omega,\nu) - v_{n\sigma}(\omega,\nu)\right] g_{i\sigma}(\nu)\,\mathrm{d}\nu, \\ (\omega + \Omega_i) s_{ni}(\omega) &= \sum_\sigma \int_0^\infty \left[u_{n\sigma}(\omega,\nu) - v_{n\sigma}(\omega,\nu)\right] g_{i\sigma}(\nu)\,\mathrm{d}\nu, \\ (\omega - \nu) u_{n\sigma}(\omega,\nu) &= \sum_i \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega) - s_{ni}(\omega)\right] g_{i\sigma}(\nu), \\ (\omega + \nu) v_{n\sigma}(\omega,\nu) &= \sum_i \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega) - s_{ni}(\omega)\right] g_{i\sigma}(\nu).\end{aligned} \tag{D.13}$$

The latter three equations have solutions in terms of $q_{ni}(\omega)$. First noting that

$$\begin{aligned}s_{ni}(\omega) &= \frac{\omega - \Omega_i}{\omega + \Omega_i}\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}(\omega) \\ &= \frac{\omega - \Omega_i}{2\Omega_i} q_{ni}(\omega),\end{aligned} \tag{D.14}$$

we can see that the most general solutions for $u_{n\sigma}(\omega,\nu)$ and $v_{n\sigma}(\omega,\nu)$ are

$$\begin{aligned}
u_{n\sigma}(\omega,\nu) &= PV\left\{\frac{1}{\omega-\nu}\right\}\sum_i g_{i\sigma}(\nu)q_{ni}(\omega) + \sum_i g_{i\sigma}(\nu)q_{ni}(\omega)x_{ni\sigma}(\omega)\delta(\omega-\nu), \\
v_{n\sigma}(\omega,\nu) &= \frac{1}{\omega+\nu}\sum_i g_{i\sigma}(\nu)q_{ni}(\omega),
\end{aligned} \tag{D.15}$$

wherein the Cauchy principal value operator PV has been introduced to handle the singularity in $u_{n\sigma}(\omega,\nu)$ at $\omega=\nu$. The functions $x_{ni\sigma}(\omega)$ are smooth, complex, and nonzero and have forms determined through physical constraints, as will be shown below.

Plugging these solutions back into the first equation of Eq. (D.13), one finds

$$\begin{aligned}
\frac{\omega^2-\Omega_i^2}{2\Omega_i}q_{ni}(\omega) = \sum_\sigma\sum_j q_{nj}(\omega)\int_0^\infty\left(PV\left\{\frac{1}{\omega-\nu}\right\}-\frac{1}{\omega+\nu}\right)g_{i\sigma}(\nu)g_{j\sigma}(\nu)\,\mathrm{d}\nu \\
+\sum_\sigma\sum_j x_{nj\sigma}(\omega)q_{nj}(\omega)g_{i\sigma}(\omega)g_{j\sigma}(\omega).
\end{aligned} \tag{D.16}$$

Note that the thermal bath terms in the above sum are identical to the photonic bath therms under the transformation $\xi_\sigma\to 0$.

Summation over i allows for the separation of this relation into matrix-vector notation. Using the unit vectors $\hat{\mathbf{e}}_i(\omega)$ to represent the orthogonal axes of the Hilbert space formed by the expansion coefficients $q_{ni}(\omega)$, we can say

$$\begin{aligned}
&\sum_i\frac{\omega^2-\Omega_i^2}{2\Omega_i}\hat{\mathbf{e}}_i\hat{\mathbf{e}}_i\cdot\sum_k q_{nk}(\omega)\hat{\mathbf{e}}_k \\
&\quad=\sum_\sigma\sum_{ij}\int_0^\infty\left(PV\left\{\frac{1}{\omega-\nu}\right\}-\frac{1}{\omega+\nu}\right)g_{i\sigma}(\nu)g_{j\sigma}(\nu)\,\mathrm{d}\nu\;\hat{\mathbf{e}}_i\hat{\mathbf{e}}_j\cdot\sum_k q_{nk}(\omega)\hat{\mathbf{e}}_k \\
&\qquad+\sum_\sigma\sum_{ij}x_{nj\sigma}(\omega)g_{i\sigma}(\omega)g_{j\sigma}(\omega)\hat{\mathbf{e}}_i\hat{\mathbf{e}}_j\cdot\sum_k q_{nk}(\omega)\hat{\mathbf{e}}_k.
\end{aligned} \tag{D.17}$$

Therefore, we can define the matrices

$$\begin{aligned}
\mathbf{A}(\omega) &= \sum_i\frac{(\omega^2-\Omega_i^2)}{2\Omega_i}\hat{\mathbf{e}}_i\hat{\mathbf{e}}_i, \\
\mathbf{B}_\sigma(\omega) &= \sum_{ij}\int_0^\infty\left(PV\left\{\frac{1}{\omega-\nu}\right\}-\frac{1}{\omega+\nu}\right)g_{i\sigma}(\nu)g_{j\sigma}(\nu)\,\mathrm{d}\nu\,\hat{\mathbf{e}}_i\hat{\mathbf{e}}_j, \\
\mathbf{C}_{n\sigma}(\omega) &= \sum_{ij}x_{nj\sigma}(\omega)g_{i\sigma}(\omega)g_{j\sigma}(\omega)\hat{\mathbf{e}}_i\hat{\mathbf{e}}_j, \\
\mathbf{D}_\sigma(\omega) &= \sum_{ij}g_{i\sigma}(\omega)g_{j\sigma}(\omega)\hat{\mathbf{e}}_i\hat{\mathbf{e}}_j,
\end{aligned} \tag{D.18}$$

and the vector

$$\mathbf{q}_n(\omega)=\sum_i q_{ni}(\omega)\hat{\mathbf{e}}_i, \tag{D.19}$$

such that the first line of Eq. (D.13) can be rewritten as the vector equation

$$\mathbf{A}(\omega)\cdot\mathbf{q}_n(\omega)=\sum_\sigma\mathbf{B}_\sigma(\omega)\cdot\mathbf{q}_n(\omega)+\sum_\sigma\mathbf{C}_\sigma(\omega)\cdot\mathbf{q}_n(\omega). \tag{D.20}$$

This process can calso be used to expand our final physical constraint, the commutator of Eq. (D.8), as a matrix-vector equation. In more detail,

$$\begin{aligned}
\left[\hat{C}_n(\omega,t),\hat{C}^\dagger_{n'}(\omega',t)\right] &= \sum_{ij}\left(\frac{(\omega+\Omega_i)(\omega'+\Omega_j)}{4\Omega_i\Omega_j}q_{ni}(\omega)q^*_{n'j}(\omega')\left[\hat{a}_i(t),\hat{a}^\dagger_j(t)\right]+s_{ni}(\omega)s^*_{n'j}(\omega')\left[\hat{a}^\dagger_i(t),\hat{a}_j(t)\right]\right)\\
&\quad+\sum_{\sigma\sigma'}\int_0^\infty\int_0^\infty\Big(u_{n\sigma}(\omega,\nu)u^*_{n'\sigma'}(\omega',\nu')\left[\hat{b}_\sigma(\nu,t),\hat{b}^\dagger_{\sigma'}(\nu',t)\right]\\
&\quad+v_{n\sigma}(\omega,\nu)v^*_{n'\sigma'}(\omega',\nu')\left[\hat{b}^\dagger_\sigma(\nu,t),\hat{b}_{\sigma'}(\nu',t)\right]\Big)\,\mathrm{d}\nu\,\mathrm{d}\nu'\\
&=\sum_i\left[\frac{(\omega+\Omega_i)(\omega'+\Omega_i)}{4\Omega_i^2}q_{ni}(\omega)q^*_{n'i}(\omega')-s_{ni}(\omega)s^*_{n'i}(\omega')\right]\\
&\quad+\sum_\sigma\int_0^\infty\left[u_{n\sigma}(\omega,\nu)u^*_{n'\sigma}(\omega',\nu)-v_{n\sigma}(\omega,\nu)v^*_{n'\sigma}(\omega',\nu)\right]\mathrm{d}\nu\\
&=\delta_{nn'}\delta(\omega-\omega')
\end{aligned}\tag{D.21}$$

leads, with the substitution of the explicit form of each expansion coefficient, to

$$\begin{aligned}
\delta_{nn'}\delta(\omega-\omega') &= \sum_i\left(\frac{(\omega+\Omega_i)(\omega'+\Omega_i)}{4\Omega_i^2}-\frac{(\omega-\Omega_i)(\omega'-\Omega_i)}{4\Omega_i^2}\right)q_{ni}(\omega)q^*_{n'i}(\omega')\\
&\quad+\sum_\sigma\sum_{ij}\int_0^\infty g_{i\sigma}(\nu)g_{j\sigma}(\nu)q_{ni}(\omega)q^*_{n'j}(\omega')\\
&\quad\times\Bigg(\left[PV\left\{\frac{1}{\omega-\nu}\right\}+x_{ni\sigma}(\omega)\delta(\omega-\nu)\right]\left[PV\left\{\frac{1}{\omega-\nu}\right\}+x^*_{n'j\sigma}(\omega')\delta(\omega'-\nu)\right]\\
&\quad-\frac{1}{\omega+\nu}\frac{1}{\omega'+\nu}\Bigg)\mathrm{d}\nu\\
&=\sum_i\frac{\omega+\omega'}{2\Omega_i}q_{ni}(\omega)q^*_{n'i}(\omega')+\sum_\sigma\sum_{ij}\int_0^\infty g_{i\sigma}(\nu)g_{j\sigma}(\nu)q_{ni}(\omega)q^*_{n'j}(\omega')\\
&\quad\times\Bigg(\left[PV\left\{\frac{1}{\omega-\nu}\right\}+x_{ni\sigma}(\omega)\delta(\omega-\nu)\right]\left[PV\left\{\frac{1}{\omega'-\nu}\right\}+x^*_{n'j\sigma}(\omega')\delta(\omega'-\nu)\right]\\
&\quad-\frac{1}{\omega+\nu}\frac{1}{\omega'+\nu}\Bigg)\mathrm{d}\nu.
\end{aligned}\tag{D.22}$$

Using the identities

$$\begin{aligned}
PV\left\{\frac{1}{x}\right\} &= \lim_{\epsilon\to0}\frac{x}{x^2+\epsilon^2},\\
\delta(x) &= \lim_{\epsilon\to0}\frac{\epsilon}{x^2+\epsilon^2},
\end{aligned}\tag{D.23}$$

and the method of partial fractions, we can expand Cauchy principal value products as

$$\begin{aligned}
PV\left\{\frac{1}{\omega-\nu}\right\}PV\left\{\frac{1}{\omega'-\nu}\right\} &= \lim_{\epsilon\to0}\frac{\omega-\nu}{(\omega-\nu)^2+\epsilon^2}\frac{\omega'-\nu}{(\omega'-\nu)^2+\epsilon^2}\\
&=\pi^2\delta(\nu-\omega')\delta(\nu-\omega)+PV\left\{\frac{1}{\omega-\omega'}\right\}\left(PV\left\{\frac{1}{\omega'-\nu}\right\}-PV\left\{\frac{1}{\omega-\nu}\right\}\right)
\end{aligned}\tag{D.24}$$

and note that, for ω, ω', and ν all nonnegative,

$$\begin{aligned}\frac{1}{\omega+\nu}\frac{1}{\omega'+\nu} &= PV\left\{\frac{1}{\omega+\nu}\right\}PV\left\{\frac{1}{\omega'+\nu}\right\} \\ &= PV\left\{\frac{1}{\omega-\omega'}\right\}\left(\frac{1}{\omega'+\nu}-\frac{1}{\omega+\nu}\right).\end{aligned} \tag{D.25}$$

Therefore,

$$\begin{aligned}\delta_{nn'}\delta(\omega-\omega') = \sum_i \frac{(\omega+\omega')}{2\Omega_i} q_{ni}(\omega)q^*_{n'i}(\omega') &+ \sum_\sigma\sum_{ij}\int_0^\infty g_{i\sigma}(\nu)g_{j\sigma}(\nu)q_{ni}(\omega)q^*_{n'j}(\omega') \\ &\times\left(\pi^2\delta(\omega'-\nu)\delta(\omega-\nu) + PV\left\{\frac{1}{\omega-\omega'}\right\}\left[PV\left\{\frac{1}{\omega'-\nu}\right\}-\frac{1}{\omega'+\nu}\right]\right. \\ &\left.-PV\left\{\frac{1}{\omega-\omega'}\right\}\left[PV\left\{\frac{1}{\omega-\nu}\right\}-\frac{1}{\omega+\nu}\right]\right)\mathrm{d}\nu \\ &-\sum_\sigma\sum_{ij} q_{ni}(\omega)q^*_{n'j}(\omega')PV\left\{\frac{1}{\omega-\omega'}\right\}x_{ni\sigma}(\omega)g_{i\sigma}(\omega)g_{j\sigma}(\omega) \\ &+\sum_\sigma\sum_{ij} q_{ni}(\omega)q^*_{n'j}(\omega')PV\left\{\frac{1}{\omega-\omega'}\right\}x^*_{n'j\sigma}(\omega')g_{i\sigma}(\omega')g_{j\sigma}(\omega') \\ &+\sum_\sigma\sum_{ij}\int_0^\infty q_{ni}(\omega)q^*_{n'j}(\omega')x_{ni\sigma}(\omega)x^*_{n'j\sigma}(\omega')g_{i\sigma}(\nu)g_{j\sigma}(\nu)\delta(\omega-\nu)\delta(\omega'-\nu)\,\mathrm{d}\nu.\end{aligned} \tag{D.26}$$

