A quantum theory of dispersion for an inhomogeneous solid is obtained, from a starting point of multipolar coupled atoms interacting with an electromagnetic field. The dispersion relations obtained are equivalent to the standard classical Sellmeir equations obtained from the Drude-Lorentz model. In the homogeneous (plane-wave) case, we obtain the detailed quantum mode structure of the coupled polariton fields, and show that the mode expansion in all branches of the dispersion relation is completely defined by the refractive index and the group-velocity for the polaritons. We demonstrate a straightforward procedure for exactly diagonalizing the Hamiltonian in one, two or three-dimensional environments, even in the presence of longitudinal phonon-exciton dispersion, and an arbitrary number of resonant transitions with different frequencies. This is essential, since it is necessary to include at least one phonon (I.R.) and one exciton (U.V.) mode, in order to accurately represent dispersion in transparent solid media. Our method of diagonalization does not require an explicit solution of the dispersion relation, but relies instead on the analytic properties of Cauchy contour integrals over all possible mode frequencies. When there is longitudinal phonon dispersion, the relevant group-velocity term is modified so that it only includes the purely electromagnetic part of the group velocity.

I. INTRODUCTION

Recent developments in quantum optics have led to the realization that it is essential to include dispersion in the quantum theory of a linear or nonlinear dielectric, as in a fiber waveguide. At the simplest level, it is clear that all dielectric solids have dispersion and absorption. There are an increasing number of experiments that allow direct access to the quantum nature of propagating radiation fields, ranging from quantum soliton experiments in optical fibers to QND measurements; as well as quantum dynamics experiments directed at reducing photon number noise levels for broad-band communications, ultra-precise measurements and other device applications [1]- [6]. Significantly, these experiments - as well as more recent theoretical proposals - have the character of fundamental tests of the quantum theory of interacting fields [7]- [10], and of quantum measurement theory. These are complementary to older accelerator-based tests, taking place in low-energy regimes where a considerable precision of control is available on the dimensionality of the interacting quantum fields. Measurements that can be used range from the usual particle-counting measurements, to interference based techniques that allow an unprecedented level of information about quantum phases. Current technologies even allow measurements of electromagnetic properties of Bose condensates.

Because the effects of propagation always involve more than one spatial mode, it is essential to include dispersion in any physical model of a linear waveguide in one or more spatial dimensions. Treating dispersion can present problems, because it arises from the fact that the response of the medium to the field is not instantaneous. The polarization at a given time depends not only on the field at that time but on the values of the field at previous times as well. This nonlocal behavior makes standard macroscopic quantization, in which the medium is represented by its susceptibilities, problematic. Even without explicit dispersion, some early treatments even resulted in Hamiltonians that did not generate the Maxwell equations at all. This problem was analyzed by Hillery and Mlodinow [11], who showed that the problem was caused by the use of an incorrect canonical description.

However, a previous macroscopic model of a nonlinear, dispersive medium resulted in a quantum theory that has a consistent Lagrangian [12]. This was accomplished by breaking the field up into narrow frequency-band components and quantizing these separately. The frequency dependent linear susceptibilities expanded up to second order in each frequency band and this results in a local Lagrangian in each band. The present paper is intended to treat the microscopic basis of the linear dispersion more precisely. Our model is the quantized version of the classical Drude-Lorentz [13] single-electron oscillator model, in a linearized continuum limit. Similar continuum models have been treated previously, generally in the minimal coupling gauge. The present approach uses the multipolar gauge, which eliminates complications due to Coulomb interactions and contact (A^2) terms. While these effects can be included in minimal coupling theories, they result in considerable complexity. The present approach includes all these effects by virtue of the transformation to a multipolar interaction, in which the coupling is to the polarization rather than...
to the electronic momentum. This has the advantage that inhomogeneous media or higher dimensions can be treated easily.

Some earlier dispersive models of a similar type are known, starting from the phonon theory of Born and Huang \[1\] and the exciton theory of Hopfield \[2\], which used minimal coupling. Hopfield considered the electromagnetic field coupled to a polarization field which has a single resonant frequency. He diagonalized the resulting Hamiltonian and found that the resulting elementary excitations, polaritons, have a dispersion relation consisting of two branches separated by a forbidden frequency band. More recent work has revisited the Hopfield model, though not always with all the terms included in Hopfield’s treatment \[3,4\]. Other treatments have added losses to the model by coupling the polarization field to loss reservoirs \[5,6\], still with just one resonant transition. This has allowed the examination of the Kramers-Kronig relations in a fully quantized model. Finally, nonlinear generalizations of of Hopfield’s model have also been studied \[7\]. In these the linear oscillators of Hopfield’s medium have been replaced by two-level atoms and the Holstein-Primakoff representation is used to develop a systematic expansion of the polarization of the medium which includes nonlinear effects.

These and other studies have elucidated the fundamental cause of the two main types of dispersion. The first, excitonic type is due to electronic transitions, and is typically resonant in the ultra-violet. These are due to relatively tightly-bound electrons that are localized to atomic sites in insulators, and somewhat delocalized in semiconductors. The resulting classical dispersion is rather well described by the harmonically-bound Drude-Lorenz model. The next type is due to phonon transitions, mostly in the infra-red. These excitations are less strongly localized, and have their own dispersion relation in the Born-Huang model. The dispersion in the transparent region between these absorption bands is caused by the off-resonant, virtual transitions of these two main forms of excitation. In effect, a photon propagates in a dielectric as a dressed particle, due to the virtual transitions - and resulting polarization - that is induced in the solid. To treat this completely, it is essential to include multiple exciton and phonon resonances, in a full three-dimensional model.

The present paper examines this problem using an approach in which the coupling to the dielectric is included through a multipolar term \[8,9\]. This has the well-known advantages that the use of atomic sum-rules is not required, since the atomic transition probability for off-resonant (high-frequency) transitions is suppressed in this gauge \[2,10\]. The new feature presented here is the inclusion of any number of atomic resonances - thus allowing us to recover the Sellmeir \[13\] dispersion equation, which is known to provide an excellent quantitative description of real dispersive, transparent media. The use of multiple resonances is essential to the correct description of the transparent region with low group velocity dispersion, that occurs between the absorption bands in most known cases. In addition, there is a very straightforward procedure for diagonalizing the Hamiltonian - even in the presence of an arbitrary number of resonant transitions with different resonant frequencies.

In the homogeneous (plane-wave) case, we find the expansion of the fields in terms of the quantized modes has an exceptionally simple form, in which only an electromagnetic group-velocity correction has to be included relative to the usual non-dispersive theory. When there is no longitudinal phonon/exciton dispersion, this correction term is identical to that found in previous phenomenological \[1,2,11\] and single-resonance microscopic models \[4,5,7,9,10\]. As an example, Blow, et. al. \[12\] based their expansion of the fields on previous work (due to Loudon \[24\]) in which it was argued, from energy transport considerations, that the group velocity should appear in the expansion of the phonon field in terms of quantized modes, when phonon dispersion is present. Blow, et. al. took this result for phonons and used it for the case of an electromagnetic field in a dispersive medium.

Our work here provides a justification for this quantization procedure from a more fundamental point of view - a multiple-resonance microscopic model. It is remarkable that the simple inclusion of a group velocity factor is sufficient to exactly diagonalize this complex interaction Hamiltonian. The mathematical technique required to prove the results involves the use of Cauchy’s theorem to carry out the required sums over the different branches of the dispersion relation. This is a significant improvement over previous approaches, since in general there is no algebraic solution - even in principle - for the polynomial expressions whose roots give the dispersion relations. In addition, we find that when there is phonon or exciton dispersion \[25\], which is a realistic feature of many dispersive media, this procedure is modified in a straightforward way. Instead of the total group velocity, only the relevant electromagnetic component is included in the expansion coefficients, suggesting that the diagonalization is intimately related to the power transport processes in the solid. The results apply to one, two or three-dimensional environments, although only isotropic dielectrics are included here, for simplicity in the derivations.

II. ONE-DIMENSIONAL MODEL

We start by considering a straight-forward one-dimensional model, without the complicating features of transverse mode structures and longitudinal phonon/exciton dispersion. This simple case illustrates the essential analytic features
of our model. It will be generalized to more realistic, higher-dimensional dielectric structures, in later sections.

A. One-dimensional Lagrangian

We start by considering a set of dipole-coupled electronic Lagrangians for oscillators with mass \( m_\nu \), charge \( q_\nu \), and \( N \) (renormalized) resonant discrete frequencies \( \Omega_{\nu_1}, \ldots, \Omega_{\nu_N} \). These transitions may correspond physically to different types of atom, to distinct transitions within each atom, or more generally to some many-body resonance. Thus,

\[
\mathcal{L}_c = \sum_\nu \left[ \frac{m_\nu}{2} \left( \dot{r}_\nu^2 - \Omega_{\nu}^2 r_\nu^2 \right) + \frac{1}{\varepsilon_0} q_\nu r_\nu D(\pi_\nu) \right].
\]  

(2.1)

Here \( r_\nu \) is the displacement of a charged particle (typically, an electron) in a multi-polar Drude-Lorentz model, from the corresponding center of charge (typically, nuclear) position \( \pi_\nu \). Generally, these are all distinct atomic resonances. For simplicity, the self-energy terms proportional to \( B^2 \), are incorporated into the definition of the resonant frequency \( \Omega_{\nu} \). The coupling in multipolar gauge is to the displacement field \( D(\pi_\nu) \) at the central position. All Coulomb terms in this gauge are carried by the photon-exchange process \([21]\).

