Macroscopic QED in Finite Particles

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1 Derivation of the Wave Equation Satisfied by the Polarization Density

Before we can quantize a theory of light-matter interactions in a rigorous way, we need to describe the classical behavior of the matter we are working with. to do so, we begin with Maxwell's equations in the Fourier regime,

$$\begin{aligned}
\nabla \cdot \mathbf{E}(\mathbf{r},\omega) &= 4\pi\rho(\mathbf{r},\omega), \\
\nabla \cdot \mathbf{D}(\mathbf{r},\omega) &= 4\pi\rho_f(\mathbf{r},\omega), \\
\nabla \cdot \mathbf{B}(\mathbf{r},\omega) &= 0, \\
\nabla \times \mathbf{E}(\mathbf{r},\omega) &= \frac{\mathrm{i}\omega}{c}\mathbf{B}(\mathbf{r},\omega), \\
\nabla \times \mathbf{B}(\mathbf{r},\omega) &= \frac{4\pi}{c}\mathbf{J}(\mathbf{r},\omega) - \frac{\mathrm{i}\omega}{c}\mathbf{E}(\mathbf{r},\omega), \\
\nabla \times \mathbf{H}(\mathbf{r},\omega) &= \frac{4\pi}{c}\mathbf{J}_f(\mathbf{r},\omega) - \frac{\mathrm{i}\omega}{c}\mathbf{D}(\mathbf{r},\omega),
\end{aligned} \tag{1}$$

where

$$\begin{aligned}
\mathbf{D}(\mathbf{r},\omega) &= \epsilon(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega), \\
\mathbf{B}(\mathbf{r},\omega) &= \mu(\mathbf{r},\omega)\mathbf{H}(\mathbf{r},\omega),
\end{aligned} \tag{2}$$

are the electric and magnetic constitutive relations, respectively, and the charge and current distributions have been separated into bound and free contributions such that $\rho(\mathbf{r},\omega) = \rho_b(\mathbf{r},\omega) + \rho_f(\mathbf{r},\omega)$ and $\mathbf{J}(\mathbf{r},\omega) = \mathbf{J}_b(\mathbf{r},\omega) + \mathbf{J}_f(\mathbf{r},\omega)$. Alternatively, the electric and magnetic constitutive relations can be written as

$$\begin{aligned}
\mathbf{D}(\mathbf{r},\omega) &= \mathbf{E}(\mathbf{r},\omega) + 4\pi\mathbf{P}(\mathbf{r},\omega), \\
\mathbf{B}(\mathbf{r},\omega) &= \mathbf{H}(\mathbf{r},\omega) + 4\pi\mathbf{M}(\mathbf{r},\omega),
\end{aligned} \tag{3}$$

with $\mathbf{P}(\mathbf{r},\omega)$ the polarization density and $\mathbf{M}(\mathbf{r},\omega)$ the magnetization density of the matter. These lead straightforwardly to the definitions of the bound charge and current:

$$\begin{aligned}
\rho_b(\mathbf{r},\omega) &= -\nabla \cdot \mathbf{P}(\mathbf{r},\omega), \\
\mathbf{J}_b(\mathbf{r},\omega) &= -\mathrm{i}\omega\mathbf{P}(\mathbf{r},\omega) + c\nabla \times \mathbf{M}(\mathbf{r},\omega).
\end{aligned} \tag{4}$$

The constitutive relations are useful for defining a corollary to Faraday's law for the $\mathbf{D}(\mathbf{r},\omega)$- and $\mathbf{H}(\mathbf{r},\omega)$-fields:

$$\begin{aligned}
\nabla \times \mathbf{D}(\mathbf{r},\omega) &= \epsilon(\mathbf{r},\omega)\nabla \times \mathbf{E}(\mathbf{r},\omega) + \nabla\epsilon(\mathbf{r},\omega) \times \mathbf{E}(\mathbf{r},\omega) \\
&= \frac{\mathrm{i}\omega}{c}\epsilon(\mathbf{r},\omega)\mu(\mathbf{r},\omega)\mathbf{H}(\mathbf{r},\omega) + \nabla\epsilon(\mathbf{r},\omega) \times \mathbf{E}(\mathbf{r},\omega),
\end{aligned} \tag{5}$$

wherein we have used the identity $\nabla \times \{f(\mathbf{r})\mathbf{F}(\mathbf{r})\} = f(\mathbf{r})\nabla \times \mathbf{F}(\mathbf{r}) + \nabla f(\mathbf{r}) \times \mathbf{F}(\mathbf{r})$. Application of the curl operator to Faraday's law and its corollary produces

$$\begin{aligned}
\nabla \times \nabla \times \mathbf{E}(\mathbf{r},\omega) &= \frac{4\pi\mathrm{i}\omega}{c^2}\mu(\mathbf{r},\omega)\mathbf{J}_f(\mathbf{r},\omega) + \epsilon(\mathbf{r},\omega)\mu(\mathbf{r},\omega)\frac{\omega^2}{c^2}\mathbf{E}(\mathbf{r},\omega) + \frac{\mathrm{i}\omega}{c}\nabla\mu(\mathbf{r},\omega) \times \mathbf{H}(\mathbf{r},\omega), \\
\nabla \times \nabla \times \mathbf{D}(\mathbf{r},\omega) &= \frac{4\pi\mathrm{i}\omega}{c^2}\epsilon(\mathbf{r},\omega)\mu(\mathbf{r},\omega)\mathbf{J}_f(\mathbf{r},\omega) + \epsilon(\mathbf{r},\omega)\mu(\mathbf{r},\omega)\frac{\omega^2}{c^2}\mathbf{D}(\mathbf{r},\omega) \\
&\quad + \frac{\mathrm{i}\omega}{c}\nabla\left\{\epsilon(\mathbf{r},\omega)\mu(\mathbf{r},\omega)\right\} \times \mathbf{H}(\mathbf{r},\omega) + \nabla \times \left\{\nabla\epsilon(\mathbf{r},\omega) \times \mathbf{E}(\mathbf{r},\omega)\right\}.
\end{aligned} \tag{6}$$

The last term on the right-hand side of the first equation and the last two terms on the right-hand side of the second equation are nonzero for any system with space-dependent (i.e. interesting) material properties. These terms also act as sources that depend on the state of the field at any given frequency, such that, on the surface, they appear to make our lives quite complicated. However, in the special case where the dielectric and diamagnetic functions are piece-wise constant in space, these terms disappear. To see this, we can begin with the first line of Eq. (6) and let

$$\mathbf{E}(\mathbf{r},\omega) = \sum_i \Theta(\mathbf{r} \in \mathbb{V}_i)\mathbf{E}_i(\mathbf{r},\omega). \tag{7}$$

Here, space is divided into a number of regions i that each contain a set of points $\mathbb{V}_i$. Each volume $\mathbb{V}_i$ borders any other number neighboring of volumes $\mathbb{V}_j$ and possibly infinity. Each volume $\mathbb{V}_i$ shares a set of points along the surface $\partial\mathbb{V}_{ij}$ with its neighbor $\mathbb{V}_j$ along which the dielectric functions, diamagnetic functions, and fields are undefined. Letting $\partial\mathbb{V}_i$ be the total set of surface points surrounding the volume $\mathbb{V}_i$, we can define the Heaviside function

$$\Theta(\mathbf{r} \in \mathbb{V}_i) = \begin{cases} \mathbf{r} \in \mathbb{V}_i \setminus \partial\mathbb{V}_i, & 1, \\ \mathbf{r} \notin \mathbb{V}_i, & 0, \\ \mathbf{r} \in \partial\mathbb{V}_i, & \text{undefined}. \end{cases} \tag{8}$$

Further,

$$\mathbf{E}_i(\mathbf{r},\omega) = \begin{cases} \mathbf{E}(\mathbf{r},\omega), & \mathbf{r} \in \mathbb{V}_i, \\ 0, & \text{otherwise}, \end{cases} \tag{9}$$

such that our piece-wise definition of the electric field serves to "stitch together" the electric field in each separate region of space by connecting it along the system's boundaries to its neighbors. The dielectric and diamagnetic functions follow as

$$\begin{aligned} \epsilon(\mathbf{r},\omega) &= \sum_i \epsilon_i(\omega)\Theta(\mathbf{r} \in \mathbb{V}_i), \\ \mu(\mathbf{r},\omega) &= \sum_i \mu_i(\omega)\Theta(\mathbf{r} \in \mathbb{V}_i), \end{aligned} \tag{10}$$

but, in contrast with the definition of the electric field, are space-independent within each region.

Using these definitions, we can apply the double-curl operator on the left-hand side of the first line of Eq. (6) to find

$$\begin{aligned} &\sum_i \Theta(\mathbf{r} \in \mathbb{V}_i)\nabla \times \nabla \times \mathbf{E}_i(\mathbf{r},\omega) + \sum_i \delta(\mathbf{r} \in \partial\mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r}) \times \nabla \times \mathbf{E}_i(\mathbf{r},\omega) \\ &\quad + \sum_i \nabla \times \{\delta(\mathbf{r} \in \mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r}) \times \mathbf{E}_i(\mathbf{r},\omega)\} = \sum_i \frac{4\pi \mathrm{i}\omega}{c}\mu_i(\omega)\mathbf{J}_{fi}(\mathbf{r},\omega)\Theta(\mathbf{r} \in \mathbb{V}_i) \\ &\quad + \frac{\omega^2}{c^2}\sum_i \mu_i(\omega)\epsilon_i(\omega)\mathbf{E}_i(\mathbf{r},\omega)\Theta(\mathbf{r} \in \mathbb{V}_i) + \sum_i \frac{\mathrm{i}\omega}{c}\mu_i(\omega)\delta(\mathbf{r} \in \partial\mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r}) \times \mathbf{H}_i(\mathbf{r},\omega). \end{aligned} \tag{11}$$

Here, we have used the identity $\nabla\Theta(\mathbf{r} \in \mathbb{V}_i) = \hat{\mathbf{n}}_i(\mathbf{r})\delta(\mathbf{r} \in \partial\mathbb{V}_i)$ to evaluate the derivatives, where $\hat{\mathbf{n}}_i(\mathbf{r})$ is the outward-facing surface-normal unit vector of the boundary of $\mathbb{V}_i$. Because the fields and material functions are not defined on the boundary exactly, we will take the Dirac deltas to imply limits as one approaches a point on a boundary from inside the associated bounded volume. In this case, we have let $\Theta(\mathbf{r} \in \mathbb{V}_i)\delta(\mathbf{r} \in \partial\mathbb{V}_i) = \delta(\mathbf{r} \in \partial\mathbb{V}_i)$. Moreover, we can see that along each shared boundary $\partial\mathbb{V}_{ij}$,

$$\hat{\mathbf{n}}_i(\mathbf{r} \in \partial\mathbb{V}_{ij}) \times \mathbf{E}_i(\mathbf{r} \in \partial\mathbb{V}_{ij},\omega) = -\hat{\mathbf{n}}_j(\mathbf{r} \in \partial\mathbb{V}_{ij}) \times \mathbf{E}_j(\mathbf{r} \in \partial\mathbb{V}_{ij},\omega) \tag{12}$$

as the surface-parallel components of the electric fields are equal and the surface-normal vectors are equal and opposite. Therefore, we can see that

$$\sum_i \delta(\mathbf{r} \in \mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r}) \times \mathbf{E}_i(\mathbf{r},\omega) = 0 \tag{13}$$

and, accordingly, $\sum_i \nabla \times \{\delta(\mathbf{r} \in \mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r}) \times \mathbf{E}_i(\mathbf{r},\omega)\} = 0$. Moreover, we can see from Faraday's law that

$$\sum_i \Theta(\mathbf{r} \in \mathbb{V}_i)\nabla \times \mathbf{E}_i(\mathbf{r},\omega) + \sum_i \delta(\mathbf{r} \in \partial\mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r}) \times \mathbf{E}_i(\mathbf{r},\omega) = \sum_i \frac{\mathrm{i}\omega}{c}\mathbf{B}_i(\mathbf{r},\omega)\Theta(\mathbf{r} \in \mathbb{V}_i). \tag{14}$$

Since the second term on the left-hand side is zero, we can equate the remaining quantities point-wise within their respective regions:

$$\nabla \times \mathbf{E}_i(\mathbf{r},\omega) = \frac{\mathrm{i}\omega}{c}\mathbf{B}_i(\mathbf{r},\omega). \tag{15}$$

Combined with the definition $\mu_i(\omega)\mathbf{H}_i(\mathbf{r},\omega) = \mathbf{B}_i(\mathbf{r},\omega)$, we can use the region-separated form of Faraday's law to see that $\nabla \times \mathbf{E}_i(\mathbf{r},\omega) = \mathrm{i}\omega\mu_i(\omega)\mathbf{H}_i(\mathbf{r},\omega)/c$. We can then cancel the remaining terms on either side of Eq. (11) proportional to Dirac deltas. Further, as the rest of the terms feature sums of independent quantities, we can drop the sums and Heaviside functions as we did with Faraday's law, such that the double curl of $\mathbf{E}(\mathbf{r},\omega)$ provides

$$\nabla \times \nabla \times \mathbf{E}_i(\mathbf{r},\omega) = \frac{4\pi\mathrm{i}\omega}{c}\mu_i(\omega)\mathbf{J}_{fi}(\mathbf{r},\omega) + \frac{\omega^2}{c^2}\mu_i(\omega)\epsilon_i(\omega)\mathbf{E}_i(\mathbf{r},\omega). \tag{16}$$

This is simply the wave equation for the electric field within region i. Therefore, we have proved that the wave equation for the electric field in Eq. (6) can be decomposed into a series of wave equations in each region that must be solved individually and do *not* contain any anomalous boundary terms proportional to $\nabla\mu(\mathbf{r},\omega)$.

A similar process can be used to find the region-separated form of the wave equation of the **D**-field. Letting

$$\begin{aligned} \mathbf{D}(\mathbf{r},\omega) &= \sum_i \Theta(\mathbf{r}\in\mathbb{V}_i)\epsilon_i(\omega)\mathbf{E}_i(\mathbf{r},\omega), \\ \epsilon(\mathbf{r},\omega)\mu(\mathbf{r},\omega) &= \sum_i \Theta(\mathbf{r}\in\mathbb{V}_i)\epsilon_i(\omega)\mu_i(\omega), \end{aligned} \tag{17}$$

we arrive at

$$\begin{aligned} &\sum_i \Theta(\mathbf{r}\in\mathbb{V}_i)\nabla\times\nabla\times\mathbf{D}_i(\mathbf{r},\omega) + \sum_i \epsilon_i(\omega)\delta(\mathbf{r}\in\partial\mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r})\times\nabla\times\mathbf{E}_i(\mathbf{r},\omega) \\ &\quad + \nabla\times\left\{\sum_i \epsilon_i(\omega)\delta(\mathbf{r}\in\partial\mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r})\times\mathbf{E}_i(\mathbf{r},\omega)\right\} = \sum_i \Theta(\mathbf{r}\in\mathbb{V}_i)\frac{4\pi\mathrm{i}\omega}{c^2}\epsilon_i(\omega)\mu_i(\omega)\mathbf{J}_{fi}(\mathbf{r},\omega) \\ &\quad + \sum_i \Theta(\mathbf{r}\in\mathbb{V}_i)\epsilon_i(\omega)\mu_i(\omega)\frac{\omega^2}{c^2}\mathbf{D}_i(\mathbf{r},\omega) + \sum_i \frac{\mathrm{i}\omega}{c}\epsilon_i(\omega)\mu_i(\omega)\delta(\mathbf{r}\in\delta\mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r})\times\mathbf{H}_i(\mathbf{r},\omega) \\ &\quad + \nabla\times\left\{\sum_i \epsilon_i(\omega)\delta(\mathbf{r}\in\partial\mathbb{V}_i)\hat{\mathbf{n}}_i(\mathbf{r})\times\mathbf{E}_i(\mathbf{r},\omega)\right\}, \end{aligned} \tag{18}$$

wherein $\mathbf{D}_i(\mathbf{r},\omega) = \epsilon_i(\omega)\mathbf{E}_i(\mathbf{r},\omega)$. We can immediately see that the last terms on both the left- and right-hand sides of the equation cancel. The second-to-last terms on either side of the equation can be seen to cancel using Faraday's law as was done in the previous paragraph, such that, after dropping sums and Heaviside functions, we find

$$\nabla \times \nabla \times \mathbf{D}_i(\mathbf{r},\omega) = \frac{4\pi\mathrm{i}\omega}{c^2}\epsilon_i(\omega)\mu_i(\omega)\mathbf{J}_{fi}(\mathbf{r},\omega) + \epsilon_i(\omega)\mu_i(\omega)\frac{\omega^2}{c^2}\mathbf{D}_i(\mathbf{r},\omega). \tag{19}$$

We can now define the polarization field in each region as $\mathbf{P}_i(\mathbf{r},\omega)$ such that

$$\mathbf{P}(\mathbf{r},\omega) = \sum_i \Theta(\mathbf{r}\in\mathbb{V}_i)\mathbf{P}_i(\mathbf{r},\omega). \tag{20}$$

This paves the wave for the identity

$$\mathbf{P}_i(\mathbf{r},\omega) = \frac{\mathbf{D}_i(\mathbf{r},\omega) - \mathbf{E}_i(\mathbf{r},\omega)}{4\pi}. \tag{21}$$

Therefore, subtracting Eq. (16) from Eq. (19) and dividing by 4π, we find

$$\nabla \times \nabla \times \mathbf{P}_i(\mathbf{r},\omega) - \epsilon_i(\omega)\mu_i(\omega)\frac{\omega^2}{c^2}\mathbf{P}_i(\mathbf{r},\omega) = \frac{\mathrm{i}\omega}{c}\mu_i(\omega)\left[\epsilon_i(\omega) - 1\right]\mathbf{J}_{fi}(\mathbf{r},\omega). \tag{22}$$

Therefore, the polarization field in each region obeys the same wave equation as do the electric and **D**-fields, modulo a space-independent prefactor in front of the free current source term on the right-hand side.

2 The First-Principles Lagrangian

To describe the quantum electrodynamical behavior of finite particles, we need to rebuild our first principles in terms of quantum-mechanical operators and organize these operators inside of a Hamiltonian formalism. To do so, we will begin with the classical Lagrangian density

$$\begin{aligned}&\mathscr{L}\left[\{\mathbf{x}_{fi}(t)\},\{\dot{\mathbf{x}}_{fi}(t)\};\mathbf{Q}_m(\mathbf{r},t),\dot{\mathbf{Q}}_m(\mathbf{r},t);\{\tilde{\mathbf{Q}}_\nu(\mathbf{r},t)\},\{\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\};\mathbf{A}(\mathbf{r},t),\dot{\mathbf{A}}(\mathbf{r},t)\right]\\&\quad=\mathscr{L}_f+\mathscr{L}_m+\mathscr{L}_r+\mathscr{L}_{\text{int}}+\mathscr{L}_{\text{bind}}+\mathscr{L}_{\text{EM}}+\mathscr{L}_{\text{NL}}\end{aligned} \tag{23}$$

where

$$\mathscr{L}_f=\sum_{i=1}^{N_f}\frac{1}{2}m_{fi}\dot{\mathbf{x}}_{fi}^2(t)\delta[\mathbf{r}-\mathbf{x}_{fi}(t)] \tag{24}$$

describes the kinetic energy of the free charges with coordinates $\mathbf{x}_{fi}(t)$ and velocities $\dot{\mathbf{x}}_{fi}(t)$,

$$\mathscr{L}_m=\frac{\mu_m(\mathbf{r})}{2}\left[\dot{\mathbf{Q}}_m^2(\mathbf{r},t)-\omega_0^2\mathbf{Q}_m^2(\mathbf{r},t)\right] \tag{25}$$

describes the kinetic and harmonic potential energies experienced by the mobile material charges with coordinate fields $\mathbf{Q}_m(\mathbf{r},t)$ (see Section 2.1 for details),

$$\mathscr{L}_r=\int_0^\infty\frac{\mu_\nu(\mathbf{r})}{2}\left[\dot{\tilde{\mathbf{Q}}}_\nu^2(\mathbf{r},t)-\nu^2\tilde{\mathbf{Q}}_\nu^2(\mathbf{r},t)\right]\,\mathrm{d}\nu \tag{26}$$

describes the kinetic and potential energies of reservoir charges that build in the system's damping,

$$\begin{aligned}\mathscr{L}_{\text{int}}=&-\left[-e\nabla\cdot\{\bar{n}_m(\mathbf{r})\mathbf{Q}_m(\mathbf{r},t)\}+\rho_f(\mathbf{r},t)\right]\Phi(\mathbf{r},t)+\frac{1}{c}\left[e\bar{n}_m(\mathbf{r})\dot{\mathbf{Q}}_m(\mathbf{r},t)+\mathbf{J}_f(\mathbf{r},t)\right]\cdot\mathbf{A}(\mathbf{r},t)\\&-\int_0^\infty\tilde{v}(\nu)\mathbf{Q}_m(\mathbf{r},t)\cdot\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\,\mathrm{d}\nu\end{aligned} \tag{27}$$

describes the interactions between the electromagnetic field, mobile charges, and free charges,

$$\mathscr{L}_{\text{bind}}=-\sum_{\alpha,\beta=m,b,r}\left[u_{\alpha\beta}^\infty(\mathbf{r})+u_{\beta\alpha}^\infty(\mathbf{r})\right]-\sum_{\alpha=m,b,r,f}u_\alpha^{\text{self}} \tag{28}$$

describes the binding potentials and self-energies experienced by each charge, and

$$\mathscr{L}_{\text{EM}}=\frac{1}{8\pi}\left[[\nabla\Phi(\mathbf{r},t)]^2+\frac{1}{c^2}\dot{\mathbf{A}}^2(\mathbf{r},t)-[\nabla\times\mathbf{A}(\mathbf{r},t)]^2+\frac{2}{c}\nabla\Phi(\mathbf{r},t)\cdot\dot{\mathbf{A}}(\mathbf{r},t)\right] \tag{29}$$

describes the energy bound in the electromagnetic fields. Finally,

$$\mathscr{L}_{\text{NL}}=-\frac{\sigma_0}{3}\mu_m(\mathbf{r})\mathbf{Q}_m(\mathbf{r},t)\cdot[\mathbf{Q}_m(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}_m(\mathbf{r},t)] \tag{30}$$

describes the anharmonic portion of the potential energy experienced by the material charges. This term can be generalized as needed to describe the interesting nonlinear dynamics of the system.

A formal derivation of this Lagrangian density from the canonical QED Lagrangian

$$\begin{aligned}\mathscr{L}_{\text{QED}}\left[\mathbf{x}_1,\ldots,\mathbf{x}_N,\dot{\mathbf{x}}_1,\ldots,\dot{\mathbf{x}}_N;\mathbf{A},\dot{\mathbf{A}}\right]=&\sum_{i=1}^N\frac{1}{2}m_i\dot{\mathbf{x}}_i^2(t)\delta[\mathbf{r}-\mathbf{x}_i(t)]+\frac{1}{8\pi}\left[\mathbf{E}^2(\mathbf{r},t)-\mathbf{B}^2(\mathbf{r},t)\right]\\&+\sum_{i=1}^N\frac{e_i}{c}\dot{\mathbf{x}}_i(t)\delta[\mathbf{r}-\mathbf{x}_i(t)]\cdot\mathbf{A}(\mathbf{r},t)-\sum_{i=1}^N e_i\delta[\mathbf{r}-\mathbf{x}_i(t)]\Phi(\mathbf{r},t).\end{aligned} \tag{31}$$

involves in-depth discussions of the atomic and molecular structure of a given material and will not be done here. Instead, we will simply define each of our terms and demonstrate that $\mathscr{L}$ reproduces the equations of motion we want for our material.

2.1 Definitions of the coordinate fields

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 $\mathscr{L}_{\mathrm{QED}}$ (see Eq. (31)) are a bad approximation and must be replaced by a more general description.

In our model, we borrow from hydrodynamic models of media and assume that four separate charge distributions $\rho_\phi(\mathbf{r},t)$ exist within the volume $\mathbb{V}$ occupied by the material 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. Giving this charge density a characteristic charge magnitude e, we can define a number density of mobile charges via $n_m(\mathbf{r},t)=\rho_m(\mathbf{r},t)/e$. Moreover, we assume that this charge density can be expanded as $n_m(\mathbf{r},t)=\bar{n}_m(\mathbf{r})+\delta n_m(\mathbf{r},t)$, with $\bar{n}_m(\mathbf{r})$ the equilibrium charge density that exists in the absence of driving and $\delta n_m(\mathbf{r},t)$ a perturbation density the encodes the material's response to electromagnetic stimuli.

These mobile charges experience motion described by a velocity field $\dot{\mathbf{Q}}_m(\mathbf{r},t)$ and form a bound current density $\mathbf{J}_m(\mathbf{r})$ given by

$$
\begin{aligned}
\mathbf{J}_m(\mathbf{r},t) &= \sum_i \mathbf{j}_i(\mathbf{r},t) \\
&= e\bar{n}_m(\mathbf{r})\dot{\mathbf{Q}}_m(\mathbf{r},t)
\end{aligned}
\tag{32}
$$

that captures the contributions of the microscopic currents $\mathbf{j}_i(\mathbf{r},t)$ that can be calculated through atomistic models. The mobile charge current obeys the hard-wall boundary condition $\mathbf{J}_m(\mathbf{r},t)\cdot\hat{\mathbf{n}}(\mathbf{r})|_{\partial\mathbb{V}}$ with $\hat{\mathbf{n}}(\mathbf{r})$ the outward-oriented surface-normal unit vector, such that no mobile charge enters or exits the material when the system is excited. More explicitly, the continuity equation

$$
\begin{aligned}
\frac{\partial\rho_m(\mathbf{r},t)}{\partial t} &= e\delta\dot{n}_m(\mathbf{r},t) \\
&= -\nabla\cdot\mathbf{J}_m(\mathbf{r},t) \\
&= -e\left[\bar{n}_m(\mathbf{r})\nabla\cdot\dot{\mathbf{Q}}_m(\mathbf{r},t)+\nabla\bar{n}_m(\mathbf{r})\cdot\dot{\mathbf{Q}}_m(\mathbf{r},t)\right]
\end{aligned}
\tag{33}
$$

contains no contribution from $\nabla\bar{n}_m(\mathbf{r})\cdot\dot{\mathbf{Q}}_m(\mathbf{r},t)|_{\mathbf{r}\in\partial\mathbb{V}}$, as $\nabla\bar{n}_m(\mathbf{r})|_{\mathbf{r}\in\partial\mathbb{V}}\sim\hat{\mathbf{n}}(\mathbf{r})$.

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

$$
\mathbf{Q}_m(\mathbf{r},t)=\int_{-\infty}^{t}\dot{\mathbf{Q}}_m(\mathbf{r},t')\,\mathrm{d}t',
\tag{34}
$$

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

$$
\begin{aligned}
\mathbf{P}_m(\mathbf{r},t) &= \int_{-\infty}^{t}\mathbf{J}_m(\mathbf{r},t')\,\mathrm{d}t' \\
&= e\bar{n}_m(\mathbf{r})\mathbf{Q}_m(\mathbf{r},t),
\end{aligned}
\tag{35}
$$

such that

$$
\begin{aligned}
\nabla\cdot\mathbf{P}_m(\mathbf{r},t) &= -e\,\delta n_m(\mathbf{r},t) \\
&= e\left[\bar{n}_m(\mathbf{r})\nabla\cdot\mathbf{Q}_m(\mathbf{r},t)+\nabla\bar{n}_m(\mathbf{r})\cdot\mathbf{Q}_m(\mathbf{r},t)\right].
\end{aligned}
\tag{36}
$$

Interestingly, this suggests that $\nabla\bar{n}_m(\mathbf{r})\cdot\mathbf{Q}_m(\mathbf{r},t)|_{\mathbf{r}\in\partial\mathbb{V}}$ cannot be time-varying. Via our initial conditions, we have disallowed the existence of a static coordinate field, such that the polarization field has no surface-normal component at the material boundaries.

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

$\rho_b(\mathbf{r}) = -en_b(\mathbf{r})$. In the case where the equilibrium density mobile and background density have identical spatial distributions, $\rho_b(\mathbf{r}) + e\bar{n}_m(\mathbf{r}) = 0$.

The reservoir charges ($\phi = r$) is 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 charge e_ν and a number density $n_\nu(\mathbf{r}, t)$ such that $\rho_\nu(\mathbf{r}, t) = e_\nu n_\nu(\mathbf{r}, t)$, and this number density is expanded around an equilibrium density such that $n_\nu(\mathbf{r}, t) = \bar{n}_\nu(\mathbf{r}) + \delta n_\nu(\mathbf{r}, t)$. In turn, the reservoir currents $\mathbf{J}_\nu(\mathbf{r}, t) = e_\nu \bar{n}_\nu(\mathbf{r})\dot{\mathbf{Q}}_\nu(\mathbf{r}, t)$ 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 $\rho_f(\mathbf{r}, t) = \sum_{i=1}^{N_f} e_{fi}\delta[\mathbf{r} - \mathbf{x}_{fi}(t)]$ such that a more specific driving charge source can be specified later.