Further, with

$$\int_0^\infty f(\nu)\delta(\omega'-\nu)\delta(\omega-\nu)\,\mathrm{d}\nu = f(\omega)\delta(\omega-\omega'), \tag{D.27}$$

we find

$$\begin{aligned}\delta_{nn'}\delta(\omega-\omega') = \sum_i \frac{(\omega+\omega')}{2\Omega_i} q_{ni}(\omega)q^*_{n'i}(\omega') &+ \sum_\sigma\sum_{ij}\pi^2\delta(\omega-\omega')g_{i\sigma}(\omega)g_{j\sigma}(\omega)q_{ni}(\omega)q^*_{n'j}(\omega') \\ +\sum_\sigma\sum_{ij} q_{ni}(\omega)q^*_{n'j}(\omega')PV\left\{\frac{1}{\omega-\omega'}\right\}&\int_0^\infty\left(PV\left\{\frac{1}{\omega'-\nu}\right\}-\frac{1}{\omega'+\nu}\right)g_{i\sigma}(\nu)g_{j\sigma}(\nu)\,\mathrm{d}\nu \\ -\sum_\sigma\sum_{ij} q_{ni}(\omega)q^*_{n'j}(\omega')PV\left\{\frac{1}{\omega-\omega'}\right\}&\int_0^\infty\left(PV\left\{\frac{1}{\omega-\nu}\right\}-\frac{1}{\omega+\nu}\right)g_{i\sigma}(\nu)g_{j\sigma}(\nu)\,\mathrm{d}\nu \\ +\sum_\sigma\sum_{ij} q_{ni}(\omega)q^*_{n'j}(\omega')PV&\left\{\frac{1}{\omega-\omega'}\right\}x^*_{n'j\sigma}(\omega')g_{i\sigma}(\omega')g_{j\sigma}(\omega') \\ -\sum_\sigma\sum_{ij} q_{ni}(\omega)q^*_{n'j}(\omega')PV&\left\{\frac{1}{\omega-\omega'}\right\}x_{ni\sigma}(\omega)g_{i\sigma}(\omega)g_{j\sigma}(\omega) \\ +\sum_\sigma\sum_{ij} q_{ni}(\omega)q^*_{n'j}(\omega')&x_{ni\sigma}(\omega)x^*_{n'j\sigma}(\omega')g_{i\sigma}(\omega)g_{j\sigma}(\omega)\delta(\omega-\omega').\end{aligned} \tag{D.28}$$

Here, we have let $\delta(\omega+\omega')=0$ for $\omega,\omega'>0$. Further, it will make our lives easier if there exists a solution for $q_{ni}(\omega)$ wherein the prefactor $x_{ni\sigma}(\omega)$ is independent of the polariton index n. Assuming a solution can be found, we will let $x_{ni\sigma}(\omega)\to x_{i\sigma}(\omega)$. Further defining a new matrix

$$\mathbf{E}_\sigma(\omega) = \sum_{ij} x_{i\sigma}(\omega)x^*_{j\sigma}(\omega)g_{i\sigma}(\omega)g_{j\sigma}(\omega)\hat{\mathbf{e}}_i\hat{\mathbf{e}}_j, \tag{D.29}$$

we can rearrange the above expression in vector form such that

$$\begin{aligned}\delta_{nn'}\delta(\omega-\omega') = \mathbf{q}_n(\omega)\cdot\sum_i \frac{\omega+\omega'}{2\Omega_i}\hat{\mathbf{e}}_i\hat{\mathbf{e}}_i\cdot\mathbf{q}^*_{n'}(\omega')&\\
+\mathbf{q}_n(\omega)\cdot\pi^2\delta(\omega-\omega')\sum_\sigma \mathbf{D}_\sigma(\omega)\cdot\mathbf{q}^*_{n'}(\omega')&\\
+\mathbf{q}_n(\omega)\cdot PV\left\{\frac{1}{\omega-\omega'}\right\}\sum_\sigma\left[\mathbf{B}_\sigma(\omega')-\mathbf{B}_\sigma(\omega)\right]\cdot\mathbf{q}^*_{n'}(\omega')&\\
+\mathbf{q}_n(\omega)\cdot PV\left\{\frac{1}{\omega-\omega'}\right\}\sum_\sigma\left[\mathbf{C}_\sigma(\omega')-\mathbf{C}^\top_\sigma(\omega)\right]\cdot\mathbf{q}^*_{n'}(\omega')&\\
+\mathbf{q}_n(\omega)\cdot\delta(\omega-\omega')\sum_\sigma \mathbf{E}_\sigma(\omega)\cdot\mathbf{q}^*_{n}(\omega'),&\end{aligned}\tag{D.30}$$

wherein we have let $\mathbf{C}_{n\sigma}(\omega)\to\mathbf{C}_\sigma(\omega)$. Use of the identity of Eq. (D.20) provides

$$\begin{aligned}\delta_{nn'}\delta(\omega-\omega') &= \mathbf{q}_n(\omega)\cdot\sum_i \frac{\omega+\omega'}{2\Omega_i}\hat{\mathbf{e}}_i\hat{\mathbf{e}}_i\cdot\mathbf{q}^*_{n'}(\omega')\\
&\quad+\mathbf{q}_n(\omega)\cdot\delta(\omega-\omega')\sum_\sigma\left[\pi^2\mathbf{D}_\sigma(\omega)+\mathbf{E}_\sigma(\omega)\right]\cdot\mathbf{q}^*_{n'}(\omega')\\
&\quad+\mathbf{q}_n(\omega)\cdot PV\left\{\frac{1}{\omega-\omega'}\right\}\sum_\sigma\left[\mathbf{B}_\sigma(\omega')-\mathbf{B}_\sigma(\omega)\right]\cdot\mathbf{q}^*_{n'}(\omega')\\
&\quad+\mathbf{q}_n(\omega)\cdot PV\left\{\frac{1}{\omega-\omega'}\right\}\left[\mathbf{A}(\omega')-\sum_\sigma\mathbf{B}_\sigma(\omega')-\mathbf{A}(\omega)+\sum_\sigma\mathbf{B}_\sigma(\omega)\right]\\
&= \mathbf{q}_n(\omega)\cdot\sum_i\left(\frac{\omega+\omega'}{2\Omega_i}+PV\left\{\frac{1}{\omega-\omega'}\right\}\left[\frac{\omega'^2-\Omega_i^2}{2\Omega_i}-\frac{\omega^2-\Omega_i^2}{2\Omega_i}\right]\right)\hat{\mathbf{e}}_i\hat{\mathbf{e}}_i\cdot\mathbf{q}^*_{n'}(\omega')\\
&\quad+\mathbf{q}_n(\omega)\cdot\delta(\omega-\omega')\sum_\sigma\left[\pi^2\mathbf{D}_\sigma(\omega)+\mathbf{E}_\sigma(\omega)\right]\cdot\mathbf{q}^*_{n'}(\omega')\\
&= \mathbf{q}_n(\omega)\cdot\sum_i\left(\frac{\omega+\omega'}{2\Omega_i}+PV\left\{\frac{1}{\omega-\omega'}\right\}\frac{(\omega'-\omega)(\omega'+\omega)}{2\Omega_i}\right)\hat{\mathbf{e}}_i\hat{\mathbf{e}}_i\cdot\mathbf{q}^*_{n'}(\omega')\\
&\quad+\mathbf{q}_n(\omega)\cdot\delta(\omega-\omega')\sum_\sigma\left[\pi^2\mathbf{D}_\sigma(\omega)+\mathbf{E}_\sigma(\omega)\right]\cdot\mathbf{q}^*_{n'}(\omega')\\
&= \delta(\omega-\omega')\mathbf{q}_n(\omega)\cdot\sum_\sigma\left[\pi^2\mathbf{D}_\sigma(\omega)+\mathbf{E}_\sigma(\omega)\right]\cdot\mathbf{q}^*_{n'}(\omega').\end{aligned}\tag{D.31}$$

Finally, it is advantageous to define

$$\mathbf{G}(\omega)=\sum_\sigma\left[\pi^2\mathbf{D}_\sigma(\omega)+\mathbf{E}_\sigma(\omega)\right]\tag{D.32}$$

as the characteristic matrix of the polariton decomposition. The bosonic commutator $[\hat{C}_n(\omega,t),\hat{C}^\dagger_{n'}(\omega',t)]=\delta_{nn'}\delta(\omega-\omega')$ is then only satisfied if $\mathbf{q}_n(\omega)$ is an eigenvector of $\mathbf{G}(\omega)$. Letting its corresponding eigenvalue be $\lambda_n(\omega)$, we have, explicitly,

$$\mathbf{G}(\omega)\cdot\mathbf{q}_n(\omega)=\lambda_n(\omega)\mathbf{q}_n(\omega).\tag{D.33}$$

Because the matrices $\mathbf{D}_\sigma(\omega)$ and $\mathbf{E}_\sigma(\omega)$ are Hermitian, $\mathbf{G}(\omega)$ is Hermitian. Therefore, the eigenvalues $\lambda_n(\omega)$ are real and the eigenvectors $\mathbf{q}_n(\omega)$ are orthogonal such that

$$\mathbf{q}_n(\omega)\cdot\mathbf{q}^*_{n'}(\omega)=|\mathbf{q}_n(\omega)|^2\delta_{nn'}\tag{D.34}$$

and

$$\begin{aligned}\delta_{nn'}\delta(\omega-\omega') &= \delta(\omega-\omega')\mathbf{q}^*_n(\omega)\cdot\mathbf{G}(\omega)\cdot\mathbf{q}_{n'}(\omega)\\
&= \delta(\omega-\omega')\lambda_n(\omega)\mathbf{q}^*_n(\omega)\cdot\mathbf{q}_{n'}(\omega)\\
&= \delta(\omega-\omega')\lambda_n(\omega)|\mathbf{q}_n(\omega)|^2\delta_{nn'}.\end{aligned}\tag{D.35}$$

We can then see

$$|\mathbf{q}_n(\omega)|^2 = \frac{1}{\lambda_n(\omega)}. \tag{D.36}$$

is the normalization condition that sets the absolute magnitude of the elements of $\mathbf{q}_n(\omega)$.

It is important to note at this point that the complex coefficients $x_{i\sigma}(\omega)$ are still not uniquely defined. This means that, for a given n and ω, a set of polariton operators $\{\hat{C}_n(\omega, t; \mathbf{X})\}$ exists in which each element $\hat{C}_n(\omega, t; \mathbf{X})$ describes the dynamics of the system with equal validity. These elements are indexed by the matrices $\mathbf{X}(\omega) = \sum_{i\sigma} x_{i\sigma}(\omega)\hat{\mathbf{e}}_i\hat{\mathbf{e}}_\sigma$ whose elements describe the degree to which the operators $\hat{b}_\sigma(\omega)$ contribute to $\hat{C}_n(\omega, t)$. These elements are nonzero, but are otherwise free to vary in the complex plane. Therefore, it is up to the author of a particular model to choose which values of $\mathbf{X}(\omega)$ are most advantageous for him/her. However, as we will see in the following, the inverse Fano transformations that reproduce the uncoupled operators $\hat{a}_i(t)$ and $\hat{b}_\sigma(\nu, t)$ as combinations of polariton operators will place additional constraints on $\mathbf{X}(\omega)$ such that particular values of $x_{i\sigma}(\omega)$ will be more convenient for use in one model or another.

To begin, we can see that the inverse Fano transformations used to define the unhybridized operators $\hat{a}_i(t)$ and $\hat{b}_\sigma(\nu, t)$ in terms of the polariton operators can be straightforwardly built from these results and the integral techniques of Appendix C. Forgoing the explicit dependence of each $\hat{C}_n(\omega, t)$ on $\mathbf{X}$, we can propose a trial solution

$$\begin{aligned}
\hat{a}_i(t) &= \frac{1}{\mathcal{N}_i}\sum_n \int_0^\infty P_{ni}(\omega)\left[\frac{\omega+\Omega_i}{2\Omega_i}q_{ni}^*(\omega)\hat{C}_n(\omega,t) - s_{ni}(\omega)\hat{C}_n^\dagger(\omega,t)\right]\mathrm{d}\omega \\
&= \frac{1}{\mathcal{N}_i}\Bigg(\sum_j \hat{a}_j(t)\sum_n\int_0^\infty P_{ni}(\omega)\left[\frac{(\omega+\Omega_i)(\omega+\Omega_j)}{4\Omega_i\Omega_j}q_{ni}^*(\omega)q_{nj}(\omega) - s_{ni}(\omega)s_{nj}^*(\omega)\right]\mathrm{d}\omega \\
&\quad + \sum_j \hat{a}_j^\dagger(t)\sum_n\int_0^\infty P_{ni}(\omega)\left[\frac{\omega+\Omega_i}{2\Omega_i}q_{ni}^*(\omega)s_{nj}(\omega) - \frac{\omega+\Omega_j}{2\Omega_j}s_{ni}(\omega)q_{nj}^*(\omega)\right]\mathrm{d}\omega \\
&\quad + \sum_\sigma\int_0^\infty \hat{b}_\sigma(\nu,t)\sum_n\int_0^\infty P_{ni}(\omega)\left[\frac{\omega+\Omega_i}{2\Omega_i}q_{ni}^*(\omega)u_{n\sigma}(\omega,\nu) - s_{ni}(\omega)v_{n\sigma}^*(\omega,\nu)\right]\mathrm{d}\omega\,\mathrm{d}\nu \\
&\quad + \sum_\sigma\int_0^\infty b_\sigma^\dagger(\nu,t)\sum_n\int_0^\infty P_{ni}(\omega)\left[\frac{\omega+\Omega_i}{2\Omega_i}q_{ni}^*(\omega)v_{n\sigma}(\omega,\nu) - s_{ni}(\omega)u_{n\sigma}^*(\omega,\nu)\right]\mathrm{d}\omega\,\mathrm{d}\nu\Bigg)
\end{aligned} \tag{D.37}$$

where $P_{ni}(\omega)$ is a characteristic prefactor and $\mathcal{N}_i$ is a normalization function. Before we being simplifying these expressions, we can see that, because $\mathbf{G}(\omega)$ is Hermitian, the matrix of its normalized column-eigenvectors,

$$\mathbf{V}(\omega) = \begin{pmatrix}\mathbf{v}_1^\top(\omega) & \mathbf{v}_2^\top(\omega) & \cdots & \mathbf{v}_M^\top(\omega)\end{pmatrix}, \tag{D.38}$$

is unitary. Here $\mathbf{v}_n(\omega) = \sqrt{\lambda_n}\mathbf{q}_n(\omega) = \sqrt{\lambda_n}\,[q_{n1}(\omega)\hat{\mathbf{e}}_1 + q_{n2}(\omega)\hat{\mathbf{e}}_2 + \ldots]$. Because both the rows and columns of a unitary matrix form an orthonormal set of vectors, we can define $\mathbf{u}_i(\omega) = \sqrt{\lambda_1}q_{1i}(\omega)\hat{\mathbf{e}}_1 + \sqrt{\lambda_2}q_{2i}(\omega)\hat{\mathbf{e}}_2 + \ldots$, such that

$$\begin{aligned}
\mathbf{v}_n^*(\omega)\cdot\mathbf{v}_{n'}(\omega) &= \lambda_n\sum_i q_{ni}^*(\omega)q_{n'i}(\omega) = \delta_{nn'}, \\
\mathbf{u}_i^*(\omega)\cdot\mathbf{u}_j(\omega) &= \sum_n \lambda_n q_{ni}^*(\omega)q_{nj}(\omega) = \delta_{ij}.
\end{aligned} \tag{D.39}$$