Next, in order to develop a simple electromagnetic Lagrangian in the multipolar gauge, we introduce a gauge field - the dual potential \( \Lambda \), so that \( D = \partial \Lambda / \partial x \) and \( B = \mu \partial \Lambda / \partial t \). This simply corresponds to a canonical transformation to the multipolar gauge \([20,21]\) of the more usual minimal coupling theory - and is simplified here by the assumption that there are no free charges. The discrete atomic positions are replaced by a continuum polarization field, together with the appropriate Lorentz-shifts in the resonant frequencies \( \nu \), due to local field corrections. This simplification is used here because it is not really necessary for us to consider the details of local-field corrections at this stage. We will show that this approach is able to generate the identical (low-frequency) Sellmeir dispersion equation that is obtained in the usual Drude-Lorentz theory, which does include local-field corrections. Of course, the approximations involved are only valid for carrier wavelengths much greater than the inter-atomic spacing.

The corresponding Lagrangian density that generates the correct electromagnetic energy and Maxwell’s equations in one dimension is, for a one-dimensional wave-guide with effective cross-section \( A \):

\[
\mathcal{L} / A = \frac{1}{2} \dot{\Lambda}^2 (x) + \frac{1}{\varepsilon_0} \left[ P(x) \partial_x \Lambda(x) - \frac{1}{2} (\partial_x \Lambda(x))^2 \right] + \sum_{\nu=1}^{N} \left[ p_\nu^2 (x) - \Omega_{\nu}^2 r_\nu^2 (x) \right] / (2\varepsilon_0 q_\nu (x)).
\]  

(2.2)

Here \( \rho_\nu (x) \) is the density of the oscillators with resonant frequency \( \nu \), and \( r_\nu (x) \) is regarded as a continuum field, with polarization \( P(x) = \sum_\nu \rho_\nu (x) = \sum_\nu q_\nu \rho_\nu (x) r_\nu (x) \), in order to allow us to use a continuum approximation in later calculations.

The coupling between the field and the polarization is entirely included in one term \( g_\nu (x) \), which effectively combines all the relevant information about charge, density, and dipole-moment:

\[
g_\nu (x) = q_\nu^2 \rho_\nu (x) / (m_\nu \varepsilon_0).
\]  

(2.3)

Noting that \( D(x) = \varepsilon_0 E(x) + P(x) \), the electric field is given by \( E(x) = [D(x) - P(x)] / \varepsilon_0 \), and the resulting Maxwell-Bloch equations have the expected form of:

\[
\partial_t^2 \Lambda - c^2 \partial_x^2 \Lambda = -c^2 \partial_x P(x)
\]

\[
\partial_t^2 \rho_\nu + \Omega_{\nu}^2 r_\nu = g_\nu \partial_x \Lambda.
\]  

(2.4)

In order to quantize the system, we simply introduce commutators for the canonical momenta and position, where:

\[
\Pi(x) = \mu \dot{\Lambda}(x) = B(x),
\]

\[
\pi_\nu (x) = \dot{p}_\nu (x) / (\varepsilon_0 q_\nu (x)).
\]  

(2.5)

The quantization rules are then the usual ones, obtained on replacing Poisson brackets with commutators, except with the atomic operators treated as a continuum field. All position-type operators of the same type of variable must commute amongst themselves, as do all momentum-type operators. We list the commutators involving cross-terms between the position and momentum operators, and between the different types of variable, for reference in the next
sections. In order that the commutators will have a familiar appearance, they are written in terms of the electric displacement and magnetic fields, rather than the canonical variables and their momenta. Since the electric field only differs from the displacement field by the polarization terms (which commute with field operators at equal times) there is no essential difference between the electromagnetic commutators written with the displacement field or the electric field. The fact that the electric displacement is the derivative of a potential is, of course, the origin of the differentiated delta function in the commutator between the electric and magnetic fields, which is also found in minimal coupling theory:

\[
\begin{align*}
(\text{I}) & \quad \left[ \hat{D}(x), \hat{B}(x') \right] = i\hbar \delta'(x-x')/(A), \\
(\text{II}) & \quad \left[ \hat{p}_\nu(x), \hat{\pi}_\nu(x') \right] = i\hbar \delta_{\nu\nu'} \delta(x-x')/A, \\
(\text{III}) & \quad \left[ \hat{D}(x), \hat{p}_\nu(x') \right] = 0, \\
(\text{IV}) & \quad \left[ \hat{B}(x), \hat{\pi}_\nu(x') \right] = 0, \\
(\text{V}) & \quad \left[ \hat{D}(x), \hat{\pi}_\nu(x') \right] = 0, \\
(\text{VI}) & \quad \left[ \hat{B}(x), \hat{p}_\nu(x') \right] = 0, \\
\end{align*}
\]

(2.6)

It is important to notice here that the commutators are essentially identical to those for the corresponding free fields and oscillators. This is a consequence of the fact that the couplings do not involve time-derivatives. It also demonstrates that the present theory is canonically equivalent to other techniques involving minimal coupling. One apparent difference is in the commutator between the displacement field and the momentum, which replaces a commutator between the electric field and a gauge-dependent canonical momentum in minimal-coupling theory. Since the electric field and displacement field differ by a term that doesn’t commute with the canonical momentum, it might seem that this introduces a difference. The explanation is due to the well-known fact that the canonical momentum in this case is just the usual mechanical momentum, and therefore differs from the minimal coupling momentum - which includes a term proportional to the vector potential A. These two effects cancel each other, so the overall commutators are unchanged.

The final Hamiltonian density, equal to the linear energy density of the coupled system, has a rather straightforward expression in which there are no explicit Coulomb interaction terms. This is a typical property of multipolar interaction Hamiltonians. Effectively, the Coulomb couplings are included partly in the oscillator self-frequency terms (i.e., in \( \nu_t \)), and partly in the photon-exchange dynamics that are implicit in the Hamiltonian. The result is that:

\[
\mathcal{H}/A = \frac{1}{2\varepsilon_0} \hat{D}^2(x) + \frac{1}{2\mu} \hat{B}^2(x) - \frac{1}{\varepsilon_0} \hat{D}(x) \hat{P}(x) \\
+ \frac{1}{2\varepsilon_0 g_\nu(x)} \sum_{\nu=1}^{N} \left[ \varepsilon_0^2 g_\nu(x)^2 \pi^2_\nu(x) + \Omega^2_\nu p^2_\nu(x) \right]
\]

(2.7)

All these equations hold for an arbitrary spatial distribution \( \rho(x) \) of the polarizable atoms. If \( \rho(x) \) is a sum of delta-function terms, a discrete lattice model is obtained; it is an unusual waveguide in which the atoms are all lined up in a one-dimensional row, but not an impossible one, with atom-trapping techniques. For simplicity, we do not treat this type of model here. Instead, we will focus on the uniform dielectric limit, in which all the local-field corrections are included via the relevant Lorentz shifts of the oscillator frequencies, to give a more tractable case.

B. Mode Structure

We now specialize to the case of a continuum model with a uniform distribution, in order to find the dispersion relations for plane-waves. We introduce a mode structure by finding the eigenmodes of the equation of motion. Suppose the solutions to Maxwell’s equations have the form:

\[
\lambda(t, x) = \begin{pmatrix} \lambda(t, x) \\ p_\nu(t, x) \end{pmatrix} = \begin{pmatrix} \bar{\lambda} \\ \bar{p}_\nu \end{pmatrix} e^{ikx-\omega t}.
\]

(2.9)

It follows that these satisfy the resulting equations in the form:
\[ (-\omega^2 + c^2 k^2) \tilde{\lambda} = -ikc^2 \sum_{\nu} \tilde{p}_\nu \]
\[ (-\omega^2 + \Omega_\nu^2)\tilde{p}_\nu = ig_\nu \tilde{\lambda}. \] (2.10)

Solving for \( \tilde{p}_\nu \), we find that:
\[ (\Omega_\nu^2 - \omega^2)(c^2 k^2 - \omega^2)\tilde{p}_\nu = c^2 k^2 g_\nu \sum_{\nu'} \tilde{p}_{\nu'} \] (2.11)
or, summing over all the oscillators and introducing \( \bar{p} = \sum_\nu \tilde{p}_\nu \), we find:
\[ (c^2 k^2 - \omega^2)\bar{p} = c^2 k^2 \sum_\nu \frac{g_\nu}{\Omega_\nu^2 - \omega^2}. \] (2.12)

Eliminating the polarization field \( \bar{p} \) leads to the eigenvalue equation:
\[ \omega^2 = \frac{c^2 k^2}{n^2(\omega)} = c^2 k^2 \left[ 1 - \sum_\nu \frac{g_\nu}{\Omega_\nu^2 - \omega^2} \right]. \] (2.13)

We find a band-structure in which there are typically \((N + 1)\) positive roots \( \omega_\mu(k) \) with \( \mu = 0, 1, \ldots N \) to the solution. To look at this differently, we can solve for the wave-number \( k \), at any given frequency \( \omega \):
\[ k = \pm \left[ \frac{\omega^2/\omega^2}{1 - \sum_\nu \frac{g_\nu}{(\Omega_\nu^2 - \omega^2)}} \right]^{1/2} = \pm k(\omega). \] (2.14)

This solution is unique for any given modal frequency, but has forbidden regions at \( \omega \approx \Omega_\mu \), where \( k^2 \to -\infty \). This indicates a resonance, or absorption band. In the transmission bands, there is a unique refractive index \( n(\omega) \), and hence a well-defined permittivity, \( \varepsilon(\omega) = n^2(\omega)\varepsilon_0 \).