2.2 Equations of motion

The derivations of the equations of motion of the generalized coordinates of $\mathscr{L}$ are straightforward. The equation of motion for the free charges is given by

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial L}{\partial \dot{\mathbf{x}}_{fi}(t)} = \frac{\partial L}{\partial \mathbf{x}_{fi}(t)}, \tag{37}$$

where $L = \int \mathscr{L}\, \mathrm{d}^3\mathbf{r}$. Before we can evaluate these variational derivatives, we need to first define the free current $\mathbf{J}_f(\mathbf{r}, t)$ that appears in $\mathscr{L}_{\text{int}}$. It is straightforward to prove that, with the Dirac delta "softened" to be a very sharply-peaked but finite even function,

$$\frac{\partial}{\partial t}\left\{\delta[\mathbf{r} - \mathbf{x}_{fi}(t)]\right\} = -\dot{\mathbf{x}}_{fi}(t) \cdot \nabla \left\{\delta[\mathbf{r} - \mathbf{x}_i(t)]\right\}, \tag{38}$$

such that, with the definition

$$\mathbf{J}_{fi}(\mathbf{r}, t) = \sum_{i=1}^{N_f} e_{fi}\dot{\mathbf{x}}_{fi}(t)\delta[\mathbf{r} - \mathbf{x}_{fi}(t)], \tag{39}$$

the continuity equation $(\partial/\partial t)\rho_f(\mathbf{r}, t) + \nabla \cdot \mathbf{J}_f(\mathbf{r}, t) = 0$ is satisfied for all $(\mathbf{r}, t)$. Using this description and noting that $\partial/\partial \mathbf{x}_{fi}(t) = \sum_{k=x,y,z} \hat{\mathbf{e}}_k \partial/\partial[\hat{\mathbf{e}}_k \cdot \mathbf{x}_{fi}(t)]$ is simply a gradient operator with respect to $\mathbf{x}_{fi}(t)$ rather than the usual $\mathbf{r}$, we can see that the Euler-Lagrange equations for the free charges give

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial}{\partial \dot{\mathbf{x}}_{fi}(t)}\left\{\sum_j \frac{1}{2}m_{fj}\dot{\mathbf{x}}_{fj}^2(t) + \frac{1}{c}\sum_j e_{fj}\dot{\mathbf{x}}_{fj}(t) \cdot \mathbf{A}[\mathbf{x}_{fj}(t), t] + \dots\right\} &= m_{fi}\ddot{\mathbf{x}}_{fi}^2(t) + \frac{1}{c}e_{fi}\frac{\mathrm{d}}{\mathrm{d}t}\{\mathbf{A}[\mathbf{x}_{fi}(t), t]\} \\ &= m_{fi}\ddot{\mathbf{x}}_{fi}^2(t) + \frac{e_{fi}}{c}\Big(\dot{\mathbf{A}}[\mathbf{x}_{fi}(t), t] + [\dot{\mathbf{x}}_{fi}(t) \cdot \nabla]\left\{\mathbf{A}[\mathbf{x}_{fi}(t), t]\right\}\Big)\end{aligned} \tag{40}$$

and

$$\begin{aligned}\frac{\partial}{\partial \mathbf{x}_{fi}(t)}\left\{-\sum_j e_{fj}\Phi[\mathbf{x}_{fj}(t), t] + \frac{1}{c}\sum_j e_{fj}\dot{\mathbf{x}}_{fj}(t) \cdot \mathbf{A}[\mathbf{x}_{fj}(t), t] + \dots\right\} &= -e_{fi}\nabla\Phi[\mathbf{x}_{fi}(t), t] \\ &+ \frac{1}{c}\left(\dot{\mathbf{x}}_{fi}(t) \times \nabla \times \mathbf{A}[\mathbf{x}_{fi}(t), t] + [\dot{\mathbf{x}}_{fi}(t) \cdot \nabla]\left\{\mathbf{A}[\mathbf{x}_{fi}(t), t]\right\}\right),\end{aligned} \tag{41}$$

wherein we have used the identities $\nabla\{\mathbf{a} \cdot \mathbf{B}(\mathbf{r})\} = \mathbf{a} \times \nabla \times \mathbf{B}(\mathbf{r}) + (\mathbf{a} \cdot \nabla)\{\mathbf{B}(\mathbf{r})\}$ and $\mathrm{d}/\mathrm{d}t\{\mathbf{F}[\mathbf{x}(t), t]\} = \partial\mathbf{F}[\mathbf{x}(t), t]/\partial t + \sum_i [\partial F_i[\mathbf{x}(t), t]/\partial x_i(t)]\partial x_i(t)/\partial t$ and assumed that the gradient operators are taken with respect to the coordinate that each field depends on. Therefore,

$$m_{fi}\ddot{\mathbf{x}}_{fi}^2(t) = e_{fi}\left(-\nabla\Phi[\mathbf{x}_{fi}(t), t] - \frac{1}{c}\dot{\mathbf{A}}[\mathbf{x}_{fi}(t), t] + \frac{1}{c}\dot{\mathbf{x}}_{fi}(t) \times \nabla \times \mathbf{A}[\mathbf{x}_{fi}(t), t]\right), \tag{42}$$

which is precisely the Maxwell-Lorentz force with $\mathbf{E}(\mathbf{r}, t) = -\nabla\Phi(\mathbf{r}, t) - (1/c)\dot{\mathbf{A}}(\mathbf{r}, t)$ and $\mathbf{B}(\mathbf{r}, t) = \nabla \times \mathbf{A}(\mathbf{r}, t)$.

The equations of motion for the mobile and reservoir charge coordinate fields are derived from $\mathscr{L}$ directly, with

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial \mathscr{L}}{\partial \dot{F}_j(\mathbf{r},t)}\right\} = \frac{\partial \mathscr{L}}{\partial F_j(\mathbf{r},t)} - \sum_{k=1}^{3}\frac{\partial}{\partial r_k}\frac{\partial \mathscr{L}}{\partial\left(\frac{\partial F_j(\mathbf{r},t)}{\partial r_k}\right)} \tag{43}$$

the Euler-Lagrange equation for the j^{th}-component of the dummy vector field $\mathbf{F}(\mathbf{r},t)$. Beginning with the mobile charges,

$$\begin{aligned}
\sum_j \hat{\mathbf{e}}_j \frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial \mathscr{L}}{\partial \dot{Q}_{mj}(\mathbf{r},t)}\right\} &= \mu_m(\mathbf{r})\ddot{\mathbf{Q}}_m(\mathbf{r},t) + \frac{e}{c}\bar{n}_m(\mathbf{r})\dot{\mathbf{A}}(\mathbf{r},t),\\
\sum_j \hat{\mathbf{e}}_j \frac{\partial \mathscr{L}}{\partial Q_{mj}(\mathbf{r},t)} &= -\mu_m(\mathbf{r})\omega_0^2\mathbf{Q}_m(\mathbf{r},t) + e\nabla\bar{n}_m(\mathbf{r}\notin\partial\mathbb{V})\Phi(\mathbf{r},t) - \int_0^\infty \tilde{v}(\nu)\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\,\mathrm{d}\nu\\
&\quad - \sigma_0\mu_m(\mathbf{r})\mathbf{Q}_m^2(\mathbf{r},t),\\
-\sum_j \hat{\mathbf{e}}_j \sum_{k=1}^{3}\frac{\partial}{\partial r_k}\frac{\partial \mathscr{L}}{\partial\left(\frac{\partial Q_{mj}(\mathbf{r},t)}{\partial r_k}\right)} &= -e\bar{n}_m(\mathbf{r})\nabla\Phi(\mathbf{r},t),
\end{aligned} \tag{44}$$

such that

$$\begin{aligned}
&\mu_m(\mathbf{r})\ddot{\mathbf{Q}}_m(\mathbf{r},t) + \int_0^\infty \tilde{v}(\nu)\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\,\mathrm{d}\nu + \mu_m(\mathbf{r})\omega_0^2\mathbf{Q}_m(\mathbf{r},t) + \mu_m(\mathbf{r})\sigma_0\mathbf{Q}_m^2(\mathbf{r},t)\\
&\qquad = e\nabla\bar{n}_m(\mathbf{r}\notin\partial\mathbb{V})\Phi(\mathbf{r},t) + e\bar{n}_m(\mathbf{r})\left[-\nabla\Phi(\mathbf{r},t) - \frac{1}{c}\dot{\mathbf{A}}(\mathbf{r},t)\right].
\end{aligned} \tag{45}$$

The reservoir charges have a similarly simple results, with

$$\begin{aligned}
\sum_j \hat{\mathbf{e}}_j \frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial \mathscr{L}}{\partial \dot{\tilde{Q}}_{\nu j}(\mathbf{r},t)}\right\} &= \mu_\nu(\mathbf{r})\ddot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t) - \tilde{v}(\nu)\dot{\mathbf{Q}}_m(\mathbf{r},t),\\
\sum_j \hat{\mathbf{e}}_j \frac{\partial \mathscr{L}}{\partial \tilde{Q}_{\nu j}(\mathbf{r},t)} &= -\mu_\nu(\mathbf{r})\nu^2\tilde{\mathbf{Q}}_\nu(\mathbf{r},t),\\
-\sum_j \hat{\mathbf{e}}_j \sum_{k=1}^{3}\frac{\partial}{\partial r_k}\frac{\partial \mathscr{L}}{\partial\left(\frac{\partial Q_{\nu j}(\mathbf{r},t)}{\partial r_k}\right)} &= 0,
\end{aligned} \tag{46}$$

such that

$$\mu_\nu(\mathbf{r})\ddot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t) + \mu_\nu(\mathbf{r})\nu^2\tilde{\mathbf{Q}}_\nu(\mathbf{r},t) = \tilde{v}(\nu)\dot{\mathbf{Q}}_m(\mathbf{r},t). \tag{47}$$

Together, Eqs. (45) and (47) can be combined to produce a simple anharmonic oscillator dielectric picture with internal losses determined by the details of the coupling function $\tilde{v}(\nu)$. For details, see Appendix D.

Finally, we can use $\mathscr{L}$ to define the equations of motion for the potentials $\Phi(\mathbf{r},t)$ and $\mathbf{A}(\mathbf{r},t)$. First, we can see that

$$\begin{aligned}
\frac{\partial \mathscr{L}}{\partial \dot{\Phi}(\mathbf{r},t)} &= 0,\\
\frac{\partial \mathscr{L}}{\partial \Phi(\mathbf{r},t)} &= e\nabla\cdot\{\bar{n}_m(\mathbf{r})\mathbf{Q}_m(\mathbf{r},t)\} - \rho_f(\mathbf{r},t) = -e\delta n_m(\mathbf{r},t) - \rho_f(\mathbf{r},t),\\
\sum_{k=1}^{3}\frac{\partial}{\partial r_k}\frac{\partial \mathscr{L}}{\partial\left(\frac{\partial \Phi(\mathbf{r},t)}{\partial r_k}\right)} &= \frac{1}{4\pi}\nabla\cdot\nabla\Phi(\mathbf{r},t),
\end{aligned} \tag{48}$$

such that

$$-\nabla\cdot\nabla\Phi(\mathbf{r},t) = 4\pi\left[e\,\delta n_m(\mathbf{r},t) + \rho_f(\mathbf{r},t)\right]. \tag{49}$$

In the limit where the equilibrium charge densities $\bar{n}_m(\mathbf{r})$ and $n_b(\mathbf{r})$ generate equal and opposite scalar potentials, our Euler-Lagrange equation for the scalar potential therefore correctly reproduces the potential formulation of Gauss' law, which can quickly be rewritten as

$$\nabla \cdot \mathbf{E}(\mathbf{r},t) = 4\pi \left[e\, \delta n_m(\mathbf{r},t) + \rho_f(\mathbf{r},t)\right]. \tag{50}$$

Further, using the identity

$$\sum_{jk} \frac{\partial}{\partial r_k} \frac{\partial}{\partial\left(\frac{\partial F_j(\mathbf{r},t)}{\partial r_k}\right)} \left\{\left[\nabla \times \mathbf{F}(\mathbf{r},t)\right]^2\right\} \hat{\mathbf{e}}_j = -2\nabla \times \nabla \times \mathbf{F}(\mathbf{r},t), \tag{51}$$

we can see that

$$\begin{aligned} \sum_j \hat{\mathbf{e}}_j \frac{\mathrm{d}}{\mathrm{d}t} \left\{\frac{\partial \mathcal{L}}{\partial \dot{A}_j(\mathbf{r},t)}\right\} &= \frac{1}{4\pi c^2}\ddot{\mathbf{A}}(\mathbf{r},t) + \frac{1}{4\pi c}\nabla\dot{\Phi}(\mathbf{r},t), \\ \sum_j \hat{\mathbf{e}}_j \frac{\partial \mathcal{L}}{\partial A_j(\mathbf{r},t)} &= \frac{1}{c}\left[\mathbf{J}_m(\mathbf{r},t) + \mathbf{J}_f(\mathbf{r},t)\right], \\ -\sum_j \hat{\mathbf{e}}_j \sum_{k=1}^{3} \frac{\partial}{\partial r_k} \frac{\partial \mathcal{L}}{\partial\left(\frac{\partial A_j(\mathbf{r},t)}{\partial r_k}\right)} &= -\frac{1}{4\pi}\nabla \times \nabla \times \mathbf{A}(\mathbf{r},t), \end{aligned} \tag{52}$$

such that

$$\nabla \times \nabla \times \mathbf{A}(\mathbf{r},t) + \frac{1}{c^2}\ddot{\mathbf{A}}(\mathbf{r},t) = \frac{4\pi}{c}\left[\mathbf{J}_m(\mathbf{r},t) + \mathbf{J}_f(\mathbf{r},t)\right] - \frac{1}{c}\nabla\dot{\Phi}(\mathbf{r},t). \tag{53}$$

From our analysis of the scalar potential we know that $-\nabla \cdot \nabla\dot{\Phi}(\mathbf{r},t) = 4\pi\left[e\,\delta\dot{n}(\mathbf{r},t) + \dot{\rho}_f(\mathbf{r},t)\right] = 4\pi\nabla \cdot \{-\mathbf{J}_m^{\parallel}(\mathbf{r},t) - \mathbf{J}_f^{\parallel}(\mathbf{r},t)\}$, where the superscipt $\parallel$ denotes the longitudinal component of a vector field. Therefore,

$$-\frac{1}{c}\nabla\dot{\Phi}(\mathbf{r},t) = -\frac{4\pi}{c}\left[\mathbf{J}_m^{\parallel}(\mathbf{r},t) + \mathbf{J}_f^{\parallel}(\mathbf{r},t)\right] \tag{54}$$

and, noting that $\mathbf{J}_\phi(\mathbf{r},t) = \mathbf{J}_\phi^{\parallel}(\mathbf{r},t) + \mathbf{J}_\phi^{\perp}(\mathbf{r},t)$, the correct wave equation for the vector potential in the Coulomb gauge,

$$\nabla \times \nabla \times \mathbf{A}(\mathbf{r},t) + \frac{1}{c^2}\ddot{\mathbf{A}}(\mathbf{r},t) = \frac{4\pi}{c}\left[\mathbf{J}_m^{\perp}(\mathbf{r},t) + \mathbf{J}_f^{\perp}(\mathbf{r},t)\right], \tag{55}$$

is reproduced.

3 Construction of a Mechanical Picture for MQED

From this point, our goal is to map our existing 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, matter field, and reservoir field into sets of normal modes and analyzing the time-dependence of those modes' amplitudes. These expansions are given by

$$\begin{aligned} \mathbf{A}(\mathbf{r},t) &= \sum_{\boldsymbol{\alpha}} \int_{-\infty}^{\infty} \mathcal{A}_{\boldsymbol{\alpha}}(k,t)\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)\,\mathrm{d}k, \\ \mathbf{Q}_m(\mathbf{r},t) &= \Theta(a-r)\sum_{\boldsymbol{\beta}}\sum_n \left[\mathcal{B}_{\boldsymbol{\beta}}^{\perp}(k_{\ell n},t)\mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k_{\ell n}) + \mathcal{B}_{\boldsymbol{\beta}}^{\parallel}(k_{\ell n},t)\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},k_{\ell n})\right], \\ \tilde{\mathbf{Q}}_\nu(\mathbf{r},t) &= \Theta(a-r)\sum_{\boldsymbol{\gamma}}\sum_n \left[\tilde{\mathcal{C}}_{\boldsymbol{\gamma}}^{\perp}(k_{\ell n},\nu;t)\mathbf{X}_{\boldsymbol{\gamma}}(\mathbf{r},k_{\ell n}) + \tilde{\mathcal{C}}_{\boldsymbol{\gamma}}^{\parallel}(k_{\ell n},\nu;t)\mathbf{L}_{\boldsymbol{\gamma}}(\mathbf{r},k_{\ell n})\right]. \end{aligned} \tag{56}$$

The mode functions $\mathbf{X}_{\boldsymbol{\beta}}$ and $\mathbf{L}_{\boldsymbol{\beta}}$ 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 (also: Appendix **??**). Specifically, the vector potential is, in the Coulomb gauge, comprised of the excitation of a series of photon modes [4], each of which has angular indices $\boldsymbol{\alpha}$ and a radial index k that determine its wave pattern in space. In free space without sources, the radial index is linked to the time evolution through the relation $k = \omega/c$ with ω the Fourier conjugate to t. However, with both free and bound charges present, this relationship will not hold in general such that each mode's time-variations will need to be accounted for separately.

The expansions for the matter and bath fields are not so obvious. However, it is useful to remember that the displacement field is simply related to the electric polarization field via $\mathbf{Q}_m(\mathbf{r}, t) = \Delta\mathbf{P}(\mathbf{r}, t)/e$. Thus, since the polarization field is always a solution to the wave equation in any region containing a homogeneous medium (or vacuum) with sharp boundaries, we can conclude that a spherical particle with sharp boundaries will contain a polarization field $\mathbf{P}(\mathbf{r}, t)$ and a corresponding displacement field $\mathbf{Q}_m(\mathbf{r}, t)$ that are solutions to the wave equation. A proof is given in Section 1.

The displacement field can therefore be completely described by a linear combination of the basis functions of the wave equation in spherical coordinates. Knowing exactly which basis functions should be included, however, requires intimate knowledge of the physical picture our mathematics is attempting to approximate. In particular, as shown in Liebsch [5] and later in Ishida and Liebsch [6], the mobile charge density inside a metal surface under the influence of an electric potential tends to "bunch" near the surface, creating a sharp feature within the charge density (averaged appropriately) with a peak within Angstroms of the boundary of the material.

This bunching agrees qualitatively with the definition of the bound charge density, $\rho_d(\mathbf{r}, t) = -(e/\Delta)\nabla \cdot \mathbf{Q}_m(\mathbf{r}, t)$, wherein the divergence of the matter displacement field is given by

$$\nabla \cdot \mathbf{Q}_m(\mathbf{r}, t) = -\frac{\Delta}{e} \left[\bar{\rho}_d(\mathbf{r}, t) + \sigma_d(\theta, \phi; t)\delta(r - a)\right]. \tag{57}$$

The first term on the right-hand side contains the bulk bound charge density $\bar{\rho}_d(\mathbf{r}, t)$ caused by discontinuities in the polarization field inside the sphere (see Appendix F.1 for details). The second contains the surface bound charge density $\sigma_d(\theta, \phi; t)$ caused by discontinuities of the polarization field at the sphere's sharp boundary. Importantly, this second term also contains the radial profile $\delta(r-a)$ that represents the infinitely-narrow limit of the sharp features discovered by Ishida and Liebsch [6].

Direct evidence of such charge bunching in materials other than metals can be seen in**[cites]**. In conjunction with the broad agreement between dielectric models of materials and optics experiments**[cites]**, this evidence suggests we can propose bound charge density model

$$\rho_d(\mathbf{r}, t) = \bar{\rho}_d(\mathbf{r}, t) + \sigma_d(\theta, \phi; t) \lim_{w \ll a} \frac{1}{w\sqrt{2\pi}} \mathrm{e}^{-\frac{(r-a)^2}{2w^2}} \tag{58}$$

for a general class of materials, where the bulk charge density $\bar{\rho}_d(\mathbf{r}, t)$ can be used to model bound charge fluctuations throughout the material and the surface charge density $\sigma_d(\theta, \phi; t) \lim_{w \ll a} \exp\left(-(r-a)^2/2w^2\right)/w\sqrt{2\pi}$ can be used to capture sharply-peaked surface features. We have replaced the Dirac delta with a very narrow Gaussian function to allow the surface peak width w to be used as a tuning parameter. Further, this model highlights the fact that

$$\frac{\partial}{\partial r} \left\{ \sigma_d(\theta, \phi; t) \lim_{w \ll a} \frac{1}{w\sqrt{2\pi}} \mathrm{e}^{-\frac{(r-a)^2}{2w^2}} \right\}_{r=a} = 0, \tag{59}$$

which is in good agreement with Liebsch [5] and Ishida and Liebsch [6].

This last property is especially useful when we examine the transport of charge density across the sphere's surface. In particular, we can appeal to quantum mechanics to describe our bound charge density via Schrödinger-picture wavefunctions $\psi_i(\mathbf{r}, t)$ such that $\rho_d(\mathbf{r}, t) = \rho_b(\mathbf{r}, t) + \sum_{i=1}^{N_m} e_{mi} |\psi_i(\mathbf{r}, t)|^2$. These wavefunctions are used only to model the mobile charges, as the background charges

not necesarily single-particle wavefunctions and are assumed to be the eigenstates of

we can define an associated probability current density

$$\mathcal{J}(\mathbf{r}, t) = \tag{60}$$

However, the behavior of $\mathbf{Q}_m(\mathbf{r},t)$ at the boundary must be considered carefully. In particular, because the surface charges of the sphere are $\sigma_b(\theta,\phi) = \hat{\mathbf{r}}\cdot\mathbf{P}(r=a,\theta,\phi;t) = (e/\Delta)\hat{\mathbf{r}}\cdot\mathbf{Q}_m(r=a,\theta,\phi;t)$, we know that the radial components of the vector harmonics that comprise $\mathbf{Q}_m(\mathbf{r},t)$ must be nonzero at the boundary. The surface-parallel components of $\mathbf{Q}_m(\mathbf{r},t)$ have no such restriction, however, such that we can let them go to zero at $r=a$.

Before inserting these new definitions of our photon and matter field into the Hamiltonian, it is convenient to rewrite any terms of our system Lagrangian/Hamiltonian involving the photon coordinates or velocities as integrals only over nonnegative wavenumbers. We can use the symmetry properties $\mathcal{A}_{\boldsymbol{\alpha}}(-k,t) = (-1)^{\ell+\delta_{TE}}\mathcal{A}_{\boldsymbol{\alpha}}(k,t)$ and $\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},-k) = (-1)^{\ell+\delta_{TE}}\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)$ to do so, such that

$$\begin{aligned}\mathbf{A}(\mathbf{r},t) &= \sum_{\boldsymbol{\alpha}}\int_{-\infty}^{\infty}\mathcal{A}_{\boldsymbol{\alpha}}(k,t)\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)\,\mathrm{d}k \\ &= \sum_{\boldsymbol{\alpha}}\int_{0}^{\infty}2\mathcal{A}_{\boldsymbol{\alpha}}(k,t)\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)\,\mathrm{d}k.\end{aligned} \tag{61}$$

Further, it will be convenient to define the matter field Helmholtz components

$$\begin{aligned}\mathbf{Q}_m^{\perp}(\mathbf{r},t) &= \Theta(a-r)\sum_{\boldsymbol{\beta}}\sum_{n}\mathcal{B}_{\boldsymbol{\beta}}^{\perp}(k_{\ell n},t)\mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k_{\ell n}), \\ \mathbf{Q}_m^{\|}(\mathbf{r},t) &= \Theta(a-r)\sum_{\boldsymbol{\beta}}\sum_{n}\mathcal{B}_{\boldsymbol{\beta}}^{\|}(k_{\ell n},t)\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},k_{\ell n}), \\ \tilde{\mathbf{Q}}_\nu^{\perp}(\mathbf{r},t) &= \Theta(a-r)\sum_{\boldsymbol{\beta}}\sum_{n}\tilde{\mathcal{C}}_{\boldsymbol{\beta}}^{\perp}(k_{\ell n},\nu;t)\mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k_{\ell n}), \\ \tilde{\mathbf{Q}}_\nu^{\|}(\mathbf{r},t) &= \Theta(a-r)\sum_{\boldsymbol{\beta}}\sum_{n}\tilde{\mathcal{C}}_{\boldsymbol{\beta}}^{\|}(k_{\ell n},\nu;t)\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},k_{\ell n}).\end{aligned} \tag{62}$$

These components are not quite the transverse and longitudinal components of each field in the traditional sense, as they aren't smooth over the entire universe. However, any terms in the Hamiltonian proportional to $\mathbf{Q}_m^2(\mathbf{r},t)$ or similar will only involve integrals over the inside of the sphere, such that the above component definitions provide a notion of Helmholtz orthogonality between the matter and bath fields that will provide some useful simplifications.

To see this, we can repeat the process of deriving the system Hamiltonian from the Lagrangian using our new matter and photon field expansions. The expansion coefficients both for the matter field and for the reservoir field can be treated as their own degrees of freedom with corresponding conjugate momenta. Taking care to note that our coordinates, as our harmonics, are zero in the case $p=1$, $m=0$, we can see that

$$\begin{aligned}L_m &= \frac{\eta_m}{2}\int\left(\dot{\mathbf{Q}}_m^2(\mathbf{r},t) - \omega_0^2\mathbf{Q}_m^2(\mathbf{r},t)\right)\mathrm{d}^3\mathbf{r} \\ &= \sum_{\boldsymbol{\beta}n}\eta_m\pi a^3 j_{\ell+1}^2(k_{\ell n}a)\left[\left(\left[\dot{\mathcal{B}}_{\boldsymbol{\beta}}^{\perp}(k_{\ell n},t)\right]^2 - \omega_0^2\left[\mathcal{B}_{\boldsymbol{\beta}}^{\perp}(k_{\ell n},t)\right]^2\right) + \left(\left[\dot{\mathcal{B}}_{\boldsymbol{\beta}}^{\|}(k_{\ell n},t)\right]^2 - \omega_0^2\left[\mathcal{B}_{\boldsymbol{\beta}}^{\|}(k_{\ell n},t)\right]^2\right)\right]\end{aligned} \tag{63}$$

and

$$\begin{aligned}L_r &= \int_{r<a}\int_0^{\infty}\frac{\eta_\nu}{2}\left(\dot{\tilde{\mathbf{Q}}}_\nu^2(\mathbf{r},t) - \nu^2\tilde{\mathbf{Q}}_\nu^2(\mathbf{r},t)\right)\mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} \\ &= \sum_{\boldsymbol{\beta}n}\int_0^{\infty}\eta_\nu\pi a^3 j_{\ell+1}^2(k_{\ell n}a)\left[\left(\left[\dot{\tilde{\mathcal{C}}}_{\boldsymbol{\beta}}^{\perp}(k_{\ell n},\nu;t)\right]^2 - \nu^2\left[\tilde{\mathcal{C}}_{\boldsymbol{\beta}}^{\perp}(k_{\ell n},\nu;t)\right]^2\right)\right. \\ &\qquad \left. + \left(\left[\dot{\tilde{\mathcal{C}}}_{\boldsymbol{\beta}}^{\|}(k_{\ell n},\nu;t)\right]^2 - \nu^2\left[\tilde{\mathcal{C}}_{\boldsymbol{\beta}}^{\|}(k_{\ell n},\nu;t)\right]^2\right)\right]\mathrm{d}\nu.\end{aligned} \tag{64}$$

Further, noting that $\Phi(\mathbf{r},t) = \Phi_f(\mathbf{r},t) + \Phi_m(\mathbf{r},t)$ where

$$
\begin{aligned}
\Phi_m(\mathbf{r},t) &= \int \frac{\rho_m(\mathbf{r'},t)}{|\mathbf{r}-\mathbf{r'}|}\ \mathrm{d}^3\mathbf{r'} \\
&= -\frac{e}{\Delta}\int \frac{\nabla'\cdot\mathbf{Q}_m^{\|}(\mathbf{r'},t)}{|\mathbf{r}-\mathbf{r'}|}\ \mathrm{d}^3\mathbf{r'} \\
&= -\frac{e}{\Delta}\int\left(\int_0^\infty \sum_{\boldsymbol{\beta}} \mathcal{Q}_{\boldsymbol{\beta}}^{\|}(\kappa,t)\nabla'\cdot\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r'},\kappa)\right)\sum_{\boldsymbol{\alpha}}\left[f_{\boldsymbol{\alpha}}^{>}(\mathbf{r})f_{\boldsymbol{\alpha}}^{<}(\mathbf{r'})\Theta(r-r') + f_{\boldsymbol{\alpha}}^{<}(\mathbf{r})f_{\boldsymbol{\alpha}}^{>}(\mathbf{r'})\Theta(r'-r)\right]\mathrm{d}^3\mathbf{r'} \\
&= \frac{e}{\Delta}\int_0^\infty \sum_{\boldsymbol{\alpha\beta}} \delta_{TE}\kappa\mathcal{Q}_{\boldsymbol{\beta}}^{\|}(\kappa,t)\sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}\left[f_{\boldsymbol{\alpha}}^{>}(\mathbf{r})\int f_{\boldsymbol{\alpha}}^{<}(\mathbf{r'})j_\ell(\kappa r')P_{\ell m}(\cos\theta')S_p(m\phi')\ \mathrm{d}^3\mathbf{r'}\right. \\
&\qquad \left. +\ f_{\boldsymbol{\alpha}}^{<}(\mathbf{r})\int f_{\boldsymbol{\alpha}}^{>}(\mathbf{r'})j_\ell(\kappa r')P_{\ell m}(\cos\theta')S_p(m\phi')\ \mathrm{d}^3\mathbf{r'}\right]\mathrm{d}\kappa \\
&= \frac{e}{\Delta}\int_0^\infty \sum_{\boldsymbol{\alpha\beta}} \delta_{TE}\kappa\mathcal{Q}_{\boldsymbol{\beta}}^{\|}(\kappa,t)\frac{4\pi}{\kappa}(1-\delta_{p1}\delta_{m0})\delta_{T'E}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}\frac{1}{\sqrt{2\ell+1}} \\
&\qquad \times\left[f_{\boldsymbol{\alpha}}^{>}(\mathbf{r})r^{\ell+2}j_{\ell+1}(\kappa t) + f_{\boldsymbol{\alpha}}^{<}(\mathbf{r})r^{-\ell+1}j_{\ell-1}(\kappa r)\right]\mathrm{d}\kappa \\
&= \frac{4\pi e}{\Delta}\int_0^\infty \sum_{\boldsymbol{\beta}} \delta_{TE}(1-\delta_{p1}\delta_{m0})\mathcal{Q}_{\boldsymbol{\beta}}^{\|}(\kappa,t)\frac{1}{\sqrt{2\ell+1}} \\
&\qquad \times\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}P_{\ell m}(\cos\theta)S_p(m\phi)\left[\frac{r^{\ell+2}}{r^{\ell+1}}j_{\ell+1}(\kappa r) + \frac{r^\ell}{r^{\ell-1}}j_{\ell-1}(\kappa r)\right]\mathrm{d}\kappa \\
&= \frac{4\pi e}{\Delta}\int_0^\infty \sum_{\boldsymbol{\beta}} \delta_{TE}(1-\delta_{p1}\delta_{m0})\mathcal{Q}_{\boldsymbol{\beta}}^{\|}(\kappa,t)\frac{1}{\sqrt{2\ell+1}} \\
&\qquad \times\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}P_{\ell m}(\cos\theta)S_p(m\phi)(2\ell+1)j_\ell(\kappa r)\ \mathrm{d}\kappa \\
&= \frac{4\pi e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \delta_{TE}(1-\delta_{p1}\delta_{m0})\mathcal{Q}_{\boldsymbol{\beta}}^{\|}(\kappa,t)\sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}j_\ell(\kappa r)P_{\ell m}(\cos\theta)S_p(m\phi)\ \mathrm{d}\kappa
\end{aligned}
\tag{65}
$$

is the electrostatic potential set up by the matter field,

$$
\begin{aligned}
L_{\mathrm{EM}} &= \frac{1}{8\pi}\int\left([\nabla\Phi(\mathbf{r},t)]^2 + \frac{1}{c^2}\dot{\mathbf{A}}^2(\mathbf{r},t) - [\nabla\times\mathbf{A}(\mathbf{r},t)]^2\right)\mathrm{d}^3\mathbf{r} \\
&= \frac{1}{8\pi}\int[\nabla\Phi(\mathbf{r},t)]^2\ \mathrm{d}^3\mathbf{r} + \sum_{\boldsymbol{\alpha}}\int_0^\infty\left(\frac{\pi}{k^2c^2}\dot{\mathcal{A}}_{\boldsymbol{\alpha}}^2(k,t) - \pi\mathcal{A}_{\boldsymbol{\alpha}}^2(k,t)\right)\mathrm{d}k,
\end{aligned}
\tag{66}
$$