Therefore, looking at the first integral of our trial solution for $\hat{a}_i(t)$, we can see upon substitution of $s_{ni}(\omega) =$

$(\omega - \Omega_i)q_{ni}(\omega)/2\Omega_i$ that

$$
\begin{aligned}
&\sum_n \int_0^\infty P_{ni}(\omega) \left[\frac{(\omega+\Omega_i)(\omega+\Omega_j)}{4\Omega_i\Omega_j} q^*_{ni}(\omega) q_{nj}(\omega) - s_{ni}(\omega) s^*_{nj}(\omega) \right] \mathrm{d}\omega \\
&= \sum_n \int_0^\infty P_{ni}(\omega) \left[\frac{(\omega+\Omega_i)(\omega+\Omega_j)}{4\Omega_i\Omega_j} q^*_{ni}(\omega) q_{nj}(\omega) - \frac{(\omega-\Omega_i)(\omega-\Omega_j)}{4\Omega_i\Omega_j} q_{ni}(\omega) q^*_{nj}(\omega) \right] \mathrm{d}\omega \\
&= \sum_n \int_0^\infty \lambda_n \bar{P}_i(\omega) \left[\frac{(\omega+\Omega_i)(\omega+\Omega_j)}{4\Omega_i\Omega_j} q^*_{ni}(\omega) q_{nj}(\omega) - \frac{(\omega-\Omega_i)(\omega-\Omega_j)}{4\Omega_i\Omega_j} q_{ni}(\omega) q^*_{nj}(\omega) \right] \mathrm{d}\omega \\
&= \int_0^\infty \bar{P}_i(\omega) \delta_{ij} \left[\frac{(\omega+\Omega_i)(\omega+\Omega_j)}{4\Omega_i\Omega_j} - \frac{(\omega-\Omega_i)(\omega-\Omega_j)}{4\Omega_i\Omega_j} \right] \mathrm{d}\omega \\
&= \int_0^\infty \delta_{ij} \frac{\omega(\Omega_i+\Omega_j)}{2\Omega_i\Omega_j} \bar{P}_i(\omega)\, \mathrm{d}\omega,
\end{aligned}
\tag{D.40}
$$

where we have let $P_{ni}(\omega) = \lambda_n \bar{P}_i(\omega)$. Finally, looking at the form of the integral in Eq. (C.46), we can propose that $\bar{P}_i(\omega) = 4\Omega_i^2 \Sigma_i^2(\omega)/|\omega^2 - \Omega_i^2 z_i(\omega)|^2$, wherein

$$
z_i(\omega) = 1 + \lim_{\epsilon \to 0} \frac{2}{\Omega_i} \int_{-\infty}^{\infty} \frac{\Sigma_i^2(\nu)}{\omega - \nu - \mathrm{i}\epsilon}\, \mathrm{d}\nu
\tag{D.41}
$$

and $\Sigma_i^2(\omega)$ is an as-yet-undertermined function with units of coupling constant squared that obeys the property $\Sigma_i^2(-\omega) = -\Sigma_i^2(\omega)$. This provides us with

$$
\begin{aligned}
\sum_j \hat{a}_j(t) \sum_n \int_0^\infty P_{ni}(\omega) & \left[\frac{(\omega+\Omega_i)(\omega+\Omega_j)}{4\Omega_i\Omega_j} q^*_{ni}(\omega) q_{nj}(\omega) - s_{ni}(\omega) s^*_{nj}(\omega) \right] \mathrm{d}\omega \\
&= \sum_j \hat{a}_j(t) \int_0^\infty \delta_{ij} \frac{\omega(\Omega_i+\Omega_j)}{2\Omega_i\Omega_j} \frac{4\Omega_i^2 \Sigma_i^2(\omega)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2}\, \mathrm{d}\omega \\
&= \hat{a}_i(t) \int_0^\infty \frac{4\Omega_i \omega \Sigma_i^2(\omega)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2}\, \mathrm{d}\omega \\
&= \hat{a}_i(t)
\end{aligned}
\tag{D.42}
$$

such that

$$
\begin{aligned}
P_{ni}(\omega) &= \lambda_n \frac{4\Omega_i^2 \Sigma_i^2(\omega)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2}, \\
\mathcal{N}_i &= 1
\end{aligned}
\tag{D.43}
$$

are candidate solutions for the prefactor and normalization constant of $\hat{a}_i(t)$.

Moving forward, we can see that

$$
\begin{aligned}
&\sum_j \hat{a}_j^\dagger(t) \sum_n \int_0^\infty P_{ni}(\omega) \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}^*(\omega) s_{nj}(\omega) - \frac{\omega+\Omega_j}{2\Omega_j} s_{ni}(\omega) q_{nj}^*(\omega) \right] \mathrm{d}\omega \\
=&\sum_j \hat{a}_j^\dagger(t) \sum_n \int_0^\infty \lambda_n \frac{4\Omega_i^2 \Sigma_i^2(\omega)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2} \\
&\quad \times \left[\frac{(\omega+\Omega_i)(\omega-\Omega_j)}{4\Omega_i\Omega_j} q_{ni}^*(\omega) q_{nj}(\omega) - \frac{(\omega-\Omega_i)(\omega+\Omega_j)}{4\Omega_i\Omega_j} q_{ni}(\omega) q_{nj}^*(\omega) \right] \mathrm{d}\omega \\
&= \sum_j \hat{a}_j^\dagger(t) \int_0^\infty \frac{4\Omega_i^2 \Sigma_i^2(\omega)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2} \frac{2\omega(\Omega_i - \Omega_j)}{4\Omega_i\Omega_j} \delta_{ij} \, \mathrm{d}\omega \, \mathrm{d}\nu \\
&= 0.
\end{aligned}
\tag{D.44}
$$

Further,

$$
\begin{aligned}
&\sum_\sigma \int_0^\infty \hat{b}_\sigma(\nu,t) \sum_n \int_0^\infty P_{ni}(\omega) \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}^*(\omega) u_{n\sigma}(\omega,\nu) - s_{ni}(\omega) v_{n\sigma}^*(\omega,\nu) \right] \mathrm{d}\omega \, \mathrm{d}\nu \\
=&\sum_\sigma \int_0^\infty \hat{b}_\sigma(\nu,t) \sum_n \int_0^\infty \lambda_n \frac{4\Omega_i^2 \Sigma_i^2(\omega)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2} \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}^*(\omega) \left(PV\left\{ \frac{1}{\omega-\nu} \right\} \sum_j g_{j\sigma}(\nu) q_{nj}(\omega) \right.\right. \\
&\quad \left.\left. + \sum_j g_{j\sigma}(\nu) q_{nj}(\omega) x_{j\sigma}(\omega) \delta(\omega-\nu) \right) - \frac{\omega-\Omega_i}{2\Omega_i} q_{ni}(\omega) \frac{1}{\omega+\nu} \sum_j g_{j\sigma}(\nu) q_{nj}^*(\omega) \right] \mathrm{d}\omega \, \mathrm{d}\nu.
\end{aligned}
\tag{D.45}
$$

To align this expression with the form of Eq. (C.48) such that it evaluates to zero, we choose the form of our final unknown quantity,

$$
x_{i\sigma}(\omega) = \mathrm{i}\pi + \frac{\omega^2 - \Omega_i^2 z_i(\omega)}{2\Omega_i \Sigma_i^2(\omega)}, \tag{D.46}
$$

such that

$$
u_{n\sigma}(\omega,\nu) = \lim_{\epsilon\to 0} \frac{1}{\omega-\nu-\mathrm{i}\epsilon} \sum_i g_{i\sigma}(\nu) q_{ni}(\omega) + \sum_i g_{i\sigma}(\nu) q_{ni}(\omega) \frac{\omega^2 - \Omega_i^2 z_i(\omega)}{2\Omega_i \Sigma_i^2(\omega)} \delta(\omega-\nu) \tag{D.47}
$$

and

$$
\begin{aligned}
&= \sum_\sigma \int_0^\infty \hat{b}_\sigma(\nu,t) \left[\int_0^\infty \frac{2\Omega_i \Sigma_i^2(\omega) g_{i\sigma}(\nu)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2} \lim_{\epsilon\to 0} \left(\frac{\omega+\Omega_i}{\omega-\nu-\mathrm{i}\epsilon} - \frac{\omega-\Omega_i}{\omega+\nu} \right) \mathrm{d}\omega + \frac{(\nu+\Omega_i) g_{i\sigma}(\nu)}{\nu^2 - \Omega_i^2 z_i^*(\nu)} \right] \mathrm{d}\nu \\
&= 0.
\end{aligned}
\tag{D.48}
$$

The final equality to zero is inferred from the form of the expression in brackets in the second-to-last line above. The bracketed expression can be seen to go to zero via the same process as Eq. (C.48). An analogous process using the logic of Eq. (C.49) can be used to show that

$$
\sum_\sigma \int_0^\infty b_\sigma^\dagger(\nu,t) \sum_n \int_0^\infty P_{ni}(\omega) \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}^*(\omega) v_{n\sigma}(\omega,\nu) - s_{ni}(\omega) u_{n\sigma}^*(\omega,\nu) \right] \mathrm{d}\omega \, \mathrm{d}\nu = 0, \tag{D.49}
$$

but we will not show it explicitly here. Therefore, we have confirmed that

$$
\hat{a}_i(t) = \sum_n \int_0^\infty \lambda_n \frac{4\Omega_i^2 \Sigma_i^2(\omega)}{|\omega^2 - \Omega_i^2 z_i(\omega)|^2} \left[\frac{\omega+\Omega_i}{2\Omega_i} q_{ni}^*(\omega) \hat{C}_n(\omega,t) - s_{ni}(\omega) \hat{C}_n^\dagger(\omega,t) \right] \mathrm{d}\omega. \tag{D.50}
$$

Note that an explicit form for $\Sigma_i^2(\omega)$ has not been determined, such that the above definition for $\hat{a}_i(t)$ is not unique. We can then regard $\Sigma_i(\omega)$ and $z_i(\omega)$ as "dummy" functions, with explicit values free to be chosen as is convenient for anyone implementing the model except where they are subject to the constraints listed above.

A trial solution for the bath operators is then

$$\hat{b}_\sigma(\nu,t) = \frac{1}{\mathcal{M}_\sigma(\nu)} \sum_n \int_0^\infty K_{n\sigma}(\omega) \left[u'^*_{n\sigma}(\omega,\nu)\hat{C}'_n(\omega,t) - v_{n\sigma}(\omega,\nu)\hat{C}'^\dagger_n(\omega,t)\right] \mathrm{d}\omega. \tag{D.51}$$

The primed expansion coefficient

$$\begin{aligned} u'_{n\sigma}(\omega,\nu) &= PV\left\{\frac{1}{\omega-\nu}\right\}\sum_i g_{i\sigma}(\nu)q_{ni}(\omega) + \sum_i g_{i\sigma}(\nu)q_{ni}(\omega)[\mathrm{i}\pi + y_{i\sigma}(\omega)]\delta(\omega-\nu) \\ &= \lim_{\epsilon\to0}\frac{1}{\omega-\nu-\mathrm{i}\epsilon}\sum_i g_{i\sigma}(\nu)q_{ni}(\omega) + \sum_i g_{i\sigma}(\nu)q_{ni}(\omega)y_{i\sigma}(\omega)\delta(\omega-\nu) \end{aligned} \tag{D.52}$$

is defined such that its Dirac-delta term, which can have any complex coefficient and still allow $u'_{n\sigma}(\omega,\nu)$ to satisfy Eq. (D.13), has a prefactor $y_{i\sigma}(\omega) + \mathrm{i}\pi$ that may or may not be equal to $x_{i\sigma}(\omega)$. The primed polariton operators are similarly defined such that $\hat{C}'_n(\omega,t) = \hat{C}_n(\omega,t)|_{u_{n\sigma}\to u'_{n\sigma}}$. Expanding, we find

$$\begin{aligned} \hat{b}_\sigma(\nu,t) = \frac{1}{\mathcal{M}_\sigma(\nu)}\Bigg(&\sum_i \hat{a}_i(t)\int_0^\infty \sum_n K_{n\sigma}(\omega)\left[\frac{\omega+\Omega_i}{2\Omega_i}q_{ni}(\omega)u'^*_{n\sigma}(\omega,\nu) - s^*_{ni}(\omega)v_{n\sigma}(\omega,\nu)\right]\mathrm{d}\omega \\ &+ \sum_i \hat{a}^\dagger_i(t)\int_0^\infty \sum_n K_{n\sigma}(\omega)\left[s_{ni}(\omega)u'^*_{n\sigma}(\omega,\nu) - \frac{\omega+\Omega_i}{2\Omega_i}q^*_{ni}(\omega)v_{n\sigma}(\omega,\nu)\right]\mathrm{d}\omega \\ &+ \sum_{\sigma'}\int_0^\infty \hat{b}_{\sigma'}(\nu',t)\sum_n\int_0^\infty K_{n\sigma}(\omega)\left[u'^*_{n\sigma}(\omega,\nu)u'_{n\sigma'}(\omega,\nu') - v_{n\sigma}(\omega,\nu)v^*_{n\sigma'}(\omega,\nu')\right]\mathrm{d}\omega\,\mathrm{d}\nu' \\ &+ \sum_{\sigma'}\int_0^\infty \hat{b}^\dagger_{\sigma'}(\nu',t)\sum_n\int_0^\infty K_{n\sigma}(\omega)\left[u'^*_{n\sigma}(\omega,\nu)v_{n\sigma'}(\omega,\nu') - v_{n\sigma}(\omega,\nu)u'^*_{n\sigma'}(\omega,\nu')\right]\mathrm{d}\omega\,\mathrm{d}\nu'\Bigg). \end{aligned} \tag{D.53}$$