It should be realized that the dispersion relation is not completely identical to the usual classical Sellmeir expansion, although it is very similar to it. The Sellmeir expansion is:
\[ n^2(\omega) = 1 + \sum_\mu \frac{\tilde{g}_\mu}{\Omega_\mu^2 - \omega^2}. \] (2.15)

This agrees with the functional form of the present result to lowest order in \( g_\nu \), if we assume that \( g_\nu = \tilde{g}_\mu \). The difference is simply due to different approaches to treating local-field corrections in the continuum approximation. If a precise local-field theory is required, then the assumption of a homogeneous polarization field must be replaced by a lattice model. In fact, the usual Drude-Lorentz model is not typically obtained from a coupled Lagrangian, so it cannot be readily quantized directly. Rather, it is obtained from an approximate theory in which the local (microscopic) \( E \) field plays the role that the \( D \) field does in the present theory \( \tilde{E} \). From microscopic considerations, both forms needs to have local-field corrections included near an absorption band, in order to give an accurate comparison with a three-dimensional crystal lattice, from first principles.

When this is done in the Drude-Lorentz model, all the low-frequency resonances are shifted by amounts known as the Lorentz shift. With these shifts included, the Sellmeir expansion is obtained, with local-field corrections included. However, the number of poles in the rational function representation derived here is finite, just as in the classical Drude-Lorentz theory. Accordingly, it is always possible to re-express our dispersion relation exactly in the Sellmeir rational-function form, using partial fraction expansions, just as in the Drude-Lorentz derivation of the Sellmeir equation. In this case, the frequencies and couplings \( \tilde{g}_\mu \) appearing in the final Sellmeir formula are not identical with the original frequencies in the Lagrangian; these shifts of course, can be regarded as evidence of the photonic coupling between the oscillators in our model.

An important, and experimentally well-tested property of the Sellmeir equation is that the refractive index approaches unity at high enough frequencies, while at low frequencies it approaches a constant value greater than one:
\[ \lim_{\omega \to 0} n^2(\omega) = 1 + \sum_\mu \frac{\tilde{g}_\mu}{\Omega_\mu^2}. \] (2.16)
Our dispersion relation from the multipolar Hamiltonian has a similar behavior, except that the algebraic form is slightly different at low frequencies:

$$\lim_{\omega \to 0} n^2(\omega) = \left[1 - \sum \frac{g_{\nu}}{\Omega_{\nu}^2}\right]^{-1}. \quad (2.17)$$

Clearly, one form can always be transformed into another, given the obvious restrictions on having distinct roots, with $\sum \frac{g_{\nu}}{\Omega_{\nu}^2} < 1$. We note that this equivalence does not apply in all other models of the dispersion relations, which may have inequivalent analytic properties - leading to a different limiting behavior at high and low frequencies. Provided the general analytic properties are equivalent, the partial-fraction procedure to transform one form into the other is not required in most cases. We shall demonstrate that only the refractive index and group velocity are needed to obtain a complete quantum theoretic description of the modes. This information is readily available from the usual Sellmeir parameters that are experimentally known for many transparent materials. A typical dispersion relation for a solid with both low and high-frequency resonances, would show multiple transmission and absorption bands - with three distinct branches to the dispersion curve - and a region of relatively low group-velocity dispersion between the absorption bands. This is the origin of the well-known zero-dispersion point (at around $\lambda = 1.5 \mu m$ in fused silica), which plays a prominent role in optical communications systems.

III. ONE-DIMENSIONAL MODE OPERATORS

Having derived the modal solutions, we now wish to expand the fields of the theory in terms of annihilation and creation operators. We develop this expansion in two stages. First, we will consider the conditions on the mode expansion which preserve the correct commutation relations for the original canonical fields. Then, we show that a mode expansion defined this way does lead to a diagonal Hamiltonian form, when the Hamiltonian is re-expressed in terms of the mode operators. That is, our goal is to find operators $\hat{a}_\mu(k)$ which have the function of diagonalizing the Hamiltonian, giving the final structure of:

$$H = \sum_{\mu=0}^{N} \int h \omega_{\mu}(k) \hat{a}^\dagger_{\mu}(t, k) \hat{a}_{\mu}(t, k) \, dk. \quad (3.1)$$

A. Commutation properties

Clearly, since the above expansion must lead to the same eigenfrequencies as the original Maxwell equations, we should define mode operators $\hat{a}_\mu$ relative to each branch of the dispersion relation (with, for the sake of simplicity, $N + 1$ distinct branches) so that:

$$\hat{\Lambda}(t, x) = \sum_{\mu=0}^{N} \int dk \left[ \Lambda_{\mu}(k) \hat{a}_{\mu}(t, k) e^{ikx} + h.c. \right]. \quad (3.2)$$

Here $\omega_{\mu}(k)$ is the inverse of $k(\omega)$, for the $\mu$-th branch. The summation is over the discrete branches in the dispersion relation, each of which correspond to a different ‘particle’ type. The time-dependence of the mode operators in the Heisenberg picture - given the desired Hamiltonian structure - must be:

$$\hat{a}_{\mu}(t, k) = \hat{a}_{\mu}(k) e^{-i\omega_{\mu}(k)t}. \quad (3.3)$$

These combined field-polarization excitations can be termed polaritons. We will suppose that the $\hat{a}_{\mu}(k)$ are chosen so that:

$$\left[ \hat{a}_{\mu}(k), \hat{a}^\dagger_{\mu'}(k') \right] = \delta_{\mu\mu'} \delta(k - k'). \quad (3.4)$$

Similarly, the momentum field can be expanded as:

$$\hat{\Pi}(t, x) = \sum_{\mu=0}^{N} \int dk \left[ \Pi_{\mu}(k) \hat{a}_{\mu}(k) e^{ikx - i\omega_{\mu}(k)t} + h.c. \right]. \quad (3.5)$$
The requirement of commutation relations means that we must have (at equal times):

\[
\hat{\Lambda}(x), \hat{\Pi}(x') = i\hbar \delta(x-x')/A = \sum_{\mu=0}^{N} dk \left[ \Lambda_{\mu}(k)\Pi^{*}_{\mu}(k)e^{ik(x-x')} - h.c. \right].
\] (3.6)

This implies the relationship that, in order to preserve commutation relations,

\[
\sum_{\mu=0}^{N} \Lambda_{\mu}(k)\Pi^{*}_{\mu}(k) = \frac{i\hbar}{4\pi A}. \tag{3.7}
\]

The Lagrangian density for this model implies that \(\Pi = \mu \partial_{t} \Lambda\). With the assumed time dependence of the annihilation operators, we can also write \(\Pi^{*}_{\mu}(k)\) in the form of:

\[
\Pi^{*}_{\mu}(k) = -\frac{i\omega_{\mu}(k)}{\omega_{\mu}(k)}\Lambda_{\mu}(k). \tag{3.8}
\]

The above result therefore reduces to an equation for the expansion coefficients \(\Lambda_{\mu}(k)\), in the form of:

\[
\sum_{\mu=0}^{N} \omega_{\mu}(k)\Lambda^{2}_{\mu}(k) = \frac{\hbar}{4\pi A \mu}. \tag{3.9}
\]

Next, we wish to obtain an expression for \(\Lambda_{\mu}(k)\), which is unknown at this stage. It is no restriction to choose \(\Lambda_{\mu}(k)\) to be real. Therefore, we can always choose an (unknown) function \(v_{\mu}(k)\) so that, in analogy to the standard vacuum expansion,

\[
\Lambda_{\mu}(k) = \left[ \frac{hv_{\mu}(k)\varepsilon_{\mu}(k)}{4\pi A k} \right]^{1/2}. \tag{3.10}
\]

If \(v_{\mu}(k) = c\) and \(\varepsilon_{\mu}(k) = \varepsilon_{0}\), this reduces to the accepted vacuum field expansion. More generally, we define \(\varepsilon_{\mu}(k) = k^{2}/[\mu \omega^{2}_{\mu}(k)]\) as the effective permittivity of the \(\mu\)-th branch. We will show later that \(v_{\mu}(k)\) must be interpreted as the group velocity, just as in an earlier narrow-band analysis of this problem, using effective Lagrangian arguments \[\text{(12)}\].