and

$$
\begin{aligned}
L_{\mathrm{int}} &= \int\left(\left[\frac{e}{\Delta}\nabla\cdot\mathbf{Q}_m(\mathbf{r},t) - \rho_f(\mathbf{r},t)\right]\Phi(\mathbf{r},t) + \frac{1}{c}\left[\frac{e}{\Delta}\dot{\mathbf{Q}}_m(\mathbf{r},t) + \mathbf{J}_f(\mathbf{r},t)\right]\cdot\mathbf{A}(\mathbf{r},t)\right. \\
&\qquad \left. -\int_0^\infty \tilde{v}(\nu)\mathbf{Q}_m(\mathbf{r},t)\cdot\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\ \mathrm{d}\nu\right)\mathrm{d}^3\mathbf{r} \\
&=
\end{aligned}
\tag{67}
$$

$$
\begin{aligned}
L = &\frac{\eta_m}{2}\int\left(\left[\dot{\mathbf{Q}}_m^{\perp}(\mathbf{r},t)\right]^2+\left[\dot{\mathbf{Q}}_m^{\parallel}(\mathbf{r},t)\right]^2\right)\mathrm{d}^3\mathbf{r}+\int_0^\infty\frac{\eta_\nu}{2}\int\left(\left[\dot{\tilde{\mathbf{Q}}}_\nu^{\perp}(\mathbf{r},t)\right]^2+\left[\dot{\tilde{\mathbf{Q}}}_\nu^{\parallel}(\mathbf{r},t)\right]^2\right)\mathrm{d}^3\mathbf{r}\,\mathrm{d}\nu\\
&+\frac{1}{8\pi}\int_0^\infty\frac{1}{c^2}\dot{\mathbf{A}}^2(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}-\int_0^\infty\tilde{v}(\nu)\int\left[\mathbf{Q}_m^{\perp}(\mathbf{r},t)\cdot\dot{\tilde{\mathbf{Q}}}_\nu^{\perp}(\mathbf{r},t)+\mathbf{Q}_m^{\parallel}(\mathbf{r},t)\cdot\dot{\tilde{\mathbf{Q}}}_\nu^{\parallel}(\mathbf{r},t)\right]\mathrm{d}^3\mathbf{r}\,\mathrm{d}\nu\\
&+\frac{e}{c\Delta}\int\dot{\mathbf{Q}}_m^{\perp}(\mathbf{r},t)\cdot\mathbf{A}(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}+\dots\\
=&\frac{\eta_m}{2}\sum_{\boldsymbol{\beta}}\int_0^\infty\frac{2\pi^2}{\kappa^2}\left(\left[\dot{\mathcal{Q}}_{\boldsymbol{\beta}}^{\perp}(\kappa,t)\right]^2+\delta_{TE}\left[\dot{\mathcal{Q}}_{\boldsymbol{\beta}}^{\parallel}(\kappa,t)\right]^2\right)\mathrm{d}\kappa\\
&+\int_0^\infty\frac{\eta_\nu}{2}\sum_{\boldsymbol{\gamma}}\int_0^\infty\frac{2\pi^2}{\kappa^2}\left(\left[\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\gamma}}^{\perp}(\kappa,\nu;t)\right]^2+\delta_{TE}\left[\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\gamma}}^{\parallel}(\kappa,\nu;t)\right]^2\right)\mathrm{d}\kappa\,\mathrm{d}\nu+\sum_{\boldsymbol{\alpha}}\int_0^\infty\frac{\pi}{k^2c^2}\dot{\mathcal{A}}_{\boldsymbol{\alpha}}^2(k,t)\,\mathrm{d}k\\
&-\sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty\frac{2\pi^2}{\kappa^2}\tilde{v}(\nu)\left[\mathcal{Q}_{\boldsymbol{\beta}}^{\perp}(\kappa,t)\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\beta}}^{\perp}(\kappa,\nu;t)+\delta_{TE}\mathcal{Q}_{\boldsymbol{\beta}}^{\parallel}(\kappa,t)\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\beta}}^{\parallel}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu\\
&+\frac{e}{c\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty\frac{4\pi^2}{\kappa^2}\dot{\mathcal{Q}}_{\boldsymbol{\beta}}^{\perp}(\kappa,t)\mathcal{A}_{\boldsymbol{\beta}}(\kappa,t)\,\mathrm{d}\kappa+\dots,
\end{aligned}
\tag{68}
$$

where the terms in the ellipsis do not contain factors of any of the system's velocities. We have let $\eta_m(\mathbf{r})\to\eta_m$ and $\eta_\nu(\mathbf{r})\to\eta_\nu$, as either of the mass densities' spatial profiles is simply a Heaviside function $\Theta(a-r)$ and therefore redundant when multiplied by the Heaviside functions within the matter and reservoir field coordinates $\mathbf{Q}_m(\mathbf{r},t)$ and $\tilde{\mathbf{Q}}_\nu(\mathbf{r},t)$ or the corresponding velocities $\dot{\mathbf{Q}}_m(\mathbf{r},t)$ and $\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)$.

The conjugate momenta then become

$$
\begin{aligned}
\mathit{\Pi}_{\boldsymbol{\alpha}}(k,t)&=\frac{\partial L}{\partial\dot{\mathcal{A}}_{\boldsymbol{\alpha}}(k,t)}=\frac{2\pi}{k^2c^2}\dot{\mathcal{A}}_{\boldsymbol{\alpha}}(k,t),\\
\mathcal{P}_{\boldsymbol{\beta}}^{\perp}(k,t)&=\frac{\partial L}{\partial\dot{\mathcal{Q}}_{\boldsymbol{\beta}}^{\perp}(k,t)}=\eta_m\frac{2\pi^2}{k^2}\dot{\mathcal{Q}}_{\boldsymbol{\beta}}^{\perp}(k,t)+\frac{e}{c\Delta}\frac{4\pi^2}{k^2}\mathcal{A}_{\boldsymbol{\beta}}(k,t),\\
\mathcal{P}_{\boldsymbol{\beta}}^{\parallel}(k,t)&=\frac{\partial L}{\partial\dot{\mathcal{Q}}_{\boldsymbol{\beta}}^{\parallel}(t)}=\eta_m\frac{2\pi^2}{k^2}\dot{\mathcal{Q}}_{\boldsymbol{\beta}}^{\parallel}(k,t),\\
\tilde{\mathcal{P}}_{\boldsymbol{\gamma}}^{\perp}(k,\nu;t)&=\frac{\partial L}{\partial\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\gamma}}^{\perp}(k,\nu;t)}=\eta_\nu\frac{2\pi^2}{k^2}\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\gamma}}^{\perp}(k,\nu;t)-\frac{2\pi^2}{k^2}\tilde{v}(\nu)\mathcal{Q}_{\boldsymbol{\gamma}}^{\perp}(k,t),\\
\tilde{\mathcal{P}}_{\boldsymbol{\gamma}}^{\parallel}(k,\nu;t)&=\frac{\partial L}{\partial\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\gamma}}^{\parallel}(k,\nu;t)}=\eta_\nu\frac{2\pi^2}{k^2}\dot{\tilde{\mathcal{Q}}}_{\boldsymbol{\gamma}}^{\parallel}(k,\nu;t)-\frac{2\pi^2}{k^2}\tilde{v}(\nu)\mathcal{Q}_{\boldsymbol{\gamma}}^{\parallel}(t),
\end{aligned}
\tag{69}
$$

such that

$$
\begin{aligned}
\mathbf{\Pi}(\mathbf{r},t) &= \sum_{\boldsymbol{\alpha}} \int_0^\infty \frac{k^2}{4\pi^2} \mathit{\Pi}_{\boldsymbol{\alpha}}(k,t)\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)\ \mathrm{d}k + \frac{1}{4\pi c}\nabla\Phi(\mathbf{r},t), \\
\mathbf{P}_m^{\perp}(\mathbf{r},t) &= \sum_{\boldsymbol{\beta}} \int_0^\infty \frac{\kappa^2}{2\pi^2} \mathscr{P}_{\boldsymbol{\beta}}^{\perp}(\kappa,t)\mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\ \mathrm{d}\kappa, \\
\mathbf{P}_m^{\parallel}(\mathbf{r},t) &= \sum_{\boldsymbol{\beta}} \int_0^\infty \frac{\kappa^2}{2\pi^2} \mathscr{P}_{\boldsymbol{\beta}}^{\parallel}(\kappa,t)\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\ \mathrm{d}\kappa, \\
\tilde{\mathbf{P}}_\nu^{\perp}(\mathbf{r},t) &= \sum_{\boldsymbol{\gamma}} \int_0^\infty \frac{\kappa^2}{2\pi^2} \tilde{\mathscr{P}}_{\boldsymbol{\gamma}}^{\perp}(\kappa,\nu;t)\mathbf{X}_{\boldsymbol{\gamma}}(\mathbf{r},\kappa)\ \mathrm{d}\kappa, \\
\tilde{\mathbf{P}}_\nu^{\parallel}(\mathbf{r},t) &= \sum_{\boldsymbol{\gamma}} \int_0^\infty \frac{\kappa^2}{2\pi^2} \tilde{\mathscr{P}}_{\boldsymbol{\gamma}}^{\parallel}(\kappa,\nu;t)\mathbf{L}_{\boldsymbol{\gamma}}(\mathbf{r},\kappa)\ \mathrm{d}\kappa.
\end{aligned} \tag{70}
$$

These definitions, along with Eqs. (56), (**??**), and (**??**) allow us to rewrite H in terms of the scalar coordinates and momenta. We first note that, with

$$
1/|\mathbf{r}-\mathbf{r}'| = \sum_{\boldsymbol{\alpha}} [f_{\boldsymbol{\alpha}}^{>}(\mathbf{r})f_{\boldsymbol{\alpha}}^{<}(\mathbf{r}')\Theta(r-r') + f_{\boldsymbol{\alpha}}^{<}(\mathbf{r})f_{\boldsymbol{\alpha}}^{>}(\mathbf{r}')\Theta(r'-r)] \tag{71}
$$

and

$$
\nabla\cdot\mathbf{L}_{\boldsymbol{\alpha}}(\mathbf{r},k) = -k\delta_{TE}\sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}j_\ell(kr)P_{\ell m}(\cos\theta)S_p(m\phi), \tag{72}
$$

the scalar potential set up by the matter field is

$$
\begin{aligned}
\Phi_m(\mathbf{r},t) &= \int \frac{\rho_m(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|}\ \mathrm{d}^3\mathbf{r}' \\
&= -\frac{e}{\Delta}\int \frac{\nabla'\cdot\mathbf{Q}_m^{\parallel}(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|}\ \mathrm{d}^3\mathbf{r}' \\
&= \frac{4\pi e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \delta_{TE}(1-\delta_{p1}\delta_{m0})\mathscr{Q}_{\boldsymbol{\beta}}^{\parallel}(\kappa,t)\sqrt{K_{\ell m}}\sqrt{\ell(\ell+1)}j_\ell(\kappa r)P_{\ell m}(\cos\theta)S_p(m\phi)\ \mathrm{d}\kappa
\end{aligned} \tag{73}
$$

such that

$$
\nabla\Phi_m(\mathbf{r},t) = \frac{4\pi e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \mathscr{Q}_{\boldsymbol{\beta}}^{\parallel}(\kappa,t)\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\ \mathrm{d}\kappa. \tag{74}
$$

This allows us to say

$$
\begin{aligned}
H_m &= \sum_{s=\perp,\parallel}\int\left(\frac{[\mathbf{P}_m^s(\mathbf{r},t)]^2}{2\eta_m} + \frac{1}{2}\eta_m\omega_0^2\left[\mathbf{Q}_m^s(\mathbf{r},t)\right]^2\right)\mathrm{d}^3\mathbf{r} + \sum_{s=\perp,\parallel}\int\int_0^\infty \frac{\tilde{v}^2(\nu)}{2\eta_\nu}\left[\mathbf{Q}_m^s(\mathbf{r},t)\right]^2\ \mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} \\
&\quad + \frac{e}{\Delta}\int \mathbf{Q}_m^{\parallel}(\mathbf{r},t)\cdot\nabla\Phi_m(\mathbf{r},t)\ \mathrm{d}^3\mathbf{r} \\
&= \sum_{\boldsymbol{\beta}}\int_0^\infty\left(\frac{\kappa^2}{2\pi^2}\frac{\left[\mathscr{P}_{\boldsymbol{\beta}}^{\perp}(\kappa,t)\right]^2}{2\eta_m} + \frac{2\pi^2}{\kappa^2}\frac{\eta_m}{2}\left[\omega_0^2+\Gamma^2\right]\left[\mathscr{Q}_{\boldsymbol{\beta}}^{\perp}(\kappa,t)\right]^2\right)\mathrm{d}\kappa \\
&\quad + \sum_{\boldsymbol{\beta}}\int_0^\infty\left(\frac{\kappa^2}{2\pi^2}\frac{\left[\mathscr{P}_{\boldsymbol{\beta}}^{\parallel}(\kappa,t)\right]^2}{2\eta_m} + \frac{2\pi^2}{\kappa^2}\frac{\eta_m}{2}\left[\omega_0^2+\Gamma^2+\frac{8\pi e^2}{\eta_m\Delta^2}\right]\left[\mathscr{Q}_{\boldsymbol{\beta}}^{\parallel}(\kappa,t)\right]^2\right)\mathrm{d}\kappa
\end{aligned} \tag{75}
$$

where

$$\Gamma^2 = \int_0^\infty \frac{\tilde{v}^2(\nu)}{\eta_\nu \eta_m} \, \mathrm{d}\nu \tag{76}$$

and we have assumed that $\mathscr{Q}^s_{\boldsymbol{\beta}}(\kappa, t)$ and $\mathscr{P}^s_{\boldsymbol{\beta}}(\kappa, t)$ are zero for $(p, m) = (1, 0)$ and that the longitudinal matter coordinates and momenta are zero when $T = M$. Additionally,

$$\begin{aligned}
H_r &= \sum_{s=\perp,\parallel} \int_0^\infty \int \left(\frac{\left[\tilde{\mathbf{P}}^s_\nu(\mathbf{r}, t)\right]^2}{2\eta_\nu} + \frac{1}{2}\eta_\nu \nu^2 \left[\tilde{\mathbf{Q}}^s_\nu(\mathbf{r}, t)\right]^2 \right) \mathrm{d}^3\mathbf{r} \, \mathrm{d}\nu \\
&= \sum_{\boldsymbol{\gamma}} \int_0^\infty \int_0^\infty \left(\frac{\kappa^2}{2\pi^2} \frac{\left[\tilde{\mathscr{P}}^\perp_{\boldsymbol{\gamma}}(\kappa, \nu; t)\right]^2}{2\eta_\nu} + \frac{2\pi^2}{\kappa^2} \frac{\eta_\nu}{2} \nu^2 \left[\tilde{\mathscr{Q}}^\perp_{\boldsymbol{\gamma}}(\kappa, \nu; t)\right]^2 \right) \mathrm{d}\kappa \, \mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\gamma}} \int_0^\infty \int_0^\infty \left(\frac{\kappa^2}{2\pi^2} \frac{\left[\tilde{\mathscr{P}}^\parallel_{\boldsymbol{\gamma}}(\kappa, \nu; t)\right]^2}{2\eta_\nu} + \frac{2\pi^2}{\kappa^2} \frac{\eta_\nu}{2} \nu^2 \left[\tilde{\mathscr{Q}}^\parallel_{\boldsymbol{\gamma}}(\kappa, \nu; t)\right]^2 \right) \mathrm{d}\kappa \, \mathrm{d}\nu
\end{aligned} \tag{77}$$

and, letting $\nabla\Phi(\mathbf{r}, t) = \nabla\Phi_f(\mathbf{r}, t) + (4\pi e/\Delta) \sum_{\boldsymbol{\beta}} \int_0^\infty \mathscr{Q}^\parallel_{\boldsymbol{\beta}}(\kappa, t) \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) \, \mathrm{d}\kappa$,

$$\begin{aligned}
H_{\mathrm{EM}} &= \int \left(2\pi c^2 \left[\mathbf{\Pi}^\perp(\mathbf{r}, t)\right]^2 + 2\pi c^2 \left[\mathbf{\Pi}^\parallel(\mathbf{r}, t)\right]^2 + \frac{1}{8\pi} \left[\nabla \times \mathbf{A}(\mathbf{r}, t)\right]^2 + \frac{e^2}{2\eta_m c^2 \Delta^2} \mathbf{A}^2(\mathbf{r}, t) \right) \mathrm{d}^3\mathbf{r} \\
&\quad + \sum_i \frac{e_i^2}{2\mu_{fi} c^2} \mathbf{A}^2[\mathbf{x}_{fi}(t), t] \\
&= \sum_{\boldsymbol{\alpha}} \int_0^\infty \left(\frac{c^2 k^2}{4\pi} \mathit{\Pi}^2_{\boldsymbol{\alpha}}(k, t) + \left[\pi + \frac{4\pi^2 e^2}{\eta_m c^2 k^2 \Delta^2}\right] \mathscr{A}^2_{\boldsymbol{\alpha}}(k, t) \right) \mathrm{d}k + \sum_{\boldsymbol{\beta}} \int_0^\infty \frac{4\pi^3 e^2}{k^2 \Delta^2} \left[\mathscr{Q}^\parallel_{\boldsymbol{\beta}}(k, t)\right]^2 \, \mathrm{d}k \\
&\quad + \frac{e}{\Delta} \sum_{\boldsymbol{\beta}} \int_0^\infty \mathscr{Q}^\parallel_{\boldsymbol{\beta}}(k, t) \int \nabla\Phi_f(\mathbf{r}, t) \cdot \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, k) \, \mathrm{d}^3\mathbf{r} \, \mathrm{d}k + \frac{1}{8\pi} \int \left[\nabla\Phi_f(\mathbf{r}, t)\right]^2 \, \mathrm{d}^3\mathbf{r} \\
&\quad + \sum_{\boldsymbol{\alpha}\boldsymbol{\alpha}'} \int_0^\infty \int_0^\infty \mathscr{A}_{\boldsymbol{\alpha}}(k, t) \mathscr{A}_{\boldsymbol{\alpha}'}(k', t) \sum_i \frac{2e_i^2}{\mu_{fi} c^2} \mathbf{X}_{\boldsymbol{\alpha}}[\mathbf{x}_{fi}(t), k] \cdot \mathbf{X}_{\boldsymbol{\alpha}'}[\mathbf{x}_{fi}(t), k'] \, \mathrm{d}k \, \mathrm{d}k'.
\end{aligned} \tag{78}$$

The interaction and driving Hamiltonian terms follow suit, such that

$$\begin{aligned}
H_{\mathrm{int}} &= -\int \frac{e}{\eta_m c \Delta} \mathbf{P}^\perp_m(\mathbf{r}, t) \cdot \mathbf{A}(\mathbf{r}, t) \, \mathrm{d}^3\mathbf{r} + \int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu} \int \left(\tilde{\mathbf{P}}^\perp_\nu(\mathbf{r}, t) \cdot \mathbf{Q}^\perp_m(\mathbf{r}, t) + \tilde{\mathbf{P}}^\parallel_\nu(\mathbf{r}, t) \cdot \mathbf{Q}^\parallel_m(\mathbf{r}, t) \right) \mathrm{d}^3\mathbf{r} \\
&\quad - c \int \mathbf{\Pi}^\parallel(\mathbf{r}, t) \cdot \nabla\Phi_m(\mathbf{r}, t) \, \mathrm{d}^3\mathbf{r} \\
&= -\frac{2e}{\eta_m c \Delta} \sum_{\boldsymbol{\alpha}} \int_0^\infty \mathscr{A}_{\boldsymbol{\alpha}}(k, t) \mathscr{P}_{\boldsymbol{\alpha}}(k, t) \, \mathrm{d}k + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu} \mathscr{Q}^\perp_{\boldsymbol{\beta}}(\kappa, t) \tilde{\mathscr{P}}^\perp_{\boldsymbol{\beta}}(\kappa, \nu; t) \, \mathrm{d}\kappa \, \mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu} \mathscr{Q}^\parallel_{\boldsymbol{\beta}}(\kappa, t) \tilde{\mathscr{P}}^\parallel_{\boldsymbol{\beta}}(\kappa, \nu; t) \, \mathrm{d}\kappa \, \mathrm{d}\nu - \frac{4\pi e^2}{\Delta^2} \sum_{\boldsymbol{\beta}} \int_0^\infty \frac{2\pi^2}{\kappa^2} \left[\mathscr{Q}^\parallel_{\boldsymbol{\beta}}(\kappa, t)\right]^2 \mathrm{d}\kappa \\
&\quad - \frac{e}{\Delta} \sum_{\boldsymbol{\beta}} \int_0^\infty \mathscr{Q}^\parallel_{\boldsymbol{\beta}}(\kappa, t) \int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) \cdot \nabla\Phi_f(\mathbf{r}, t) \, \mathrm{d}^3\mathbf{r} \, \mathrm{d}\kappa
\end{aligned} \tag{79}$$

and, letting $\sum_i e_i \Phi_m[\mathbf{x}_{fi}, t] = \int \rho_f(\mathbf{r}, t)\Phi_m(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r}$ and $\rho_f(\mathbf{r}, t) = -\nabla \cdot \nabla\Phi_f(\mathbf{r}, t)/4\pi$ and using integration by parts, one finds

$$\begin{aligned}
H_{\text{drive}} &= -\sum_i \frac{e_i}{\mu_{fi}c}\mathbf{p}_{fi}(t) \cdot \mathbf{A}[\mathbf{x}_{fi}(t), t] - c\int \mathbf{\Pi}^{\parallel}(\mathbf{r}, t) \cdot \nabla\Phi_f(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r} \\
&\quad - \int \frac{e}{\Delta}\nabla \cdot \mathbf{Q}^{\parallel}_m(\mathbf{r}, t)\Phi_f(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r} + \int \rho_f(\mathbf{r}, t)\Phi_m(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r} \\
&= -\sum_i \frac{e_i}{\mu_{fi}c}\mathbf{p}_{fi}(t) \cdot \mathbf{A}[\mathbf{x}_{fi}(t), t] - c\int \frac{1}{4\pi c}\left[\nabla\Phi_f(\mathbf{r}, t) + \nabla\Phi_m(\mathbf{r}, t)\right] \cdot \nabla\Phi_f(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r} \\
&\quad + \int \frac{e}{\Delta}\mathbf{Q}^{\parallel}_m(\mathbf{r}, t) \cdot \nabla\Phi_f(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r} + \frac{1}{4\pi}\int \nabla\Phi_f(\mathbf{r}, t) \cdot \nabla\Phi_m(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r} \\
&= -\sum_i \frac{e_i}{\mu_{fi}c}\mathbf{p}_{fi}(t) \cdot \sum_{\boldsymbol{\alpha}} \int_0^\infty 2\mathcal{A}_{\boldsymbol{\alpha}}(k, t)\mathbf{X}_{\boldsymbol{\alpha}}[\mathbf{x}_{fi}(t), k]\ \mathrm{d}k - \frac{1}{4\pi}\int \left[\nabla\Phi_f(\mathbf{r}, t)\right]^2\mathrm{d}^3\mathbf{r} \\
&\quad + \frac{e}{\Delta}\sum_{\boldsymbol{\beta}} \int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa, t)\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) \cdot \nabla\Phi_f(\mathbf{r}, t)\ \mathrm{d}^3\mathbf{r}\,\mathrm{d}\kappa
\end{aligned} \tag{80}$$

Finally, the nonlinear Hamiltonian is

$$\begin{aligned}
H_{\text{NL}} &= \frac{1}{3}\sigma_0\eta_m \int \sum_{\boldsymbol{\beta}} \int_0^\infty \left[\mathcal{Q}^{\perp}_{\boldsymbol{\beta}}(\kappa, t)\mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) + \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa, t)\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa)\right]\mathrm{d}\kappa \\
&\quad \cdot \left(\sum_{\boldsymbol{\beta}'} \int_0^\infty \left[\mathcal{Q}^{\perp}_{\boldsymbol{\beta}'}(\kappa', t)\mathbf{X}_{\boldsymbol{\beta}'}(\mathbf{r}, \kappa') + \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}'}(\kappa', t)\mathbf{L}_{\boldsymbol{\beta}'}(\mathbf{r}, \kappa')\right]\mathrm{d}\kappa' \cdot \mathbf{1}_3\right. \\
&\quad \left. \cdot \sum_{\boldsymbol{\beta}''} \int_0^\infty \left[\mathcal{Q}^{\perp}_{\boldsymbol{\beta}''}(\kappa'', t)\mathbf{X}_{\boldsymbol{\beta}''}(\mathbf{r}, \kappa'') + \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}''}(\kappa'', t)\mathbf{L}_{\boldsymbol{\beta}''}(\mathbf{r}, \kappa'')\right]\mathrm{d}\kappa''\right)\mathrm{d}^3\mathbf{r} \\
&= \frac{1}{3}\sigma_0\eta_m \sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}'''} \left[\int_0^\infty\int_0^\infty\int_0^\infty \mathcal{Q}^{\perp}_{\boldsymbol{\beta}}(\kappa, t)\mathcal{Q}^{\perp}_{\boldsymbol{\beta}'}(\kappa', t)\mathcal{Q}^{\perp}_{\boldsymbol{\beta}''}(\kappa'', t)I^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'')\ \mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''\right. \\
&\quad + 3\int_0^\infty\int_0^\infty\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa, t)\mathcal{Q}^{\perp}_{\boldsymbol{\beta}'}(\kappa', t)\mathcal{Q}^{\perp}_{\boldsymbol{\beta}''}(\kappa'', t)I^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'')\ \mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&\quad + 3\int_0^\infty\int_0^\infty\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa, t)\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}'}(\kappa', t)\mathcal{Q}^{\perp}_{\boldsymbol{\beta}''}(\kappa'', t)I^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'')\ \mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&\quad \left. + \int_0^\infty\int_0^\infty\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa, t)\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}'}(\kappa', t)\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}''}(\kappa'', t)I^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'')\ \mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''\right]
\end{aligned} \tag{81}$$

where

$$\begin{aligned}
I^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'') &= \int \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) \cdot \left[\mathbf{X}_{\boldsymbol{\beta}'}(\mathbf{r}, \kappa') \cdot \mathbf{1}_3 \cdot \mathbf{X}_{\boldsymbol{\beta}''}(\mathbf{r}, \kappa'')\right]\mathrm{d}^3\mathbf{r}, \\
I^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'') &= \int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) \cdot \left[\mathbf{X}_{\boldsymbol{\beta}'}(\mathbf{r}, \kappa') \cdot \mathbf{1}_3 \cdot \mathbf{X}_{\boldsymbol{\beta}''}(\mathbf{r}, \kappa'')\right]\mathrm{d}^3\mathbf{r}, \\
I^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'') &= \int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) \cdot \left[\mathbf{L}_{\boldsymbol{\beta}'}(\mathbf{r}, \kappa') \cdot \mathbf{1}_3 \cdot \mathbf{X}_{\boldsymbol{\beta}''}(\mathbf{r}, \kappa'')\right]\mathrm{d}^3\mathbf{r}, \\
I^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa, \kappa', \kappa'') &= \int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r}, \kappa) \cdot \left[\mathbf{L}_{\boldsymbol{\beta}'}(\mathbf{r}, \kappa') \cdot \mathbf{1}_3 \cdot \mathbf{L}_{\boldsymbol{\beta}''}(\mathbf{r}, \kappa'')\right]\mathrm{d}^3\mathbf{r},
\end{aligned} \tag{82}$$

and the last three terms of our Hamiltonian are unchanged:

$$\begin{aligned} H_f &= \sum_i \frac{\mathbf{p}_{fi}^2(t)}{2\mu_{fi}} + \sum_i e_i \Phi_f[\mathbf{x}_{fi}(t), t], \\ H_{\text{bind}} &= \sum_{\alpha,\beta=m,b,r} \left[u_{\alpha\beta}^{\infty}(\mathbf{r}) + u_{\beta\alpha}^{\infty}(\mathbf{r}) \right], \\ H_{\text{self}} &= \sum_{\alpha=m,b,r,f} u_{\alpha}^{\text{self}}. \end{aligned} \tag{83}$$

4 Quantization with Classical Free Charges

To make the process of quantization easier, we can begin by demoting the free charges from dynamical objects with trajectories dependent on the system motion to "pump" sources with predestined positions and velocities. In doing so, we can remove dependence on the free charge momenta $\mathbf{p}_{fi}(t)$ from the Hamiltonian H and, in doing so, produce some dramatic simplification of the mathematics. Explicitly, one can see that, before $\mathbf{A}[\mathbf{x}_{fi}(t), t]$ is expanded in terms of its mode amplitudes and mode functions, three terms of the Hamiltonian depend on either or both of $\mathbf{p}_{fi}(t)$ or $\mathbf{A}[\mathbf{x}_{fi}(t), t]$:

$$\begin{aligned} H_f &= \sum_i \frac{\mathbf{p}_{fi}^2(t)}{2\mu_{fi}} + \ldots \\ &= \sum_i \frac{1}{2}\mu_{fi}\dot{\mathbf{x}}_{fi}^2(t) + \sum_i \frac{e_i}{c}\dot{\mathbf{x}}_{fi}(t)\cdot\mathbf{A}[\mathbf{x}_{fi}(t),t] + \sum_i \frac{e_i^2}{2\mu_{fi}c^2}\mathbf{A}^2[\mathbf{x}_{fi}(t),t] + \ldots \\ H_{\text{drive}} &= -\sum_i \frac{e_i}{\mu_{fi}c}\mathbf{p}_{fi}(t)\cdot\mathbf{A}[\mathbf{x}_{fi}(t),t] + \ldots \\ &= -\sum_i \frac{e_i}{c}\dot{\mathbf{x}}_{fi}(t)\cdot\mathbf{A}[\mathbf{x}_{fi}(t),t] - \sum_i \frac{e_i^2}{\mu_{fi}c^2}\mathbf{A}^2[\mathbf{x}_{fi}(t),t] + \ldots \\ H_{\text{EM}} &= \sum_i \frac{e_i^2}{2\mu_{fi}c^2}\mathbf{A}^2[\mathbf{x}_{fi}(t),t] + \ldots . \end{aligned} \tag{84}$$

Here, the ellipses contain all of the terms within the three Hamiltonian terms that don't depend on $\mathbf{p}_{fi}(t)$ or $\mathbf{A}[\mathbf{x}_{fi}(t), t]$. Conveniently, $H_f + H_{\text{drive}} + H_{\text{EM}} = \sum_i \mu_{fi}\dot{\mathbf{x}}_{fi}^2(t)/2 + \ldots$, such that we can ignore all of the complicated free-charge-related terms of H in the case where $\mathbf{p}_{fi}(t)$ does not need to be explicitly represented within the formalism.