With

$$\begin{aligned} K_{n\sigma}(\omega) &= \lambda_n\frac{4\Omega^2\Sigma^2_\sigma(\omega)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}, \\ z_\sigma(\omega) &= 1 + \lim_{\epsilon\to0}\frac{2}{\Omega}\int_{-\infty}^\infty \frac{\Sigma^2_\sigma(\nu)}{\omega-\nu-\mathrm{i}\epsilon}\,\mathrm{d}\nu, \end{aligned} \tag{D.54}$$

in which Ω and $\Sigma_\sigma(\omega)$ are left to be constrained by downstream requirements, we can see that

$$
\begin{aligned}
&\sum_n \int_0^\infty K_{n\sigma}(\omega)\left[u'^*_{n\sigma}(\omega,\nu)u'_{n\sigma'}(\omega,\nu') - v_{n\sigma}(\omega,\nu)v^*_{n\sigma'}(\omega,\nu')\right]\mathrm{d}\omega \\
=&\sum_n \int_0^\infty \lambda_n \frac{4\Omega^2\Sigma^2_\sigma(\omega)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}\left[\left(\frac{1}{\omega-\nu+\mathrm{i}\epsilon}\sum_i g_{i\sigma}(\nu)q_{ni}(\omega) + \sum_i g_{i\sigma}(\nu)q_{ni}(\omega)y^*_{i\sigma}(\omega)\delta(\omega-\nu)\right)\right. \\
&\quad\times\left(\frac{1}{\omega-\nu'-\mathrm{i}\epsilon}\sum_j g_{j\sigma'}(\nu')q_{nj}(\omega) + \sum_j g_{j\sigma'}(\nu')q_{nj}(\omega)y_{j\sigma'}(\omega)\delta(\omega-\nu')\right) \\
&\quad\left.-\frac{1}{\omega+\nu}\frac{1}{\omega+\nu'}\sum_i g_{i\sigma}(\nu)q_{ni}(\omega)\sum_j g_{j\sigma'}(\nu')q_{nj}(\omega)\right]\mathrm{d}\omega \\
=&\sum_n \lambda_n\left[\int_0^\infty \frac{4\Omega^2\Sigma^2_\sigma(\omega)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}\left(\frac{1}{\omega-\nu+\mathrm{i}\epsilon}\frac{1}{\omega-\nu'-\mathrm{i}\epsilon} - \frac{1}{\omega+\nu}\frac{1}{\omega+\nu'}\right)\right. \\
&\quad\times\sum_{ij} g_{i\sigma}(\nu)g_{j\sigma'}(\nu')q_{ni}(\omega)q_{nj}(\omega)\,\mathrm{d}\omega \\
&\quad+\frac{4\Omega^2\Sigma^2_\sigma(\nu')}{|\nu'^2-\Omega^2 z_\sigma(\nu')|^2}\frac{1}{\nu'-\nu+\mathrm{i}\epsilon}\sum_{ij} y_{j\sigma'}(\nu')g_{i\sigma}(\nu)g_{j\sigma'}(\nu')q_{ni}(\nu')q_{nj}(\nu') \\
&\quad+\frac{4\Omega^2\Sigma^2_\sigma(\nu)}{|\nu^2-\Omega^2 z_\sigma(\nu)|^2}\frac{1}{\nu-\nu'-\mathrm{i}\epsilon}\sum_{ij} y^*_{i\sigma}(\nu)g_{i\sigma}(\nu)g_{j\sigma'}(\nu')q_{ni}(\nu)q_{nj}(\nu) \\
&\quad\left.+\frac{4\Omega^2\Sigma^2_\sigma(\nu)}{|\nu^2-\Omega^2 z_\sigma(\nu)|^2}\sum_{ij} y^*_{i\sigma}(\nu)y_{j\sigma'}(\nu)g_{i\sigma}(\nu)g_{j\sigma'}(\nu')q_{ni}(\nu)q_{nj}(\nu)\delta(\nu-\nu')\right] \\
=&\sum_i g_{i\sigma}(\nu)g_{i\sigma'}(\nu')\int_0^\infty \frac{4\Omega^2\Sigma^2_\sigma(\omega)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}\left(\frac{1}{\omega-\nu+\mathrm{i}\epsilon}\frac{1}{\omega-\nu'-\mathrm{i}\epsilon} - \frac{1}{\omega+\nu}\frac{1}{\omega+\nu'}\right)\mathrm{d}\omega \\
&\quad+\frac{4\Omega^2\Sigma^2_\sigma(\nu')}{|\nu'^2-\Omega^2 z_\sigma(\nu')|^2}\frac{1}{\nu'-\nu+\mathrm{i}\epsilon}\sum_i y_{i\sigma'}(\nu')g_{i\sigma}(\nu)g_{i\sigma'}(\nu') \\
&\quad+\frac{4\Omega^2\Sigma^2_\sigma(\nu)}{|\nu^2-\Omega^2 z_\sigma(\nu)|^2}\frac{1}{\nu-\nu'-\mathrm{i}\epsilon}\sum_i y^*_{i\sigma}(\nu)g_{i\sigma}(\nu)g_{i\sigma'}(\nu') \\
&\quad+\frac{4\Omega^2\Sigma^2_\sigma(\nu)}{|\nu^2-\Omega^2 z_\sigma(\nu)|^2}\sum_i y^*_{i\sigma}(\nu)y_{i\sigma'}(\nu')g_{i\sigma}(\nu)g_{i\sigma'}(\nu')\delta(\nu-\nu').
\end{aligned}
\tag{D.55}
$$

At this point, it is useful to note that $y_{i\sigma}(\omega)$ can be an operator instead of a function. To see why this is, one can analyze a simplified version of the definition of $u_{n\sigma}(\omega,\nu)$ (Eq. [D.15]). Specifically, if $f(x)$ is a real function of a real variable x, then the solution of $xf(x)=1$ is $f(x)=PV\{1/x\}+C\delta(x)$, where C can be any constant in the complex plane. We can generalize this problem by noting that the solution of

$$xf(x) = g(y,z,\ldots) \tag{D.56}$$

is given by

$$f(x) = PV\left\{\frac{1}{x}\right\}g(y,z,\ldots) + \mathcal{C}\{g(y,z,\ldots)\}\delta(x) \tag{D.57}$$

for nonzero $g(y,z,\ldots)$. Here, $\mathcal{C}$ is an operator on $g(y,z,\ldots)$. The solution exists for any $\mathcal{C}$ that transforms $g(y,z,\ldots)$ to a finite value, as is easily confirmed by multiplying by x and using the identities $x\,PV\{1/x\}=1$

and $x\delta(x) = 0$. Using this logic, we will define

$$y_{i\sigma}(\omega) = \frac{\omega^2 - \Omega^2 z_\sigma(\omega)}{2\Omega\Sigma_\sigma^2(\omega)} \mathcal{U}_{i\sigma}(\omega), \tag{D.58}$$

where $\mathcal{U}_{i\sigma}(\omega)$ are operators defined by

$$\begin{aligned} \mathcal{U}_{i\sigma}(\omega) f(x, y, z, \ldots) &= \mathcal{U}_{i\sigma} \left\{ f(x, y, z, \ldots) \right\} (\omega) \\ &= \begin{cases} f(x, y, z \ldots), & f(x, y, z, \ldots) \neq f(\omega), \\ \frac{w_{i\sigma}}{g_{i\sigma}(\omega)} \Sigma_\sigma(\omega) f(\omega), & f(x, y, z \ldots) = f(\omega). \end{cases} \end{aligned} \tag{D.59}$$

In other words, $\mathcal{U}_{i\sigma}(\omega)$ is an operator that acts on any function it is multiplied by and either a) reproduces the function unaltered if the the function is multivariate or does not depend on the operator argument ω, or b) produces a rescaled version of the function if the function is a univariate function of ω. Finally, $w_{i\sigma}$ is an element of the vector $\mathbf{w}_\sigma$ defined such that $\mathbf{w}_\sigma \cdot \mathbf{w}_{\sigma'} = \delta_{\sigma\sigma'}$. The simplest convention for these vectors is to define them as orthonormal basis vectors $\mathbf{w}_\sigma = \sum_i \delta_{i\sigma} \hat{\mathbf{e}}_i$, which also makes clear that, for $N > M$, $\mathcal{U}_{i\sigma}(\omega) f(\omega) = 0$ for some values of σ.

Substitution into the above expression provides

$$\begin{aligned} &\sum_n \int_0^\infty K_{n\sigma}(\omega) \left[u_{n\sigma}'^*(\omega, \nu) u_{n\sigma'}'(\omega, \nu') - v_{n\sigma}(\omega, \nu) v_{n\sigma'}^*(\omega, \nu') \right] \mathrm{d}\omega \\ &= \sum_i g_{i\sigma}(\nu) g_{j\sigma'}(\nu') \int_0^\infty \frac{4\Omega^2 \Sigma_\sigma^2(\omega)}{|\omega^2 - \Omega^2 z_\sigma(\omega)|^2} \left(\frac{1}{\omega - \nu + \mathrm{i}\epsilon} \frac{1}{\omega - \nu' - \mathrm{i}\epsilon} - \frac{1}{\omega + \nu} \frac{1}{\omega + \nu'} \right) \mathrm{d}\omega \\ &\quad + \sum_i \frac{2\Omega g_{i\sigma}(\nu) g_{i\sigma'}(\nu')}{\nu^2 - \Omega^2 z^*(\nu)} \frac{1}{\nu - \nu' - \mathrm{i}\epsilon} + \sum_i \frac{2\Omega g_{i\sigma}(\nu) g_{i\sigma'}(\nu')}{\tilde{\nu}_{\sigma'}'^2 - \Omega^2 z_\sigma(\nu')} \frac{1}{\nu' - \nu + \mathrm{i}\epsilon} \\ &\quad + \frac{1}{\Sigma_\sigma^2(\nu)} \sum_i \frac{w_{i\sigma}}{g_{i\sigma}(\nu)} \Sigma_\sigma(\nu) \frac{w_{i\sigma'}}{g_{i\sigma'}(\nu')} \Sigma_\sigma(\nu') g_{i\sigma}(\nu) g_{i\sigma'}(\nu') \delta(\nu - \nu') \\ &= 0 + \frac{\Sigma_\sigma(\nu')}{\Sigma_\sigma(\nu)} \sum_i w_{i\sigma} w_{i\sigma'} \delta(\nu - \nu') \end{aligned} \tag{D.60}$$

where we have recognized that the last term on the right-hand side is either a function of a single variable (ν or ν', equivalently) or zero, as enforced by the Dirac delta. The terms on the right-hand side not proportional to a Dirac delta sum to zero via the process shown in Eq. (C.53) such that

$$\begin{aligned} &\sum_n \int_0^\infty K_{n\sigma}(\omega) \left[u_{n\sigma}'^*(\omega, \nu) u_{n\sigma'}'(\omega, \nu') - v_{n\sigma}(\omega, \nu) v_{n\sigma'}^*(\omega, \nu') \right] \mathrm{d}\omega \\ &= \frac{\Sigma_\sigma(\nu')}{\Sigma_\sigma(\nu)} \delta_{\sigma\sigma'} \delta(\nu - \nu') \\ &= \delta_{\sigma\sigma'} \delta(\nu - \nu') \end{aligned} \tag{D.61}$$

and

$$\begin{aligned} \sum_{\sigma'} \int_0^\infty \hat{b}_{\sigma'}(\nu', t) \sum_n \int_0^\infty K_{n\sigma}(\omega) \left[u_{n\sigma}'^*(\omega, \nu) u_{n\sigma'}'(\omega, \nu') - v_{n\sigma}(\omega, \nu) v_{n\sigma'}^*(\omega, \nu') \right] \mathrm{d}\omega \, \mathrm{d}\nu \\ = \sum_{\sigma'} \int_0^\infty \hat{b}_{\sigma'}(\nu', t) \delta_{\sigma\sigma'} \delta(\nu - \nu') \, \mathrm{d}\nu' \\ = \hat{b}_\sigma(\nu, t). \end{aligned} \tag{D.62}$$