In order to demonstrate this, we first substitute the above expression for \(\Lambda_{\mu}(k)\) into the equation for the consistency of the field and mode-operator commutation relations (i.e. for \([\hat{\Lambda}, \hat{\Pi}]\) and \([\hat{a}, \hat{a}^{\dagger}]\)). This leads to the very simple result that:

\[
(1) \quad \sum_{\mu=0}^{N} \frac{k v_{\mu}(k)}{\omega_{\mu}(k)} = 1. \tag{3.11}
\]

As explained above, we have to determine a function \(v_{\mu}(k)\) which satisfies this condition, and we intend to demonstrate that the choice of \(v_{\mu}(k)\) as the group-velocity of the relevant polariton branch is sufficient; no other correction factors are needed in this simple model. In order to verify this, we can differentiate both sides of Eq. \[\text{(2.13)}\] with respect to \(k\). This gives a group velocity of:

\[
v_{\mu}(k) = \frac{d\omega_{\mu}(k)}{dk} = \frac{\omega_{\mu}(k)}{k} \left( 1 + \sum_{\nu} \frac{k^2 c^2 g_{\nu}}{(\Omega_{\nu}^2 - \omega_{\mu}(k))^2} \right)^{-1}, \tag{3.12}
\]

which is the function we propose to use in the mode expansion.

However, it is clear that the mode function expansion of \(\hat{p}_{\nu}\) and \(\hat{A}_{\nu}\) are also needed, for a complete demonstration of consistency. Using Maxwell’s equations, we note that for a Fourier component of \(p_{\nu}\) at frequency \(\omega\), wave-vector \(k\), we must have:

\[
\tilde{p}_{\nu} = \frac{ikg_{\nu}\tilde{\lambda}}{\Omega_{\nu}^2 - \omega^2}. \tag{3.13}
\]

Thus, if we expand \(\tilde{p}_{\nu}\) as:
\[ \hat{p}_\nu = \sum_{\mu=0}^{N} \int dk \left[ p^\nu_{\mu}(k) \hat{a}_{\mu}(k) e^{ikx - i\omega_{\nu}(k)t} + \text{h.c.} \right], \]  

(3.14)

it follows that the expansion coefficient for in the \( j \)-th frequency band is:

\[ p^\nu_{\mu}(k) = \frac{ikg_{\nu} \Lambda_{\mu}(k)}{(\Omega_{\nu}^2 - \omega_{\mu}^2(k))}. \]  

(3.15)

Similarly, if the canonical momentum for the atomic polarization field is:

\[ \hat{\pi}_\nu(t, x) = \sum_{\mu=0}^{N} \int dk \left[ \pi^\nu_{\mu}(k) \hat{a}_{\mu}(k) e^{ikx - i\omega_{\nu}(k)t} + \text{h.c.} \right], \]  

(3.16)

then the corresponding expansion coefficient is:

\[ \pi^\nu_{\mu}(k) = \frac{k \omega_{\nu}(k) \Lambda_{\mu}(k)}{\varepsilon_0(\Omega_{\nu}^2 - \omega_{\mu}^2(k))}. \]  

(3.17)

For these operators to have the correct equal-time commutators, the different oscillator position operators \( \hat{p}_\nu \) must commute amongst themselves at equal times, as must the different momentum operators \( \hat{\pi}_\nu \). This is trivial from the form of the mode operator expansion. However, the commutation relations (II) between the position and momentum operators are non-trivial. The relevant commutation conditions are therefore:

\[ [\hat{p}_\nu(x), \hat{\pi}_\nu(x')] = i\hbar \delta_{\nu\nu'} \delta(x - x')/A = \sum_{\mu=0}^{N} \int dk \left[ p^\nu_{\mu}(k) \pi^\nu_{\mu}^{*}(k) e^{ik(x-x')} - \text{h.c.} \right]. \]  

(3.18)

This in turn implies that:

\[ \sum_{\mu=0}^{N} p^\nu_{\mu}(k) \pi^\nu_{\mu}^{*}(k) = \frac{i\hbar}{4\pi A} \delta_{\nu\nu'}. \]  

(3.19)

Expanding the coefficients gives the new equation:

\[ \sum_{\mu=0}^{N} \frac{\omega_{\mu}(k)k^2g_{\nu} \Lambda^2_{\mu}(k)}{\varepsilon_0(\Omega_{\nu}^2 - \omega_{\mu}^2(k))(\Omega_{\nu}^2 - \omega_{\mu}^2(k))} = \frac{\hbar}{4\pi A} \delta_{\nu\nu'}. \]  

(3.20)

However, since \( \varepsilon_{\mu}(k) = c^2k^2\varepsilon_0/|\omega_{\mu}(k)| \), and hence

\[ \Lambda^2_{\mu}(k) = \frac{\hbar c^2 \varepsilon_0 k v_{\mu}(k)}{4\pi A \omega_{\mu}^2(k)}, \]  

(3.21)

this simplifies to the form:

\[ (\text{II}) \sum_{\mu=0}^{N} \frac{c^2k^3v_{\mu}(k)g_{\nu}}{\omega_{\mu}(k)(\omega_{\mu}^2(k) - \Omega_{\nu}^2)} = \delta_{\nu\nu'}. \]  

(3.22)

Finally, to ensure that there are correct field-atom commutators, we must satisfy the commutators III-VI. For these cross-terms between the oscillators and field variables, we find that conditions (III) and (IV), involving commutators between the field and the particle position (or the field momentum and particle momentum) are automatically satisfied. This occurs for the same reason that commutators like \([\hat{\Lambda}(x), \hat{\Lambda}(x')]\) or \([\hat{\Pi}(x), \hat{\Pi}(x')]\) must equal zero in our mode expansion. In all these cases involving pairs of canonical position-type operators or pairs of momentum-type operators, the commutator reduces to an integral over \( k \), which is an odd function of \( k \) and hence vanishes when integrated over all positive and negative \( k \)-values.

This leaves the requirements (V) and (VI), which are that \( \hat{\Lambda} \) and \( \hat{\pi}_\nu \) must commute at equal times, as well as \( \hat{\Pi} \) and \( \delta_{\nu} \). These two requirements both imply the same restriction on the expansion coefficients, and hence on \( v_{\mu}(k) \), which is that for all \( k \) and \( \nu \) we must have the conditions:
\[ \sum_{\mu=0}^{N} \frac{k v_{\mu}(k)}{\omega_{\mu}^2(k)\left(\omega_{\mu}^2(k) - \Omega_{\nu}^2\right)} = 0 . \] (3.23)

Despite the complex nature of each of these conditions - which involve sums over all the roots of the dispersion equation, and must be satisfied for all the resonant frequencies \( \nu \), as well all momenta \( k \) - we will show that each of these sums can be analytically evaluated without requiring an algebraic solution for the roots.

**B. Conditions on expansion coefficients**

From the previous results, we have shown that the condition on the expansion coefficient of mode operators is that we must find a function \( v_{\mu}(k) \), such that:

\[ S^{(I)} = \sum_{\mu=0}^{N} \frac{k v_{\mu}(k)}{\omega_{\mu}(k)} = 1 , \] (3.24)

Together with an orthogonality condition:

\[ S^{(II)}_{\nu\nu'} = \sum_{\mu=0}^{N} \frac{c^2 k^3 v_{\mu}(k) g_{\nu'}}{\omega_{\mu}(k)(\omega_{\mu}^2(k) - \Omega_{\nu}^2)(\omega_{\mu}^2(k) - \Omega_{\nu'}^2)} = \delta_{\nu\nu'} . \] (3.25)

In addition, to ensure commutation between the particle and electromagnetic fields, we should impose the condition:

\[ S^{(III)}_{\nu} = \sum_{\mu=0}^{N} \frac{k v_{\mu}(k)}{\omega_{\mu}^2(k)\left(\omega_{\mu}^2(k) - \Omega_{\nu}^2\right) - \Omega_{\nu}^2} = 0 . \] (3.26)

Earlier work [12] on more phenomenological narrow-band quantum models of dispersion led to the conclusion that, for an expansion of modes to be consistent with both Maxwell’s equations and the known dispersive energy, it is necessary to choose \( v_{\mu}(k) \) equal to the group velocity. Thus, the use of \( v_{\mu}(k) = \frac{\partial \omega_{\mu}(k)}{\partial k} \) is an obvious choice, but it is necessary to demonstrate that this still leads to a complete set of consistent commutation relations.