In addition, it turns out that the most convenient way to rearrange the terms within H is to define $H = H'_f + H'_m + H_r + H'_{\text{EM}} + H'_{\text{int}} + H'_{\text{drive}} + H_{\text{NL}} + H_{\text{bind}} + H_{\text{self}}$ due to some convenient cancellations between the terms of Eqs. (75)–(81). The new Hamiltonian terms are given by

$$\begin{aligned} H'_f = H_f &- \sum_i \frac{e_i}{\mu_{fi}c}\mathbf{p}_{fi}(t)\cdot\mathbf{A}[\mathbf{x}_{fi}(t),t] + \sum_i \frac{e_i^2}{2\mu_{fi}c^2}\mathbf{A}^2[\mathbf{x}_{fi}(t),t] + \frac{1}{8\pi}\int \left[\nabla\Phi_f(\mathbf{r},t)\right]^2 \mathrm{d}^3\mathbf{r} \\ &- \frac{1}{4\pi}\int \left[\nabla\Phi_f(\mathbf{r},t)\right]^2 \,\mathrm{d}^3\mathbf{r} \\ = &\sum_i \frac{1}{2}\mu_{fi}\dot{\mathbf{x}}_{fi}^2(t) + \frac{1}{8\pi}\int \left[\nabla\Phi_f(\mathbf{r},t)\right]^2 \mathrm{d}^3\mathbf{r} \end{aligned} \tag{85}$$

and

$$\begin{aligned} H'_m &= H_m + \sum_{\boldsymbol{\beta}} \int_0^\infty \frac{4\pi^3 e^2}{k^2\Delta^2}\left[\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(k,t)\right]^2 \mathrm{d}k - \frac{4\pi e^2}{\Delta^2}\sum_{\boldsymbol{\beta}} \int_0^\infty \frac{2\pi^2}{\kappa^2}\left[\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right]^2 \mathrm{d}\kappa \\ &= \sum_{\boldsymbol{\beta}} \int_0^\infty \left(\frac{\kappa^2}{2\pi^2}\frac{\left[\mathcal{P}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\right]^2}{2\eta_m} + \frac{2\pi^2}{\kappa^2}\frac{\eta_m}{2}\left[\omega_0^2 + \Gamma^2\right]\left[\mathcal{Q}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\right]^2 \right) \mathrm{d}\kappa \\ &\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty \left(\frac{\kappa^2}{2\pi^2}\frac{\left[\mathcal{P}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right]^2}{2\eta_m} + \frac{2\pi^2}{\kappa^2}\frac{\eta_m}{2}\left[\omega_0^2 + \Gamma^2 + \frac{4\pi e^2}{\eta_m\Delta^2}\right]\left[\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right]^2 \right) \mathrm{d}\kappa \end{aligned} \tag{86}$$

and

$$
\begin{aligned}
H'_{\mathrm{EM}} &= H_{\mathrm{EM}} - \sum_i \frac{e_i^2}{2\mu_{fi}c^2}\mathbf{A}^2[\mathbf{x}_{fi}(t),t] - \frac{1}{8\pi}\int [\nabla\Phi_f(\mathbf{r},t)]^2\,\mathrm{d}^3\mathbf{r} \\
&\quad - \frac{e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(k,t)\int \nabla\Phi_f(\mathbf{r},t)\cdot\mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},k)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}k - \sum_{\boldsymbol{\beta}}\int_0^\infty \frac{4\pi^3e^2}{k^2\Delta^2}\left[\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(k,t)\right]^2\mathrm{d}k \\
&= \sum_{\boldsymbol{\alpha}}\int_0^\infty \left(\frac{c^2k^2}{4\pi}\mathit{\Pi}^2_{\boldsymbol{\alpha}}(k,t) + \left[\pi + \frac{4\pi^2e^2}{\eta_m c^2k^2\Delta^2}\right]\mathcal{A}^2_{\boldsymbol{\alpha}}(k,t)\right)\mathrm{d}k
\end{aligned} \tag{87}
$$

and

$$
\begin{aligned}
H'_{\mathrm{int}} &= H_{\mathrm{int}} + \frac{4\pi e^2}{\Delta^2}\sum_{\boldsymbol{\beta}}\int_0^\infty \frac{2\pi^2}{\kappa^2}\left[\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right]^2\mathrm{d}\kappa + \frac{e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}\kappa \\
&= -\frac{2e}{\eta_m c\Delta}\sum_{\boldsymbol{\alpha}}\int_0^\infty \mathcal{A}_{\boldsymbol{\alpha}}(k,t)\mathcal{P}^{\perp}_{\boldsymbol{\alpha}}(k,t)\,\mathrm{d}k + \sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu}\mathcal{Q}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\tilde{\mathcal{P}}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu}\mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\tilde{\mathcal{P}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu
\end{aligned} \tag{88}
$$

and

$$
\begin{aligned}
H'_{\mathrm{drive}} &= H_{\mathrm{drive}} + \sum_i \frac{e_i}{\mu_{fi}c}\mathbf{p}_{fi}(t)\cdot\mathbf{A}[\mathbf{x}_{fi}(t),t] + \frac{1}{4\pi}\int [\nabla\Phi_f(\mathbf{r},t)]^2\,\mathrm{d}^3\mathbf{r} \\
&\quad + \frac{e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}\kappa \\
&\quad - \frac{e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(k,t)\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},k)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}k \\
&= \frac{e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \mathcal{Q}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}\kappa.
\end{aligned} \tag{89}
$$

We are now ready to begin the quantization process. This can be formally conducted with the transformation

$$H \to \hat{H} \tag{90}$$

alongside

$$
\begin{aligned}
\mathcal{Q}^{\perp,\parallel}_{\boldsymbol{\beta}} &\to \hat{\mathcal{Q}}^{\perp,\parallel}_{\boldsymbol{\beta}}, \\
\mathcal{P}^{\perp,\parallel}_{\boldsymbol{\beta}} &\to \hat{\mathcal{P}}^{\perp,\parallel}_{\boldsymbol{\beta}}, \\
\tilde{\mathcal{Q}}^{\perp,\parallel}_{\boldsymbol{\gamma}} &\to \hat{\tilde{\mathcal{Q}}}^{\perp,\parallel}_{\boldsymbol{\gamma}}, \\
\tilde{\mathcal{P}}^{\perp,\parallel}_{\boldsymbol{\gamma}} &\to \hat{\tilde{\mathcal{P}}}^{\perp,\parallel}_{\boldsymbol{\gamma}}, \\
\mathcal{A}_{\boldsymbol{\alpha}} &\to \hat{\mathcal{A}}_{\boldsymbol{\alpha}}, \\
\mathit{\Pi}_{\boldsymbol{\alpha}} &\to \hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}.
\end{aligned} \tag{91}
$$

Each of our new coordinate and momentum operators obeys an equal-time commutation relation with its conjugate operator, such that

$$\begin{aligned}
\left[\hat{\mathscr{A}}_{\boldsymbol{\alpha}}(k,t), \hat{\Pi}_{\boldsymbol{\alpha}'}(k',t)\right] &= \mathrm{i}\hbar\delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'}\delta(k-k'), \\
\left[\hat{\mathscr{Q}}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,t), \hat{\mathscr{P}}^{\perp,\parallel}_{\boldsymbol{\beta}'}(\kappa',t)\right] &= \mathrm{i}\hbar\delta_{\boldsymbol{\beta}\boldsymbol{\beta}'}\delta(\kappa-\kappa'), \\
\left[\hat{\tilde{\mathscr{Q}}}^{\perp,\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t), \hat{\tilde{\mathscr{P}}}^{\perp,\parallel}_{\nu'\boldsymbol{\gamma}'}(\kappa',\nu';t)\right] &= \mathrm{i}\hbar\delta_{\boldsymbol{\gamma}\boldsymbol{\gamma}'}\delta(\kappa-\kappa')\delta(\nu-\nu').
\end{aligned} \tag{92}$$

Further, we can define boson ladder operators for each of the coordinate-momentum pairs. Beginning with the operators corresponding to the field variables, we can define

$$\hat{a}_{\boldsymbol{\alpha}}(k,t) = P(k)\left[u(k)\hat{\mathscr{A}}_{\boldsymbol{\alpha}}(k,t) + \mathrm{i}v(k)\hat{\mathscr{P}}_{\boldsymbol{\alpha}}(k,t)\right], \tag{93}$$

where the prefactors $P(k)$, $u(k)$, and $v(k)$ must be determined from the constraints of the system. In particular, we want the ladder operators to provide an EM Hamiltonian

$$\hat{H}'_{\mathrm{EM}} = \sum_{\boldsymbol{\alpha}}\int_0^\infty \frac{1}{2}\hbar\tilde{k}c\left[\hat{a}_{\boldsymbol{\alpha}}(k,t)\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t) + \hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\hat{a}_{\boldsymbol{\alpha}}(k,t)\right]\mathrm{d}k \tag{94}$$

and to have commutators

$$\begin{aligned}
[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}^\dagger_{\boldsymbol{\alpha}'}(k',t)], &= \delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'}\delta(k-k') \\
[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}_{\boldsymbol{\alpha}'}(k',t)] &= [\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t), \hat{a}^\dagger_{\boldsymbol{\alpha}'}(k',t)] = 0,
\end{aligned} \tag{95}$$

wherein $\hbar\tilde{k}c$ is a characteristic eigenenergy of the photon mode of wavenumber k. The form of $\tilde{k}$ can be found by plugging the explicit form of $\hat{a}_{\boldsymbol{\alpha}}(k,t)$ into Eqs. (94) and (95) such that

$$\hat{H}'_{\mathrm{EM}} = \sum_{\boldsymbol{\alpha}}\int_0^\infty \hbar\tilde{k}c\left[P^2(k)u^2(k)\hat{\mathscr{A}}^2_{\boldsymbol{\alpha}}(k,t) + P^2(k)v^2(k)\hat{\Pi}^2_{\boldsymbol{\alpha}}(k,t)\right]\mathrm{d}k \tag{96}$$

and

$$[\hat{a}_{\boldsymbol{\alpha}}(k,t), \hat{a}^\dagger_{\boldsymbol{\alpha}'}(k',t)] = 2\hbar P^2(k)u(k)v(k)\delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'}\delta(k-k'). \tag{97}$$

Comparing Eq. (96) with Eq. (87) and Eq. (97) with Eq. (95), we can see that the definitions $A(k) = \pi + 4\pi^2e^2/\eta_m c^2k^2\Delta^2$ and $B(k) = c^2k^2/4\pi$ result in the system of equations

$$\begin{aligned}
\hbar\tilde{k}cP^2(k)u^2(k) &= A(k), \\
\hbar\tilde{k}cP^2(k)v^2(k) &= B(k), \\
2\hbar P^2(k)u(k)v(k) &= 1.
\end{aligned} \tag{98}$$

This system of equations contains three equalities and four unknowns [$u(k)$, $v(k)$, $P(k)$, and $\tilde{k}$], such that we have some freedom in defining each quantity. However, we can pin down $\tilde{k}$ immediately. We find from the first and second lines above that $u(k) = [1/P(k)]\sqrt{A(k)/\hbar\tilde{k}c}$ and $v(k) = [1/P(k)]\sqrt{B(k)/\hbar\tilde{k}c}$. Plugging these results into the third line we find that $2\sqrt{A(k)B(k)}/\tilde{k}c = 1$. Therefore, substituting the full forms of $A(k)$ and $B(k)$, we find

$$\tilde{k} = k\sqrt{1 + \frac{4\pi e^2}{\eta_m c^2k^2\Delta^2}}. \tag{99}$$

Finally, we can see that Eq. (98) has no solutions except in the case $P(k) = \sqrt{1/2\hbar u(k)v(k)}$, which quickly leads us to the final constraint

$$\frac{u(k)}{v(k)} = 2\pi\frac{\tilde{k}c}{k^2c^2}. \tag{100}$$

We are free to choose $u(k) = \tilde{k}c$ such that $v(k) = k^2c^2/2\pi$ and $P(k) = \sqrt{\pi/\hbar\tilde{k}ck^2c^2}$, giving us

$$\hat{a}_{\boldsymbol{\alpha}}(k,t) = \sqrt{\frac{\pi}{\hbar\tilde{k}ck^2c^2}}\left[\tilde{k}c\hat{\mathscr{A}}_{\boldsymbol{\alpha}}(k,t) + \mathrm{i}\frac{k^2c^2}{2\pi}\hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}(k,t)\right]. \tag{101}$$

We can repeat this process for H'_m and H_r, the ladder-operator forms of which we take to be

$$\begin{aligned}
\hat{H}'_m &= \sum_{\boldsymbol{\beta}}\int_0^\infty \frac{1}{2}\hbar\Omega_\perp\left[\hat{b}^\perp_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^\perp_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa \\
&\quad + \sum_{\boldsymbol{\beta}}\int_0^\infty \frac{1}{2}\hbar\Omega_\parallel\left[\hat{b}^\parallel_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^\parallel_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa, \\
\hat{H}_r &= \sum_{\boldsymbol{\gamma}}\int_0^\infty\int_0^\infty \frac{1}{2}\hbar\nu\left[\hat{c}^\perp_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t) + \hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^\perp_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\gamma}}\int_0^\infty\int_0^\infty \frac{1}{2}\hbar\nu\left[\hat{c}^\parallel_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t) + \hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^\parallel_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu
\end{aligned} \tag{102}$$

with

$$\begin{aligned}
\Omega_\perp^2 &= \omega_0^2 + \Gamma^2, \\
\Omega_\parallel^2 &= \omega_0^2 + \Gamma^2 + \frac{4\pi e^2}{\Delta^2\eta_m}.
\end{aligned} \tag{103}$$

The resulting definitions of the ladder operators are

$$\begin{aligned}
\hat{b}^\perp_{\boldsymbol{\beta}}(\kappa,t) &= \sqrt{\frac{\pi^2\eta_m}{\kappa^2\hbar\Omega_\perp}}\left[\Omega_\perp\hat{\mathscr{Q}}^\perp_{\boldsymbol{\beta}}(\kappa,t) + \mathrm{i}\frac{\kappa^2}{2\pi^2\eta_m}\hat{\mathscr{P}}^\perp_{\boldsymbol{\beta}}(\kappa,t)\right], \\
\hat{b}^\parallel_{\boldsymbol{\beta}}(\kappa,t) &= \sqrt{\frac{\pi^2\eta_m}{\kappa^2\hbar\Omega_\parallel}}\left[\Omega_\parallel\hat{\mathscr{Q}}^\parallel_{\boldsymbol{\beta}}(\kappa,t) + \mathrm{i}\frac{\kappa^2}{2\pi^2\eta_m}\hat{\mathscr{P}}^\parallel_{\boldsymbol{\beta}}(\kappa,t)\right], \\
\hat{c}^\perp_{\boldsymbol{\gamma}}(\kappa,\nu;t) &= \sqrt{\frac{\pi^2\eta_\nu}{\kappa^2\hbar\nu}}\left[-\mathrm{i}\nu\hat{\tilde{\mathscr{Q}}}^\perp_{\boldsymbol{\gamma}}(\kappa,\nu;t) + \frac{\kappa^2}{2\pi^2\eta_\nu}\hat{\tilde{\mathscr{P}}}^\perp_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right], \\
\hat{c}^\parallel_{\boldsymbol{\gamma}}(\kappa,\nu;t) &= \sqrt{\frac{\pi^2\eta_\nu}{\kappa^2\hbar\nu}}\left[-\mathrm{i}\nu\hat{\tilde{\mathscr{Q}}}^\parallel_{\boldsymbol{\gamma}}(\kappa,\nu;t) + \frac{\kappa^2}{2\pi^2\eta_\nu}\hat{\tilde{\mathscr{P}}}^\parallel_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right],
\end{aligned} \tag{104}$$

which can be straightforwardly be seen through the last line of Eq. (98) to obey the commutation relations

$$\begin{aligned}
\left[\hat{b}^\perp_{\boldsymbol{\beta}}(\kappa,t), \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right] &= \delta_{\boldsymbol{\beta}\boldsymbol{\beta}'}\delta(\kappa-\kappa'), \\
\left[\hat{b}^\parallel_{\boldsymbol{\beta}}(\kappa,t), \hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right] &= \delta_{\boldsymbol{\beta}\boldsymbol{\beta}'}\delta(\kappa-\kappa'), \\
\left[\hat{c}^\perp_{\boldsymbol{\gamma}}(\kappa,\nu;t), \hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right] &= \delta_{\boldsymbol{\gamma}\boldsymbol{\gamma}'}\delta(\kappa-\kappa')\delta(\nu-\nu'), \\
\left[\hat{c}^\parallel_{\boldsymbol{\gamma}}(\kappa,\nu;t), \hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right] &= \delta_{\boldsymbol{\gamma}\boldsymbol{\gamma}'}\delta(\kappa-\kappa')\delta(\nu-\nu').
\end{aligned} \tag{105}$$

In total, the alternate forms of our generalized coordinates and momenta are

$$\begin{aligned}
\hat{\mathscr{A}}_{\boldsymbol{\alpha}}(k,t) &= \sqrt{\frac{\hbar k^2c^2}{4\pi\tilde{k}c}}\left[\hat{a}_{\boldsymbol{\alpha}}(k,t) + \hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\right], \\
\hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}(k,t) &= -\mathrm{i}\sqrt{\frac{\pi\hbar\tilde{k}c}{k^2c^2}}\left[\hat{a}_{\boldsymbol{\alpha}}(k,t) - \hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\right],
\end{aligned} \tag{106}$$

and

$$
\begin{aligned}
\hat{\mathcal{Q}}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) &= \sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_m\Omega_\perp}}\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right],\\
\hat{\mathcal{P}}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) &= -\mathrm{i}\sqrt{\frac{\pi^2\eta_m\hbar\Omega_\perp}{\kappa^2}}\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)-\hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right],\\
\hat{\mathcal{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t) &= \sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_m\Omega_\parallel}}\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right],\\
\hat{\mathcal{P}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t) &= -\mathrm{i}\sqrt{\frac{\pi^2\eta_m\hbar\Omega_\parallel}{\kappa^2}}\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)-\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right],
\end{aligned}
\tag{107}
$$

and

$$
\begin{aligned}
\hat{\tilde{\mathcal{Q}}}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t) &= \mathrm{i}\sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_\nu\nu}}\left[\hat{c}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t)-\hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right],\\
\hat{\tilde{\mathcal{P}}}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t) &= \sqrt{\frac{\pi^2\eta_\nu\hbar\nu}{\kappa^2}}\left[\hat{c}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t)+\hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right],\\
\hat{\tilde{\mathcal{Q}}}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t) &= \mathrm{i}\sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_\nu\nu}}\left[\hat{c}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t)-\hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right],\\
\hat{\tilde{\mathcal{P}}}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t) &= \sqrt{\frac{\pi^2\eta_\nu\hbar\nu}{\kappa^2}}\left[\hat{c}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t)+\hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right].
\end{aligned}
\tag{108}
$$

The last interesting linear parts of our Hamiltonian to be quantized are the interaction and driving Hamiltonians H'_{int} and H'_{drive}. The former is given by

$$
\begin{aligned}
\hat{H}'_{\text{int}} &= -\frac{2e}{\eta_m c\Delta}\sum_{\boldsymbol{\alpha}}\int_0^\infty \hat{\mathcal{A}}_{\boldsymbol{\alpha}}(k,t)\hat{\mathcal{P}}^{\perp}_{\boldsymbol{\alpha}}(k,t)\,\mathrm{d}k + \sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu}\hat{\mathcal{Q}}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\hat{\tilde{\mathcal{P}}}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu\\
&\quad + \sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu}\hat{\mathcal{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{\tilde{\mathcal{P}}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu\\
&= \mathrm{i}\frac{\hbar}{2}\sum_{\boldsymbol{\alpha}}\int_0^\infty \Lambda(k)\left[\hat{a}_{\boldsymbol{\alpha}}(k,t)+\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\right]\left[\hat{b}^{\perp}_{\boldsymbol{\alpha}}(k,t)-\hat{b}^{\perp\dagger}_{\boldsymbol{\alpha}}(k,t)\right]\mathrm{d}k\\
&\quad + \frac{\hbar}{2}\sum_{\boldsymbol{\beta}}\int_0^\infty V_\perp(\nu)\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\perp,\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{c}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{c}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu\\
&\quad + \frac{\hbar}{2}\sum_{\boldsymbol{\beta}}\int_0^\infty V_\parallel(\nu)\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{c}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{c}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu
\end{aligned}
\tag{109}
$$

wherein the coupling functions are

$$
\begin{aligned}
\Lambda(\kappa) &= \frac{2e}{\Delta}\sqrt{\frac{\pi\Omega_\perp}{\eta_m\tilde{\kappa}c}},\\
V_\perp(\nu) &= \tilde{v}(\nu)\sqrt{\frac{\nu}{\eta_m\eta_\nu\Omega_\perp}},\\
V_\parallel(\nu) &= \tilde{v}(\nu)\sqrt{\frac{\nu}{\eta_m\eta_\nu\Omega_\parallel}},
\end{aligned}
\tag{110}
$$

and $\tilde{\kappa} = \kappa\sqrt{1 + 4\pi e^2/\eta_m c^2\kappa^2\Delta^2}$. The latter is

$$
\begin{aligned}
\hat{H}'_{\text{drive}} &= \frac{e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}\kappa \\
&= \frac{e}{\Delta}\sum_{\boldsymbol{\beta}}\int_0^\infty \sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_m\Omega_\parallel}}\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right]\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}\,\mathrm{d}\kappa \\
&= \sum_{\boldsymbol{\beta}}\int_0^\infty F_{\boldsymbol{\beta}}(\kappa,t)\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa
\end{aligned}
\tag{111}
$$

where

$$
F_{\boldsymbol{\beta}}(\kappa,t) = \sqrt{\frac{e^2}{\Delta^2}\frac{\hbar\kappa^2}{4\pi^2\eta_m\Omega_\parallel}}\int \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},\kappa)\cdot\nabla\Phi_f(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r}
\tag{112}
$$

4.1 Diagonalization of the Linear Hamiltonian

We will ignore the exact forms of $\hat{H}_{\text{bind}}$ and $\hat{H}_{\text{self}}$, as they will not contribute meaningfully to the dynamics of our system. Further, to describe the nonlinear dynamics stemming from $\hat{H}_{\text{NL}}$ in a useful and convenient fashion, it is best to first focus on the linear Hamiltonian,

$$
\hat{H}_0 = \hat{H}'_{\text{EM}} + \hat{H}'_m + \hat{H}_r + \hat{H}'_{\text{int}} + \hat{H}'_{\text{drive}},
\tag{113}
$$

where we have implicitly defined $\hat{H} = \hat{H}_0 + \hat{H}_{\text{NL}} + \hat{H}_{\text{bind}} + \hat{H}_{\text{self}}$. This linear Hamiltonian can be diagonalized using the Fano diagonalization method as shown in Huttner and Barnett [3]. This diagonalization is most simply done by breaking the linear Hamiltonian into its transverse and longitudinal pieces, which are uncoupled. In other words,

$$
\hat{H}_0 = \hat{H}_0^{\perp} + \hat{H}_0^{\parallel},
\tag{114}
$$

wherein

$$
\hat{H}_0^{i} = \hat{H}'^{i}_{\text{EM}} + \hat{H}'^{i}_m + \hat{H}^{i}_r + \hat{H}'^{i}_{\text{int}} + \hat{H}'^{i}_{\text{drive}}
\tag{115}
$$

with $i \in \{\perp, \parallel\}$. In detail, we find that

$$
\begin{aligned}
\hat{H}'^{\perp}_{\text{EM}} &= \hat{H}'_{\text{EM}} = \sum_{\boldsymbol{\alpha}}\int_0^\infty \frac{1}{2}\hbar\tilde{k}c\left[\hat{a}_{\boldsymbol{\alpha}}(k,t)\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)+\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\hat{a}_{\boldsymbol{\alpha}}(k,t)\right]\mathrm{d}k \\
\hat{H}'^{\parallel}_{\text{EM}} &= 0,
\end{aligned}
\tag{116}
$$

and

$$
\begin{aligned}
\hat{H}'^{\perp}_m &= \sum_{\boldsymbol{\beta}}\int_0^\infty \frac{1}{2}\hbar\Omega_\perp\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa \\
\hat{H}'^{\parallel}_m &= \sum_{\boldsymbol{\beta}}\int_0^\infty \frac{1}{2}\hbar\Omega_\parallel\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)+\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa,
\end{aligned}
\tag{117}
$$

and

$$
\begin{aligned}
\hat{H}'^{\perp}_r &= \sum_{\boldsymbol{\gamma}}\int_0^\infty\int_0^\infty \frac{1}{2}\hbar\nu\left[\hat{c}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)+\hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu \\
\hat{H}'^{\parallel}_r &= \sum_{\boldsymbol{\gamma}}\int_0^\infty\int_0^\infty \frac{1}{2}\hbar\nu\left[\hat{c}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)+\hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu,
\end{aligned}
\tag{118}
$$

and

$$\hat{H}'^{\perp}_{\text{int}} = \sum_{\boldsymbol{\beta}} \int_0^\infty \mathrm{i}\frac{\hbar}{2}\Lambda(\kappa)\left[\hat{a}_{\boldsymbol{\beta}}(\kappa,t) + \hat{a}^\dagger_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) - \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa$$
$$+ \sum_{\boldsymbol{\beta}} \int_0^\infty\int_0^\infty \frac{\hbar}{2}V_{\perp}(\nu)\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{c}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t) + \hat{c}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu, \tag{119}$$

$$\hat{H}'^{\parallel}_{\text{int}} = \sum_{\boldsymbol{\beta}} \int_0^\infty\int_0^\infty \frac{\hbar}{2}V_{\parallel}(\nu)\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{c}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t) + \hat{c}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu,$$

and

$$\begin{aligned}
\hat{H}'^{\perp}_{\text{drive}} &= 0, \\
\hat{H}'^{\parallel}_{\text{drive}} &= \sum_{\boldsymbol{\beta}} \int_0^\infty F_{\boldsymbol{\beta}}(\kappa,t)\left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa.
\end{aligned} \tag{120}$$

We can further separate the transverse interaction Hamiltonian as $\hat{H}'^{\perp}_{\text{int}} = \hat{H}^{\perp}_{mr} + \hat{H}_{mp}$, where

$$\begin{aligned}
\hat{H}^{\perp}_{mr} &= \sum_{\boldsymbol{\beta}} \int_0^\infty\int_0^\infty \frac{\hbar}{2}V_{\perp}(\nu)\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{c}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t) + \hat{c}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu, \\
\hat{H}_{mp} &= \sum_{\boldsymbol{\beta}} \int_0^\infty \mathrm{i}\frac{\hbar}{2}\Lambda(\kappa)\left[\hat{a}_{\boldsymbol{\beta}}(\kappa,t) + \hat{a}^\dagger_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) - \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa.
\end{aligned} \tag{121}$$

This allows us to diagonalize the matter-reservoir part of the linear Hamiltonian such that

$$\begin{aligned}
\hat{H}^{\perp}_{\text{mat}} &= \hat{H}'^{\perp}_{m} + \hat{H}'^{\perp}_{r} + \hat{H}^{\perp}_{mr} \\
&= \sum_{\boldsymbol{\beta}} \int_0^\infty \frac{1}{2}\hbar\Omega_{\perp}\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\right]\mathrm{d}\kappa \\
&\quad + \sum_{\boldsymbol{\gamma}} \int_0^\infty\int_0^\infty \frac{1}{2}\hbar\nu\left[\hat{c}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t) + \hat{c}^{\perp\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\perp}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty\int_0^\infty \frac{\hbar}{2}V_{\perp}(\nu)\left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right]\left[\hat{c}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t) + \hat{c}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu \\
&= \sum_{\boldsymbol{\beta}} \int_0^\infty\int_0^\infty \hbar\nu\hat{B}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\hat{B}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu
\end{aligned} \tag{122}$$

and

$$
\begin{aligned}
\hat{H}^{\parallel}_{\text{mat}} &= \hat{H}'^{\parallel}_{m} + \hat{H}'^{\parallel}_{r} + \hat{H}^{\parallel}_{mr} \\
&= \sum_{\boldsymbol{\beta}} \int_0^\infty \frac{1}{2}\hbar\Omega_\parallel \left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\right] \mathrm{d}\kappa \\
&\quad + \sum_{\boldsymbol{\gamma}} \int_0^\infty \int_0^\infty \frac{1}{2}\hbar\nu \left[\hat{c}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t) + \hat{c}^{\parallel\dagger}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\hat{c}^{\parallel}_{\boldsymbol{\gamma}}(\kappa,\nu;t)\right] \mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \frac{\hbar}{2} V_\parallel(\nu) \left[\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t) + \hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right] \left[\hat{c}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t) + \hat{c}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right] \mathrm{d}\kappa\,\mathrm{d}\nu \\
&= \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\nu \hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu,
\end{aligned}
\tag{123}
$$

where we have let $\hat{H}^{\parallel}_{mr} = \hat{H}'^{\parallel}_{\text{int}}$ for consistency. The explicit forms of the hybrid operators $\hat{B}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t)$ and $\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)$ are

$$
\begin{aligned}
\hat{B}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t) &= q_\perp(\nu)\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) + s_\perp(\nu)\hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t) + \int_0^\infty \left[u_\perp(\nu,\nu')\hat{c}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu';t) + v_\perp(\nu,\nu')\hat{c}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu';t)\right] \mathrm{d}\nu', \\
\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t) &= q_\parallel(\nu)\hat{b}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t) + s_\parallel(\nu)\hat{b}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,t) + \int_0^\infty \left[u_\parallel(\nu,\nu')\hat{c}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu';t) + v_\parallel(\nu,\nu')\hat{c}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu';t)\right] \mathrm{d}\nu',
\end{aligned}
\tag{124}
$$

where the coefficients $q_{\perp,\parallel}(\nu)$, $s_{\perp,\parallel}(\nu)$, $u_{\perp,\parallel}(\nu,\nu')$, and $v_{\perp,\parallel}(\nu,\nu')$ can be found via the Fano diagonalization process shown in Appendix C by making the substitutions $g(\nu) \to V_{\perp,\parallel}(\nu)/2$, $\Omega \to \{\Omega_\perp, \Omega_\parallel\}$, $\hat{a}_{\boldsymbol{\beta}}(k,t) \to \hat{b}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,t)$, $\hat{b}_{\boldsymbol{\beta}}(k,\nu;t) \to \hat{c}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)$, and $\hat{B}_{\boldsymbol{\beta}}(k,\nu;t) \to \hat{B}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)$, respectively. The coefficients are too cumbersome to write here, but guarantee the inverse transformations

$$
\begin{aligned}
\hat{b}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,t) &= \int_0^\infty \left[q^*_{\perp,\parallel}(\nu)\hat{B}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t) - s_{\perp,\parallel}(\nu)\hat{B}^{\{\perp,\parallel\}\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right] \mathrm{d}\nu, \\
\hat{c}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t) &= \int_0^\infty \left[u^*_{\perp,\parallel}(\omega,\nu)\hat{B}^{\perp,\parallel}_{\boldsymbol{\beta}}(\kappa,\omega;t) - v_{\perp,\parallel}(\omega,\nu)\hat{B}^{\{\perp,\parallel\}\dagger}_{\boldsymbol{\beta}}(\kappa,\omega;t)\right] \mathrm{d}\omega.
\end{aligned}
\tag{125}
$$

We can then rewrite our linear Hamiltonians as

$$
\begin{aligned}
\hat{H}^{\perp}_0 &= \hat{H}'^{\perp}_{\text{EM}} + \hat{H}^{\perp}_{\text{mat}} + \hat{H}^{\perp}_{mp} \\
&= \sum_{\boldsymbol{\alpha}} \int_0^\infty \frac{\hbar\tilde{k}c}{2} \left[\hat{a}_{\boldsymbol{\alpha}}(k,t)\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t) + \hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\hat{a}_{\boldsymbol{\alpha}}(k,t)\right] \mathrm{d}k + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\nu \hat{B}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\hat{B}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty \mathrm{i}\frac{\hbar}{2}\Lambda(\kappa) \left[\hat{a}_{\boldsymbol{\beta}}(\kappa,t) + \hat{a}^\dagger_{\boldsymbol{\beta}}(\kappa,t)\right] \left[\hat{b}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) - \hat{b}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,t)\right] \mathrm{d}\kappa, \\
&= \sum_{\boldsymbol{\alpha}} \int_0^\infty \frac{\hbar\tilde{k}c}{2} \left[\hat{a}_{\boldsymbol{\alpha}}(k,t)\hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t) + \hat{a}^\dagger_{\boldsymbol{\alpha}}(k,t)\hat{a}_{\boldsymbol{\alpha}}(k,t)\right] \mathrm{d}k + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\nu \hat{B}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\hat{B}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \mathrm{i}\frac{\hbar}{2}\Lambda(\kappa)\left[q_\perp(\nu) + s_\perp(\nu)\right] \left[\hat{a}_{\boldsymbol{\beta}}(\kappa,t) + \hat{a}^\dagger_{\boldsymbol{\beta}}(\kappa,t)\right] \left[\hat{B}^{\perp}_{\boldsymbol{\beta}}(\kappa,\nu;t) - \hat{B}^{\perp\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right] \mathrm{d}\kappa\,\mathrm{d}\nu
\end{aligned}
\tag{126}
$$

and

$$
\begin{aligned}
\hat{H}_0^{\|} &= \hat{H}_{\text{mat}}^{\|} + \hat{H}_{\text{drive}}^{\|} \\
&= \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\nu \hat{B}_{\boldsymbol{\beta}}^{\|\dagger}(\kappa,\nu;t)\hat{B}_{\boldsymbol{\beta}}^{\|}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu + \sum_{\boldsymbol{\beta}} \int_0^\infty F_{\boldsymbol{\beta}}(\kappa,t)\left[\hat{b}_{\boldsymbol{\beta}}^{\|}(\kappa,t) + \hat{b}_{\boldsymbol{\beta}}^{\|\dagger}(\kappa,t)\right]\mathrm{d}\kappa \\
&= \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\nu \hat{B}_{\boldsymbol{\beta}}^{\|\dagger}(\kappa,\nu;t)\hat{B}_{\boldsymbol{\beta}}^{\|}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty F_{\boldsymbol{\beta}}(\kappa,t)\left[q_{\|}(\nu) - s_{\|}(\nu)\right]\left[\hat{B}_{\boldsymbol{\beta}}^{\|}(\kappa,\nu;t) + \hat{B}_{\boldsymbol{\beta}}^{\|\dagger}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu.
\end{aligned}
\tag{127}
$$

The longitudinal Hamiltonian is, at this point, fully hybridized, but not yet diagonal. An exact diagonalization procedure exists[7], but is only useful in the case where the driving amplitude $F_{\boldsymbol{\beta}}(\kappa,t)[q_{\|}(\nu) - s_{\|}(\nu)]$ is large with respect to $\hbar\nu$ at any time t. For weaker driving, the nondiagonal term in $\hat{H}_0^{\|}$ can be sufficiently handled via perturbation theory such that it does not need to be addressed at the moment.