Next, we can look at the integral

$$
\begin{aligned}
&\sum_n \int_0^\infty K_{n\sigma}(\omega)\left[u'^*_{n\sigma}(\omega,\nu)v_{n\sigma'}(\omega,\nu') - v_{n\sigma}(\omega,\nu)u'^*_{n\sigma'}(\omega,\nu')\right]\mathrm{d}\omega \\
&= \int_0^\infty \sum_i g_{i\sigma}(\nu)g_{i\sigma'}(\nu')\frac{4\Omega^2\Sigma^2_\sigma(\omega)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}\left(\frac{1}{\omega-\nu+\mathrm{i}\epsilon}\frac{1}{\omega+\nu'} - \frac{1}{\omega-\nu'-\mathrm{i}\epsilon}\frac{1}{\omega+\nu}\right) \\
&\qquad + \sum_i g_{i\sigma}(\nu)g_{j\sigma'}(\nu')\frac{2\Omega}{\nu^2-\Omega^2 z_\sigma(\nu)}\frac{1}{\nu+\nu'} - \sum_i g_{i\sigma}(\nu)g_{j\sigma'}(\nu')\frac{2\Omega}{\nu'^2-\Omega^2 z_\sigma(\nu')}\frac{1}{\nu+\nu'} \\
&= 0,
\end{aligned}
\tag{D.63}
$$

where the equality with zero is inferred by the analogous forms of the above expression and that of Eq. (C.52). Finally, we can see that

$$
\begin{aligned}
&\sum_n \int_0^\infty K_{n\sigma}(\omega)\left[\frac{\omega+\Omega_i}{2\Omega_i}q_{ni}(\omega)u'^*_{n\sigma}(\omega,\nu) - s^*_{ni}(\omega)v_{n\sigma}(\omega,\nu)\right]\mathrm{d}\omega \\
&= \int_0^\infty \frac{4\Omega^2\Sigma^2_\sigma(\omega)g_{i\sigma}(\nu)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}\left[\frac{\omega+\Omega_i}{2\Omega_i}\frac{1}{\omega-\nu+\mathrm{i}\epsilon} - \frac{\omega-\Omega_i}{2\Omega_i}\frac{1}{\omega+\nu}\right]\mathrm{d}\omega + \frac{\nu+\Omega_i}{2\Omega_i}\frac{2\Omega g_{i\sigma}(\nu)}{\nu^2-\Omega^2 z_\sigma(\nu)} \\
&= 0
\end{aligned}
\tag{D.64}
$$

via the process of Eq. (C.48) (note the overall complex conjugate) and

$$
\begin{aligned}
&\sum_n \int_0^\infty K_{n\sigma}(\omega)\left[s_{ni}(\omega)u'^*_{n\sigma}(\omega,\nu) - \frac{\omega+\Omega_i}{2\Omega_i}q^*_{ni}(\omega)v_{n\sigma}(\omega,\nu)\right]\mathrm{d}\omega \\
&= \int_0^\infty \frac{4\Omega^2\Sigma^2_\sigma(\omega)g_{i\sigma}(\nu)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}\left[\frac{\omega-\Omega_i}{2\Omega_i}\frac{1}{\omega-\nu+\mathrm{i}\epsilon} - \frac{\omega+\Omega_i}{2\Omega_i}\frac{1}{\omega+\nu}\right]\mathrm{d}\omega + \frac{\nu-\Omega_i}{2\Omega_i}\frac{2\Omega g_{i\sigma}(\nu)}{\nu^2-\Omega^2 z_\sigma(\nu)} \\
&= 0
\end{aligned}
\tag{D.65}
$$

via the logic of Eq. (C.49) (note the added complex conjugate and negative sign). Therefore, we have shown that, with

$$\mathcal{m}_\sigma(\nu) = 1, \tag{D.66}$$

the reservoir operators can be retrieved via

$$\hat{b}_\sigma(\nu,t) = \sum_n \int_0^\infty \lambda_n \frac{4\Omega^2\Sigma^2_\sigma(\omega)}{|\omega^2-\Omega^2 z_\sigma(\omega)|^2}\left[u'^*_{n\sigma}(\omega,\nu)\hat{C}'_n(\omega,t) - v_{n\sigma}(\omega,\nu)\hat{C}'^\dagger_n(\omega,t)\right]\mathrm{d}\omega. \tag{D.67}$$

Note that, aside from the constraint that $\Sigma^2_\sigma(\omega)$ has units of coupling strength squared and obeys the property $\Sigma^2_\sigma(-\omega) = -\Sigma^2_\sigma(\omega)$, the values of Ω and $\Sigma^2_\sigma(\omega)$ can be freely chosen. Therefore, the values of $z_\sigma(\omega)$ and $\hat{b}_\sigma(\nu,t)$ are not unique.

E Details of the Helmholtz Expansion of the Dyadic Dirac Delta in Spherical Coordinates

Consider the Laplace equation

$$\nabla^2\psi_{p\ell m}(\mathbf{r},k) = -k^2\psi_{p\ell m}(\mathbf{r},k) \tag{E.1}$$

with the corresponding boundary condition $\lim_{r\to\infty} \psi_{p\ell m}(\mathbf{r}, k) \to 0$. The solutions, which take the form

$$\psi_{p\ell m}(\mathbf{r}, k) = \sqrt{K_{\ell m}} j_\ell(kr) P_{\ell m}(\cos\theta) S_p(m\phi), \tag{E.2}$$

are therefore defined here as eigenfunctions of the Laplacian operator in spherical coordinates with eigenvalues $-k^2$. The individual factors of the eigenfunctions are defined in Appendix B.

These eigenfunctions possess a special completeness property that is crucial to our ability to reconstruct complicated three-dimensional functions as linear combinations of simpler harmonic functions. To see how this completeness property arises, one can expand the Laplacian in spherical coordinates to find

$$\nabla^2\{\cdot\} = \frac{1}{r^2}\frac{\partial}{\partial r}\left\{r^2 \frac{\partial}{\partial r}\{\cdot\}\right\} + \frac{1}{r^2 \sin\theta}\frac{\partial}{\partial\theta}\left\{\sin\theta\frac{\partial}{\partial\theta}\{\cdot\}\right\} + \frac{1}{r^2\sin^2\theta}\frac{\partial^2}{\partial\phi^2}\{\cdot\}. \tag{E.3}$$

Therefore, dropping the indices (p, ℓ, m) for a moment, the action of $\nabla^2 + k^2$ on any separable eigenfunction $\psi(\mathbf{r}) = R(r)\Theta(\theta)\Phi(\phi)$ can be seen to produce zero such that

$$\begin{aligned}
&\left(\nabla^2 + k^2\right)\{\psi(\mathbf{r})\} = \frac{r^2}{\psi(\mathbf{r})}\left(\nabla^2 + k^2\right)\{\psi(\mathbf{r})\}, \\
&= \frac{r}{R(r)}\left(k^2 r R(r) + 2\frac{\partial R(r)}{\partial r} + r\frac{\partial^2 R(r)}{\partial r^2}\right) + \frac{1}{\tan(\theta)\Theta(\theta)}\left(\frac{\partial\Theta(\theta)}{\partial\theta} + \frac{\partial^2\Theta(\theta)}{\partial\theta^2}\right) + \frac{1}{\sin^2(\theta)\Phi(\phi)}\frac{\partial^2\Phi(\phi)}{\partial\phi^2} \\
&= 0.
\end{aligned} \tag{E.4}$$

For any fixed values of θ and ϕ, the above differential equation in r is a particular form of Sturm-Liouville equation known as the spherical Bessel equation. As is discussed for the one-dimensional case in Titchmarsh,[2] solutions to Sturm-Liouville equations with specified boundary conditions form complete sets of orthogonal eigenfunctions from which any twice-differentiable function can be exactly reconstructed within the boundaries. Therefore, the set of functions $\{R(r)\}$ that are solutions to the above equation can completely describe any one-dimensional function of r on the interval $(-\infty, \infty)$.

The other two terms of Eq. (E.4) also form Sturm-Liouville equations, the associated Legendre and harmonic differential equations, when only θ or ϕ is allowed to vary, respectively. Therefore, $\{\Theta(\theta)\}$ and $\{\Phi(\phi)\}$ are complete sets on $\theta \in [0, \pi)$ and $\phi \in [0, 2\pi)$, respectively, and the eigenfunctions $\{\psi(\mathbf{r})\}$ are guaranteed to be able to reproduce any twice-differentiable scalar field in $\mathbb{R}^3$.

To extend our logic to the reconstruction of vector fields, we can note that three sets of vector eigenfunctions $\hat{\mathbf{e}}_i\psi_{p\ell m}(\mathbf{r}, k)$ with $i = \{x, y, z\}$ can be defined that clearly are able to reproduce any vector field on $\mathbb{R}^3$. More precisely, these three sets of vector eigenfunctions are non-collinear[4] and contain independent eigenfunctions of (the Sturm-Liouville operator) ∇^2. The first property can be deduced from the nonzero cross-products between any two eigenfunctions from two different sets, and the second property can be deduced from the fact that ∇^2 and $\hat{\mathbf{e}}_i$ commute. In other words, $\nabla^2\{\hat{\mathbf{e}}_i\psi_{p\ell m}(\mathbf{r}, k)\} = \hat{\mathbf{e}}_i\nabla^2\{\psi_{p\ell m}(\mathbf{r}, k)\} = -k^2\hat{\mathbf{e}}_i\psi_{p\ell m}(\mathbf{r}, k)$.

Because Cartesian vector functions are inconvenient to use for problems with spherical symmetry, Hansen[5] invented, as an alternative, the three functions $\mathbf{L}_{p\ell m}(\mathbf{r}, k)$, $\mathbf{M}_{p\ell m}(\mathbf{r}, k)$, and $\mathbf{N}_{p\ell m}(\mathbf{r}, k)$ as an alternate basis. These three functions satisfy the two criteria above, the latter of which can be directly confirmed via $\nabla^2\{\nabla\psi_{p\ell m}(\mathbf{r}, k)\} = \nabla\{\nabla^2\psi_{p\ell m}(\mathbf{r}, k)\}$, $\nabla^2\{\nabla \times \{\mathbf{r}\psi_{p\ell m}(\mathbf{r}, k)\}\} = \nabla \times \{\mathbf{r}\nabla^2\psi_{p\ell m}(\mathbf{r}, k)\}$, and $\nabla^2\{\nabla \times \nabla \times \{\mathbf{r}\psi_{p\ell m}(\mathbf{r}, k)\}\} = \nabla \times \nabla \times \{\mathbf{r}\nabla^2\psi_{p\ell m}(\mathbf{r}, k)\}$. The non-collinearity of the three sets of functions can be seen by taking the cross-product of any two basis functions and noting that it is nonzero.

We are thus free to expand in terms of $\mathbf{L}_{p\ell m}(\mathbf{r}, k)$, $\mathbf{M}_{p\ell m}(\mathbf{r}, k)$, and/or $\mathbf{N}_{p\ell m}(\mathbf{r}, k)$ any of the vector fields of our system that are smooth throughout the universe. As a matter of clarity, it is useful to show that we can expand the dyadic Dirac delta in terms of these basis functions as well. Assuming the Dirac delta to be the sharp limit of a smooth bump function, we can let

$$\mathbf{1}_2\delta(\mathbf{r} - \mathbf{r}') = \sum_{p\ell m}\int_0^\infty \left[\mathbf{A}_{p\ell m}(\mathbf{r}', k)\mathbf{L}_{p\ell m}(\mathbf{r}, k) + \mathbf{B}_{p\ell m}(\mathbf{r}', k)\mathbf{M}_{p\ell m}(\mathbf{r}, k) + \mathbf{C}_{p\ell m}(\mathbf{r}', k)\mathbf{N}_{p\ell m}(\mathbf{r}, k)\right] \mathrm{d}k, \tag{E.5}$$

wherein $\mathbf{A}_{p\ell m}(\mathbf{r}', k)$, $\mathbf{B}_{p\ell m}(\mathbf{r}', k)$, and $\mathbf{C}_{p\ell m}(\mathbf{r}', k)$ are unknown vector fields. Using the orthogonality condition of Eq. (B.11), we can see that

$$\begin{aligned}
\int \mathbf{1}_2\delta(\mathbf{r}-\mathbf{r}')\cdot\mathbf{L}_{p'\ell'm'}(\mathbf{r},k')\,\mathrm{d}^3\mathbf{r} = \mathbf{L}_{p'\ell'm'}(\mathbf{r}',k')& \\
&= \sum_{p\ell m}\int_0^\infty\int \mathbf{A}_{p\ell m}(\mathbf{r}',k)\mathbf{L}_{p\ell m}(\mathbf{r},k)\cdot\mathbf{L}_{p'\ell'm'}(\mathbf{r},k')\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}k \\
&= \sum_{p\ell m}\int_0^\infty \mathbf{A}_{p\ell m}(\mathbf{r}',k)\frac{2\pi^2}{k^2}\delta(k-k')(1-\delta_{p0}\delta_{m0})\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'} \\
&= \frac{2\pi^2}{k'^2}(1-\delta_{p'1}\delta_{m'0})\mathbf{A}_{p'\ell'm'}(\mathbf{r}',k').
\end{aligned} \tag{E.6}$$

Further, with $\mathbf{L}_{p'\ell'm'}(\mathbf{r}',k') = \mathbf{L}_{p'\ell'm'}(\mathbf{r}',k')(1-\delta_{p'1}\delta_{m'0})$, we can see that

$$\mathbf{A}_{p\ell m}(\mathbf{r}',k) = \frac{k^2}{2\pi^2}\mathbf{L}_{p\ell m}(\mathbf{r}',k). \tag{E.7}$$

Repeating this process for the transverse vector spherical harmonics using (B.11), we can see that

$$\begin{aligned}
\mathbf{B}_{p\ell m}(\mathbf{r}',k) &= \frac{k^2}{2\pi^2}\mathbf{M}_{p\ell m}(\mathbf{r}',k), \\
\mathbf{C}_{p\ell m}(\mathbf{r}',k) &= \frac{k^2}{2\pi^2}\mathbf{N}_{p\ell m}(\mathbf{r}',k),
\end{aligned} \tag{E.8}$$

such that

$$\mathbf{1}_2\delta(\mathbf{r}-\mathbf{r}') = \sum_{p\ell m}\int_0^\infty \frac{k^2}{2\pi^2}\left[\mathbf{L}_{p\ell m}(\mathbf{r}',k)\mathbf{L}_{p\ell m}(\mathbf{r},k)+\mathbf{M}_{p\ell m}(\mathbf{r}',k)\mathbf{M}_{p\ell m}(\mathbf{r},k)+\mathbf{N}_{p\ell m}(\mathbf{r}',k)\mathbf{N}_{p\ell m}(\mathbf{r},k)\right]\mathrm{d}k. \tag{E.9}$$

This in turn can be separated into its transverse and longitudinal parts as

$$\begin{aligned}
[\mathbf{1}_2\delta(\mathbf{r}-\mathbf{r}')]_\perp &= \sum_{p\ell m}\int_0^\infty \frac{k^2}{2\pi^2}\left[\mathbf{M}_{p\ell m}(\mathbf{r}',k)\mathbf{M}_{p\ell m}(\mathbf{r},k)+\mathbf{N}_{p\ell m}(\mathbf{r}',k)\mathbf{N}_{p\ell m}(\mathbf{r},k)\right]\mathrm{d}k, \\
[\mathbf{1}_2\delta(\mathbf{r}-\mathbf{r}')]_\parallel &= \sum_{p\ell m}\int_0^\infty \frac{k^2}{2\pi^2}\mathbf{L}_{p\ell m}(\mathbf{r}',k)\mathbf{L}_{p\ell m}(\mathbf{r},k)\,\mathrm{d}k.
\end{aligned} \tag{E.10}$$

Finally, we also deal in this manuscript with vector fields that are smooth within a finite radial boundary a but are discontinuous across the boundary. This problem can be attacked in two ways, both of which will produce the same outcome. The more direct but mathematically complicated route is to represent the sharp boundary as the steep limit of a sigmoid function. This route has nice symmetry with our previous discussion as it involves the expansion of the vector field in the full eigenfunction basis of $\mathbb{R}^3$. However, it will require numerical evaluation of complicated overlap integrals involving spherical Bessel functions and will not, in general, produce expressions with advantageous explanatory power.