**C. Single Oscillator Case**

As an example, we consider the single-oscillator case, where the dispersion relation can be treated algebraically as the solution of a quadratic equation. In this case the refractive index is given by:

\[ n(\omega)^2 = \left[1 - \frac{g}{\Omega_{\nu}^2 - \omega^2}\right]^{-1} . \] (3.27)

In order to show how the Sellmeir form can be regained, we define a new frequency \( \tilde{\Omega}_{\nu}^2 = \Omega_{\nu}^2 - g \). As long as \( \Omega_{\nu}^2 > g \), the above equation is equivalent to a Sellmeir type of dispersion relation, which is simply:

\[ n(\omega)^2 = 1 + \frac{g}{\Omega_{\nu}^2 - \omega^2} . \] (3.28)

Either equation leads to a quadratic for \( \omega^2 \), having the form:

\[ \omega^4 - \omega^2\left(c^2 k^2 + \Omega_{\nu}^2\right) + \Omega_{\nu}^2 - g = 0 . \] (3.29)

The resonant frequencies at any given wavenumber \( k \), are then:

\[ \omega = \frac{1}{2} \left(c^2 k^2 + \Omega_{\nu}^2 \pm \sqrt{(c^2 k^2 + \nu^2)^2 - 4c^2 k^2(\Omega_{\nu}^2 - g)}\right) . \] (3.30)

We now identify \( v_{\mu}(k) \) with the group-velocity on each of the two branches, by taking derivatives with respect to \( k \). Thus, assuming \( \Omega_{\nu}^2 > g \) (to have distinct branches):
where the quantity $\Delta$ is given by

$$\Delta = \sqrt{(c^2 k^2 + \Omega^2) - 4c^2 k^2 (\Omega^2 - g)} .$$

(3.32)

Clearly it is necessary to have $\Omega^2 > g$ in order to have distinct real branches to the dispersion relation, each with positive frequency $\omega$. This is precisely the condition under which the Sellmeir expansion is valid, as one might have expected.

Summing over the two branches, we note that (defining $\bar{K} = c^2 k^2 + \Omega^2$):

$$\sum_{\pm} \frac{k v_\pm(k)}{\omega_\pm(k)} = \frac{1}{4} \sum_{\pm} \left( 1 \pm \frac{\bar{K} + 2(g - \Omega^2)}{\Delta} \right) \frac{\bar{K} + \Delta}{\Omega^2 - g} .$$

(3.33)

On taking the sum, this reduces to the required result of:

$$S^{(1)} = \sum_{\pm} \frac{k v_\pm(k)}{\omega_\pm(k)} = 1 .$$

(3.34)

This indicates that the use of group-velocity expansion coefficients appears correct in this case, although we have only calculated the simplest of the commutators. However, this algebraic technique is rather clumsy to use for the other identities. Even worse, it is not able to be used at all for an arbitrary number of branches; in these more general cases there is no closed form expression for the solution to the dispersion equation.

IV. ANALYTIC PROPERTIES OF DISPERSION RELATIONS

For the other, more complex, commutation relation identities - or more oscillators - it is preferable to use techniques from complex function theory, which transform the sums over roots of the dispersion relation to complex contour integrals of related meromorphic functions. The dispersion relations considered here have the general structure of:

$$\frac{\omega^2}{c^2 k^2} = 1 - \sum_{\nu} \frac{g_{\nu}}{\Omega_{\nu}^2 - \omega^2} .$$

(4.1)

This can be written in the form of roots of a polynomial in $z = \omega^2$, so that:

$$\kappa a(z_\mu) = b(z_\mu) ,$$

(4.2)

where $z_\mu = \omega^2_\mu$, $\kappa = c^2 k^2$ and:

$$a(z) = \prod_{\nu} (\Omega_{\nu}^2 - z) - \sum_{\nu, \nu'} g_{\nu, \nu'} \prod_{\nu \neq \nu'} (\Omega_{\nu'}^2 - z) ,$$

(4.3)

$$b(z) = z \prod_{\nu} (\Omega_{\nu}^2 - z) .$$

(4.4)

Next, in order to test the assumption that the expansion coefficients involve group velocities, we must consider the slope of the dispersion relations:

$$\kappa a'(z) + \frac{\partial \kappa}{\partial z} a(z) = b'(z) .$$

(4.5)

Hence,

$$v_\mu(k) = \frac{\partial \omega_\mu(k)}{\partial k} = \frac{c^2 k a(z)}{\omega_\mu(k)(b'(z) - \kappa a')} .$$

(4.6)

A simpler way to write this expression - entirely equivalent to the above definition - is Eq. (4.14). While this form is more transparent, the above expression is a rational function, which is amenable to analysis using Cauchy’s theorem.
A. Condition I

The sum \( S^{(I)} \) has the form:

\[
S^{(I)} = \sum_{\mu} \frac{kv_{\mu}(k)}{\omega_{\mu}(k)} = \sum_{\mu} \left[ \frac{\kappa a(z)}{z} \frac{1}{b'(z) - \kappa a'(z)} \right]_{z = z_{\mu}(\kappa)}. \tag{4.7}
\]

Next, consider the complex function, defined for the analytic continuation of \( z \) into complex values:

\[
f^{(I)}(z) = \frac{\kappa a(z)}{z [b(z) - \kappa a(z)]}. \tag{4.8}
\]

This generally has \( (N + 2) \) poles; and has the property that \( \lim_{|z| \to \infty} f^{(I)}(z) \sim 1/z^2 \). For example, in the trivial case of no oscillators \((N = 0)\), we find that:

\[
f^{(I)}(z) = \frac{\kappa}{z(z - \kappa)}. \tag{4.9}
\]

In this case, the identity \((I)\) is satisfied trivially, since it reduces to \( \text{Res}[f^{(I)}(z = c^2 k^2)] = 1 \). The sum of residues of \( f^{(I)}(z) \) is zero in this case, which must be true in general for a meromorphic function behaving as \( f^{(I)}(z) \sim 1/z^2 \) as \( |z| \to \infty \). As usual in complex function theory of the inverse variable \((u = 1/z)\), a contour integral around all the finite poles turns into a contour integral around zero poles at infinity, and hence must equal zero. Thus, we have the general result that:

\[
0 = \sum \text{Res} \left[ f^{(I)}(z) \right] = -1 + \sum_{\mu} \left[ \frac{\kappa a(z_{\mu})}{z_{\mu} (b'(z_{\mu}) - \kappa a'(z_{\mu}))} \right]. \tag{4.10}
\]

However, this is precisely condition \((I)\), for the \( N \)-oscillator case, since:

\[
S^{(I)} = \sum_{\mu} \left[ \frac{\kappa a(z_{\mu})}{z_{\mu} (b'(z_{\mu}) - \kappa a'(z_{\mu}))} \right] = \sum_{\mu} \frac{kv_{\mu}(k)}{\omega_{\mu}(k)} = 1. \tag{4.11}
\]

Thus, the use of complex function theory shows that \((I)\) is always satisfied, provided there are \((N + 1)\) distinct roots.

B. Condition II

Similarly, we can prove the other relations. For example, to prove relation \((II)\) we consider:

\[
f^{(II)}_{\nu \nu'}(z) = \frac{\kappa g_{\nu \nu'} f^{(I)}(z)}{(z - \Omega_{\nu}^2)(z - \Omega_{\nu'}^2)}. \tag{4.12}
\]

Summing the residues of this function, and noting that \( \lim_{z \to 0} f^{(I)}(z) = -1/z \), we find (for \( \nu \neq \nu' \)):

\[
0 = S^{(II)}_{\nu \nu'} = -\kappa g_{\nu \nu'} \frac{\Omega_{\nu}^2}{\Omega_{\nu}^2 + \Omega_{\nu'}^2} + \kappa g_{\nu \nu'} \frac{f^{(I)}(\Omega_{\nu}^2)}{(\Omega_{\nu}^2 - \Omega_{\nu'}^2)} + \kappa g_{\nu \nu'} f^{(I)}(\Omega_{\nu'}^2). \tag{4.13}
\]

Examining the RHS of the required identity, we must obtain the value of \( f^{(I)}(\Omega_{\nu}^2) \), evaluated at each resonance:

\[
f^{(I)}(\Omega_{\nu}^2) = \frac{\kappa a(\Omega_{\nu}^2)}{\Omega_{\nu}^2 [b(\Omega_{\nu}^2) - \kappa a(\Omega_{\nu}^2)]}. \tag{4.14}
\]

However, \( b(\Omega_{\nu}^2) = 0 \) at each resonance, so that \( f^{(I)}(\Omega_{\nu}^2) = -1/\Omega_{\nu}^2 \). Hence, the RHS of the required identity becomes:

\[
S^{(II)}_{\nu \nu'} = \kappa g_{\nu \nu'} \left[ \frac{1}{\Omega_{\nu}^2 \Omega_{\nu'}^2} + \frac{1}{\Omega_{\nu}^2 (\Omega_{\nu}^2 - \Omega_{\nu'}^2)} + \frac{1}{\Omega_{\nu'}^2 (\Omega_{\nu}^2 - \Omega_{\nu'}^2)} \right] = 0. \tag{4.15}
\]
In the case that \( \nu = \nu' \), a double pole is found, so the residue is obtained on differentiating \( f^{(I)}(z) \). We can perform this operation most simply in the neighborhood of the double root at \( z = \Omega^2_{\nu} \), by using the definition of \( f^{(I)}(z) \) to show that:

\[
f^{(I)}(z) = -\frac{1}{z} + \frac{1}{z - \kappa \left(1 - \sum_{\nu} g_{\nu}/(\Omega^2_{\nu} - z)\right)}.
\] (4.16)

Thus, as \( z \to \Omega^2_{\nu} \), we find the second term is dominated by the pole in the denominator:

\[
\lim_{z \to \Omega^2_{\nu}} f^{(I)}(z) = -\frac{1}{z} + \frac{\Omega^2_{\nu} - z}{\kappa g_{\nu}}.
\] (4.17)