The transverse Hamiltonian, however, requires one more hybridization step. Its structure is entirely parallel to that of the transverse matter Hamiltonian, such that the same hybridization scheme can be devised. First defining polariton operators $\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)$ as

$$
\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t) = \tilde{q}(\kappa,\omega)\hat{a}_{\boldsymbol{\beta}}(\kappa,t) + \tilde{s}(\kappa,\omega)\hat{a}_{\boldsymbol{\beta}}^{\dagger}(\kappa,t) + \int_0^\infty \left[\tilde{u}(\kappa;\omega,\nu)\hat{B}_{\boldsymbol{\beta}}^{\perp}(\kappa,\nu;t) + \tilde{v}(\kappa;\omega,\nu)\hat{B}_{\boldsymbol{\beta}}^{\perp\dagger}(\kappa,\nu;t)\right]\mathrm{d}\nu, \tag{128}
$$

we can use the procedure of Appendix C with the substitutions $g(\nu) \to \mathrm{i}\Lambda(\kappa)[q_{\perp}(\nu) + s_{\perp}(\nu)]/2$, $\Omega \to \tilde{k}c$, $\hat{b}_{\boldsymbol{\beta}}(k,\nu;t) \to \hat{B}_{\boldsymbol{\beta}}^{\perp}(\kappa,\nu;t)$, and $\hat{B}_{\boldsymbol{\beta}}(k,\nu;t) \to \hat{C}_{\boldsymbol{\beta}}(\kappa,\nu;t)$ to show that

$$
\begin{aligned}
\hat{a}_{\boldsymbol{\beta}}(\kappa,t) &= \int_0^\infty \left[\tilde{q}^*(\kappa,\omega)\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t) - \tilde{s}(\kappa,\omega)\hat{C}_{\boldsymbol{\beta}}^{\dagger}(\kappa,\omega;t)\right]\mathrm{d}\omega, \\
\hat{B}_{\boldsymbol{\beta}}^{\perp}(\kappa,\nu;t) &= \int_0^\infty \left[\tilde{u}^*(\kappa;\omega,\nu)\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t) - \tilde{v}(\kappa;\omega,\nu)\hat{C}_{\boldsymbol{\beta}}^{\dagger}(\kappa,\omega;t)\right]\mathrm{d}\omega,
\end{aligned}
\tag{129}
$$

and

$$
\hat{H}_0^{\perp} = \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\omega \hat{C}_{\boldsymbol{\beta}}^{\dagger}(\kappa,\omega;t)\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)\,\mathrm{d}\kappa\,\mathrm{d}\omega. \tag{130}
$$

Therefore, we can write a fully hybridized linear Hamiltonian as

$$
\begin{aligned}
\hat{H}_0 &= \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\omega \hat{C}_{\boldsymbol{\beta}}^{\dagger}(\kappa,\omega;t)\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)\,\mathrm{d}\kappa\,\mathrm{d}\omega + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty \hbar\nu \hat{B}_{\boldsymbol{\beta}}^{\|\dagger}(\kappa,\nu;t)\hat{B}_{\boldsymbol{\beta}}^{\|}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu \\
&\quad + \sum_{\boldsymbol{\beta}} \int_0^\infty \int_0^\infty F_{\boldsymbol{\beta}}(\kappa,t)\left[q_{\|}(\nu) - s_{\|}(\nu)\right]\left[\hat{B}_{\boldsymbol{\beta}}^{\|}(\kappa,\nu;t) + \hat{B}_{\boldsymbol{\beta}}^{\|\dagger}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu.
\end{aligned}
\tag{131}
$$

4.2 Finishing the Rest of it

We also have the tools to expand the nonlinear Hamiltonian in the basis of polaritons we have just defined. Quantizing in the usual way, we have

$$
\begin{aligned}
\hat{H}_{\mathrm{NL}} = \frac{1}{3}\sigma_0\eta_m \sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}'''} \Bigg[&\int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&+ 3\int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&+ 3\int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&+ \int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \Bigg]
\end{aligned} \tag{132}
$$

From here, we can rewrite the coordinate operators in terms of the dressed raising and lowering operators such that

$$
\begin{aligned}
\hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t) &= \int_0^\infty \sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_m\Omega_\parallel}}[q_\parallel(\nu)-s_\parallel(\nu)]\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\nu, \\
\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}}(\kappa,t) &= \int_0^\infty\int_0^\infty \sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_m\Omega_\perp}}[q_\perp(\nu)-s_\perp(\nu)][\tilde{u}(\kappa;\omega,\nu)-\tilde{v}(\kappa;\omega,\nu)]\left[\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}}(\kappa,\omega;t)\right]\mathrm{d}\nu\,\mathrm{d}\omega \\
&= \int_0^\infty \sqrt{\frac{\hbar\kappa^2}{4\pi^2\eta_m\Omega_\perp}}K(\kappa,\omega)\left[\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}}(\kappa,\omega;t)\right]\mathrm{d}\omega,
\end{aligned} \tag{133}
$$

where

$$
\begin{aligned}
K(\kappa,\omega) &= \int_0^\infty [q_\perp(\nu)-s_\perp(\nu)][\tilde{u}(\kappa;\omega,\nu)-\tilde{v}(\kappa;\omega,\nu)]\mathrm{d}\nu \\
&= -\frac{2\mathrm{i}}{\pi V_\perp(\omega)} - \frac{2V_\perp(\omega)}{\pi^2\omega}\int_0^\infty \frac{\nu}{V_\perp^2(\nu)}\left(PV\left\{\frac{1}{\omega-\nu}\right\} - PV\left\{\frac{1}{\omega+\nu}\right\}\right)\mathrm{d}\nu.
\end{aligned} \tag{134}
$$

With these definitions, we can see that

$$
\begin{aligned}
&\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}\int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&= \sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}\int_0^\infty\int_0^\infty\int_0^\infty I^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\perp^3}}\int_0^\infty\int_0^\infty\int_0^\infty K(\omega)K(\omega')K(\omega'') \\
&\quad\times\left[\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}}(\kappa,\omega;t)\right]\left[\hat{C}_{\boldsymbol{\beta}'}(\kappa',\omega';t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}'}(\kappa',\omega';t)\right]\left[\hat{C}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)\right] \\
&\quad\times\mathrm{d}\omega\,\mathrm{d}\omega'\,\mathrm{d}\omega''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''.
\end{aligned} \tag{135}
$$

Distribution of the binomials produces a cumbersome expression, so we'll leave this here for now. We can now immediately see that

$$
\begin{aligned}
&\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''} \int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&= \sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''} \int_0^\infty\int_0^\infty\int_0^\infty I^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\parallel^3}}\int_0^\infty\int_0^\infty\int_0^\infty \left[q_\parallel(\nu)-s_\parallel(\nu)\right]\left[q_\parallel(\nu')-s_\parallel(\nu')\right] \\
&\quad\times\left[q_\parallel(\nu'')-s_\parallel(\nu'')\right]\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',\nu';t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}'}(\kappa',\nu';t)\right] \\
&\quad\times\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}''}(\kappa'',\nu'';t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}''}(\kappa'',\nu'';t)\right]\mathrm{d}\nu\,\mathrm{d}\nu'\,\mathrm{d}\nu''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'',
\end{aligned}
\tag{136}
$$

and

$$
\begin{aligned}
&\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''} \int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&= \sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''} \int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty \frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\parallel\Omega_\perp^2}}\left[q_\parallel(\nu)-s_\parallel(\nu)\right]K(\omega')K(\omega'')I^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'') \\
&\quad\times\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\left[\hat{C}_{\boldsymbol{\beta}'}(\kappa',\omega';t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}'}(\kappa',\omega';t)\right]\left[\hat{C}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)\right] \\
&\quad\times\mathrm{d}\nu\,\mathrm{d}\omega'\,\mathrm{d}\omega''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'',
\end{aligned}
\tag{137}
$$

and

$$
\begin{aligned}
&\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''} \int_0^\infty\int_0^\infty\int_0^\infty \hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}}(\kappa,t)\hat{\mathfrak{Q}}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',t)\hat{\mathfrak{Q}}^{\perp}_{\boldsymbol{\beta}''}(\kappa'',t) I^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'')\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa'' \\
&= \sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''} \int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty \frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\parallel^2\Omega_\perp}}\left[q_\parallel(\nu)-s_\parallel(\nu)\right]\left[q_\parallel(\nu')-s_\parallel(\nu')\right]K(\omega'')I^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\kappa,\kappa',\kappa'') \\
&\quad\times\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',\nu';t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}'}(\kappa',\nu';t)\right]\left[\hat{C}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)+\hat{C}^{\dagger}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)\right] \\
&\quad\times\mathrm{d}\nu\,\mathrm{d}\nu'\,\mathrm{d}\omega''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''.
\end{aligned}
\tag{138}
$$

Finally, since the free-charge, binding, and self-energy terms of the Hamiltonian do not contain any canonical coordinates or momenta, we can say

$$
\begin{aligned}
\hat{H}'_f &= H'_f, \\
\hat{H}_{\mathrm{bind}} &= H_{\mathrm{bind}}, \\
\hat{H}_{\mathrm{self}} &= H_{\mathrm{self}}.
\end{aligned}
\tag{139}
$$

Therefore, if we introduce for brevity the coupling functions

$$
\begin{aligned}
J^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa'') &= \frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\perp^3}}K(\omega)K(\omega')K(\omega'')I^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa''),\\
J^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa'') &= \frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\perp^2\Omega_\parallel}}\left[q_\parallel(\omega)-s_\parallel(\omega)\right]K(\omega')K(\omega'')\\
&\qquad\times I^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa''),\\
J^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa'') &= \frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\perp\Omega_\parallel^2}}\left[q_\parallel(\omega)-s_\parallel(\omega)\right]\left[q_\parallel(\omega')-s_\parallel(\omega')\right]K(\omega'')\\
&\qquad\times I^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa''),\\
J^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa'') &= \frac{\kappa\kappa'\kappa''}{8\pi^3}\sqrt{\frac{\hbar^3}{\eta_m^3\Omega_\parallel^3}}\left[q_\parallel(\omega)-s_\parallel(\omega)\right]\left[q_\parallel(\omega')-s_\parallel(\omega')\right]\left[q_\parallel(\omega'')-s_\parallel(\omega'')\right]\\
&\qquad\times I^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa''),
\end{aligned}
\tag{140}
$$

we can write the complete Hamiltonian as

$$
\begin{aligned}
\hat{H} &= \hat{H}_0 + \hat{H}_{\mathrm{NL}} + \hat{H}'_f + \hat{H}_{\mathrm{bind}} + \hat{H}_{\mathrm{self}}\\
&= \sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty \hbar\omega\hat{C}^\dagger_{\boldsymbol{\beta}}(\kappa,\omega;t)\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)\,\mathrm{d}\kappa\,\mathrm{d}\omega + \sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty \hbar\nu\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)\,\mathrm{d}\kappa\,\mathrm{d}\nu\\
&\quad+\sum_{\boldsymbol{\beta}}\int_0^\infty\int_0^\infty F_{\boldsymbol{\beta}}(\kappa,t)\left[q_\parallel(\nu)-s_\parallel(\nu)\right]\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\mathrm{d}\kappa\,\mathrm{d}\nu\\
&\quad+\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty J^{(1)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\omega,\omega',\omega'';\kappa,\kappa',\kappa'')\left[\hat{C}_{\boldsymbol{\beta}}(\kappa,\omega;t)+\hat{C}^\dagger_{\boldsymbol{\beta}}(\kappa,\omega;t)\right]\\
&\qquad\times\left[\hat{C}_{\boldsymbol{\beta}'}(\kappa',\omega';t)+\hat{C}^\dagger_{\boldsymbol{\beta}'}(\kappa',\omega';t)\right]\left[\hat{C}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)+\hat{C}^\dagger_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)\right]\mathrm{d}\omega\,\mathrm{d}\omega'\,\mathrm{d}\omega''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''\\
&\quad+\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty J^{(2)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\nu,\omega',\omega'';\kappa,\kappa',\kappa'')\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\\
&\qquad\times\left[\hat{C}_{\boldsymbol{\beta}'}(\kappa',\omega';t)+\hat{C}^\dagger_{\boldsymbol{\beta}'}(\kappa',\omega';t)\right]\left[\hat{C}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)+\hat{C}^\dagger_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)\right]\mathrm{d}\nu\,\mathrm{d}\omega'\,\mathrm{d}\omega''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''\\
&\quad+\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty J^{(3)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\nu,\nu',\omega'';\kappa,\kappa',\kappa'')\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\\
&\qquad\times\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',\nu';t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}'}(\kappa',\nu';t)\right]\left[\hat{C}_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)+\hat{C}^\dagger_{\boldsymbol{\beta}''}(\kappa'',\omega'';t)\right]\mathrm{d}\nu\,\mathrm{d}\nu'\,\mathrm{d}\omega''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''\\
&\quad+\sum_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty\int_0^\infty J^{(4)}_{\boldsymbol{\beta}\boldsymbol{\beta}'\boldsymbol{\beta}''}(\nu,\nu',\nu'';\kappa,\kappa',\kappa'')\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}}(\kappa,\nu;t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}}(\kappa,\nu;t)\right]\\
&\qquad\times\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}'}(\kappa',\nu';t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}'}(\kappa',\nu';t)\right]\left[\hat{B}^{\parallel}_{\boldsymbol{\beta}''}(\kappa'',\nu'';t)+\hat{B}^{\parallel\dagger}_{\boldsymbol{\beta}''}(\kappa'',\nu'';t)\right]\mathrm{d}\nu\,\mathrm{d}\nu'\,\mathrm{d}\nu''\,\mathrm{d}\kappa\,\mathrm{d}\kappa'\,\mathrm{d}\kappa''\\
&\quad+\sum_i\frac{1}{2}\mu_{fi}\dot{\mathbf{x}}^2_{fi}(t)+\frac{1}{8\pi}\int\left[\nabla\Phi_f(\mathbf{r},t)\right]^2\,\mathrm{d}^3\mathbf{r}+\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.
\end{aligned}
\tag{141}
$$

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
Δ	$[L]^3$	cm^3
μ_{fi}, μ_{mi}, μ_{ri}	$[M]$	g
η_m, η_ν	$\frac{[M]}{[L]^3}$	$\frac{\text{g}}{\text{cm}^3}$
$\mathbf{Q}_m(\mathbf{r},t)$	$[L]$	cm
$\tilde{\mathbf{Q}}_\nu(\mathbf{r},t)$	$[L][T]^{\frac{1}{2}}$	$\text{cm}\cdot\text{s}^{\frac{1}{2}}$
$\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)$, $\mathbf{Y}_{\boldsymbol{\alpha}}(\mathbf{r},k)$, $\mathbf{L}_{\boldsymbol{\alpha}}(\mathbf{r};s)$	1	no units
$f_{\boldsymbol{\alpha}}^{>,<}(\mathbf{r})$	$[L]^{-\ell-1}$ (for $>$) or $[L]^\ell$ (for $<$)	$\text{cm}^{-\ell-1}$ or cm^ℓ
$\mathbf{G}_0(\mathbf{r},t)$	$[L]^{-1}$	cm^{-1}
$\tilde{v}(\nu)$	$\frac{[M]}{[L]^3[T]^{\frac{1}{2}}}$	$\frac{\text{g}}{\text{cm}^3\text{s}^{\frac{1}{2}}}$
σ_0	$\frac{1}{[T]^2[L]}$	$\text{s}^{-2}\text{cm}^{-1}$
$\chi^{(1)}(\mathbf{r},\omega)$	1	no units
$\chi^{(2)}(\mathbf{r};\omega',\omega-\omega')$	$\frac{[L]^2}{[C]}$	$\frac{\text{cm}}{\text{statV}} = \frac{\text{cm}^2}{\text{statC}}$
k, κ	$[L]^{-1}$	cm^{-1}
$\mathbf{A}(\mathbf{r},t)$, $\Phi(\mathbf{r},t)$	$\frac{[C]}{[L]}$	$\frac{\text{statC}}{\text{cm}}$
$\mathbf{p}_{fi}(t)$	$\frac{[M][L]}{[T]}$	$\frac{\text{g}\cdot\text{cm}}{\text{s}}$
$\mathbf{P}_m(\mathbf{r},t)$	$\frac{[M]}{[L]^2[T]}$	$\frac{\text{g}}{\text{cm}^2\text{s}}$
$\tilde{\mathbf{P}}_\nu(\mathbf{r},t)$	$\frac{[M]}{[L]^2[T]^{\frac{1}{2}}}$	$\frac{\text{g}}{\text{cm}^2\cdot\text{s}^{\frac{1}{2}}}$
$\mathbf{\Pi}(\mathbf{r},t)$	$\frac{[C][T]}{[L]^3}$	$\frac{\text{statC}\cdot\text{s}}{\text{cm}^3}$
$\mathscr{A}_{\boldsymbol{\alpha}}(k,t)$, $\hat{\mathscr{A}}_{\boldsymbol{\alpha}}(k,t)$	$[C]$	statC
$\mathfrak{Q}_{\boldsymbol{\beta}}^{\perp,\parallel}(\kappa,t)$, $\hat{\mathfrak{Q}}_{\boldsymbol{\beta}}^{\perp,\parallel}(\kappa,t)$	$[L]^2$	cm^2
$\tilde{\mathfrak{Q}}_{\boldsymbol{\gamma}}^{\perp,\parallel}(\kappa,t)$, $\hat{\tilde{\mathfrak{Q}}}_{\boldsymbol{\gamma}}^{\perp,\parallel}(\kappa,t)$	$[L]^2[T]^{\frac{1}{2}}$	$\text{cm}^2\cdot\text{s}^{\frac{1}{2}}$
$\mathit{\Pi}_{\boldsymbol{\alpha}}(k,t)$, $\hat{\mathit{\Pi}}_{\boldsymbol{\alpha}}(k,t)$	$[C][T]$	$\text{statC}\cdot\text{s}$
$\mathscr{P}_{\boldsymbol{\beta}}^{\perp,\parallel}(k,t)$, $\hat{\mathscr{P}}_{\boldsymbol{\beta}}^{\perp,\parallel}(k,t)$	$\frac{[M][L]}{[T]}$	$\frac{\text{g}\cdot\text{cm}}{\text{s}}$
$\tilde{\mathscr{P}}_{\boldsymbol{\gamma}}^{\perp,\parallel}(k,\nu;t)$, $\hat{\tilde{\mathscr{P}}}_{\boldsymbol{\gamma}}^{\perp,\parallel}(k,\nu;t)$	$\frac{[M][L]}{[T]^{\frac{1}{2}}}$	$\frac{\text{g}\cdot\text{cm}}{\text{s}^{\frac{1}{2}}}$
ω_0, Γ, $\Omega_\perp$, $\Omega_\parallel$	$\frac{1}{[T]}$	s^{-1}
$\hat{a}_{\boldsymbol{\alpha}}(k,t)$	$[L]^{\frac{1}{2}}$	$\text{cm}^{\frac{1}{2}}$
$\hat{b}_{\boldsymbol{\beta}}^{\perp,\parallel}(\kappa,t)$	$[L]^{\frac{1}{2}}$	$\text{cm}^{\frac{1}{2}}$
$\hat{c}_{\boldsymbol{\gamma}}^{\perp,\parallel}(\kappa,\nu;t)$	$[L]^{\frac{1}{2}}[T]^{\frac{1}{2}}$	$\text{cm}^{\frac{1}{2}}\text{s}^{\frac{1}{2}}$

B Harmonic Definitions

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

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

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

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

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

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

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 the transverse vector harmonics more compactly as

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

Here, we have finally introduced the fourth "type" index to the transverse harmonic notation. 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 $\mathbf{L}_{\boldsymbol{\alpha}}(\mathbf{r},k)$, $\boldsymbol{\mathcal{L}}_{\boldsymbol{\alpha}}(\mathbf{r},k)$, $\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)$, and $\boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}(\mathbf{r},k)$. By convention, we'll let $\mathbf{L}_{Mp\ell m}(\mathbf{r},k) = 0$ and $\boldsymbol{\mathcal{L}}_{Mp\ell m}(\mathbf{r},k) = 0$ as well as $\mathbf{L}_{Ep\ell m}(\mathbf{r},k) = \mathbf{L}_{p\ell m}(\mathbf{r},k)$ and $\boldsymbol{\mathcal{L}}_{Ep\ell m}(\mathbf{r},k) = \boldsymbol{\mathcal{L}}_{p\ell m}(\mathbf{r},k)$.

The symmetries of the transverse vector spherical harmonics are such that

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

with identical relations for the respective exterior transverse harmonics. Further, they 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 transverse harmonics can be defined as

$$
\begin{aligned}
\mathbf{Y}_{\boldsymbol{\alpha}}(\mathbf{r},k) &= \frac{1}{k}\nabla\times\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k), \\
\boldsymbol{\mathcal{Y}}_{\boldsymbol{\alpha}}(\mathbf{r},k) &= \frac{1}{k}\nabla\times\boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}(\mathbf{r},k).
\end{aligned}
\tag{B.9}
$$

These obey the same conjugation symmetry as $\mathbf{X}_{\boldsymbol{\alpha}}$ and $\boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}$ but with an extra factor of -1 such that

$$
\begin{aligned}
\boldsymbol{\mathcal{X}}^*_{\boldsymbol{\alpha}}(\mathbf{r},k) &= \begin{cases}(-1)^\ell\boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}(\mathbf{r},-k), & T = M,\\ (-1)^{\ell+1}\boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}(\mathbf{r},-k), & T = E;\end{cases} \\
\boldsymbol{\mathcal{Y}}^*_{\boldsymbol{\alpha}}(\mathbf{r},k) &= \begin{cases}(-1)^{\ell+1}\boldsymbol{\mathcal{Y}}_{\boldsymbol{\alpha}}(\mathbf{r},-k), & T = M,\\ (-1)^\ell\boldsymbol{\mathcal{Y}}_{\boldsymbol{\alpha}}(\mathbf{r},-k), & T = E;\end{cases}
\end{aligned}
\tag{B.10}
$$

and similar for the interior transverse harmonics. The longitudinal vector spherical harmonics obey the symmetries

$$\begin{aligned}\mathbf{L}_{p\ell m}(\mathbf{r},k) &= (-1)^{\ell+1}\mathbf{L}_{p\ell m}(\mathbf{r},-k),\\ \boldsymbol{\mathcal{L}}^{*}_{p\ell m}(\mathbf{r},k) &= (-1)^{\ell+1}\boldsymbol{\mathcal{L}}_{p\ell m}(\mathbf{r},-k).\end{aligned}\tag{B.11}$$

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

$$\int \mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)\cdot\mathbf{X}_{\boldsymbol{\alpha}'}(\mathbf{r},k')\,\mathrm{d}^3\mathbf{r} = \frac{2\pi^2}{k^2}\delta(k-k')(1-\delta_{p1}\delta_{m0})\delta_{TT'}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}\tag{B.12}$$

for $k,k'>0$ and integration across the entire universe. The simpler orthogonality relations that contribute to this simple result are given in Appendix B.1. The longitudinal interior harmonics obey the orthogonality condition

$$\int \mathbf{L}_{\boldsymbol{\alpha}}(\mathbf{r},k)\cdot\mathbf{L}_{\boldsymbol{\alpha}'}(\mathbf{r},k')\,\mathrm{d}^3\mathbf{r} = \frac{2\pi^2}{k^2}\delta(k-k')(1-\delta_{p1}\delta_{m0})\delta_{TE}\delta_{TT'}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}.\tag{B.13}$$

Accordingly, the transverse and longitudinal harmonics also satisfy convenient orthogonality relations when integrated over finite regions in r. Explicitly,

$$\begin{aligned}\int_0^{2\pi}\int_0^{\pi}\int_a^{b}\mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k)\cdot\mathbf{X}_{\boldsymbol{\alpha}'}(\mathbf{r},k')r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi &= 4\pi(1-\delta_{p1}\delta_{m0})\delta_{\boldsymbol{\alpha\alpha}'}R^{\ll}_{T\ell}(k,k';a,b),\\ \int_0^{2\pi}\int_0^{\pi}\int_a^{b}\mathbf{L}_{\boldsymbol{\alpha}}(\mathbf{r},k)\cdot\mathbf{L}_{\boldsymbol{\alpha}'}(\mathbf{r},k')r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi &= 4\pi(1-\delta_{p1}\delta_{m0})\delta_{\boldsymbol{\alpha\alpha}'}R^{\ll}_{L\ell}(k,k';a,b)\end{aligned}\tag{B.14}$$

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

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

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.

From their opposing Helmholtz symmetries, we can conclude that integrals of the inner product of $\mathbf{M}$ or $\mathbf{N}$ and $\mathbf{L}$ over all space are null. However, the same is not true when integrating over finite regions of space. Again choosing the region of integration to be finite in r, we can see that

$$\begin{aligned}\int_0^{2\pi}\int_0^{\pi}\int_a^{b}\mathbf{L}_{\boldsymbol{\alpha}}(\mathbf{r},k)\cdot\mathbf{X}_{\boldsymbol{\alpha}'}(\mathbf{r},k')r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi &= 4\pi(1-\delta_{p1}\delta_{m0})\sqrt{\ell(\ell+1)}\delta_{TE}\delta_{T'E}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}\\ &\quad\times\frac{1}{kk'}\left[bj_\ell(kb)j_\ell(k'b)-aj_\ell(ka)j_\ell(k'a)\right].\end{aligned}\tag{B.16}$$

Note that integration of the longitudinal and transverse magnetic harmonics always produces zero, even when carried out over finite regions in r. Integration of the longitudinal and transverse electric harmonics can be seen to produce zero in the limit where the integration region is allowed to become infinite. Explicitly, with $a\to 0$ and $b\to\infty$, one can see that $\lim_{r\to 0} rj_\ell(kr)j_\ell(k'r) = \lim_{r\to\infty} rj_\ell(kr)j_\ell(k'r) = 0$ for all real, positive k and k' if ℓ is an integer greater than 0.