The second route begins by setting our Sturm-Liouville boundary conditions such that we only use a subset of the total number vector spherical harmonics. More specifically, we will only use vector spherical harmonics in our expansion that have one or more components that go to zero at $r = a$, with the precise boundary requirements set by physical arguments. Because our boundary condition is a function of r, it is the radial index k that will be restricted to certain values, and the completeness of the harmonic expansion of the sphere will depend on whether a set of vector harmonics with the "allowed" values of k can be generated from a restricted set of scalar eigenfunctions $\psi_{p\ell m}(\mathbf{r}, k)$ that is nonetheless complete within $r < a$.

Conveniently, this set of scalar eigenfunctions is simple to define. The physical boundary conditions on the vector harmonics, as described in the main text, require their boundary-parallel components to go to zero at $r = a$ and their radial components to be nonzero. This behavior can be reproduced from scalar eigenfunctions $\psi_{p\ell m}(\mathbf{r}, k_{\ell n})$, where $k_{\ell n}a$ is the n^{th} nonzero root of the ℓ^{th} spherical Bessel function. These scalar eigenfunctions therefore all go to zero at the boundary. This is a valid Sturm-Liouville boundary condition, such that they form a complete set within the sphere. Therefore, we can rebuild our Dirac delta from the discrete set of vector harmonics that have the permitted wavenumbers $k_{\ell n}$. Explicitly,

$$\begin{aligned}\mathbf{1}_2\Theta(a-r)\delta(\mathbf{r}-\mathbf{r}') = \Theta(a-r)\sum_{p\ell mn}&[\mathbf{A}_{p\ell mn}(\mathbf{r}')\mathbf{L}_{p\ell m}(\mathbf{r}, k_{\ell n}) \\ &+\mathbf{B}_{p\ell mn}(\mathbf{r}')\mathbf{M}_{p\ell m}(\mathbf{r}, k_{\ell n}) + \mathbf{C}_{p\ell mn}(\mathbf{r}')\mathbf{N}_{p\ell m}(\mathbf{r}, k_{\ell n})]\,.\end{aligned} \tag{E.11}$$

Projecting as before, we can see that, by the orthonormality conditions of Appendix B,

$$\begin{aligned}\mathbf{A}_{p\ell mn}(\mathbf{r}') &= \frac{1}{2\pi a^3 j_{\ell+1}^2(k_{\ell n}a)}\mathbf{L}_{p\ell m}(\mathbf{r}, k_{\ell n}), \\ \mathbf{B}_{p\ell mn}(\mathbf{r}') &= \frac{1}{2\pi a^3 j_{\ell+1}^2(k_{\ell n}a)}\mathbf{M}_{p\ell m}(\mathbf{r}, k_{\ell n}), \\ \mathbf{C}_{p\ell mn}(\mathbf{r}') &= \frac{1}{2\pi a^3 j_{\ell+1}^2(k_{\ell n}a)}\mathbf{N}_{p\ell m}(\mathbf{r}, k_{\ell n}).\end{aligned} \tag{E.12}$$

Therefore,

$$\begin{aligned}\mathbf{1}_2\Theta(a-r)\delta(\mathbf{r}-\mathbf{r}') = \Theta(a-r)\sum_{p\ell mn}&\frac{1}{2\pi a^3 j_{\ell+1}^2(k_{\ell n}a)} \\ \times\,[\mathbf{L}_{p\ell m}(\mathbf{r}', k_{\ell n})\mathbf{L}_{p\ell m}(\mathbf{r}, k_{\ell n}) + \mathbf{M}_{p\ell m}(\mathbf{r}', k_{\ell n})&\mathbf{M}_{p\ell m}(\mathbf{r}, k_{\ell n}) + \mathbf{N}_{p\ell m}(\mathbf{r}', k_{\ell n})\mathbf{N}_{p\ell m}(\mathbf{r}, k_{\ell n})]\,.\end{aligned} \tag{E.13}$$

F Dependent Matter Degrees of Freedom: the Dielectric Picture

We have touched on this picture already in the preceding sections, but it is useful to reiterate that the Euler-Lorentz equations derivable from the system Lagrangian L state clearly that the degrees of freedom of the matter and electromagnetic field are coupled. More explicitly, there are four Euler-Lorentz equations of the form

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial\mathscr{L}}{\partial\dot{F}(\mathbf{r},t)}\right\} = \frac{\partial\mathscr{L}}{\partial F(\mathbf{r},t)} - \sum_{i=1}^{3}\frac{\partial}{\partial r_i}\frac{\partial\mathscr{L}}{\partial\left(\frac{\partial F(\mathbf{r},t)}{\partial r_i}\right)}, \tag{F.1}$$

where $F(\mathbf{r}, t)$ takes the place of the scalar potential or a component of the vector potential, matter displacement field, or reservoir displacement field.

In the following derivations, we will analyze the Euler-Lorentz equations of the modified Lagrangian density

$$\bar{\mathscr{L}} = \mathscr{L} - \frac{1}{4\pi c}\nabla\Phi(\mathbf{r}, t)\cdot\dot{\mathbf{A}}(\mathbf{r}, t) + \mathscr{L}_{NL}. \tag{F.2}$$

The second term on the right-hand side simplifies our mathematics without changing the motion of any observables. Its integral over all space is zero due to the Helmholtz orthogonality of its longitudinal first factor and transverse second factor such that it cannot contribute to the Lagrangian. The third term on the right-hand side builds in nonlinear material motion via

$$\mathscr{L}_{\text{NL}} = -\frac{\sigma_0}{3}\mu\Theta(\mathbf{r}\in\mathbb{V})\mathbf{Q}(\mathbf{r}, t)\cdot[\mathbf{Q}(\mathbf{r}, t)\cdot\mathbf{1}_3\cdot\mathbf{Q}(\mathbf{r}, t)]\,, \tag{F.3}$$

and is useful for providing a slight generalization of the dielectric picture.

The equations of motion the four Euler-Lagrange equations lead to are

$$\begin{aligned}
&\mu\ddot{\mathbf{Q}}(\mathbf{r},t) + \eta\int_0^\infty v(\nu)\dot{\mathbf{Q}}_\nu(\mathbf{r},t)\,\mathrm{d}\nu + \mu\omega_0^2\mathbf{Q}(\mathbf{r},t) + \mu\sigma_0\mathbf{Q}(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}(\mathbf{r},t) \\
&\qquad\qquad = \Theta(\mathbf{r}\in\mathbb{V})e\eta\left(-\nabla\Phi(\mathbf{r},t) - \frac{1}{c}\dot{\mathbf{A}}(\mathbf{r})\right), \\
&\mu\ddot{\mathbf{Q}}_\nu(\mathbf{r},t) + \mu\nu^2\mathbf{Q}_\nu(\mathbf{r},t) = \eta v(\nu)\dot{\mathbf{Q}}(\mathbf{r},t), \\
&-\nabla\cdot\nabla\Phi(\mathbf{r},t) = 4\pi\rho_f(\mathbf{r},t) - 4\pi e\eta\nabla\cdot\{\Theta(\mathbf{r}\in\mathbb{V})\mathbf{Q}(\mathbf{r},t)\}, \\
&\nabla\times\nabla\times\mathbf{A}(\mathbf{r},t) - \frac{1}{c^2}\ddot{\mathbf{A}}(\mathbf{r},t) = \frac{4\pi}{c}\left[\Theta(\mathbf{r}\in\mathbb{V})e\eta\dot{\mathbf{Q}}(\mathbf{r},t) + \mathbf{J}_f(\mathbf{r},t)\right] - \frac{\nabla\dot{\Phi}(\mathbf{r},t)}{c} \\
&\qquad\qquad = \frac{4\pi}{c}\left[\Theta(\mathbf{r}\in\mathbb{V})e\eta\dot{\mathbf{Q}}^\perp(\mathbf{r},t) + \mathbf{J}_f^\perp(\mathbf{r},t)\right].
\end{aligned} \tag{F.4}$$

The most pertient equation to analyze here is the second. Using the Green's function (valid for $r < a$)

$$G(t-t') = \frac{\Theta(t-t')}{\mu\nu}\sin(\nu[t-t']) \tag{F.5}$$

of the oscillator differential equation

$$\mu\ddot{G}(t-t') + \mu\nu^2 G(t-t') = \begin{cases}\delta(t-t'), & t > t'; \\ 0, & t < t';\end{cases} \tag{F.6}$$

we can see that

$$\begin{aligned}
\dot{\mathbf{Q}}_\nu(\mathbf{r},t) &= \dot{\mathbf{Q}}_\nu^{(0)}(\mathbf{r},t) + \frac{\mathrm{d}}{\mathrm{d}t}\int_{-\infty}^{\infty} G(t-t')\eta v(\nu)\dot{\mathbf{Q}}(\mathbf{r},t')\,\mathrm{d}t' \\
&= \dot{\mathbf{Q}}_\nu^{(0)}(\mathbf{r},t) + \int_{-\infty}^{\infty}\frac{\Theta(t-t')}{\mu}\cos(\nu[t-t'])\eta v(\nu)\dot{\mathbf{Q}}(\mathbf{r},t')\,\mathrm{d}t'.
\end{aligned} \tag{F.7}$$

where $\dot{\mathbf{Q}}_\nu^{(0)}(\mathbf{r},t)$ is the ν^{th} velocity field at t generated by stimuli far in the past (formally, at the initial time $-\infty$). We can then define a loss function

$$\gamma_0(t-t') = \int_0^\infty \frac{\eta^2 v^2(\nu)}{\mu^2}\cos(\nu[t-t'])\,\mathrm{d}\nu \tag{F.8}$$

and a thermal noise field (dimensions of force per unit volume)

$$\mathbf{N}_{\text{therm}}(\mathbf{r},t) = -\int_0^\infty \eta v(\nu)\dot{\mathbf{Q}}_\nu^{(0)}(\mathbf{r},t)\,\mathrm{d}\nu \tag{F.9}$$

such that the first equation of Eq. (F.4) becomes

$$\begin{aligned}
&\mu\ddot{\mathbf{Q}}(\mathbf{r},t) + \mu\int_{-\infty}^{t}\gamma_0(t-t')\dot{\mathbf{Q}}(\mathbf{r},t')\,\mathrm{d}t' + \mu\omega_0^2\mathbf{Q}(\mathbf{r},t) \\
&\quad + \mu\sigma_0\mathbf{Q}(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}(\mathbf{r},t) = \Theta(\mathbf{r}\in\mathbb{V})e\eta\left(-\nabla\Phi(\mathbf{r},t) - \frac{1}{c}\dot{\mathbf{A}}(\mathbf{r})\right) + \mathbf{N}_{\text{therm}}(\mathbf{r},t).
\end{aligned} \tag{F.10}$$

Solutions to Eq. (F.10) are simplest in the case where σ_0 is a "small" quantity. We will not belabor the exact definition of "small" here, other than to say that we will assume σ_0 to be linearly proportional to a characteristically small number λ that can be used to expand the matter displacement field as a perturbation series:

$$\begin{aligned}\mathbf{Q}(\mathbf{r},t) &= \mathbf{Q}^{(1)}(\mathbf{r},t) + \mathbf{Q}^{(2)}(\mathbf{r},t) + \ldots \\ &= \sum_{n=1}^{\infty} \lambda^{n-1} \mathfrak{Q}^{(n)}(\mathbf{r},t).\end{aligned} \tag{F.11}$$

Here, the functions $\mathfrak{Q}^{(n)}(\mathbf{r},t)$ are of roughly the same order of magnitude at each order n such that it is the different powers of λ that separate the terms in the series. Using this expansion, we find that

$$\begin{aligned}&\mu\ddot{\mathbf{Q}}^{(1)}(\mathbf{r},t) + \mu \int_{-\infty}^{t} \gamma_0(t-t')\dot{\mathbf{Q}}^{(1)}(\mathbf{r},t')\,\mathrm{d}t' + \mu\omega_0^2\mathbf{Q}^{(1)}(\mathbf{r},t) = \Theta(\mathbf{r}\in\mathbb{V})e\eta\mathbf{E}(\mathbf{r},t) + \mathbf{N}_{\text{therm}}(\mathbf{r},t), \\ &\mu\ddot{\mathbf{Q}}^{(2)}(\mathbf{r},t) + \mu \int_{-\infty}^{t} \gamma_0(t-t')\dot{\mathbf{Q}}^{(2)}(\mathbf{r},t')\,\mathrm{d}t' + \mu\omega_0^2\mathbf{Q}^{(2)}(\mathbf{r},t) \\ &\qquad\qquad = -\mu\sigma_0\mathbf{Q}^{(1)}(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}^{(1)}(\mathbf{r},t)\end{aligned} \tag{F.12}$$

where we have let $\mathbf{E}(\mathbf{r},t) = -\nabla\Phi(\mathbf{r},t) - \dot{\mathbf{A}}(\mathbf{r},t)/c$.