Hence, on differentiating to obtain the residue,

\[
\frac{\partial}{\partial z} f^{(I)}(z) \bigg|_{z = \Omega^2_{\nu}} = -\frac{1}{\kappa g_{\nu}} + \frac{1}{\Omega^2_{\nu}}.
\] (4.18)

This is sufficient to complete the proof of the second relation, which is

\[
S^{(II)}_{\nu\nu'} = \sum \text{Res} \left[ f^{(II)}_{\nu\nu'}(z) \right] + \delta_{\nu\nu'} = \delta_{\nu\nu'}.
\] (4.19)

C. Condition III-VI

As shown previously, the conditions (III) - (IV) are straightforward consequences of the general type of expansion chosen here, while conditions (V) and (VI) reduce to an identical summation identity. To obtain this last identity, we can now introduce a third analytic function,

\[
f^{(III)}_{\nu} = \frac{f^{(I)}(z)}{z - \Omega^2_{\nu}}.
\] (4.20)

As well as the poles at \( z = 0 \) and the \((N+1)\) roots of the dispersion relation, this has a pole at \( z = \Omega^2_{\nu} \). However, the residues at \( z = 0 \) and \( z = \Omega^2_{\nu} \) cancel each other, so the sum over the remaining zeros must give zero, as required. In summary, we find that summing over the residues gives:

\[
S^{(III)}_{\nu\nu'} = \sum \text{Res} \left[ f^{(III)}_{\nu}(z) \right] = 0.
\] (4.21)

This proves the last sum-rule requirement on the commutators.

V. HAMILTONIAN

We now wish to show that when the Hamiltonian is expressed in terms of the operators \( \hat{a}_{\mu}(k) \) and \( \hat{a}^\dagger_{\mu}(k) \), \( \mu = 0, \ldots, N \), it is of diagonal form. Our first step is to derive an orthogonality relation for the classical modes. This will allow us to show that the Hamiltonian contains only terms of the form \( \hat{a}^\dagger_{\mu}(k)\hat{a}_{\mu}(k) \). The next step is to examine the normalization of the modes. Once this has been done we find that the Hamiltonian is given by

\[
H = \sum_{\mu=0}^{N} \int dk \hbar \omega_{\mu}(k) \hat{a}^\dagger_{\mu}(k)\hat{a}_{\mu}(k).
\] (5.1)

In order to find the proper orthogonality relation for the modes, let us first define the \( N+1 \) component vector:

\[
\vec{\lambda} = \left( \begin{array}{c} \tilde{\lambda} \\ \tilde{p}_{\nu} \end{array} \right),
\] (5.2)

or \( \lambda_0 = \tilde{\lambda} \) and \( \lambda_s = \tilde{p}_{\nu} \) for \( s \geq 1 \). The eigenvalue equations can be expressed in the form (for each value of \( k \)
\[ M \overline{\lambda} = \omega^2 \overline{\lambda}, \]  
(5.3)

where the \((N + 1) \times (N + 1)\) matrix \(M\) is given by

\[
M = \begin{pmatrix}
  k^2 c^2 & i k c^2 & i k c^2 & \ldots \\
  -i k g_{v_1} & \Omega_{v_1}^2 & 0 & \ldots \\
  -i k g_{v_2} & 0 & \Omega_{v_2}^2 & \ldots \\
  \vdots & \vdots & \vdots & \ddots
\end{pmatrix},
\]
(5.4)

The matrix \(M\) is not hermitian, but if it is multiplied by the positive, diagonal, \((N + 1) \times (N + 1)\) matrix \(G\),

\[
G_{rs} = \delta_{rs} G_{ss},
\]

\[
G_{ss} = \begin{cases} 1 & s = 0 \\ c^2 / g_s & s \geq 1 \end{cases},
\]
(5.5)

then the combination \(GM\) is hermitian. Therefore, if

\[
M \lambda^{(1)} = \omega_1^2 \lambda^{(1)}, \quad M \lambda^{(2)} = \omega_2^2 \lambda^{(2)},
\]
(5.6)

then

\[
\langle \lambda^{(2)} | GM \lambda^{(1)} \rangle = \omega_1^2 \langle \lambda^{(2)} | \lambda^{(1)} \rangle,
\]
(5.7)

\[
\langle \lambda^{(2)} | GM \lambda^{(2)} \rangle = \omega_2^2 \langle \lambda^{(2)} | \lambda^{(1)} \rangle.
\]
(5.8)

This implies that if \(\omega_1^2 \neq \omega_2^2\), then

\[
\langle \lambda^{(2)} | G \lambda^{(1)} \rangle = 0,
\]
(5.9)

and we have the desired orthogonality relation. Expressing this in slightly more generality, we note that for each value of \(k\) there are \(N + 1\) eigenvectors \(\lambda^{(\mu)}\), \(\mu = 0, \ldots, N\), each corresponding to a different eigenvalue \(\omega^{(\mu)}(k)\). As a result we have that

\[
\langle \lambda^{(\mu)} | G \lambda^{(\mu')} \rangle = \delta_{\mu \mu'} \langle \lambda^{(\mu)} | \lambda^{(\mu)} \rangle.
\]
(5.10)

We now express the fields in terms of the eigenvectors and substitute them into the Hamiltonian, which is given by integrating the Hamiltonian density in Eq. (2.8) over \(x\). In particular, we have that

\[
\Lambda^{(\mu)} = \lambda^{(\mu)}(0), \quad P^{(\mu)} = \lambda^{(\mu)}(0) \\
\Pi_{\mu} = -i \mu \omega^{(\mu)}(0), \quad \pi_{\mu} = \frac{-i \mu \lambda^{(\mu)}(0)}{\varepsilon_{0} g_{\nu}}.
\]
(5.11)

This allows us to use Eqs. (5.4) and (5.10) when calculating the Hamiltonian, and we find that

\[
H = 4 \pi \mu A \sum_{\mu=0}^{N} \int dk \langle \lambda^{(\mu)}(k) | G \lambda^{(\mu)}(k) \rangle \omega^{2} \mu(k) \hat{a}^{\dagger}_{\mu}(k) \hat{a}_{\mu}(k).
\]
(5.13)

In order to show that the Hamiltonian assumes the form given in Eq. (5.4), and to justify our assumption that \(i \hat{a}_{\mu} = \omega_{\mu} \hat{a}_{\mu}\), we need to prove that

\[
4 \pi \mu A \langle \lambda^{(\mu)}(k) | G \lambda^{(\mu)}(k) \rangle \omega_{\mu}(k) = \hbar.
\]
(5.14)

Noting that

\[
\lambda^{(\mu)}(k) = \frac{i k g_{\nu} \lambda^{(\mu)}(0)}{\Omega^{2}_{\nu} - \omega_{\mu}^{2}} \lambda^{(\mu)}(0), \quad \lambda^{(\mu)}(0) = \Lambda_{\mu} = \sqrt{\frac{\hbar v_{\mu} k}{4 \pi \mu A \omega_{\mu}^{2}}},
\]
(5.15)

we see that Eq. (5.14) will be true if
This implies that that \( v_\mu = d\omega_\mu / dk \), from Eq. (3.12). We can then conclude that the expression for the Hamiltonian given in Eq. (5.1) is correct.

Summarizing, our theory of a linear medium with \( N \) resonances is described by the Hamiltonian in Eq. (5.1) and the corresponding field operators have the expansion:

\[
\hat{\mathcal{D}}(t, x) = i \sum_\mu \int dk \left[ \frac{\hbar \varepsilon(\omega_\mu(k)) v_\mu(k)}{4\pi \varepsilon_0 k A} \right]^{1/2} \hat{a}_\mu(k) e^{i k x - i \omega_\mu(k) t} + h.c.,
\]

(5.17)

\[
\hat{\mathcal{E}}(t, x) = i \sum_\mu \int dk \left[ \frac{\hbar k v_\mu(k)}{4\pi \varepsilon(\omega_\mu(k)) A} \right]^{1/2} \hat{a}_\mu(k) e^{i k x - i \omega_\mu(k) t} + h.c.,
\]

(5.18)

\[
\hat{\mathcal{B}}(t, x) = -i \sum_\mu \int dk \left[ \frac{\hbar k v_\mu(k)}{4\pi A} \right]^{1/2} \hat{a}_\mu(k) e^{i k x - i \omega_\mu(k) t} + h.c.
\]

(5.19)

Here we have also included the electric field expansion (which is obtained by including the polarization term), for comparison with more familiar results. As one might expect, the only difference between the electric field and displacement field expansions, is a factor proportional to the dielectric permittivity \( \varepsilon(\omega_\mu(k)) \) in each branch of the dispersion relation. It is important to notice here that the two fields cannot be related by one, frequency-independent displacement field expansions, is a factor proportional to the dielectric permittivity \( \varepsilon(\omega_\mu(k)) \) in each branch of the dispersion relation.