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 $n = 1, 2, \ldots$. With these values of k and k' fixed, we can easily see that $R^{\lll}_{M\ell}(z_{\ell n}/a, z_{\ell n'}/a; 0, a) = 0$ for $n \neq n'$. Further, with

$$\lim_{k' \to k} R^{\lll}_{M\ell}(k, k'; 0, a) = \frac{a^3}{2}\left[j_\ell^2(ka) - j_{\ell-1}(ka)j_{\ell+1}(ka)\right] \tag{B.17}$$

by L'Hopital's rule, we can see that

$$R^{\lll}_{M\ell}\left(\frac{z_{\ell n}}{a}, \frac{z_{\ell n'}}{a}; 0, a\right) = -\delta_{nn'}\frac{a^3}{2} j_{\ell-1}(z_{\ell n})j_{\ell+1}(z_{\ell n}). \tag{B.18}$$

Finally, using the recursion identity $j_\ell(x) = [x/(2\ell+1)][j_{\ell-1}(x) + j_{\ell 1}(x)]$, we can see that $j_{\ell-1}(z_{n\ell}) = -j_{\ell+1}(z_{n\ell})$. Therefore, letting $k_{\boldsymbol{\alpha} n} = z_{\ell n}/a$ and $k_{\boldsymbol{\alpha}' n'} = z_{\ell' n'}/a$, we have

$$\begin{aligned}
\int_0^{2\pi}\int_0^{\pi}\int_0^{a} \mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r}, k_{\boldsymbol{\alpha} n}) \cdot \mathbf{X}_{\boldsymbol{\alpha}'}(\mathbf{r}, k_{\boldsymbol{\alpha}' n'}) r^2 \sin\theta \, \mathrm{d}r \, \mathrm{d}\theta \, \mathrm{d}\phi &= 2\pi a^3(1 - \delta_{p1}\delta_{m0})\delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'}\delta_{nn'} j_{\ell+1}^2(z_{\ell n}), \\
\int_0^{2\pi}\int_0^{\pi}\int_0^{a} \mathbf{L}_{\boldsymbol{\alpha}}(\mathbf{r}, k_{\boldsymbol{\alpha} n}) \cdot \mathbf{L}_{\boldsymbol{\alpha}'}(\mathbf{r}, k_{\boldsymbol{\alpha}' n'}) r^2 \sin\theta \, \mathrm{d}r \, \mathrm{d}\theta \, \mathrm{d}\phi &= 2\pi a^3(1 - \delta_{p1}\delta_{m0})\delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'}\delta_{nn'} j_{\ell+1}^2(z_{\ell n}).
\end{aligned} \tag{B.19}$$

For calculations involving one or more *exterior* harmonics, one simply replaces one or both of the spherical Bessel functions with spherical Hankel functions. More explicitly, if we define

$$z_\ell^i(x) = \begin{cases} j_\ell(x), & i = <, \\ h_\ell(x), & i = >, \end{cases} \tag{B.20}$$

then we can generalize Eq. (B.15) with

$$\begin{aligned}
R^{ij}_{M\ell}(k, k'; a, b) &= \int_a^b z_\ell^i(kr) z_\ell^j(k'r) r^2 \, \mathrm{d}r \\
&= \lim_{\kappa \to k'} \frac{r^2}{k^2 - \kappa^2}\left[\kappa z_{\ell-1}^j(\kappa r) z_\ell^i(kr) - k z_{\ell-1}^i(kr) z_\ell^j(\kappa r)\right]_a^b
\end{aligned} \tag{B.21}$$

and $R^{ij}_{E\ell}(k, k'; a, b) = (\ell+1)R^{ij}_{M,\ell-1}(k, k'; a, b)/(2\ell+1) + \ell R^{ij}_{M,\ell+1}(k, k'; a, b)/(2\ell+1)$. For $k' \neq k$, the limits within both the magnetic and electric radial integrals can be evaluated with simple substitution, but for $k' = k$ it is necessary to evaluate the limit carefully.

and Laplacian vector spherical harmonics

$$\begin{aligned}
\mathbf{F}_{Tp\ell m}(\mathbf{r}) &= \nabla\left\{\delta_{TE}\sqrt{(2 - \delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}} r^\ell P_{\ell m}(\cos\theta) S_p(m\phi)\right\}, \\
&= \delta_{TE}\sqrt{(2 - \delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}\left[\ell r^{\ell-1} P_{\ell m}(\cos\theta) S_p(m\phi)\hat{\mathbf{r}}\right. \\
&\quad \left. + r^{\ell-1}\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta} S_p(m\phi)\hat{\boldsymbol{\theta}} + \frac{(-1)^{p+1}m}{\sin\theta} r^{\ell-1} P_{\ell m}(\cos\theta) S_{p+1}(m\phi)\hat{\boldsymbol{\phi}}\right]
\end{aligned} \tag{B.22}$$

which have neither curl ($\nabla \times \mathbf{F}_{Tp\ell m}(\mathbf{r}) = 0$) or divergence ($\nabla \cdot \mathbf{F}_{Tp\ell m}(\mathbf{r}) = 0$).

$$
\begin{aligned}
\mathbf{M}_{p\ell m}(\mathbf{r},k) &= \sqrt{K_{\ell m}}\left[\frac{(-1)^{p+1}m}{\sin\theta}j_\ell(kr)P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\theta}} - j_\ell(kr)\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\phi}}\right], \\
\mathbf{N}_{p\ell m}(\mathbf{r},k) &= \sqrt{K_{\ell m}}\left[\frac{\ell(\ell+1)}{kr}j_\ell(kr)P_{\ell m}(\cos\theta)S_p(m\phi)\hat{\mathbf{r}}\right. \\
&\qquad \left. + \frac{1}{kr}\frac{\partial\{rj_\ell(kr)\}}{\partial r}\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\theta}} + \frac{(-1)^{p+1}m}{\sin\theta}P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\phi}}\right)\right],
\end{aligned} \tag{B.23}
$$

$$
\begin{aligned}
\boldsymbol{\mathcal{F}}_{Tp\ell m}(\mathbf{r},k) &= \nabla\left\{\delta_{TE}\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}\frac{1}{r^{\ell+1}}P_{\ell m}(\cos\theta)S_p(m\phi)\right\}, \\
&= \delta_{TE}\sqrt{(2-\delta_{m0})\frac{(\ell-m)!}{(\ell+m)!}}\left[(-\ell-1)\frac{1}{r^{\ell+2}}P_{\ell m}(\cos\theta)S_p(m\phi)\hat{\mathbf{r}}\right. \\
&\qquad \left. + \frac{1}{r^{\ell+2}}\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}S_p(m\phi)\hat{\boldsymbol{\theta}} + \frac{(-1)^{p+1}m}{\sin\theta}\frac{1}{r^{\ell+2}}P_{\ell m}(\cos\theta)S_{p+1}(m\phi)\hat{\boldsymbol{\phi}}\right]
\end{aligned} \tag{B.24}
$$

The Laplacian harmonics are either irregular at infinity (r^ℓ) or at the origin ($1/r^{\ell+1}$) for $\ell > 0$, such that their orthogonality relations can only be defined across finite regions in r:

$$
\begin{aligned}
&\int_0^{2\pi}\int_0^{\pi}\int_a^b \mathbf{F}_{\boldsymbol{\alpha}}(\mathbf{r})\cdot\mathbf{F}_{\boldsymbol{\alpha}'}(\mathbf{r})r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = 4\pi\frac{\ell}{2\ell+1}\left(b^{2\ell+1}-a^{2\ell+1}\right)\delta_{TE}\delta_{TT'}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}, \\
&\int_0^{2\pi}\int_0^{\pi}\int_a^b \boldsymbol{\mathcal{F}}_{\boldsymbol{\alpha}}(\mathbf{r})\cdot\boldsymbol{\mathcal{F}}_{\boldsymbol{\alpha}'}(\mathbf{r})r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = -4\pi\frac{\ell+1}{2\ell+1}\left(b^{-2\ell-1}-a^{-2\ell-1}\right)\delta_{TE}\delta_{TT'}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}, \\
&\int_0^{2\pi}\int_0^{\pi}\int_a^b \mathbf{F}_{\boldsymbol{\alpha}}(\mathbf{r})\cdot\boldsymbol{\mathcal{F}}_{\boldsymbol{\alpha}'}(\mathbf{r})r^2\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi = 0.
\end{aligned} \tag{B.25}
$$

A useful mixed Laplacian-transverse orthogonality conditions also exists:

$$
\begin{aligned}
&\int_0^{2\pi}\int_0^{\pi}\left[\hat{\mathbf{r}}\cdot\mathbf{N}_{p\ell m}(r,\theta,\phi;k)\right]f^i_{p'\ell'm'}(r,\theta,\phi)\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi \\
&\qquad = 4\pi\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}(1-\delta_{p1}\delta_{m0})\sqrt{\frac{\ell(\ell+1)}{2\ell+1}}\frac{j_\ell(kr)}{kr}\left[r^\ell\delta_{i,<}+\frac{\delta_{i,>}}{r^{\ell+1}}\right],
\end{aligned} \tag{B.26}
$$

as does the mixed Laplacian-longitudinal orthogonality condition

$$
\begin{aligned}
&\int_0^{2\pi}\int_0^{\pi}\left[\hat{\mathbf{r}}\cdot\mathbf{L}_{p\ell m}(r,\theta,\phi;k)\right]f^i_{p'\ell'm'}(r,\theta,\phi)\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi \\
&\qquad = 4\pi\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}(1-\delta_{p1}\delta_{m0})\frac{1}{\sqrt{2\ell+1}}\frac{1}{k}\frac{\partial j_\ell(kr)}{\partial r}\left[r^\ell\delta_{i,<}+\frac{\delta_{i,>}}{r^{\ell+1}}\right].
\end{aligned} \tag{B.27}
$$

$$
\begin{aligned}
\int\limits_{r<a} \mathbf{F}_{\boldsymbol{\alpha}}(\mathbf{r}) \cdot \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k)\, \mathrm{d}^3\mathbf{r} &= \int\limits_{r<a} \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k) \cdot \nabla f^{<}_{\boldsymbol{\alpha}}(\mathbf{r})\, \mathrm{d}^3\mathbf{r} \\
&= \int\limits_{r<a} \nabla \cdot \left\{ \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k) f^{<}_{\boldsymbol{\alpha}}(\mathbf{r}) \right\} \, \mathrm{d}^3\mathbf{r} - \int\limits_{r<a} \nabla \cdot \mathbf{X}_{\boldsymbol{\beta}}(\mathbf{r},k) f^{<}_{\boldsymbol{\alpha}}(\mathbf{r})\, \mathrm{d}^3\mathbf{r} \\
&= \int\limits_0^{2\pi}\int\limits_0^{\pi} \mathbf{X}_{\boldsymbol{\beta}}(a,\theta,\phi;k) f^{<}_{\boldsymbol{\alpha}}(a,\theta,\phi) \cdot \hat{\mathbf{r}} a^2 \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\phi - 0 \\
&= \delta_{TE}\delta_{T'E} \int\limits_0^{2\pi}\int\limits_0^{\pi} \left[\mathbf{N}_{p\ell m}(a,\theta,\phi;k) \cdot \hat{\mathbf{r}} \right] f^{<}_{p'\ell' m'}(a,\theta,\phi) a^2 \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\phi \\
&= 4\pi \delta_{TE}\delta_{T'E}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}(1-\delta_{p1}\delta_{m0}) \sqrt{\frac{\ell(\ell+1)}{2\ell+1}} \frac{j_\ell(ka)}{ka} a^{\ell+2}
\end{aligned}
\tag{B.28}
$$

$$
\begin{aligned}
\int\limits_{a<r<b} \mathbf{F}_{\boldsymbol{\alpha}}(\mathbf{r}) \cdot \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},k)\, \mathrm{d}^3\mathbf{r} &= 4\pi \delta_{TE}\delta_{T'E}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}(1-\delta_{p1}\delta_{m0}) \frac{\ell}{\sqrt{2\ell+1}} \left(\frac{j_\ell(kb)}{kb} b^{\ell+2} - \frac{j_\ell(ka)}{ka} a^{\ell+2} \right), \\
\int\limits_{a<r<b} \boldsymbol{\mathcal{F}}_{\boldsymbol{\alpha}}(\mathbf{r}) \cdot \mathbf{L}_{\boldsymbol{\beta}}(\mathbf{r},k)\, \mathrm{d}^3\mathbf{r} &= 4\pi \delta_{TE}\delta_{T'E}\delta_{pp'}\delta_{\ell\ell'}\delta_{mm'}(1-\delta_{p1}\delta_{m0}) \frac{\ell+1}{\sqrt{2\ell+1}} \frac{1}{k} \left(\frac{1}{a^\ell} j_\ell(ka) - \frac{1}{b^\ell} j_\ell(kb) \right).
\end{aligned}
\tag{B.29}
$$

The associated completeness relations to these orthogonality conditions can be derived from the Helmholtz expansion of $\mathbf{1}_2\delta(\mathbf{r}-\mathbf{r}')$, which states that

$$
\begin{aligned}
\mathbf{1}_2\delta(\mathbf{r}-\mathbf{r}') = \frac{1}{2\pi^2} \int\limits_0^{\infty} \sum_{\boldsymbol{\alpha}} k^2 \mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r},k) \mathbf{X}_{\boldsymbol{\alpha}}(\mathbf{r}',k)\, \mathrm{d}k \\
+ \frac{1}{4\pi} \sum_{\boldsymbol{\alpha}} \nabla\nabla' \left\{ f^{>}_{\boldsymbol{\alpha}}(\mathbf{r}) f^{<}_{\boldsymbol{\alpha}}(\mathbf{r}')\Theta(r-r') + f^{<}_{\boldsymbol{\alpha}}(\mathbf{r}) f^{>}_{\boldsymbol{\alpha}}(\mathbf{r}')\Theta(r'-r) \right\}.
\end{aligned}
\tag{B.30}
$$

Here, the scalar harmonics

$$
f^{i}_{\boldsymbol{\alpha}}(\mathbf{r}) = \delta_{TE} \sqrt{(2-\delta_{m0}) \frac{(\ell-m)!}{(\ell+m)!}} P_{\ell m}(\cos\theta) S_p(m\phi) \times \begin{cases} r^\ell, & i = <, \\ \dfrac{1}{r^{\ell+1}}, & i = > \end{cases}
\tag{B.31}
$$

are defined such that $\mathbf{F}_{\boldsymbol{\alpha}}(\mathbf{r}) = \nabla f^{<}_{\boldsymbol{\alpha}}(\mathbf{r})$ and $\boldsymbol{\mathcal{F}}_{\boldsymbol{\alpha}}(\mathbf{r}) = \nabla f^{>}_{\boldsymbol{\alpha}}(\mathbf{r})$. Note that, as is clear form the derivation of Section **??**, the second term is curl-free in both the $\mathbf{r}$ and $\mathbf{r}'$ and the first term is likewise transverse in both coordinate systems.

The transverse exterior harmonics also satisfy the convenient relation at large radii

$$
\begin{aligned}
\lim_{r\to\infty} \int\limits_0^{2\pi}\int\limits_0^{\pi} \boldsymbol{\mathcal{X}}_{\boldsymbol{\alpha}}(\mathbf{r},k) \times \boldsymbol{\mathcal{Y}}_{\boldsymbol{\alpha}'}(\mathbf{r},k) \cdot \hat{\mathbf{r}} r^2 \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\phi = \\
(1+\delta_{p0}\delta_{m0}-\delta_{p1}\delta_{m0}) 2\pi \frac{(-1)^{\ell+1} \mathrm{i}^{2\ell+1}}{k^2} \frac{\ell(\ell+1)}{2\ell+1} \frac{(\ell+m)!}{(\ell-m)!} \delta_{\boldsymbol{\alpha}\boldsymbol{\alpha}'}.
\end{aligned}
\tag{B.32}
$$

It is also useful from time to time to use the unitless regularized longitudinal harmonics

$$
\begin{aligned}
\mathbf{Z}_{\boldsymbol{\alpha}}(\mathbf{r};s) &= s^{-\ell+1} \nabla f^{<}_{\boldsymbol{\alpha}}(\mathbf{r})\Theta(s-r) + s^{\ell+2} \nabla f^{>}_{\boldsymbol{\alpha}}(\mathbf{r})\Theta(r-s) \\
&= \nabla \left\{ s^{-\ell+1} f^{<}_{\boldsymbol{\alpha}}(\mathbf{r})\Theta(s-r) + s^{\ell+2} f^{>}_{\boldsymbol{\alpha}}(\mathbf{r})\Theta(r-s) \right\} \\
&= s^{-\ell+1} \mathbf{F}_{\boldsymbol{\alpha}}(\mathbf{r})\Theta(s-r) + s^{\ell+2} \boldsymbol{\mathcal{F}}_{\boldsymbol{\alpha}}(\mathbf{r})\Theta(r-s)
\end{aligned}
\tag{B.33}
$$

which obey the orthogonality conditions

$$\begin{aligned}\int \mathbf{Z}_{\boldsymbol{\alpha}}(\mathbf{r};a)\cdot\mathbf{Z}_{\boldsymbol{\beta}}(\mathbf{r};b)\ \mathrm{d}^3\mathbf{r} &= 4\pi\delta_{TE}\delta_{\boldsymbol{\alpha\beta}}(1-\delta_{p1}\delta_{m0})\left[\Theta(a-b)\frac{b^{\ell+2}}{a^{\ell-1}}+\Theta(b-a)\frac{a^{\ell+2}}{b^{\ell-1}}\right],\\ \int \mathbf{Z}_{\boldsymbol{\alpha}}(\mathbf{r};s)\cdot\mathbf{Z}_{\boldsymbol{\beta}}(\mathbf{r};s)\ \mathrm{d}^3\mathbf{r} &= 4\pi s^3\delta_{TE}\delta_{\boldsymbol{\alpha\beta}}(1-\delta_{p1}\delta_{m0}).\end{aligned} \tag{B.34}$$

B.1 Useful Orthogonality Relations

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

$$\int_0^{2\pi} S_p(m\phi)S_{p'}(m'\phi)\ \mathrm{d}\phi = \pi(1+\delta_{p0}\delta_{m0}-\delta_{p1}\delta_{m0})\delta_{pp'}\delta_{mm'}, \tag{B.35}$$

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 $n=0,1,2,\dots$ 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:

$$\int_0^{\pi} P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\sin\theta\ \mathrm{d}\theta = \frac{2}{(2\ell+1)}\frac{(\ell+m)!}{(\ell-m)!}\delta_{\ell\ell'} \tag{B.36}$$

and

$$\begin{aligned}&\int_0^{\pi}\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta}+m^2\frac{P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)}{\sin^2\theta}\right)\sin\theta\ \mathrm{d}\theta\\ &\qquad = \int_0^{\pi}\ell(\ell+1)P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\sin\theta\ \mathrm{d}\theta\\ &\qquad = \frac{2\ell(\ell+1)}{(2\ell+1)}\frac{(\ell+m)!}{(\ell-m)!}\delta_{\ell\ell'},\end{aligned} \tag{B.37}$$

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

$$\begin{aligned}&\frac{\partial}{\partial\theta}\left\{\sin\theta\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta}P_{\ell m}(\cos\theta)+\sin\theta\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}P_{\ell' m}(\cos\theta)\right\}\\ &= 2\sin\theta\left(\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta}+m^2\frac{P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)}{\sin^2\theta}\right)-2\ell(\ell+1)\sin\theta P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta),\end{aligned} \tag{B.38}$$

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

$$\frac{\partial^2}{\partial\theta^2}P_{\ell m}(\cos\theta)+\frac{\cos\theta}{\sin\theta}\frac{\partial}{\partial\theta}P_{\ell m}(\cos\theta)+\left(\ell(\ell+1)-\frac{m^2}{\sin^2\theta}\right)P_{\ell m}(\cos\theta)=0. \tag{B.39}$$

Further,

$$\begin{aligned}\int_0^{\pi}\left[\frac{P_{\ell m}(\cos\theta)}{\sin\theta}\frac{\partial P_{\ell' m}(\cos\theta)}{\partial\theta}+\frac{P_{\ell' m}(\cos\theta)}{\sin\theta}\frac{\partial P_{\ell m}(\cos\theta)}{\partial\theta}\right]\sin\theta\,\mathrm{d}\theta &= \int_0^{\pi}\frac{\partial}{\partial\theta}\left\{P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\right\}\mathrm{d}\theta\\ &= P_{\ell m}(\cos\theta)P_{\ell' m}(\cos\theta)\big|_0^{\pi}\\ &= 2\delta_{m0}\delta_{\mathrm{par}(\ell),1},\end{aligned} \tag{B.40}$$

where

$$\mathrm{par}(n)=\begin{cases}0, & |n|\ \text{even},\\ 1, & |n|\ \text{odd}\end{cases} \tag{B.41}$$

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

$$
\begin{aligned}
z_\ell(x) &= \frac{x}{2\ell+1}\left[z_{\ell-1}(x) + z_{\ell+1}(x)\right], \\
\frac{\partial z_\ell(x)}{\partial x} &= z_{\ell-1}(x) - \frac{\ell+1}{x} z_\ell(x), \\
\frac{\partial z_\ell(x)}{\partial x} &= -z_{\ell+1}(x) + \frac{\ell}{x} z_\ell(x),
\end{aligned}
\tag{B.42}
$$

and

$$
\frac{\partial\{x z_\ell(x)\}}{\partial x} = \frac{x}{2\ell+1}\left[(\ell+1) z_{\ell-1}(x) - \ell z_{\ell+1}(x)\right] \tag{B.43}
$$

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

$$
\begin{aligned}
\ell(\ell+1)\frac{w_\ell(x_1) z_\ell(x_2)}{x_1 x_2} &+ \frac{1}{x_1 x_2}\frac{\partial\{x_1 w_\ell(x_1)\}}{\partial x_1}\frac{\partial\{x_2 z_\ell(x_2)\}}{\partial x_2} \\
&= \frac{1}{2\ell+1}\left[(\ell+1) w_{\ell-1}(x_1) z_{\ell-1}(x_2) + \ell w_{\ell+1}(x_1) z_{\ell+1}(x_2)\right]
\end{aligned}
\tag{B.44}
$$

and

$$
\frac{\partial z_\ell(x_1)}{\partial x_1}\frac{\partial w_\ell(x_2)}{\partial x_2} = \frac{\ell+1}{2\ell+1} z_{\ell+1}(x_1) w_{\ell+1}(x_2) + \frac{\ell}{2\ell+1} z_{\ell-1}(x_1) w_{\ell-1}(x_2) - \frac{\ell(\ell+1)}{x_1 x_2} z_\ell(x_1) w_\ell(x_2) \tag{B.45}
$$

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

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

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

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

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

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

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

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

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

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

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

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

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

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

We can then define a new function

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

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

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

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 $g^2(a) \neq 0$, 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 case considered by Huttner and Barnett[3] is the case in which $g^2(\omega)$ is only zero at $\omega = 0$. In this case, with $g^2(a) = 0$ only at $a = 0$, and only one zero of $\nu^2 - \Omega^2 z(\nu)$ exists at $a + \mathrm{i}b = \mathrm{i}\epsilon$.

- 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

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

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 sceond term on the RHS is always *less* than the first, i.e.

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

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

$$2 \int_{-\infty}^{\infty} \frac{\omega g^2(\omega)}{\omega^2 + \epsilon^2}\,\mathrm{d}\omega = 4 \int_{0}^{\infty} PV\left\{\frac{g^2(\omega)}{\omega}\right\}\mathrm{d}\omega < \Omega. \tag{C.40}$$

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

$$\int_{-\infty}^{\infty} \frac{4\Omega\nu g^2(\nu)}{|\nu^2 - \Omega^2 z(\nu)|^2}\,\mathrm{d}\nu = -\int_{C_R^-} \frac{4\Omega\nu g^2(\nu)}{|\nu^2 - \Omega^2 z(\nu)|^2}\,\mathrm{d}\nu \tag{C.41}$$

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

$$\frac{\mathrm{i}}{[x - (a+\mathrm{i}b)][x - (a-\mathrm{i}b)]} = -\frac{1}{2b}\frac{1}{x - (a+\mathrm{i}b)} + \frac{\mathrm{i}}{2b}\frac{1}{x - (a-\mathrm{i}b)} \tag{C.42}$$

such that

$$\begin{aligned}\frac{1}{|\nu^2 - \Omega^2 z(\nu)|^2} &= \frac{1}{[\nu^2 - \Omega^2 z(\nu)][\nu^2 - \Omega^2 z^*(\nu)]} \\ &= -\frac{\mathrm{i}}{4\pi\Omega g^2(\nu)}\frac{1}{\nu^2 - \Omega^2 z(\nu)} + \frac{\mathrm{i}}{4\pi\Omega g^2(\nu)}\frac{1}{\nu^2 - \Omega^2 z^*(\nu)}.\end{aligned} \tag{C.43}$$

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

$$\begin{aligned} z(\nu) &= 1 + \frac{2}{\Omega} \int\limits_{-\infty}^{\infty} \frac{g^2(\omega)}{\nu - \omega - \mathrm{i}\epsilon} \mathrm{d}\omega \\ &= 1 + \frac{2}{\Omega} \left[\int\limits_{0}^{\infty} \frac{g^2(\omega)}{\nu - \omega - \mathrm{i}\epsilon}\, \mathrm{d}\omega + \int\limits_{0}^{\infty} \frac{g^2(\omega)}{-\nu - \omega + \mathrm{i}\epsilon}\, \mathrm{d}\omega \right], \end{aligned} \tag{C.44}$$

we can say

$$\begin{aligned} \int\limits_{0}^{\infty} \frac{4\Omega\nu g^2(\nu)}{|\nu^2 - \Omega^2 z(\nu)|^2}\, \mathrm{d}\nu &= -\frac{\mathrm{i}}{\pi} \int\limits_{0}^{\infty} \left(\frac{\nu}{\nu^2 - \Omega^2 z(\nu)} - \frac{\nu}{\nu^2 - \Omega^2 z^*(\nu)} \right) \mathrm{d}\nu \\ &= \lim_{R\to\infty} \frac{\mathrm{i}}{\pi} \int\limits_{-\pi}^{0} \frac{R\mathrm{e}^{\mathrm{i}\theta}}{R^2\mathrm{e}^{2\mathrm{i}\theta} - \Omega^2 z(R\mathrm{e}^{\mathrm{i}\theta})} R\mathrm{i}\mathrm{e}^{\mathrm{i}\theta}\, \mathrm{d}\theta \\ &= 1, \end{aligned} \tag{C.45}$$

wherein we have noticed that $|z[R\exp(\mathrm{i}\theta)]| \ll R^2$ for large R. Therefore, as long as Eq. (C.40) is satisfied, we have

$$\int\limits_{0}^{\infty} \left(|q_\perp(\nu)|^2 - |s_\perp(\nu)|^2 \right) \mathrm{d}\nu = 1. \tag{C.46}$$

Next, we can see that

$$\begin{aligned} \int\limits_{0}^{\infty} [q^*(\nu)u(\nu,\nu') - s(\nu)v^*(\nu,\nu')]\, \mathrm{d}\nu = \int\limits_{0}^{\infty} \Bigg(&\left[\delta(\nu - \nu') + \frac{2\Omega g(\nu)}{\nu^2 - \Omega^2 z(\nu)} \frac{g(\nu')}{\nu - \nu' - \mathrm{i}\epsilon} \right] \frac{(\nu+\Omega)g(\nu)}{\nu^2 - \Omega^2 z^*(\nu)} \\ &- \frac{(\nu-\Omega)g(\nu)}{\nu^2 - \Omega^2 z(\nu)} \frac{2\Omega g(\nu)}{\nu^2 - \Omega^2 z^*(\nu)} PV \left\{ \frac{g(\nu')}{\nu + \nu'} \right\} \Bigg) \mathrm{d}\nu \\ = \\ = 0 \end{aligned} \tag{C.47}$$

and

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

such that

$$\int\limits_{0}^{\infty} \left[q^*(\nu)\hat{B}_{\boldsymbol{\beta}}(k,\nu;t) - s(\nu)\hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu;t) \right] \mathrm{d}\nu = \hat{a}_{\boldsymbol{\beta}}(k,t). \tag{C.49}$$

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

$$
\begin{aligned}
\int_0^\infty \left[u^*(\nu',\nu)\hat{B}_{\boldsymbol{\beta}}(k,\nu';t) - v(\nu',\nu)\hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\right] \mathrm{d}\nu' &= \hat{a}_{\boldsymbol{\beta}}(k,t) \int_0^\infty \left[q(\nu')u^*(\nu',\nu) - s^*(\nu')v(\nu',\nu)\right] \mathrm{d}\nu' \\
&+ \hat{a}^\dagger_{\boldsymbol{\beta}}(k,t) \int_0^\infty \left[s(\nu')u^*(\nu',\nu) - q^*(\nu')v(\nu',\nu)\right] \mathrm{d}\nu' \\
&+ \int_0^\infty \int_0^\infty \left[u^*(\nu',\nu)u(\nu',\omega) - v(\nu',\nu)v^*(\nu',\omega)\right] \hat{b}_{\boldsymbol{\beta}}(k,\omega;t)\, \mathrm{d}\nu'\, \mathrm{d}\omega \\
&+ \int_0^\infty \int_0^\infty \left[u^*(\nu',\nu)v(\nu',\omega) - v(\nu',\nu)u^*(\nu',\omega)\right] \hat{b}^\dagger_{\boldsymbol{\beta}}(k,\omega;t)\, \mathrm{d}\nu'\, \mathrm{d}\omega,
\end{aligned}
\tag{C.50}
$$

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

$$
\begin{aligned}
\int_0^\infty \left[u^*(\nu',\nu)v(\nu',\omega) - v(\nu',\nu)u^*(\nu',\omega)\right] \mathrm{d}\nu' &= \\
&= \\
&= 0.
\end{aligned}
\tag{C.51}
$$

Finally, the integral prefactor of the third term gives

$$
\begin{aligned}
\int_0^\infty \left[u^*(\nu',\omega)u(\nu',\omega) - v(\nu',\nu)v^*(\nu',\omega)\right] \mathrm{d}\nu' &= \\
&= \\
&= \delta(\nu-\omega)
\end{aligned}
\tag{C.52}
$$

such that

$$
\begin{aligned}
\int_0^\infty \left[u^*(\nu',\nu)\hat{B}_{\boldsymbol{\beta}}(k,\nu';t) - v(\nu',\nu)\hat{B}^\dagger_{\boldsymbol{\beta}}(k,\nu';t)\right] \mathrm{d}\nu' &= \int_0^\infty \delta(\nu-\omega)\hat{b}_{\boldsymbol{\beta}}(k,\omega;t)\, \mathrm{d}\omega \\
&= \hat{b}_{\boldsymbol{\beta}}(k,\nu;t).
\end{aligned}
\tag{C.53}
$$

C.1 Hybridized Operator Expansion Coefficients

The first hybridization step from the main text produces expansion coefficients

$$
\begin{aligned}
q_{\perp,\parallel}(\nu) &= \frac{\nu+\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega^2_{\perp,\parallel}z_{\perp,\parallel}(\nu)}, \\
s_{\perp,\parallel}(\nu) &= \frac{\nu-\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega^2_{\perp,\parallel}z_{\perp,\parallel}(\nu)}, \\
u_{\perp,\parallel}(\nu,\nu') &= \delta(\nu-\nu') + \frac{\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega^2_{\perp,\parallel}z_{\perp,\parallel}(\nu)}\frac{V_{\perp,\parallel}(\nu')}{\nu-\nu'-\mathrm{i}\epsilon}, \\
v_{\perp,\parallel}(\nu,\nu') &= \frac{\Omega_{\perp,\parallel}}{2}\frac{V_{\perp,\parallel}(\nu)}{\nu^2-\Omega^2_{\perp,\parallel}z_{\perp,\parallel}(\nu)}PV\left\{\frac{V_{\perp,\parallel}(\nu)}{\nu+\nu'}\right\},
\end{aligned}
\tag{C.54}
$$

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

$$z(\nu) \to z_{\perp,\parallel}(\nu) = 1 + \frac{1}{2\Omega_{\perp,\parallel}} \int\limits_{-\infty}^{\infty} \frac{V_{\perp,\parallel}^2(\omega)}{\nu - \omega - \mathrm{i}\epsilon}\, \mathrm{d}\omega. \tag{C.55}$$

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

$$\begin{aligned} g(\nu) &\to \frac{\mathrm{i}}{2}\Lambda(k)\left[q_\perp(\nu) + s_\perp(\nu)\right] \\ &= \frac{\mathrm{i}}{2}\Lambda(k)\left(\frac{\Omega_\perp V_\perp(\nu)}{\nu^2 - \Omega_\perp^2 z_\perp(\nu)}\right), \end{aligned} \tag{C.56}$$

and

$$\begin{aligned} z(\nu) \to \tilde{z}(\nu) &= 1 + \frac{2}{\tilde{k}c} \int\limits_{-\infty}^{\infty} \frac{\left(\frac{\mathrm{i}}{2}\Lambda(k)[q_\perp(\omega) + s_\perp(\omega)]\right)^2}{\nu - \omega - \mathrm{i}\epsilon}\, \mathrm{d}\omega \\ &= 1 - \frac{\Lambda^2(k)\Omega_\perp^2}{2\tilde{k}c} \int\limits_{-\infty}^{\infty} \frac{V_\perp^2(\omega)}{[\omega^2 - \Omega_\perp^2 z_\perp(\omega)]^2} \frac{1}{\nu - \omega - \mathrm{i}\epsilon}\, \mathrm{d}\omega \end{aligned} \tag{C.57}$$

to produce

$$\begin{aligned} \tilde{q}(k,\nu) &= \mathrm{i}\frac{(\nu + \tilde{k}c)\Lambda(k)}{2} \frac{\Omega_\perp V_\perp(\nu)}{\nu^2 - \Omega_\perp^2 z_\perp(\nu)} \frac{1}{\nu^2 - \tilde{k}^2c^2\tilde{z}(\nu)}, \\ \tilde{s}(k,\nu) &= \mathrm{i}\frac{(\nu - \tilde{k}c)\Lambda(k)}{2} \frac{\Omega_\perp V_\perp(\nu)}{\nu^2 - \Omega_\perp^2 z_\perp(\nu)} \frac{1}{\nu^2 - \tilde{k}^2c^2\tilde{z}(\nu)}, \\ \tilde{u}(k;\nu,\nu') &= \delta(\nu - \nu') + \frac{2\tilde{k}c}{\nu^2 - \tilde{k}c\tilde{z}(\nu)} \frac{\mathrm{i}}{2}\Lambda(k) \frac{\Omega_\perp V_\perp(\nu)}{\nu^2 - \Omega_\perp^2 z_\perp(\nu)} \frac{\mathrm{i}}{2}\Lambda(k) \frac{\Omega_\perp V_\perp(\nu')}{\nu'^2 - \Omega_\perp^2 z_\perp(\nu')} \frac{1}{\nu - \nu' - \mathrm{i}\epsilon} \\ &= \delta(\nu - \nu') - \frac{\tilde{k}c\Lambda^2(k)\Omega_\perp^2}{2} \frac{V_\perp(\nu)}{\left[\nu^2 - \tilde{k}^2c^2\tilde{z}(\nu)\right][\nu^2 - \Omega_\perp^2 z_\perp(\nu)][\nu'^2 - \Omega_\perp^2 z_\perp(\nu')]} \frac{V_\perp(\nu')}{\nu - \nu' - \mathrm{i}\epsilon}, \\ \tilde{v}(k;\nu,\nu') &= -\frac{\tilde{k}c\Lambda^2(k)\Omega_\perp^2}{2} \frac{V_\perp(\nu)}{\left[\nu^2 - \tilde{k}^2c^2\tilde{z}(\nu)\right][\nu^2 - \Omega_\perp^2 z_\perp(\nu)][\nu'^2 - \Omega_\perp^2 z_\perp(\nu')]} PV\left\{\frac{V_\perp(\nu')}{\nu + \nu'}\right\}. \end{aligned} \tag{C.58}$$

D Dependent matter: 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

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial \mathcal{L}'}{\partial \dot{F}(\mathbf{r},t)}\right\} = \frac{\partial \mathcal{L}'}{\partial F(\mathbf{r},t)} - \sum_{i=1}^{3} \frac{\partial}{\partial r_i} \frac{\partial \mathcal{L}'}{\partial \left(\frac{\partial F(\mathbf{r},t)}{\partial r_i}\right)}, \tag{D.1}$$

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. The modified Lagrangian density $\mathcal{L}' = \mathcal{L} - (1/4\pi c)\nabla\Phi(\mathbf{r},t)\cdot\dot{\mathbf{A}}(\mathbf{r},t)$ provides the same Lagrangian, i.e. $\int \mathcal{L}\ \mathrm{d}^3\mathbf{r} = \int \mathcal{L}'\ \mathrm{d}^3\mathbf{r} = L$, due to the orthogonality under integration (see Eq. [**??**]) of the longitudinal gradient of the scalar potential and transverse (Coulomb gauge) vector potential. Here, the use of $\mathcal{L}'$ provides mathematical simplicity.