Further, we can simplify our analysis by choosing the simple loss function $\gamma_0(t-t') = \gamma_0\delta(t-t')$. We can see that this is achievable with the choice of coupling function

$$v(\nu) = \frac{\mu}{\eta}\sqrt{\frac{\gamma_0}{\pi}}, \tag{F.13}$$

as can be seen through the application of the identity $\delta(t-t') = \int_0^\infty 2\cos(\nu[t-t'])\,\mathrm{d}\nu/2\pi$. Using this simplification, we can take the Fourier transform of both lines of Eq. (F.12) to see

$$\begin{aligned}&\mathbf{Q}^{(1)}(\mathbf{r},\omega)\mu\left[-\omega^2 - \mathrm{i}\omega\gamma_0 + \omega_0^2\right] = \Theta(\mathbf{r}\in\mathbb{V})e\eta\mathbf{E}(\mathbf{r},\omega) + \mathbf{N}_{\text{therm}}(\omega), \\ &\mathbf{Q}^{(2)}(\mathbf{r},\omega)\mu\left[-\omega^2 - \mathrm{i}\omega\gamma_0 + \omega_0^2\right] = -\mu\sigma_0\int_{-\infty}^{\infty} \mathbf{Q}^{(1)}(\mathbf{r},\omega')\cdot\mathbf{1}_3\cdot\mathbf{Q}^{(1)}(\mathbf{r},\omega-\omega')\,\mathrm{d}\omega'.\end{aligned} \tag{F.14}$$

This form of the equations of motion of the matter displacement fields is simple to connect back to a dielectric picture through the definition of the polarization field $\mathbf{P}(\mathbf{r},\omega) = e\eta\Theta(\mathbf{r}\in\mathbb{V})\mathbf{Q}(\mathbf{r},\omega)$. Generally, this is done without thermal fluctuations, so we'll let $\mathbf{N}_{\text{therm}}(\mathbf{r},\omega)\to 0$ for now. The polarization field can be expanded as a perturbation series analogous to that of the displacement field, such that $\mathbf{P}(\mathbf{r},\omega) = \sum_n \mathbf{P}^{(n)}(\mathbf{r},\omega)$. This expansion can further be connected to a dielectric model through the definitions $\mathbf{P}^{(1)}(\mathbf{r},\omega) = \chi^{(1)}(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega)$ and $\mathbf{P}^{(2)}(\mathbf{r},\omega) = \int_{-\infty}^{\infty} \mathbf{E}(\mathbf{r},\omega')\cdot\boldsymbol{\chi}^{(2)}(\mathbf{r};\omega',\omega-\omega')\cdot\mathbf{E}(\mathbf{r},\omega-\omega')\,\mathrm{d}\omega'$. Explicitly, we can use these definitions along with a substitution within Eq. (F.14) to see that

$$\begin{aligned}\chi^{(1)}(\mathbf{r},\omega) &= \Theta(\mathbf{r}\in\mathbb{V})\frac{e^2\eta^2}{\mu}\frac{1}{\omega_0^2-\omega^2-\mathrm{i}\omega\gamma_0}, \\ \boldsymbol{\chi}^{(2)}(\mathbf{r};\omega',\omega-\omega') &= -\frac{3e^3\eta^3\sigma_0}{\mu^2}\frac{\Theta(\mathbf{r}\in\mathbb{V})\mathbf{1}_3}{(\omega_0^2-\omega^2-\mathrm{i}\omega\gamma_0)(\omega_0^2-\omega'^2-\mathrm{i}\omega'\gamma_0)(\omega_0^2-[\omega-\omega']^2-\mathrm{i}[\omega-\omega']\gamma_0)}.\end{aligned} \tag{F.15}$$

We are now in a position to connect our microscopic model back to a Green's-function solution to the system. Explicitly, all we need to do is redefine the last two lines of Eq. (F.4) in terms of the polarization field $\mathbf{P}(\mathbf{r},\omega) = e\eta\Theta(\mathbf{r}\in\mathbb{V})\mathbf{Q}(\mathbf{r},t)$. As is suggested by the susceptibility definitions above, this turns out to be most convenient to do in Fourier space, such that

$$\begin{aligned}-\nabla\cdot\nabla\Phi(\mathbf{r},\omega) &= 4\pi\rho_f(\mathbf{r},\omega) - 4\pi\nabla\cdot\mathbf{P}(\mathbf{r},\omega), \\ \nabla\times\nabla\times\mathbf{A}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\mathbf{A}(\mathbf{r},\omega) &= -\frac{4\pi\mathrm{i}\omega}{c}\mathbf{P}^{\perp}(\mathbf{r},\omega) + \frac{4\pi}{c}\mathbf{J}_f^{\perp}(\mathbf{r},\omega).\end{aligned} \tag{F.16}$$

We can then use our definitions for the susceptibilities to build the canonical Helmholtz equations for inhomogeneous dielectrics. In particular, the first equation above, in combination with the charge continuity equation, provides us with

$$\nabla\Phi(\mathbf{r},\omega) = \frac{4\pi\mathrm{i}}{\omega}\mathbf{J}^{\|}(\mathbf{r},\omega) + 4\pi\mathbf{P}^{\|}(\mathbf{r},\omega) \tag{F.17}$$

such that

$$\begin{aligned}
&\nabla\times\nabla\times\mathbf{A}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\mathbf{A}(\mathbf{r},\omega) - \frac{\mathrm{i}\omega}{c}\nabla\Phi(\mathbf{r},\omega)\\
&= \nabla\times\nabla\times\mathbf{A}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\left[\mathbf{A}(\mathbf{r},\omega) + \frac{\mathrm{i}c}{\omega}\nabla\Phi(\mathbf{r},\omega)\right]\\
&= \nabla\times\nabla\times\left[\mathbf{A}(\mathbf{r},\omega) + \frac{\mathrm{i}c}{\omega}\nabla\Phi(\mathbf{r},\omega)\right] - \frac{\omega^2}{c^2}\left[\mathbf{A}(\mathbf{r},\omega) + \frac{\mathrm{i}c}{\omega}\nabla\Phi(\mathbf{r},\omega)\right]\\
&= -\frac{\mathrm{i}c}{\omega}\left[\nabla\times\nabla\times\mathbf{E}(\mathbf{r},t) - \frac{\omega^2}{c^2}\mathbf{E}(\mathbf{r},\omega)\right]\\
&= -\frac{4\pi\mathrm{i}\omega}{c}\mathbf{P}^{\perp}(\mathbf{r},\omega) - \frac{4\pi\mathrm{i}\omega}{c}\mathbf{P}^{\|}(\mathbf{r},\omega) + \frac{4\pi}{c}\mathbf{J}_f^{\perp}(\mathbf{r},\omega) + \frac{4\pi}{c}\mathbf{J}_f^{\|}(\mathbf{r},\omega)
\end{aligned} \tag{F.18}$$

and, after recombination of the Helmholtz components of the source terms,

$$\nabla\times\nabla\times\mathbf{E}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\mathbf{E}(\mathbf{r},\omega) = \frac{4\pi\omega^2}{c^2}\mathbf{P}(\mathbf{r},\omega) + \frac{4\pi\mathrm{i}\omega}{c}\mathbf{J}_f(\mathbf{r},\omega). \tag{F.19}$$

Expanding $\mathbf{P}(\mathbf{r},\omega)$ up to second order and rearranging gives

$$\begin{aligned}
\nabla\times\nabla\times\mathbf{E}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\left[1+4\pi\chi^{(1)}(\mathbf{r},\omega)\right]\mathbf{E}(\mathbf{r},\omega) = \frac{4\pi\omega^2}{c^2}&\int\limits_{-\infty}^{\infty}\mathbf{E}(\mathbf{r},\omega')\cdot\boldsymbol{\chi}^{(2)}(\mathbf{r};\omega-\omega')\cdot\mathbf{E}(\mathbf{r},\omega-\omega')\,\mathrm{d}\omega'\\
&+\frac{4\pi\mathrm{i}\omega}{c}\mathbf{J}_f(\mathbf{r},\omega),
\end{aligned} \tag{F.20}$$

which, with the definition $\epsilon(\mathbf{r},\omega) = 1+4\pi\chi^{(1)}(\mathbf{r},\omega)$, is the usual Helmholtz equation for the electric field and thus is solved through the usual Green's function methods.

G Analyzing the Photon-Matter Coupling

A challenging step in the derivation of the polariton operators is the calculation of the integrals

$$I_{\boldsymbol{\alpha\beta\beta'}nn'} = \int\limits_0^{\infty}\frac{c^3}{\pi \mathrm{e}^2\omega}\lambda_{\boldsymbol{\alpha\beta}n}(\omega)\lambda_{\boldsymbol{\alpha\beta'}n'}(\omega)\,\mathrm{d}\omega, \tag{G.1}$$

where

$$\lambda_{\boldsymbol{\alpha\beta}n}(\omega) = \frac{4\pi e^2\omega^{\frac{3}{2}}\eta}{c^3}\left[\delta_{TT_\alpha}R^{\lessdot}_{T\ell}\left(\frac{z_{\ell n}}{a},\frac{\omega}{c};0,a\right) + \delta_{TL}\delta_{T_\alpha E}\sqrt{\ell(\ell+1)}\frac{a^2c}{w_{\ell n}\omega}j_\ell(w_{\ell n})j_\ell\left(\frac{\omega a}{c}\right)\right]\delta_{pp_\alpha}\delta_{\ell\ell_\alpha}\delta_{mm_\alpha} \tag{G.2}$$

are the coupling coefficients that characterize the strength of the interaction between the matter oscillator coordinates and the electromagnetic field. We'll define the collective indices as $\boldsymbol{\beta} = (T,p,\ell,m)$, $\boldsymbol{\beta} = (T',p',\ell',m')$, and $\boldsymbol{\alpha} = (T_\alpha,p_\alpha,\ell_\alpha,m_\alpha)$ in this appendix. To make the notation easier to read, we will define

$$
I_{\boldsymbol{\alpha\beta\beta'}nn'} = \delta_{p_\alpha p}\delta_{\ell_\alpha \ell}\delta_{m_\alpha m}\delta_{p_\alpha p'}\delta_{\ell_\alpha \ell'}\delta_{m_\alpha m'} \begin{cases} I^{(M)}_{\ell nn'}, & T_\alpha = T = T' = M, \\ I^{(E)}_{\ell nn'}, & T_\alpha = T = T' = E, \\ I^{(L)}_{\ell nn'}, & T_\alpha = E,\ T = T' = L, \\ I^{(EL)}_{\ell nn'}, & T_\alpha = T = E, T' = L, \\ I^{(LE)}_{\ell nn'}, & T_\alpha = T' = E, T = L, \end{cases} \tag{G.3}
$$

where

$$
\begin{aligned}
I^{(M)}_{\ell nn'} &= \int_0^\infty \frac{16\pi e^2\omega^2\eta^2}{c^3} R_{M\ell}\Big(\frac{z_{\ell n}}{a}, \frac{\omega}{c}; 0, a\Big)\, R_{M\ell}\Big(\frac{z_{\ell n'}}{a}, \frac{\omega}{c}; 0, a\Big)\, \mathrm{d}\omega, \\
I^{(E)}_{\ell nn'} &= \int_0^\infty \frac{16\pi e^2\omega^2\eta^2}{c^3} R_{E\ell}\Big(\frac{z_{\ell n}}{a}, \frac{\omega}{c}; 0, a\Big)\, R_{E\ell}\Big(\frac{z_{\ell n'}}{a}, \frac{\omega}{c}; 0, a\Big)\, \mathrm{d}\omega, \\
I^{(L)}_{\ell nn'} &= \int_0^\infty \frac{16\pi e^2 a^4\eta^2}{c w_{\ell n} w_{\ell n'}} \ell(\ell+1) j_\ell(w_{\ell n}) j_\ell(w_{\ell n'}) j_\ell^2\Big(\frac{\omega a}{c}\Big)\, \mathrm{d}\omega
\end{aligned} \tag{G.4}
$$

describe coupling between modes of the same type and

$$
\begin{aligned}
I^{(EL)}_{\ell nn'} &= \int_0^\infty \frac{16\pi e^2\omega a^2\eta^2}{c^2 w_{\ell n'}} \sqrt{\ell(\ell+1)} R_{E\ell}\Big(\frac{z_{\ell n}}{a}, \frac{\omega a}{c}; 0, a\Big)\, j_\ell(w_{\ell n'}) j_\ell\Big(\frac{\omega a}{c}\Big)\, \mathrm{d}\omega, \\
I^{(LE)}_{\ell nn'} &= \int_0^\infty \frac{16\pi e^2\omega a^2\eta^2}{c^2 w_{\ell n}} \sqrt{\ell(\ell+1)} R_{E\ell}\Big(\frac{z_{\ell n'}}{a}, \frac{\omega}{c}; 0, a\Big)\, j_\ell(w_{\ell n}) j_\ell\Big(\frac{\omega a}{c}\Big)\, \mathrm{d}\omega
\end{aligned} \tag{G.5}
$$

describe cross-coupling between electric and longitudinal modes. Further noting that

$$
R_{M\ell}\Big(\frac{z_{\ell n}}{a}, \frac{\omega}{c}; 0, a\Big) = -\frac{a^2}{\big(\frac{z_{\ell n}}{a}\big)^2 - \big(\frac{\omega}{c}\big)^2} \frac{z_{\ell n}}{a} j_{\ell-1}(z_{\ell n})\, j_\ell\Big(\frac{\omega a}{c}\Big) \tag{G.6}
$$

and

$$
\begin{aligned}
R_{E\ell}\Big(\frac{z_{\ell n}}{a}, \frac{\omega}{c}; 0, a\Big) = {} & \frac{\ell+1}{2\ell+1} \frac{a^2}{\big(\frac{z_{\ell n}}{a}\big)^2 - \big(\frac{\omega}{c}\big)^2} \Big[\frac{\omega}{c} j_{\ell-2}\Big(\frac{\omega a}{c}\Big)\, j_{\ell-1}(z_{\ell n}) - \frac{z_{\ell n}}{a} j_{\ell-2}(z_{\ell n})\, j_{\ell-1}\Big(\frac{\omega a}{c}\Big)\Big] \\
& + \frac{\ell}{2\ell+1} \frac{a^2}{\big(\frac{z_{\ell n}}{a}\big)^2 - \big(\frac{\omega}{c}\big)^2} \frac{\omega}{c} j_\ell\Big(\frac{\omega a}{c}\Big)\, j_{\ell+1}(z_{\ell n})\,,
\end{aligned} \tag{G.7}
$$

we can see that a series of integrals involving products of spherical Bessel functions of argument $\omega a/c$ need to be calculated in order to determine $I_{\boldsymbol{\alpha\beta\beta'}nn'}$ for any set of indices. First noting that each of the above integrals is even in ω, we can rewrite each as an integral over the entire real line and then use contour integration to find an analytical solution.