In order to provide a more physical understanding of this result, the summation over the branches in one dimension can be replaced by an integral over the mode frequency, in each propagation direction, i.e., define:

\[
\hat{\mathcal{D}}(t, x) = \hat{\mathcal{D}}^{(+)}(t, x) + \hat{\mathcal{D}}^{(-)}(t, x).
\]

(5.20)

Now, since the mode frequency has a bounded range for each root, we can define a frequency dependent mode operator as:

\[
\hat{a}(\omega) = \hat{a}_\mu(k) / \sqrt{|v_\mu(\omega)|},
\]

(5.21)

where the appropriate root \( \mu \) is chosen in each case to correspond to the mode frequency argument - except, of course, in the forbidden bands. The commutators of the new mode operators are:

\[
[\hat{a}(\omega), \hat{a}^\dagger(\omega')] = \delta(\omega - \omega'),
\]

(5.22)

and the mode expansion is now the same as it would be in a non-dispersive case - except that no modes exist in the forbidden bands:

\[
\hat{\mathcal{D}}^{(\pm)}(t, x) = \pm i \int_0^\infty d\omega \left[ \frac{\hbar k(\omega)\varepsilon(\omega)}{4\pi A} \right]^{1/2} \hat{a}(\omega) e^{\pm i k(\omega)x - i \omega t} + h.c.
\]

(5.23)

Similar equations hold for the other fields; for example, the electric and magnetic field expansions are just:

\[
\hat{\mathcal{E}}^{(\pm)}(t, x) = \pm i \int_0^{\infty'} d\omega \left[ \frac{\hbar k(\omega)}{4\pi \varepsilon(\omega) A} \right]^{1/2} \hat{a}(\omega) e^{\pm i k(\omega)x - i \omega t} + h.c.
\]

(5.24)

and:

\[
\hat{\mathcal{B}}^{(\pm)}(t, x) = -i \int_0^{\infty'} d\omega \left[ \frac{\hbar k(\omega)}{4\pi \varepsilon(\omega) A} \right]^{1/2} \hat{a}(\omega) e^{\pm i k(\omega)x - i \omega t} + h.c.
\]

(5.25)

The important point is that we can exactly absorb the group velocity factor into the frequency integral - which, however, is only defined in the range of allowed mode frequencies. This result (also obtained in earlier narrow-band Lagrangian approach [1, 12], and in a single-resonance model [13]), was most clearly emphasized in the single-resonance theory of Huttner and Barnett [14]. It implies that the two-time correlation function for narrow-band fields in the transmission band, is essentially identical to those of the corresponding vacuum fields, apart from the usual reflectivity factors. This is a necessary ingredient of any theory of the interface properties of the fields, and will be explored in more detail in a subsequent paper. The above mode expansion has been widely used in quantum optics, and the present result shows that it is exact for a realistic, multiple-resonance model of a dispersive medium - provided we recognize that there are no modes in the forbidden bands.
VI. HIGHER-DIMENSIONAL MODELS

We next consider an n-dimensional Lagrangian for oscillators with mass \( m_\nu \), displacement \( r_\nu \), effective charge \( q_\nu \), and oscillation frequencies \( \Omega_\nu \) about their center of charge position \( \mathbf{x}_\nu \):

\[
\mathcal{L}_n = \sum_\nu \left\{ \frac{m_\nu}{2} \left( \dot{r}_\nu^2 - \Omega_\nu^2 r_\nu^2 \right) + \frac{q_\nu}{\varepsilon_0} r_\nu \cdot \mathbf{D}(\mathbf{x}_\nu) \right\} .
\]  

(6.1)

Here \([r_\nu q_\nu]\) is the dipole moment of a charged particle in a multi-polar Drude-Lorentz model. In this general case, the labels \( \nu \) may correspond either to distinct resonances of one atom or to distinct positions. Each resonance has its own corresponding effective charge - and hence dipole moment. Any sum rules are incorporated into the definitions of the effective charges involved in a given transition. For simplicity, the self-energy terms proportional to \( \mathbf{P}^2 \) are included in the definition of the resonant frequencies, which are defined to diagonalize the individual charge-cell Hamiltonians in the multipolar gauge. The coupling in multipolar gauge is to the displacement field \( \mathbf{D}(\mathbf{x}_\nu) \) at the central position \( \mathbf{x}_\nu \), used as an origin for defining a local polarization. All inter-atomic Coulomb terms in this gauge, are carried by the photon-exchange process, which couples atoms at distinct spatial positions.

We introduce a vector gauge field - the dual potential \( \Lambda \), so that \( \mathbf{D} = \nabla \times \Lambda \) and \( \mathbf{B} = \mu \partial \Lambda / \partial t \). The discrete cell positions are now replaced by a continuum polarization field as before, together with the appropriate local field corrections. To account for more general dielectric structures that may have local interactions not included in the Coulomb corrections, we now include a quadratic dispersion term \( \alpha_\nu \) to describe the residual phonon and exciton dispersion [22], that exists in the absence of long wavelength electromagnetic couplings.

The simplest Lagrangian density that generates the correct electromagnetic energy and Maxwell’s equations for an n-dimensional wave-guide with effective cross-section \( A \simeq d^3 \) is:

\[
\mathcal{L} / A = \frac{1}{2\mu} \dot{\Lambda}^2(\mathbf{x}) + \frac{1}{\varepsilon_0} \left[ \mathbf{P}(\mathbf{x}) \cdot \nabla \times \Lambda(\mathbf{x}) - \frac{1}{2} (\nabla \times \Lambda(\mathbf{x}))^2 \right] + \sum_{\nu=1}^N \left[ \dot{p}_\nu^2(\mathbf{x}) - \Omega_\nu^2 p_\nu^2(\mathbf{x}) - \alpha_\nu(\mathbf{x})|\nabla_i p_\nu(\mathbf{x})|^2 \right] / (2\varepsilon_0 g_\nu(\mathbf{x})) .
\]  

(6.2)

Here the polarization density due to all the dipoles is \( \mathbf{P}(\mathbf{x}) = \sum_\nu \mathbf{p}_\nu(\mathbf{x}) = \sum_\nu r_\nu q_\nu \rho_\nu(\mathbf{x}) \), where \( \rho_\nu(\mathbf{x}) \) is the number density of the oscillators with resonant frequency \( \Omega_\nu \). The partial polarization \( \mathbf{p}_\nu(\mathbf{x}) \) is regarded as a continuum field, with \( \nu = 1, \ldots, N \) labeling the bare frequency of the elementary phonon and exciton resonances, in the absence of coupling to the long wavelength photons. We have assumed that the dispersion of phonon and exciton modes are the same for longitudinal and transverse modes. The transverse dispersion will ultimately be modified by the coupling between the field and the polarization, which is entirely included in the term \( g_\nu(\mathbf{x}) \), as defined in Eq. (2.3).

We impose the usual gauge constraint that \( \nabla \cdot \Lambda = 0 \), so that the field variable only has transverse degrees of freedom; this does not apply to the polarization. In addition, we can impose wave-guiding conditions that \( \Lambda \) is restricted to a one, two or three dimensional manifold. In practise, dispersion occurs in the electromagnetic mode functions (which are wavelength dependent), so that it is necessary to solve for the complete three-dimensional mode structure to rigorously treat a fiber waveguide, for example. However, a simple low-dimensional wave-guiding theory is still useful as a guide to the behavior of a complete theory. In the full three-dimensional case, the area term \( A \) is simply omitted, as \( A = 1 \).

The resulting generalized Maxwell-Bloch equations are:

\[
[\partial_t^2 - c^2 \nabla^2] \Lambda = c^2 \nabla \times \mathbf{P}(\mathbf{x}) \\
[\partial_t^2 + \Omega_\nu^2(k)] \mathbf{p}_\nu = g_\nu(\mathbf{x}) \nabla \times \Lambda + \nabla_i (\alpha_\nu(\mathbf{x}) \nabla_i \mathbf{p}_\nu(\mathbf{x})) .
\]  

(6.3)

In order to quantize the system, we introduce the canonical momenta , \( \Pi(\mathbf{x}) \) and \( \pi_\nu(\mathbf{x}) \), where:

\[
\Pi(\mathbf{x}) = \mu \dot{\Lambda}(\mathbf{x}) = \mathbf{B}(\mathbf{x}) ,
\]

\[
\pi_\nu(\mathbf{x}) = \frac{1}{\varepsilon_0 g_\nu(\mathbf{x})} \dot{p}_\nu(\mathbf{x}) \propto m \mathbf{v} .
\]  

(6.4)

The quantization rules are the usual ones obtained on replacing Poisson brackets with operator commutators. Scaling by \( A \) is introduced so that the field units are independent of waveguide dimension, and the delta-functions are n-dimensional. All position-type operators of the same type of variable must commute amongst themselves, as do all
momentum-type operators. The commutators involving cross-terms between the position and momentum operators, and between the different types of variable, are:

\[
\begin{align*}
(\text{I}) & \quad \left[ \hat{D}_i(x), \hat{B}_j(x') \right] = i\hbar \nabla_x \times \delta^i_j(x-x') / A , \\
(\text{II}) & \quad \left[ \hat{\rho}_{\mu}(x), \pi_{\mu}(x') \right] = i\hbar \delta_{ij} \delta_{\mu\nu} \delta(x-x') / A , \\
(\text{III}) & \quad \left[ \hat{D}_i(x), \hat{\rho}_{\mu}(x') \right] = 0 , \\
(\text{IV}) & \quad \left[ \hat{B}_i(x), \pi_{\mu}(x') \right] = 0 , \\
(\text{V}) & \quad \left[ \hat{D}_i(x), \pi_{\mu}(x') \right] = 0 , \\
(\text{VI}) & \quad \left[ \hat{B}_i(x), \hat{\rho}_{\mu}(x') \right] = 0 .
\end{align*}
\]