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

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

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

$$
G(t-t') = \frac{\Theta(t-t')}{\eta_\nu(\mathbf{r})\nu}\sin(\nu[t-t']) \tag{D.3}
$$

of the oscillator differential equation

$$
\eta_\nu(\mathbf{r})\ddot{G}(t-t') + \eta_\nu(\mathbf{r})\nu^2 G(t-t') = \begin{cases}\delta(t-t'), & t > t'; \\ 0, & t < t';\end{cases} \tag{D.4}
$$

we can see that

$$
\begin{aligned}
\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t) &= \dot{\tilde{\mathbf{Q}}}_\nu^{(0)}(\mathbf{r},t) + \frac{\mathrm{d}}{\mathrm{d}t}\int_{-\infty}^{\infty} G(t-t')\tilde{v}(\nu)\dot{\mathbf{Q}}_m(\mathbf{r},t')\,\mathrm{d}t' \\
&= \dot{\tilde{\mathbf{Q}}}_\nu^{(0)}(\mathbf{r},t) + \int_{-\infty}^{\infty} \frac{\Theta(t-t')}{\eta_\nu(\mathbf{r})}\cos(\nu[t-t'])\tilde{v}(\nu)\dot{\mathbf{Q}}_m(\mathbf{r},t')\,\mathrm{d}t'.
\end{aligned}
\tag{D.5}
$$

where $\dot{\tilde{\mathbf{Q}}}_\nu^{(0)}(\mathbf{r},t)$ is the ν^{th} velocity field at t generated by stimuli far in the past (formally, at the initial time $-\infty$). The inverse of the reservoir mass-density, $1/\eta_\nu(\mathbf{r}) = \eta_\nu^{-1}(\mathbf{r})$, is taken to imply use of the function $\eta_\nu^{-1}(\mathbf{r}) = \Theta(a-r)\Delta/\sum_{i=1}^{N_r}\mu_{ri}\Theta[\mathbf{r}\in\mathbb{V}(\mathbf{r}_i)]$, i.e. is assumed to be equal to Δ/μ_{ri} within each small region $\mathbb{V}(\mathbf{r}_i)$ inside the sphere and zero for all $r > a$. We can then define a loss function

$$
\gamma_0(t-t') = \frac{1}{\eta_m(\mathbf{r})}\int_0^\infty \frac{\tilde{v}^2(\nu)}{\eta_\nu(\mathbf{r})}\cos(\nu[t-t'])\,\mathrm{d}\nu \tag{D.6}
$$

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

$$
\mathbf{N}_{\text{therm}}(\mathbf{r},t) = -\int_0^\infty \tilde{v}(\nu)\dot{\tilde{\mathbf{Q}}}_\nu^{(0)}(\mathbf{r},t)\,\mathrm{d}\nu \tag{D.7}
$$

using an identical assumption for the inverse of the mass-density of the matter field (simply let $\mu_{ri}\to\mu_{mi}$, etc.) such that the first equation of Eq. (D.2) becomes

$$
\begin{aligned}
&\eta_m(\mathbf{r})\ddot{\mathbf{Q}}_m(\mathbf{r},t) + \eta_m(\mathbf{r})\int_{-\infty}^{t}\gamma_0(t-t')\dot{\mathbf{Q}}_m(\mathbf{r},t')\,\mathrm{d}t' + \eta_m(\mathbf{r})\omega_0^2\mathbf{Q}_m(\mathbf{r},t) \\
&\qquad + \eta_m(\mathbf{r})\sigma_0\mathbf{Q}_m(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}_m(\mathbf{r},t) = \frac{e}{\Delta}\left(-\nabla\Phi(\mathbf{r},t) - \frac{1}{c}\dot{\mathbf{A}}(\mathbf{r})\right) + \mathbf{N}_{\text{therm}}(\mathbf{r},t).
\end{aligned}
\tag{D.8}
$$

Solutions to Eq. (D.8) 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:

$$\begin{aligned}\mathbf{Q}_m(\mathbf{r},t) &= \mathbf{Q}_m^{(1)}(\mathbf{r},t) + \mathbf{Q}_m^{(2)}(\mathbf{r},t) + \dots \\ &= \sum_{n=1}^{\infty} \lambda^{n-1} \mathcal{Q}_m^{(n)}(\mathbf{r},t).\end{aligned} \tag{D.9}$$

Here, the functions $\mathcal{Q}_m^{(n)}(\mathbf{r},t)$ 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

$$\begin{aligned}&\eta_m(\mathbf{r})\ddot{\mathbf{Q}}_m^{(1)}(\mathbf{r},t) + \eta_m(\mathbf{r})\int_{-\infty}^{t} \gamma_0(t-t')\dot{\mathbf{Q}}_m^{(1)}(\mathbf{r},t')\,\mathrm{d}t' + \eta_m(\mathbf{r})\omega_0^2\mathbf{Q}_m^{(1)}(\mathbf{r},t) = \frac{e}{\Delta}\mathbf{E}(\mathbf{r},t) + \mathbf{N}_{\text{therm}}(\mathbf{r},t), \\ &\eta_m(\mathbf{r})\ddot{\mathbf{Q}}_m^{(2)}(\mathbf{r},t) + \eta_m(\mathbf{r})\int_{-\infty}^{t} \gamma_0(t-t')\dot{\mathbf{Q}}_m^{(2)}(\mathbf{r},t')\,\mathrm{d}t' + \eta_m(\mathbf{r})\omega_0^2\mathbf{Q}_m^{(2)}(\mathbf{r},t) \\ &\qquad\qquad\qquad\qquad = -\eta_m(\mathbf{r})\sigma_0\mathbf{Q}_m^{(1)}(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}_m^{(1)}(\mathbf{r},t)\end{aligned} \tag{D.10}$$

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

$$\tilde{v}(\nu) = \sqrt{\frac{\gamma_0}{\pi}\eta_m(\mathbf{r})\eta_\nu(\mathbf{r})}, \tag{D.11}$$

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. (D.10) to see

$$\begin{aligned}\mathbf{Q}_m^{(1)}(\mathbf{r},\omega)\eta_m(\mathbf{r})\left[-\omega^2 - \mathrm{i}\omega\gamma_0 + \omega_0^2\right] &= \frac{e}{\Delta}\mathbf{E}(\mathbf{r},\omega) + \mathbf{N}_{\text{therm}}(\omega), \\ \mathbf{Q}_m^{(2)}(\mathbf{r},\omega)\eta_m(\mathbf{r})\left[-\omega^2 - \mathrm{i}\omega\gamma_0 + \omega_0^2\right] &= -\eta_m(\mathbf{r})\sigma_0\int_{-\infty}^{\infty}\mathbf{Q}_m^{(1)}(\mathbf{r},\omega')\cdot\mathbf{1}_3\cdot\mathbf{Q}_m^{(1)}(\mathbf{r},\omega-\omega')\,\mathrm{d}\omega'.\end{aligned} \tag{D.12}$$

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 $\mathbf{P}(\mathbf{r},\omega) = (e/\Delta)\mathbf{Q}_m(\mathbf{r},\omega)$. 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. (D.12) to see that

$$\begin{aligned}\chi^{(1)}(\mathbf{r},\omega) &= \frac{e^2}{\Delta^2\eta_m(\mathbf{r})}\frac{1}{\omega_0^2-\omega^2-\mathrm{i}\omega\gamma_0}, \\ \boldsymbol{\chi}^{(2)}(\mathbf{r};\omega',\omega-\omega') &= -\frac{3e^3\sigma_0}{\Delta^3\eta_m^2(\mathbf{r})}\frac{\mathbf{1}_3}{(\omega_0^2-\omega^2-\mathrm{i}\omega\gamma_0)(\omega_0^2-\omega'^2-\mathrm{i}\omega'\gamma_0)(\omega_0^2-[\omega-\omega']^2-\mathrm{i}[\omega-\omega']\gamma_0)}.\end{aligned} \tag{D.13}$$

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. (D.2) in Fourier space,

$$\begin{aligned}-\nabla\cdot\nabla\Phi(\mathbf{r},\omega) &= 4\pi\rho_f(\mathbf{r},\omega) - 4\pi\nabla\cdot\mathbf{P}(\mathbf{r},\omega), \\ \nabla\times\nabla\times\mathbf{A}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\mathbf{A}(\mathbf{r},\omega) &= -\frac{4\pi\omega^2}{c}\mathbf{P}^{\perp}(\mathbf{r},t) - \frac{4\pi\mathrm{i}\omega}{c}\mathbf{J}_f^{\perp}(\mathbf{r},\omega),\end{aligned} \tag{D.14}$$

then use our definitions for the susceptibilities to build the canonical wave Helmholtz equations for inhomogeneous dielectrics.

E The Power-Zienau-Woolley Lagrangian and Hamiltonian

We begin with a set of N classical particles with positions $\mathbf{x}_i(t)$, velocities $\dot{\mathbf{x}}_i(t)$, and charges q_i that are well-separated from a second set of N_0 particles with positions $\mathbf{x}_{0j}(t)$, velocities $\dot{\mathbf{x}}_{0j}(t)$, and charges q_{0j}. The first set of particles will be assumed to be sensitive to the influence of the system's electromagnetic fields, while the second set will be assumed to have their trajectories fixed, to good approximation, by forces outside of the system. The simplest Lagrangian density that defines such a system is

$$\begin{aligned}\mathscr{L}\Big[\mathbf{x}_1,\dots,\mathbf{x}_N,\dot{\mathbf{x}}_1,\dots,\dot{\mathbf{x}}_N;\mathbf{A},\dot{\mathbf{A}},\Phi,\dot{\Phi};\mathbf{A}_0,\dot{\mathbf{A}}_0,\Phi_0,\dot{\Phi}_0;\mathbf{r},t\Big] &= \sum_{i=1}^{N}\frac{1}{2}m_i\dot{\mathbf{x}}_i^2(t)\delta[\mathbf{r}-\mathbf{x}_i(t)]\\ &+\frac{1}{8\pi}\left[\mathbf{E}^2(\mathbf{r},t)-\mathbf{B}^2(\mathbf{r},t)\right]+\frac{1}{8\pi}\left[\mathbf{E}_0^2(\mathbf{r},t)-\mathbf{B}_0^2(\mathbf{r},t)\right]\\ &+\sum_{i=1}^{N}\frac{q_i}{c}\dot{\mathbf{x}}_i(t)\delta[\mathbf{r}-\mathbf{x}_i(t)]\cdot[\mathbf{A}(\mathbf{r},t)+\mathbf{A}_0(\mathbf{r},t)]-\sum_{i=1}^{N}q_i\delta[\mathbf{r}-\mathbf{x}_i(t)]\left[\Phi(\mathbf{r},t)+\Phi_0(\mathbf{r},t)\right]\\ &+\sum_{j=1}^{N_0}\frac{q_{0j}}{c}\dot{\mathbf{x}}_{0j}(t)\delta[\mathbf{r}-\mathbf{x}_{0j}(t)]\cdot\mathbf{A}_0(\mathbf{r},t)-\sum_{j=1}^{N_0}q_{0j}\Phi_0(\mathbf{r},t).\end{aligned}\tag{E.1}$$

Here, $\mathbf{A}(\mathbf{r},t)$ and $\Phi(\mathbf{r},t)$ are the vector and scalar potentials of the dynamical set of particles ($\{i\}$), respectively, with associated electric and magnetic fields $\mathbf{E}(\mathbf{r},t)=-\nabla\Phi(\mathbf{r},t)-(1/c)\dot{\mathbf{A}}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t)=\nabla\times\mathbf{A}(\mathbf{r},t)$. The analogous fields of the nondynamical particles ($\{j\}$) are $\mathbf{A}_0(\mathbf{r},t)$, $\Phi_0(\mathbf{r},t)$, $\mathbf{E}_0(\mathbf{r},t)=-\nabla\Phi_0(\mathbf{r},t)-(1/c)\dot{\mathbf{A}}_0(\mathbf{r},t)$, and $\mathbf{B}_0(\mathbf{r},t)=\nabla\times\mathbf{A}_0(\mathbf{r},t)$. Importantly, however, in contrast to the fields of the nondynamical particles, the particles' positions and momenta $\mathbf{x}_{0j}(t)$ and $\dot{\mathbf{x}}_{0j}(t)$ are not included in the Lagrangian density's list of independent variables. Therefore, $\partial\mathscr{L}/\partial\mathbf{x}_{0j}(t)=0$ and $\partial\mathscr{L}/\partial\dot{\mathbf{x}}_{0j}(t)=0$.

These last two properties are important for understanding how the equations of motion for the various quantities of the system are constructed. In particular, the Maxwell-Lorentz equations for the dynamical particles can be found via the Lagrangian $L[\dots;t]=\int\mathscr{L}[\dots;\mathbf{r},t]\,\mathrm{d}^3\mathbf{r}$ via the Euler-Lagrange equations

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial L}{\partial\dot{\mathbf{x}}_i(t)}\right\}=\frac{\partial L}{\partial\mathbf{x}_i(t)}.\tag{E.2}$$

Explicitly, these imply

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\Big\{m_i\dot{\mathbf{x}}_i(t)+\frac{q_i}{c}\left(\mathbf{A}[\mathbf{x}_i(t),t]+\mathbf{A}_0[\mathbf{x}_i(t),t]\right)\Big\} &= \frac{q_i}{c}\frac{\partial}{\partial\mathbf{x}_i(t)}\left\{\dot{\mathbf{x}}_i(t)\cdot\left(\mathbf{A}[\mathbf{x}_i(t),t]+\mathbf{A}_0[\mathbf{x}_i(t),t]\right)\right\}\\ &\quad-q_i\frac{\partial}{\partial\mathbf{x}_i(t)}\left\{\Phi[\mathbf{x}_i(t),t]+\Phi_0[\mathbf{x}_i(t),t]\right\},\end{aligned}\tag{E.3}$$

where $\partial/\partial\mathbf{z}$ is simply another way to write $\nabla_{\mathbf{z}}=\sum_{k=1}^{3}\hat{\mathbf{e}}_k\partial/\partial z_k$. Further, we have used the property $(\partial/\partial\mathbf{x})\{\mathbf{a}\cdot\mathbf{x}\}=\mathbf{a}$. Using the identities

$$\begin{aligned}\nabla\left\{\mathbf{a}\cdot\mathbf{F}(\mathbf{r})\right\}&=\mathbf{a}\times\left\{\nabla\times\mathbf{F}(\mathbf{r})\right\}+\left(\mathbf{a}\cdot\nabla\right)\left\{\mathbf{F}(\mathbf{r})\right\},\\ \frac{\mathrm{d}}{\mathrm{d}t}\left\{\mathbf{A}[\mathbf{x}_i(t),t]\right\}&=\dot{\mathbf{A}}[\mathbf{x}_i(t),t]+\left(\dot{\mathbf{x}}_i(t)\cdot\nabla_{\mathbf{x}_i(t)}\right)\mathbf{A}[\mathbf{x}_i(t),t],\end{aligned}\tag{E.4}$$

Eq. (E.3) simplifies to

$$\begin{aligned}m_i\ddot{\mathbf{x}}_i(t)+\frac{q_i}{c}\Big(\dot{\mathbf{A}}[\mathbf{x}_i(t),t]+\dot{\mathbf{A}}_0[\mathbf{x}_i(t),t]\Big) &= -\frac{q_i}{c}\left(\left[\dot{\mathbf{x}}_i(t)\cdot\nabla_{\mathbf{x}_i(t)}\right]\left\{\mathbf{A}[\mathbf{x}_i(t),t]+\mathbf{A}_0[\mathbf{x}_i(t),t]\right\}\right)\\ &+\frac{q_i}{c}\nabla_{\mathbf{x}_i(t)}\left\{\dot{\mathbf{x}}_i(t)\cdot\left(\mathbf{A}[\mathbf{x}_i(t),t]+\mathbf{A}_0[\mathbf{x}_i(t),t]\right)\right\}-q_i\nabla_{\mathbf{x}_i(t)}\left\{\Phi[\mathbf{x}_i(t),t]+\Phi_0[\mathbf{x}_i(t),t]\right\}\\ &=\frac{q_i}{c}\dot{\mathbf{x}}_i(t)\times\left(\nabla_{\mathbf{x}_i(t)}\times\left\{\mathbf{A}[\mathbf{x}_i(t),t]+\mathbf{A}_0[\mathbf{x}_i(t),t]\right\}\right)-q_i\nabla_{\mathbf{x}_i(t)}\left\{\Phi[\mathbf{x}_i(t),t]+\Phi_0[\mathbf{x}_i(t),t]\right\}\end{aligned}\tag{E.5}$$

such that, after rearranging to construct the electric and magnetic fields

$$\begin{aligned}-\nabla_{\mathbf{x}_i(t)}\left\{\Phi[\mathbf{x}_i(t),t]+\Phi_0[\mathbf{x}_i(t),t]\right\}-\frac{1}{c}\Big(\dot{\mathbf{A}}[\mathbf{x}_i(t),t]+\dot{\mathbf{A}}_0[\mathbf{x}_i(t),t]\Big)&=\mathbf{E}[\mathbf{x}_i(t),t]+\mathbf{E}_0[\mathbf{x}_i(t),t],\\ \nabla_{\mathbf{x}_i(t)}\times\left\{\mathbf{A}[\mathbf{x}_i(t),t]+\mathbf{A}_0[\mathbf{x}_i(t),t]\right\}&=\mathbf{B}[\mathbf{x}_i(t),t]+\mathbf{B}_0[\mathbf{x}_i(t),t],\end{aligned}\tag{E.6}$$

one finds the correct Maxwell-Lorentz force for the dynamical particle i:

$$m_i\ddot{\mathbf{x}}_i(t) = q_i\left(\mathbf{E}[\mathbf{x}_i(t),t] + \frac{\dot{\mathbf{x}}_i(t)}{c}\times\mathbf{B}[\mathbf{x}_i(t),t]\right). \tag{E.7}$$

In contrast, the Euler-Lagrange equations return the tautology $0 = 0$ when searching for the equation of motion of $\mathbf{x}_{0j}(t)$ such that the motion of the nondynamical particles is beyond the scope of our Lagrangian.

The motion of the nondynamical particles' fields, however, *is* described by the Lagrangian. To see this, we can use the Euler-Lagrange equations for a general field $\mathbf{F}(\mathbf{r},t)$,

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial\mathscr{L}}{\partial\dot{F}_\mu(\mathbf{r},t)}\right\} = \frac{\partial\mathscr{L}}{\partial F_\mu(\mathbf{r},t)} - \sum_{\nu=1}^{3}\frac{\partial}{\partial r_\nu}\frac{\partial\mathscr{L}}{\partial\left(\frac{\partial F_\mu(\mathbf{r},t)}{\partial r_\nu}\right)}, \tag{E.8}$$

where $\mu\in\{1,2,3\}$ is a Cartesian index. The Euler-Lagrange equations for the components of $\mathbf{A}_0(\mathbf{r},t)$ give

$$\begin{aligned}\sum_\mu\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\partial}{\partial\dot{A}_{0\mu}(\mathbf{r},t)}\left\{\frac{1}{8\pi}\left[\frac{2}{c}\nabla\Phi_0(\mathbf{r},t)\cdot\dot{\mathbf{A}}_0(\mathbf{r},t) + \frac{1}{c^2}\dot{\mathbf{A}}_0^2(\mathbf{r},t)\right]\right\}\right)\hat{\mathbf{e}}_\mu &= \sum_i\frac{q_i}{c}\dot{\mathbf{x}}_i(t)\delta[\mathbf{r}-\mathbf{x}_i(t)]\\ +\sum_j\frac{q_{0j}}{c}\dot{\mathbf{x}}_{0j}(t)\delta[\mathbf{r}-\mathbf{x}_{0j}(t)] - \sum_{\mu,\nu}\frac{\partial}{\partial r_\nu}\frac{\partial}{\partial\left(\frac{\partial A_{0\mu}(\mathbf{r},t)}{\partial r_\nu}\right)}&\left\{-\frac{1}{8\pi}\left[\nabla\times\mathbf{A}(\mathbf{r},t)\right]^2\right\}\hat{\mathbf{e}}_\mu.\end{aligned} \tag{E.9}$$

The equations simplify with the identity

$$\sum_{\mu,\nu}\frac{\partial}{\partial r_\nu}\frac{\partial}{\partial\left(\frac{\partial F_\mu(\mathbf{r},t)}{\partial r_\nu}\right)}\left\{\left[\nabla\times\mathbf{F}(\mathbf{r},t)\right]^2\right\}\hat{\mathbf{e}}_\mu = -2\nabla\times\nabla\times\mathbf{F}(\mathbf{r},t), \tag{E.10}$$

such that

$$\frac{1}{4\pi c}\nabla\dot{\Phi}_0(\mathbf{r},t)+\frac{1}{4\pi c^2}\ddot{\mathbf{A}}_0(\mathbf{r},t) = \sum_i\frac{q_i}{c}\dot{\mathbf{x}}_i(t)\delta[\mathbf{r}-\mathbf{x}_i(t)]+\sum_j\frac{q_{0j}}{c}\dot{\mathbf{x}}_{0j}(t)\delta[\mathbf{r}-\mathbf{x}_{0j}(t)]-\frac{1}{4\pi}\nabla\times\nabla\times\mathbf{A}_0(\mathbf{r},t). \tag{E.11}$$

With the definitions of the bound and free currents

$$\begin{aligned}\mathbf{J}_b(\mathbf{r},t) &= \sum_i q_i\dot{\mathbf{x}}_i(t)\delta[\mathbf{r}-\mathbf{x}_i(t)],\\ \mathbf{J}_0(\mathbf{r},t) &= \sum_j q_{0j}\dot{\mathbf{x}}_{0j}(t)\delta[\mathbf{r}-\mathbf{x}_{0j}(t)],\end{aligned} \tag{E.12}$$

one can rearrange Eq. (E.11) to find the Ampère-Maxwell equation

$$\nabla\times\mathbf{B}_0(\mathbf{r},t) = \frac{4\pi}{c}\left[\mathbf{J}_b(\mathbf{r},t)+\mathbf{J}_0(\mathbf{r},t)\right] + \frac{1}{c}\dot{\mathbf{E}}_0(\mathbf{r},t). \tag{E.13}$$

We will, in what follows, use the approximation $\mathbf{J}_0(\mathbf{r},t)\gg\mathbf{J}_b(\mathbf{r},t)$ in the driving term of the fields of the nondynamical particles. In other words, we will assume that these fields are influenced minimally by the dynamical charges of the system such that

$$\nabla\times\mathbf{B}_0(\mathbf{r},t)\approx\frac{4\pi}{c}\mathbf{J}_0(\mathbf{r},t)+\frac{1}{c}\dot{\mathbf{E}}_0(\mathbf{r},t). \tag{E.14}$$

Further, we will call thee fields of nondynamical particles “free fields” from here on to emphasize their independence from the complicated system motion.

The Euler-Lagrange equation for the free scalar potential is simpler than the equations of the free vector potential:

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\partial\mathscr{L}}{\partial\dot{\Phi}_0(\mathbf{r},t)}\right\} = \frac{\partial\mathscr{L}}{\partial\Phi_0(\mathbf{r},t)} - \sum_{\nu=1}^{3}\frac{\partial}{\partial r_\nu}\frac{\partial\mathscr{L}}{\partial\left(\frac{\partial\Phi_0(\mathbf{r},t)}{\partial r_\nu}\right)}. \tag{E.15}$$

It simplifies to

$$
\begin{aligned}
0 &= -\sum_i q_i \delta[\mathbf{r}-\mathbf{x}_i(t)] - \sum_j q_{0j}\delta[\mathbf{r}-\mathbf{x}_{0j}(t)] - \nabla\cdot\frac{\partial}{\partial(\nabla\Phi_0(\mathbf{r},t))}\left\{\frac{1}{8\pi}\left([\nabla\Phi_0(\mathbf{r},t)]^2 + \frac{2}{c}\nabla\Phi_0(\mathbf{r},t)\cdot\dot{\mathbf{A}}_0(\mathbf{r},t)\right)\right\} \\
&= -\sum_i q_i \delta[\mathbf{r}-\mathbf{x}_i(t)] - \sum_j q_{0j}\delta[\mathbf{r}-\mathbf{x}_{0j}(t)] - \nabla\cdot\left\{\frac{1}{4\pi}\nabla\Phi_0(\mathbf{r},t) + \frac{1}{4\pi c}\dot{\mathbf{A}}_0(\mathbf{r},t)\right\}
\end{aligned}
\tag{E.16}
$$

which, using the identities of the bound and free charge densities

$$
\begin{aligned}
\rho_b(\mathbf{r},t) &= \sum_i q_i\delta[\mathbf{r}-\mathbf{x}_i(t)], \\
\rho_0(\mathbf{r},t) &= \sum_j q_{0j}\delta[\mathbf{r}-\mathbf{x}_{0j}(t)],
\end{aligned}
\tag{E.17}
$$

becomes Gauss' law,

$$\nabla\cdot\mathbf{E}_0(\mathbf{r},t) = 4\pi\left[\rho_b(\mathbf{r},t)+\rho_0(\mathbf{r},t)\right]. \tag{E.18}$$

Under the same approximation as above, the bound charges are assumed to contribute negligibly to the free field's divergence such that

$$\nabla\cdot\mathbf{E}_0(\mathbf{r},t) \approx 4\pi\rho_0(\mathbf{r},t). \tag{E.19}$$

The Euler-Lagrange equations for the system potentials provides through an entirely analogous process the Ampère-Maxwell and Gauss laws for the system fields,

$$
\begin{aligned}
\nabla\times\mathbf{B}(\mathbf{r},t) &= \frac{4\pi}{c}\mathbf{J}_b(\mathbf{r},t) + \frac{1}{c}\dot{\mathbf{E}}(\mathbf{r},t), \\
\nabla\cdot\mathbf{E}(\mathbf{r},t) &= 4\pi\rho_b(\mathbf{r},t).
\end{aligned}
\tag{E.20}
$$

As opposed to the free fields, we will allow the system fields to be driven both the bound and free (dynamical and nondynamical) sources. The other two Maxwell equations, i.e. Faraday's law and the magnetic nondivergence law, are satisfied identically by our choice of potentials, so we can omit their derivations.