In each integral, a useful trick is to notice that the integrals from $-\infty$ to ∞ of the even products $j_\ell(x)j_{\ell+2k}(x)$ and $xj_\ell(x)j_{\ell+k}(x)$ are equal to the integrals of $j_\ell(x)h^{(1)}_{\ell+2k}(x)$ and $xj_\ell(x)h_{\ell+k}(x)$, respectively, over the same bounds. Here, $h^{(1)}_\ell(x) = j_\ell(x) + \mathrm{i}y_\ell(x)$ is a spherical Hankel function of the first kind, $y_\ell(x)$ is a spherical Bessel function of the second kind, and $k \in \{0, \pm 1, \pm 2, \ldots\}$. This replacement is made possible by the fact that the products $j_\ell(x)y_{\ell+2k}(x)$ and $xj_\ell(x)y_{\ell+k}(x)$ are odd in x, such that integrands of these forms (perhaps multiplied by an even power of x) contribute nothing to an integral. Further, while $j_\ell(x) \to \infty$ as x goes to complex infinity, $h^{(1)}_\ell(x) \to 0$ for large x in the upper half-plane, allowing for the formation of great-arc contours that contribute nothing to a contour integral but allow for use of the residue theorem.

Using the standard toolkit of complex analysis from here, one can see our important integrals can be calculated straightforwardly and form three groups. The first group is comprised of integrals with simple results:

$$
\begin{aligned}
I_\ell^{(1)} &= \int_{-\infty}^{\infty} j_\ell^2(x)\,\mathrm{d}x = \frac{\pi}{2\ell+1}, \\
I_{\ell n}^{(2)} &= \int_{-\infty}^{\infty} \frac{x^2}{z_{\ell n}^2 - x^2} j_\ell^2(x)\,\mathrm{d}x = 0, \\
I_{\ell n}^{(3)} &= \int_{-\infty}^{\infty} \frac{x}{z_{\ell n}^2 - x^2} j_\ell(x) j_{\ell-1}(x)\,\mathrm{d}x = 0, \\
I_{\ell n}^{(4)} &= \int_{-\infty}^{\infty} \frac{x^2}{z_{\ell n}^2 - x^2} j_\ell(x) j_{\ell-2}(x)\,\mathrm{d}x = 0.
\end{aligned}
\tag{G.8}
$$

The second is comprised of integrals that evaluate to quantities proportional to the Kronecker delta $\delta_{nn'}$,

$$
\begin{aligned}
I_{\ell n n'}^{(5)} &= \int_{-\infty}^{\infty} x^2 \frac{1}{z_{\ell n}^2 - x^2} \frac{1}{z_{\ell n'}^2 - x^2} j_\ell^2(x)\,\mathrm{d}x = \delta_{nn'} \frac{\pi}{2} j_{\ell+1}(z_{\ell n}) y_\ell(z_{\ell n}), \\
I_{\ell n n'}^{(6)} &= PV \int_{-\infty}^{\infty} x^3 \frac{1}{z_{\ell n}^2 - x^2} \frac{1}{z_{\ell n'}^2 - x^2} j_\ell(x) j_{\ell-1}(x)\,\mathrm{d}x = \delta_{nn'} \frac{\pi}{2} z_{\ell n} j_{\ell+1}(z_{\ell n}) y_{\ell-1}(z_{\ell n}), \\
I_{\ell n n'}^{(7)} &= \int_{-\infty}^{\infty} x^4 \frac{1}{z_{\ell n}^2 - x^2} \frac{1}{z_{\ell n'}^2 - x^2} j_\ell^2(x)\,\mathrm{d}x = \delta_{nn'} \frac{\pi}{2} z_{\ell n}^2 j_{\ell+1}(z_{\ell n}) y_\ell(z_{\ell n}), \\
I_{\ell n n'}^{(8)} &= PV \int_{-\infty}^{\infty} x^4 \frac{1}{z_{\ell n}^2 - x^2} \frac{1}{z_{\ell n'}^2 - x^2} j_\ell(x) j_{\ell-2}(x)\,\mathrm{d}x = \delta_{nn'} \frac{\pi}{2} z_{\ell n}^2 j_{\ell+1}(z_{\ell n}) y_{\ell-2}(z_{\ell n}),
\end{aligned}
\tag{G.9}
$$

and the third contains integrals that need to be handled carefully when $n = n'$:

$$\begin{aligned}
I^{(9)}_{\ell n n'} &= PV \int_{-\infty}^{\infty} x^2 \frac{1}{z^2_{\ell n} - x^2} \frac{1}{z^2_{\ell n'} - x^2} j^2_{\ell-1}(x)\,\mathrm{d}x \\
&= \begin{cases} \dfrac{\pi}{z^2_{\ell n'} - z^2_{\ell n}} \left[z_{\ell n} j_{\ell-1}(z_{\ell n}) y_{\ell-1}(z_{\ell n}) - z_{\ell n'} j_{\ell-1}(z_{\ell n'}) y_{\ell-1}(z_{\ell n'}) \right], & n \neq n'; \\ -\dfrac{\pi}{4} \dfrac{j_{\ell-1}(z_{\ell n})}{z_{\ell n}} \left[z_{\ell n} y_{\ell-2}(z_{\ell n}) + (2\ell-1) y_{\ell-1}(z_{\ell n}) - z_{\ell n} y_{\ell}(z_{\ell n}) \right], & n = n'; \end{cases} \\
I^{(10)}_{\ell n n'} &= PV \int_{-\infty}^{\infty} x^3 \frac{1}{z^2_{\ell n} - x^2} \frac{1}{z^2_{\ell n'} - x^2} j_{\ell-1}(x) j_{\ell-2}(x)\,\mathrm{d}x \\
&= \begin{cases} \dfrac{\pi}{z^2_{\ell n'} - z^2_{\ell n}} \left[z^2_{\ell n} j_{\ell-1}(z_{\ell n}) y_{\ell-2}(z_{\ell n}) - z^2_{\ell n'} j_{\ell-1}(z_{\ell n'}) y_{\ell-2}(z_{\ell n'}) \right], & n \neq n'; \\ -\dfrac{\pi}{4} j_{\ell-1}(z_{\ell n}) \left[z_{\ell n} y_{\ell-3}(z_{\ell n}) + (2\ell+1) y_{\ell-2}(z_{\ell n}) - z_{\ell n} y_{\ell-1}(z_{\ell n}) \right], & n = n'; \end{cases} \\
I^{(11)}_{\ell n n'} &= PV \int_{-\infty}^{\infty} x^4 \frac{1}{z^2_{\ell n} - x^2} \frac{1}{z^2_{\ell n'} - x^2} j^2_{\ell-2}(x)\,\mathrm{d}x \\
&= \begin{cases} \dfrac{\pi}{z^2_{\ell n'} - z^2_{\ell n}} \left[z^3_{\ell n} j_{\ell-2}(z_{\ell n}) y_{\ell-2}(z_{\ell n}) - z^3_{\ell n'} j_{\ell-2}(z_{\ell n'}) y_{\ell-2}(z_{\ell n'}) \right], & n \neq n; \\ \dfrac{\pi}{2} z_{\ell n} \left[z_{\ell n} j_{\ell-1}(z_{\ell n}) y_{\ell-2}(z_{\ell n}) - 2 j_{\ell-2}(z_{\ell n}) y_{\ell-2}(z_{\ell n}) - z_{\ell n} j_{\ell-2}(z_{\ell n}) y_{\ell-3}(z_{\ell n}) \right], & n = n. \end{cases}
\end{aligned} \tag{G.10}$$

Here, the prefix PV denotes a Cauchy principal value integral. The integrals above that need to be carefully evaluated using principal value methods arise only in cases where singularities generated by the factors $1/(z^2_{\ell n} - x^2)$ are eliminated by subtraction. The use of principal value integrals simply allows us to analyze these integrals one-by-one.

Moving forward, we can let $x = \omega a/c$ to find

$$\begin{aligned}
I^{(M)}_{\ell n n'} &= 8\pi e^2 a^3 \eta^2 z_{\ell n'} z_{\ell n} j_{\ell-1}(z_{\ell n}) j_{\ell-1}(z_{\ell n'}) I^{(5)}_{\ell n n'} \\
&= 4\pi^2 \delta_{nn'} e^2 a^3 \eta^2 j^2_{\ell-1}(z_{\ell n}),
\end{aligned} \tag{G.11}$$

where we have used the identity $z^2_{\ell n} j_{\ell+1}(z_{\ell n}) y_\ell(z_{\ell n}) = 1$. Further, using the identities

$$\begin{aligned}
[j_{\ell-1}(z_{\ell n}) y_{\ell-2}(z_{\ell n}) - j_{\ell-2}(z_{\ell n}) y_{\ell-1}(z_{\ell n})] &= \frac{1}{z^2_{\ell n}}, \\
j_{\ell-2}(z_{\ell n}) &= \frac{2\ell-1}{z_{\ell n}} j_{\ell-1}(z_{\ell n}), \\
y_{\ell-3}(z_{\ell n}) &= \frac{2\ell-3}{z_{\ell n}} y_{\ell-2}(z_{\ell n}) - y_{\ell-1}(z_{\ell n}), \\
j_{\ell-1}(z_{\ell n}) &= -j_{\ell+1}(z_{\ell n}),
\end{aligned} \tag{G.12}$$

we can show that $I^{(E)}_{\ell n n'}$ takes the same form:

$$\begin{aligned}
I^{(E)}_{\ell n n'} = \frac{8\pi e^2 a^3 \eta^2}{(2\ell+1)^2} \Big[& (\ell+1)^2 j_{\ell-1}(z_{\ell n}) j_{\ell-1}(z_{\ell n'}) I^{(11)}_{\ell n n'} - (\ell+1)^2 z_{\ell n'} j_{\ell-1}(z_{\ell n}) j_{\ell-2}(z_{\ell n'}) I^{(10)}_{\ell n n'} \\
& - (\ell+1)^2 z_{\ell n} j_{\ell-1}(z_{\ell n'}) j_{\ell-2}(z_{\ell n}) I^{(10)}_{\ell n n'} + (\ell+1)^2 z_{\ell n} z_{\ell n'} j_{\ell-2}(z_{\ell n}) j_{\ell-2}(z_{\ell n'}) I^{(9)}_{\ell n n'} \\
& + \ell(\ell+1) j_{\ell-1}(z_{\ell n}) j_{\ell+1}(z_{\ell n'}) I^{(8)}_{\ell n n'} + \ell(\ell+1) j_{\ell-1}(z_{\ell n'}) j_{\ell+1}(z_{\ell n}) I^{(8)}_{\ell n n'} \\
& - \ell(\ell+1) z_{\ell n} j_{\ell+1}(z_{\ell n'}) j_{\ell-2}(z_{\ell n}) I^{(6)}_{\ell n n'} - \ell(\ell+1) z_{\ell n'} j_{\ell+1}(z_{\ell n}) j_{\ell-2}(z_{\ell n'}) I^{(6)}_{\ell n n'} \\
& + \ell^2 j_{\ell+1}(z_{\ell n}) j_{\ell+1}(z_{\ell n'}) I^{(7)}_{\ell n n'} \Big] \\
= 4\pi^2 \delta_{nn'} e^2 a^3 \eta^2 j^2_{\ell-1}(z_{\ell n}). &
\end{aligned} \tag{G.13}$$

In a straightforward manner, we can then show that the integrals $I^{(EL)}_{\ell nn'}$ and $I^{(EL)}_{\ell nn'}$ evaluate to zero:

$$\begin{aligned}
I^{(EL)}_{\ell nn'} &= \frac{16\pi e^2 a^3 \eta^2}{w_{\ell n'}} \sqrt{\ell(\ell+1)} j_\ell(w_{\ell n'}) \\
&\quad \times \left(\frac{\ell+1}{2\ell+1} j_{\ell-1}(z_{\ell n}) I^{(4)}_{\ell n} - \frac{\ell+1}{2\ell+1} j_{\ell-2}(z_{\ell n}) z_{\ell n} I^{(3)}_{\ell n} + \frac{\ell}{2\ell+1} j_{\ell+1}(z_{\ell n}) I^{(2)}_{\ell n} \right) \\
&= 0,
\end{aligned} \tag{G.14}$$

and $I^{(LE)}_{\ell nn'}$ follows from the transformation $I^{(LE)}_{\ell nn'} = I^{(EL)}_{\ell nn'}|_{n \leftrightarrow n'}$. Finally, we have a simple result for $I^{(L)}_{\ell nn'}$:

$$\begin{aligned}
I^{(L)}_{\ell nn'} &= 8\pi e^2 a^3 \eta^2 \ell(\ell+1) \frac{j_\ell(w_{\ell n}) j_\ell(w_{\ell n'})}{w_{\ell n} w_{\ell n'}} I^{(1)}_\ell \\
&= 8\pi^2 e^2 a^3 \eta^2 \frac{\ell(\ell+1)}{2\ell+1} \frac{j_\ell(w_{\ell n}) j_\ell(w_{\ell n'})}{w_{\ell n} w_{\ell n'}}.
\end{aligned} \tag{G.15}$$

References

[1] Claude Cohen-Tannoudji, Jacques Dupont-Roc, and Gilbert Grynberg. *Photons and Atoms: Introduction to Quantum Electrodynamics*. Physics textbook. Wiley-VCH, Weinheim, 2004.

[2] E. C. Titchmarsh. *Eigenfunction Expansions Associated With Second Order Differential Equations*. Oxford at the Clarendon Press, 1946.

[3] Bruno Huttner and Stephen M. Barnett. Quantization of the electromagnetic field in dielectrics. *Phys. Rev. A*, 46(7):4306–4322, 1992.

[4] Julius Adams Stratton. *Electromagnetic Theory*. John Wiley & Sons, Ltd, 1941.

[5] W. W. Hansen. A New Type of Expansion in Radiation Problems. *Physical Review*, 47(2):139–143, 1935.