In systems where the total charge of all of the particles is zero, i.e. where $\sum_i q_i = 0$, it is often convenient to represent the bound current density as a polarization density $\mathbf{P}(\mathbf{r},t)$ rather than through the individual motion of each charge. In general, this can be done through

$$\mathbf{J}_b(\mathbf{r},t) = \dot{\mathbf{P}}(\mathbf{r},t) + c\nabla\times\mathbf{M}(\mathbf{r},t), \tag{E.21}$$

however in nonmagnetic systems the last term containing the magnetization density $\mathbf{M}(\mathbf{r},t)$ is often neglected. To rewrite our Lagrangian in terms of a polarization density, it is simplest to use the (slightly modified) Power-Zienau-Woolley transformation

$$L_{PZW} = L - \frac{\mathrm{d}}{\mathrm{d}t}\left\{\int \mathbf{P}(\mathbf{r},t)\cdot[\mathbf{A}(\mathbf{r},t)+\mathbf{A}_0(\mathbf{r},t)]\,\mathrm{d}^3\mathbf{r}\right\}. \tag{E.22}$$

Because adding a total time derivative to a Lagrangian leaves the action invariant and does not modify the Euler-Lagrange equations or equations of motion of the system, we can be confident that both the Maxwell-Lorentz forces and Maxwell equations implied by the Lagrangian are unchanged by the new term. Using the relationships between $\dot{\mathbf{x}}_i(t)$, $\mathbf{J}_b(\mathbf{r},t)$, and $\mathbf{P}(\mathbf{r},t)$, the PZW Lagrangian density simplifies to

$$
\begin{aligned}
\mathscr{L}_{PZW} = \sum_i \frac{1}{2}m_i\dot{\mathbf{x}}_i^2(t)\delta[\mathbf{r}-\mathbf{x}_i(t)] &+ \frac{1}{8\pi}\left[\mathbf{E}^2(\mathbf{r},t)-\mathbf{B}^2(\mathbf{r},t)\right] + \frac{1}{8\pi}\left[\mathbf{E}_0^2(\mathbf{r},t)-\mathbf{B}_0^2(\mathbf{r},t)\right] \\
&- \rho_b(\mathbf{r},t)\left[\Phi(\mathbf{r},t)+\Phi_0(\mathbf{r},t)\right] + \frac{\mathbf{J}_0(\mathbf{r},t)}{c}\cdot\mathbf{A}_0(\mathbf{r},t) - \rho_0(\mathbf{r},t)\Phi_0(\mathbf{r},t) \\
&- \frac{1}{c}\mathbf{P}(\mathbf{r},t)\cdot\left[\dot{\mathbf{A}}(\mathbf{r},t)+\dot{\mathbf{A}}_0(\mathbf{r},t)\right]
\end{aligned}
\tag{E.23}
$$

with the assumption that $\mathbf{M}(\mathbf{r}, t) = 0$.

The conjugate momenta associated with L_{PZW} are

$$\begin{aligned}
\mathbf{p}_i(t) &= \frac{\partial L_{PZW}}{\partial \dot{\mathbf{x}}(t)} = m_i \dot{\mathbf{x}}_i(t), \\
\boldsymbol{\pi}(\mathbf{r}, t) &= \frac{\partial \mathcal{L}_{PZW}}{\partial \dot{\mathbf{A}}(\mathbf{r}, t)} = -\frac{1}{4\pi c}\mathbf{E}(\mathbf{r}, t) - \frac{1}{c}\mathbf{P}(\mathbf{r}, t), \\
\boldsymbol{\pi}_0(\mathbf{r}, t) &= \frac{\partial \mathcal{L}_{PZW}}{\partial \dot{\mathbf{A}}_0(\mathbf{r}, t)} = -\frac{1}{4\pi c}\mathbf{E}_0(\mathbf{r}, t) - \frac{1}{c}\mathbf{P}(\mathbf{r}, t),
\end{aligned} \tag{E.24}$$

such that, using the identities $\dot{\mathbf{A}}(\mathbf{r}, t) = -c[\mathbf{E}(\mathbf{r}, t) + \nabla\Phi(\mathbf{r}, t)]$ and $\dot{\mathbf{A}}_0(\mathbf{r}, t) = -c[\mathbf{E}_0(\mathbf{r}, t) + \nabla\Phi_0(\mathbf{r}, t)]$, the associated Hamiltonian is

$$\begin{aligned}
H_{PZW} &= \sum_i \mathbf{p}_i(t) \cdot \dot{\mathbf{x}}_i(t) + \int \boldsymbol{\pi}(\mathbf{r}, t) \cdot \dot{\mathbf{A}}(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} + \int \boldsymbol{\pi}_0(\mathbf{r}, t) \cdot \dot{\mathbf{A}}_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} - L_{PZW} \\
&= \sum_i \frac{1}{2} m_i \dot{\mathbf{x}}_i^2(t) + \frac{1}{8\pi}\int \left[\mathbf{E}^2(\mathbf{r}, t) + \mathbf{B}^2(\mathbf{r}, t)\right] \mathrm{d}^3\mathbf{r} + \frac{1}{8\pi}\int \left[\mathbf{E}_0^2(\mathbf{r}, t) + \mathbf{B}_0^2(\mathbf{r}, t)\right] \mathrm{d}^3\mathbf{r} \\
&\quad + \int \mathbf{P}(\mathbf{r}, t) \cdot [\mathbf{E}(\mathbf{r}, t) + \mathbf{E}_0(\mathbf{r}, t) + \nabla\Phi(\mathbf{r}, t) + \nabla\Phi_0(\mathbf{r}, t)]\, \mathrm{d}^3\mathbf{r} \\
&\quad + \frac{1}{4\pi}\int \mathbf{E}(\mathbf{r}, t) \cdot \nabla\Phi(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} + \frac{1}{4\pi}\int \mathbf{E}_0(\mathbf{r}, t) \cdot \nabla\Phi_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} \\
&\quad + \int \rho_b(\mathbf{r}, t)\, [\Phi(\mathbf{r}, t) + \Phi_0(\mathbf{r}, t)]\, \mathrm{d}^3\mathbf{r} + \int \rho_0(\mathbf{r}, t)\Phi_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} \\
&\quad - \frac{1}{c}\int \mathbf{J}_0(\mathbf{r}, t) \cdot \mathbf{A}_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} + \frac{1}{c}\int \mathbf{P}(\mathbf{r}, t) \cdot \left[\dot{\mathbf{A}}(\mathbf{r}, t) + \dot{\mathbf{A}}_0(\mathbf{r}, t)\right] \mathrm{d}^3\mathbf{r}.
\end{aligned} \tag{E.25}$$

One can see through substitution of the potential form of the electric field that the fourth term of the second sum above cancels with the last term, i.e.

$$\int \mathbf{P}(\mathbf{r}, t) \cdot [\mathbf{E}(\mathbf{r}, t) + \mathbf{E}_0(\mathbf{r}, t) + \nabla\Phi(\mathbf{r}, t) + \nabla\Phi_0(\mathbf{r}, t)]\, \mathrm{d}^3\mathbf{r} = -\frac{1}{c}\int \mathbf{P}(\mathbf{r}, t) \cdot \left[\dot{\mathbf{A}}(\mathbf{r}, t) + \dot{\mathbf{A}}_0(\mathbf{r}, t)\right] \mathrm{d}^3\mathbf{r}. \tag{E.26}$$

Further, integration by parts and use of the identities $\nabla \cdot \mathbf{E}(\mathbf{r}, t) = 4\pi\rho_b(\mathbf{r}, t)$ and $\nabla \cdot \mathbf{E}_0(\mathbf{r}, t) \approx 4\pi\rho_0(\mathbf{r}, t)$ can be used to show that

$$\begin{aligned}
\frac{1}{4\pi}\int \mathbf{E}(\mathbf{r}, t) \cdot \nabla\Phi(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} &= -\int \rho_b(\mathbf{r}, t)\Phi(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r}, \\
\frac{1}{4\pi}\int \mathbf{E}_0(\mathbf{r}, t) \cdot \nabla\Phi_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} &= -\int \rho_0(\mathbf{r}, t)\Phi_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r}.
\end{aligned} \tag{E.27}$$

Finally, with $\mathbf{E}^2(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}, t) \cdot \mathbf{D}(\mathbf{r}, t) - 4\pi\mathbf{E}(\mathbf{r}, t) \cdot \mathbf{P}(\mathbf{r}, t)$, the Hamiltonian simplifies to

$$\begin{aligned}
H_{PZW} &= \sum_i \frac{1}{2} m_i \dot{\mathbf{x}}_i^2(t) - \frac{1}{2}\int \mathbf{E}(\mathbf{r}, t) \cdot \mathbf{P}(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} \\
&\quad + \frac{1}{8\pi}\int \left[\mathbf{E}(\mathbf{r}, t) \cdot \mathbf{D}(\mathbf{r}, t) + \mathbf{B}^2(\mathbf{r}, t)\right] \mathrm{d}^3\mathbf{r} + \frac{1}{8\pi}\int \left[\mathbf{E}_0^2(\mathbf{r}, t) + \mathbf{B}_0^2(\mathbf{r}, t)\right] \mathrm{d}^3\mathbf{r} \\
&\quad + \int \rho_b(\mathbf{r}, t)\Phi_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r} - \frac{1}{c}\int \mathbf{J}_0(\mathbf{r}, t) \cdot \mathbf{A}_0(\mathbf{r}, t)\, \mathrm{d}^3\mathbf{r}.
\end{aligned} \tag{E.28}$$

This form of the Hamiltonian is conceptually convenient because it includes the energy bound in the macroscopic electromagnetic fields, $(1/8\pi)\int[\mathbf{E}(\mathbf{r}, \mathbf{t}) \cdot \mathbf{D}(\mathbf{r}, t) + \mathbf{B}^2(\mathbf{r}, t)]\, \mathrm{d}^3\mathbf{r}$, of a system that includes materials. However, as the conjugate momentum of the system vector potential is proportional to the displacement field, $\boldsymbol{\pi}(\mathbf{r}, t) = -(1/4\pi c)\mathbf{D}(\mathbf{r}, t)$, we need to eliminate the extraneous factor of $\mathbf{E}(\mathbf{r}, t)$ inside the integral to bring the Hamiltonian into its canonical form.

F Miscellaneous Proofs and Derivations

F.1 Analysis of the Divergence of the Matter Displacement Field

The divergence of the matter displacement field within the sphere is given by

$$\begin{aligned}
\nabla \cdot \mathbf{Q}_m(\mathbf{r},t) &= \sum_{\beta=x,y,z} \frac{\partial}{\partial r_\beta} \left\{ \hat{\mathbf{e}}_\beta \cdot \Theta(a-r) \sum_i \mathbf{q}_{mi}(t) \Theta[\mathbf{r} \in \mathbb{V}(\mathbf{r}_i)] \right\} \\
&= \Theta(a-r) \sum_{\beta i} \hat{\mathbf{e}}_\beta \cdot \mathbf{q}_{mi}(t) \left[\delta(r_\beta - [r_{i\beta} - \Delta^{1/3}/2]) - \delta(r_\beta - [r_{i\beta} + \Delta^{1/3}/2]) \right] \\
&\times \prod_{\gamma \neq \beta} \Theta(r_\gamma - [r_{i\gamma} - \Delta^{1/3}/2]) \Theta([r_{i\gamma} + \Delta^{1/3}/2] - r_\gamma) - \hat{\mathbf{r}} \delta(r-a) \cdot \sum_{i=1}^{N_m} \mathbf{q}_{mi}(t) \Theta[\mathbf{r} \in \mathbb{V}(\mathbf{r}_i)].
\end{aligned} \tag{F.1}$$

The first term on the right-hand side highlights that unit cells within the sphere's bulk that have relative charge displacements $\mathbf{q}_{mi}(t)$ unequal to those of their neighbors will give rise to coordinate field divergences. To see that, one can analyze two neighboring terms j and k in the sums over i above. These terms will have opposite signs attached to the Dirac deltas $\delta(r_\beta - [r_{j\beta} \mp \Delta^{1/3}/2])$ and $\delta(r_\beta - [r_{k\beta} \pm \Delta^{1/3}/2])$ that are otherwise equal along the boundary shared by the corresponding volumes $\mathbb{V}(\mathbf{r}_j)$ and $\mathbb{V}(\mathbf{r}_k)$. Therefore, if they have identical displacement projections $\hat{\mathbf{e}}_\beta \cdot \mathbf{q}_{mj}(t) = \hat{\mathbf{e}}_\beta \cdot \mathbf{q}_{mk}(t)$, they will cancel, such that the divergence of the matter field is only nonzero where the "head-to-tail" cancellation of neighboring displacement vectors is broken.

This logic seems to suggest that only a uniform matter displacement field will be free of divergences. Since the polarization field inside the sphere is not uniform in general, such a limitation would present real difficulties. However, one can see that smooth variations in $\mathbf{Q}_m(\mathbf{r},t)$ will not produce divergences and that our model for $\mathbf{Q}_m(\mathbf{r},t)$ is more flexible. To see this, we can recognize that, as the side-length $\Delta^{1/3}$ of each unit-cell goes to zero, infinitesimal differences between the displacement vectors of each cell proportional to this length will also go to zero. For example, if $\hat{\mathbf{e}}_\beta \cdot \mathbf{q}_{mj}(t) = \hat{\mathbf{e}}_\beta \cdot \mathbf{q}_{mk}(t) + a\Delta^{1/3}$ with a some constant, the cancellation condition described above will still be satisfied as $\Delta \to 0$ and $\mathbf{Q}_m(\mathbf{r},t)$ will remain divergence-free.

The second term on the right-hand side describes a similar broken-cancellation effect that occurs at the particle's surface. In particular, projections of the displacement vectors $\mathbf{q}_{mi}(t)$ along a given axis represent the infinitesimal flow of charge along that axis. When this projection occurs across the surface, as is represented by $\hat{\mathbf{r}} \cdot \mathbf{q}_{mi}(t)\delta(r-a)$, this flow produces infinitesimally thin layers of unbalanced positive or negative charge along the particle's surface. Therefore, as divergences of the matter displacement field within the sphere's bulk represent regions of nonzero bulk bound charge density, the surface-divergences represent regions of nonzero surface bound charge density.

G Miscellaneous Work In-Progress

Generally, it is easiest to construct this picture in a Hamiltonian formalism, especially if we are to quantize it later. We must therefore begin by identifying the momenta and momentum densities associated with our particle coordinates and fields, respectively. Following the usual procedure, we find

$$\begin{aligned}
\mathbf{p}_{fi}(t) &= \sum_k \frac{\partial L}{\partial \left(\dot{\mathbf{x}}_{fi}(t) \cdot \hat{\mathbf{e}}_k \right)} \hat{\mathbf{e}}_k = \mu_{fi} \dot{\mathbf{x}}_{fi}(t) + \frac{e_i}{c} \mathbf{A}[\mathbf{x}_{fi}(t), t], \\
\mathbf{P}_m(\mathbf{r},t) &= \sum_k \frac{\partial \mathcal{L}}{\partial \left(\dot{\mathbf{Q}}_m(\mathbf{r},t) \cdot \hat{\mathbf{e}}_k \right)} \hat{\mathbf{e}}_k = \eta_m(\mathbf{r}) \dot{\mathbf{Q}}_m(\mathbf{r},t) + \frac{e}{c\Delta} \mathbf{A}(\mathbf{r},t), \\
\tilde{\mathbf{P}}_\nu(\mathbf{r},t) &= \sum_k \frac{\partial \mathcal{L}}{\partial \left(\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t) \cdot \hat{\mathbf{e}}_k \right)} \hat{\mathbf{e}}_k = \eta_\nu(\mathbf{r}) \dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t) - \tilde{v}(\nu) \mathbf{Q}_m(\mathbf{r},t), \\
\mathbf{\Pi}(\mathbf{r},t) &= \sum_k \frac{\partial \mathcal{L}}{\partial \left(\dot{\mathbf{A}}(\mathbf{r},t) \cdot \hat{\mathbf{e}}_k \right)} \hat{\mathbf{e}}_k = \frac{1}{4\pi c} \left[\frac{1}{c} \dot{\mathbf{A}}(\mathbf{r},t) + \nabla \Phi(\mathbf{r},t) \right],
\end{aligned} \tag{G.1}$$

such that

$$
\begin{aligned}
H &= \sum_i \mathbf{p}_{fi}(t)\cdot\dot{\mathbf{x}}_{fi}(t) + \int \left[\mathbf{P}_m(\mathbf{r},t)\cdot\dot{\mathbf{Q}}_m(\mathbf{r},t) + \int_0^\infty \tilde{\mathbf{P}}_\nu(\mathbf{r},t)\cdot\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\,\mathrm{d}\nu + \mathbf{\Pi}(\mathbf{r},t)\cdot\dot{\mathbf{A}}(\mathbf{r},t)\right]\mathrm{d}^3\mathbf{r} - L \\
&= \sum_i \left(\mu_{fi}\dot{\mathbf{x}}_{fi}^2(t) + \frac{e_i}{c}\dot{\mathbf{x}}_{fi}(t)\cdot\mathbf{A}\left[\mathbf{x}_{fi}(t),t\right]\right) + \int\left[\eta_m(\mathbf{r})\dot{\mathbf{Q}}_m^2(\mathbf{r},t) + \frac{e}{c\Delta}\dot{\mathbf{Q}}_m(\mathbf{r},t)\cdot\mathbf{A}(\mathbf{r},t)\right]\mathrm{d}^3\mathbf{r} \\
&+ \int\int_0^\infty \left[\eta_\nu(\mathbf{r})\dot{\tilde{\mathbf{Q}}}_\nu^2(\mathbf{r},t) - \tilde{v}(\nu)\mathbf{Q}_m(\mathbf{r},t)\cdot\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\right]\mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} + \frac{1}{4\pi c}\int\left[\frac{1}{c}\dot{\mathbf{A}}^2(\mathbf{r},t) + \nabla\Phi(\mathbf{r},t)\cdot\dot{\mathbf{A}}(\mathbf{r},t)\right]\,\mathrm{d}^3\mathbf{r} \\
&- \sum_i \frac{1}{2}\mu_{fi}\dot{\mathbf{x}}_{fi}^2(t) + \int\frac{1}{2}\eta_m(\mathbf{r})\left(-\dot{\mathbf{Q}}_m^2(\mathbf{r},t) + \omega_0^2\mathbf{Q}_m^2(\mathbf{r},t) + \frac{2}{3}\sigma_0\mathbf{Q}_m(\mathbf{r},t)\cdot\left[\mathbf{Q}_m(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}_m(\mathbf{r},t)\right]\right)\mathrm{d}^3\mathbf{r} \\
&+ \int\int_0^\infty\left(-\frac{1}{2}\eta_\nu(\mathbf{r})\dot{\tilde{\mathbf{Q}}}_\nu^2(\mathbf{r},t) + \frac{1}{2}\eta_\nu(\mathbf{r})\nu^2\tilde{\mathbf{Q}}_\nu^2(\mathbf{r},t)\right)\mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} + \int\int_0^\infty \tilde{v}(\nu)\mathbf{Q}_m(\mathbf{r},t)\cdot\dot{\tilde{\mathbf{Q}}}_\nu(\mathbf{r},t)\,\mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} \\
&+ \int\left(-\frac{e}{\Delta}\nabla\cdot\mathbf{Q}_m(\mathbf{r},t) + \sum_i e_i\delta[\mathbf{x}_{fi}(t)-\mathbf{r}]\right)\Phi(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} \\
&- \int\left(\frac{e}{c\Delta}\dot{\mathbf{Q}}_m(\mathbf{r},t) + \sum_i\frac{e_i}{c}\dot{\mathbf{x}}_{fi}(t)\delta[\mathbf{x}_{fi}(t)-\mathbf{r}]\right)\cdot\mathbf{A}(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} + \sum_{\alpha,\beta=m,b,r}\left[u_{\alpha\beta}^\infty(\mathbf{r}) + u_{\beta\alpha}^\infty(\mathbf{r})\right] \\
&+ \sum_{\alpha=m,b,r,f} u_\alpha^{\mathrm{self}} - \frac{1}{8\pi}\int\left([\nabla\Phi(\mathbf{r},t)]^2 + \frac{1}{c^2}\dot{\mathbf{A}}^2(\mathbf{r},t) - [\nabla\times\mathbf{A}(\mathbf{r},t)]^2 + \frac{2}{c}\nabla\Phi(\mathbf{r},t)\cdot\dot{\mathbf{A}}(\mathbf{r},t)\right)\mathrm{d}^3\mathbf{r}.
\end{aligned} \tag{G.2}
$$

After eliminating any terms that include integration of Helmholtz-orthogonal fields and cancelling any reverse duplicates, the Hamiltonian simplifies to

$$
\begin{aligned}
H &= \sum_i \frac{1}{2}\mu_{fi}\dot{\mathbf{x}}_{fi}^2(t) + \int\frac{1}{2}\eta_m(\mathbf{r})\left[\dot{\mathbf{Q}}_m^2(\mathbf{r},t) + \omega_0^2\mathbf{Q}_m^2(\mathbf{r},t)\right]\mathrm{d}^3\mathbf{r} \\
&+ \int\frac{1}{3}\eta_m(\mathbf{r})\sigma_0\mathbf{Q}_m(\mathbf{r},t)\cdot\left[\mathbf{Q}_m(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}_m(\mathbf{r},t)\right]\mathrm{d}^3\mathbf{r} + \int\int_0^\infty\frac{1}{2}\eta_\nu(\mathbf{r})\left[\dot{\tilde{\mathbf{Q}}}_\nu^2(\mathbf{r},t) + \nu^2\tilde{\mathbf{Q}}_\nu^2(\mathbf{r},t)\right]\mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} \\
&+ \frac{1}{8\pi}\int\left(\frac{1}{c^2}\dot{\mathbf{A}}^2(\mathbf{r},t) + [\nabla\times\mathbf{A}(\mathbf{r},t)]^2\right)\mathrm{d}^3\mathbf{r} - \frac{1}{8\pi}\int[\nabla\Phi(\mathbf{r},t)]^2\,\mathrm{d}^3\mathbf{r} - \int\frac{e}{\Delta}\nabla\cdot\mathbf{Q}_m(\mathbf{r},t)\Phi(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} \\
&+ \sum_i e_i\Phi[\mathbf{x}_{fi}(t),t] + \sum_{\alpha,\beta=m,b,r}\left[u_{\alpha\beta}^\infty(\mathbf{r}) + u_{\beta\alpha}^\infty(\mathbf{r})\right] + \sum_{\alpha=m,b,r,f} u_\alpha^{\mathrm{self}}.
\end{aligned} \tag{G.3}
$$

Eliminating the velocities and velocity densities in favor of momenta and momentum densities, one finds

$$
\begin{aligned}
H = & \sum_i \frac{1}{2}\mu_{fi}\left(\frac{\mathbf{p}_{fi}(t)}{\mu_{fi}} - \frac{e_i}{\mu_{fi}c}\mathbf{A}\left[\mathbf{x}_{fi}(t),t\right]\right)^2 \\
& + \int \frac{1}{2}\eta_m(\mathbf{r})\left[\left(\frac{\mathbf{P}_m(\mathbf{r},t)}{\eta_m(\mathbf{r})} - \frac{e\mathbf{A}(\mathbf{r},t)}{\eta_m(\mathbf{r})c\Delta}\right)^2 + \omega_0^2\mathbf{Q}_m^2(\mathbf{r},t)\right] \mathrm{d}^3\mathbf{r} \\
& + \int \frac{1}{3}\eta_m(\mathbf{r})\sigma_0\mathbf{Q}_m(\mathbf{r},t)\cdot\left[\mathbf{Q}_m(\mathbf{r},t)\cdot\mathbf{1}_3\cdot\mathbf{Q}_m(\mathbf{r},t)\right] \mathrm{d}^3\mathbf{r} \\
& + \int\int_0^\infty \frac{1}{2}\eta_\nu(\mathbf{r})\left[\left(\frac{\tilde{\mathbf{P}}_\nu(\mathbf{r},t)}{\eta_\nu(\mathbf{r})} + \frac{\tilde{v}(\nu)}{\eta_\nu(\mathbf{r})}\mathbf{Q}_m(\mathbf{r},t)\right)^2 + \nu^2\tilde{\mathbf{Q}}_\nu^2(\mathbf{r},t)\right] \mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} \\
& + \frac{1}{8\pi}\int\left(\frac{1}{c^2}\left[4\pi c^2\mathbf{\Pi}(\mathbf{r},t) - c\nabla\Phi(\mathbf{r},t)\right]^2 + \left[\nabla\times\mathbf{A}(\mathbf{r},t)\right]^2\right)\mathrm{d}^3\mathbf{r} - \frac{1}{8\pi}\int\left[\nabla\Phi(\mathbf{r},t)\right]^2\mathrm{d}^3\mathbf{r} \\
& - \int\frac{e}{\Delta}\nabla\cdot\mathbf{Q}_m(\mathbf{r},t)\Phi(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} + \sum_i e_i\Phi[\mathbf{x}_{fi}(t),t] + \sum_{\alpha,\beta=m,b,r}\left[u_{\alpha\beta}^\infty(\mathbf{r}) + u_{\beta\alpha}^\infty(\mathbf{r})\right] + \sum_{\alpha=m,b,r,f} u_\alpha^{\mathrm{self}}.
\end{aligned}
\tag{G.4}
$$

From here, we can simplify by separating the scalar potentials of the free and bound charges. Since $\partial\mathcal{L}/\partial\dot{\Phi}(\mathbf{r},t) = 0$, the scalar potentials are not dynamical variables and do not carry their own degrees of freedom. Therefore, the scalar potentials can be considered to instantaneously track the motion of the free and bound charges such that

$$\Phi(\mathbf{r},t) = \Phi_f(\mathbf{r},t) + \Phi_m(\mathbf{r},t) \tag{G.5}$$

where $\Phi_\alpha(\mathbf{r},t) = \int \rho_\alpha(\mathbf{r}',t)/|\mathbf{r}-\mathbf{r}'|\,\mathrm{d}^3\mathbf{r}'$. The total Hamiltonian can then be expanded as

$$H = H_f + H_m + H_r + H_{\mathrm{EM}} + H_{\mathrm{int}} + H_{\mathrm{drive}} + H_{\mathrm{bind}} + H_{\mathrm{self}} + H_{\mathrm{NL}}. \tag{G.6}$$

The first term is the Hamiltonian of the free particles,

$$H_f = \sum_i \frac{\mathbf{p}_{fi}^2(t)}{2\mu_{fi}} + \sum_i e_i\Phi_f[\mathbf{x}_{fi}(t),t]. \tag{G.7}$$

The second is the Hamiltonian of the matter resonance,

$$
\begin{aligned}
H_m = & \int\left[\frac{\mathbf{P}_m^2(\mathbf{r},t)}{2\eta_m(\mathbf{r})} + \frac{1}{2}\eta_m(\mathbf{r})\omega_0^2\mathbf{Q}_m^2(\mathbf{r},t)\right]\mathrm{d}^3\mathbf{r} + \int\int_0^\infty \frac{\tilde{v}^2(\nu)}{2\eta_\nu(\mathbf{r})}\mathbf{Q}_m^2(\mathbf{r},t)\,\mathrm{d}\nu\,\mathrm{d}^3\mathbf{r} \\
& - \int\frac{e}{\Delta}\nabla\cdot\mathbf{Q}_m(\mathbf{r},t)\Phi_m(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r},
\end{aligned}
\tag{G.8}
$$

the third is the Hamiltonian of the bath,

$$H_r = \int\int_0^\infty\left[\frac{\tilde{\mathbf{P}}_\nu^2(\mathbf{r},t)}{2\eta_\nu(\mathbf{r})} + \frac{1}{2}\eta_\nu(\mathbf{r})\nu^2\tilde{\mathbf{Q}}_\nu^2(\mathbf{r},t)\right]\mathrm{d}\nu\,\mathrm{d}^3\mathbf{r}, \tag{G.9}$$

and the fourth is the Hamiltonian of the transverse electromagnetic field,

$$
\begin{aligned}
H_{\mathrm{EM}} = & \int\left(2\pi c^2\mathbf{\Pi}^2(\mathbf{r},t) + \frac{1}{8\pi}\left[\nabla\times\mathbf{A}(\mathbf{r},t)\right]^2\right)\mathrm{d}^3\mathbf{r} + \int\frac{e^2}{2\eta_m(\mathbf{r})c^2\Delta^2}\mathbf{A}^2(\mathbf{r},t)\,\mathrm{d}^3\mathbf{r} \\
& + \sum_i\frac{e_i^2}{2\mu_{fi}c^2}\mathbf{A}^2[\mathbf{x}_{fi}(t),t].
\end{aligned}
\tag{G.10}
$$

The fifth is the interaction Hamiltonian that couples the matter, bath, and EM field together,

$$\begin{aligned} H_{\text{int}} = &- \int \frac{e}{\eta_m(\mathbf{r}) c \Delta} \mathbf{P}_m(\mathbf{r},t) \cdot \mathbf{A}(\mathbf{r},t)\ \mathrm{d}^3\mathbf{r} + \int \int_0^\infty \frac{\tilde{v}(\nu)}{\eta_\nu(\mathbf{r})} \tilde{\mathbf{P}}_\nu(\mathbf{r},t) \cdot \mathbf{Q}_m(\mathbf{r},t)\ \mathrm{d}\nu\, \mathrm{d}^3\mathbf{r} \\ &- c \int \mathbf{\Pi}(\mathbf{r},t) \cdot \nabla \Phi_m(\mathbf{r},t)\ \mathrm{d}^3\mathbf{r}, \end{aligned} \tag{G.11}$$

and the sixth is the driving Hamiltonian that details the interactions of the free charges with the other resonances of the system,

$$\begin{aligned} H_{\text{drive}} = &- \sum_i \frac{e_i}{\mu_{fi} c} \mathbf{p}_{fi}(t) \cdot \mathbf{A}[\mathbf{x}_{fi}(t), t] - c \int \mathbf{\Pi}(\mathbf{r},t) \cdot \nabla \Phi_f(\mathbf{r},t)\ \mathrm{d}^3\mathbf{r} \\ &- \int \frac{e}{\Delta} \nabla \cdot \mathbf{Q}_m(\mathbf{r},t) \Phi_f(\mathbf{r},t)\ \mathrm{d}^3\mathbf{r} + \sum_i e_i \Phi_m[\mathbf{x}_{fi}(t), t]. \end{aligned} \tag{G.12}$$

The seventh and eighth terms are the binding and self-energy Hamiltonians

$$\begin{aligned} H_{\text{bind}} &= \sum_{\alpha,\beta=m,b,r} \left[u^\infty_{\alpha\beta}(\mathbf{r}) + u^\infty_{\beta\alpha}(\mathbf{r}) \right], \\ H_{\text{self}} &= \sum_{\alpha=m,b,r,f} u^{\text{self}}_\alpha, \end{aligned} \tag{G.13}$$

while the ninth is the nonlinear term in the potential energy of the matter resonance,

$$H_{\text{NL}} = \int \frac{1}{3} \eta_m(\mathbf{r}) \sigma_0 \mathbf{Q}_m(\mathbf{r},t) \cdot [\mathbf{Q}_m(\mathbf{r},t) \cdot \mathbf{1}_3 \cdot \mathbf{Q}_m(\mathbf{r},t)]\ \mathrm{d}^3\mathbf{r}. \tag{G.14}$$

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