Here we have introduced the usual notation of a transverse delta function defined so that:

\[
\delta^i_j(x) = \frac{1}{(2\pi)^6} \int d^6k \delta^i_j(k)e^{ik \cdot x},
\]

where \(\delta^i_j(k) \equiv (\delta_{ij} - k_i k_j / |k|^2)\) is the transverse projector in momentum space. In three dimensions, the first commutator can also be written in the more familiar form of:

\[
\text{(I) } \quad \left[ \hat{E}_i(x), \hat{B}_j(x') \right] = i\hbar \nabla_x \times \delta^i_j(x-x') / \varepsilon_0 .
\]

The final quantum Hamiltonian density is (using normal ordering):

\[
\mathcal{H} / A = \frac{1}{2\varepsilon_0} \widehat{D}^2(x) + \frac{1}{2\mu} \widehat{B}^2(x) - \frac{1}{\varepsilon_0} \hat{D}(x) \cdot \hat{P}(x) + \frac{1}{2\varepsilon_0 g_0(x)} \sum_{\nu=1}^N \left[ \epsilon^{2}_{\nu} \hat{g}^2_{\nu}(x) \hat{\pi}^2_{\nu}(x) + \Omega^2_{\nu} \hat{\pi}^2_{\nu}(x) + \alpha_{\nu}(x) |\nabla \cdot \hat{P}_{\nu}(x)|^2 \right] .
\]

These equations hold for an arbitrary spatial distribution \(\rho(x)\) of the continuum of polarizable atoms.

**VII. THREE-DIMENSIONAL QUANTUM MODE OPERATORS**

In this section, we specialize to the case of a continuum model with uniform couplings and velocities, as in the one-dimensional case, in order to find the dispersion relations for plane-waves. As in the one-dimensional case as well, we introduce a mode structure by finding the eigenmodes to Maxwell’s equations in the form:

\[
\begin{pmatrix}
\lambda(t, \mathbf{x}) \\
\mathbf{p}_\nu(t, \mathbf{x})
\end{pmatrix} = \begin{pmatrix}
\tilde{\lambda}(\omega, \mathbf{k}) \\
\tilde{\mathbf{p}}_\nu(\omega, \mathbf{k})
\end{pmatrix} e^{i \mathbf{k} \cdot \mathbf{x} - i \omega t} .
\]

Defining \(k = |\mathbf{k}|\), it follows that these satisfy the Maxwell-Bloch equations in the form:

\[
\begin{align*}
(-\omega^2 + c^2 k^2) \tilde{\lambda} = i c^2 \mathbf{k} \times \sum_{\nu} \tilde{\mathbf{p}}_\nu , \\
(-\omega^2 + \Omega^2_{\nu}(k)) \tilde{\mathbf{p}}_\nu = i g_\nu \mathbf{k} \times \tilde{\lambda} .
\end{align*}
\]

Our model now includes simple phonon/exciton dispersion. These effects cause the Fourier-domain equations for the polarization to be modified, so that \(\Omega^2_{\nu}(k) = \Omega^2_{\nu} + k^2 \alpha_{\nu}\), where we define \(k = |\mathbf{k}|\) in this section. The phonon/exciton dispersion can be generalized to more complicated cases if desired, with higher order \(k\)-dependences - although in many cases only the relatively long wavelength (small \(k\)) phonons are relevant to optical properties. In terms of the coupled equations given above, only the transverse part of the polarization couples to the photons, hence we can define:

\[
\tilde{\mathbf{p}}_\nu^{-1}(\mathbf{k}) = \delta^i_j(\mathbf{k}) \tilde{p}_j(\mathbf{k}) .
\]
Solving for $\tilde{\mathbf{P}}^\perp$ by summing over the transverse polarizations of all the oscillators, we find:

$$
(c^2 k^2 - \omega^2)\tilde{\mathbf{P}}^\perp = \tilde{\mathbf{P}}^\perp c^2 k^2 \sum_{\nu} \frac{g_{\nu}}{\Omega^\perp_{\nu}(k) - \omega^2}.
$$

(7.4)

The eigenvalues of the longitudinal part of the polarization field are not changed by the coupling, while eliminating the transverse polarization field $\tilde{\mathbf{P}}^\perp$ leads to the eigenvalue equation:

$$
\omega^2 = \frac{c^2 k^2}{n^2(\omega)} \equiv c^2 k^2 \left[1 - \sum_{\nu} \frac{g_{\nu}}{\Omega^\perp_{\nu}(k) - \omega^2} \right].
$$

(7.5)

For any wave-number $k = |\mathbf{k}|$, we find a band-structure in which there are typically $(2(N+1))$ positive roots $\omega_{0\sigma}(k), \omega_{N\sigma}(k)$ to the transverse eigenvalue equations for each $k$ value, with $\sigma = 1, 2$, and $\omega_{1\lambda}(k) = \omega_{2\lambda}(k)$ due to the isotropy of our model. Each mode therefore has two orthogonal polarization unit vectors $\mathbf{u}_{\sigma}(k)$, such that $\mathbf{k} \cdot \mathbf{u}_{\sigma}(k) = 0$. Similarly, there are $(N)$ longitudinal roots, which are labeled $\sigma = 0$, and are unchanged by the long wavelength electromagnetic couplings. In the case where phonon/exciton dispersion is omitted, the transverse dispersion relation is precisely the same as in the one-dimensional model.

As before, the solution is unique for any given modal frequency, but has forbidden regions which indicate a resonance, or absorption band. Typical dispersion relations for this model also demonstrate the existence of transmission and absorption bands - but with additional structure in each branch, whose origin is in the phonon (or exciton) dispersion. From now on, we use the notation $\omega_{\mu\sigma}(k)$ with $\sigma = 1, 2$, to indicate a solution to the full transverse equations. The notation $\omega_{\mu0}(k) = \Omega_{\mu}(k)$ indicates the longitudinal eigenvalue, which of course is undefined for $\mu = 0$, in the absence of longitudinal photons.

A. Three-dimensional commutators

Having derived the modal solutions, we now wish to expand the fields of the theory in terms of annihilation and creation operators in the three-dimensional model. These have the function of diagonalizing the Hamiltonian, which we anticipate will have the final structure of:

$$
H = \sum_{\mu=0}^{N} \sum_{\sigma=0'}^{2} \int d^2k \hbar \omega_{\mu\sigma}(k) \hat{a}_{\mu\sigma}^\dagger(k) \hat{a}_{\mu\sigma}(k).
$$

(7.6)

Here the lower limit notation of $\sigma = 0'$ is defined to exclude the unphysical combination of $\mu = 0, \sigma = 0$. As before, the modal frequency $\omega_{\mu\sigma}(k)$ is the inverse of $k(\Omega)$, for the $\mu$-th transverse branch. This expansion requires that we define mode operators $\hat{a}_{\mu\sigma}$ in the $\mu$-th branch of the dispersion relation so that:

$$
\hat{\Lambda}(t, \mathbf{x}) = \sum_{\mu=0}^{N} \sum_{\sigma=1}^{2} \int d^2k \left[ \Lambda_{\mu\sigma}(k) \hat{a}_{\mu\sigma}(k)e^{i\mathbf{k} \cdot \mathbf{x} - i\omega_{\mu\sigma}(k)t} + h.c. \right].
$$

(7.7)

The summation here is just over the transverse branches in the dispersion relation. These combined transverse field-polarization excitations can be termed polaritons, and we notice here that there can be longitudinal excitations that propagate, as is usual in solid-state physics. The commutation properties of the annihilation and creation operators $\hat{a}_{\mu\sigma}(k)$ are chosen so that:

$$
\left[ \hat{a}_{\mu\sigma}(k), \hat{a}_{\mu'\sigma'}^\dagger(k') \right] = \delta_{\mu\mu'} \delta_{\sigma\sigma'} \delta(\mathbf{k} - \mathbf{k}') .
$$

(7.8)

Similarly, the momentum field can be expanded as:

$$
\tilde{\Pi}(t, \mathbf{x}) = \sum_{\mu=0}^{N} \sum_{\sigma=1}^{2} \int d^2k \left[ \Pi_{\mu\sigma}(k) \hat{a}_{\mu\sigma}(k)e^{i\mathbf{k} \cdot \mathbf{x} - i\omega_{\mu\sigma}(k)t} + h.c. \right].
$$

(7.9)

Thus, at equal times:

$$
\left[ \hat{\Lambda}_i(\mathbf{x}), \hat{\Pi}_j(\mathbf{x}') \right] = i\hbar \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}')/A = \sum_{\mu=0}^{N} \sum_{\sigma=1}^{2} \int d^2k \left[ \Lambda_{i\mu\sigma}(k) \Pi_{j\mu\sigma}(k)e^{i\mathbf{k} \cdot (\mathbf{x} - \mathbf{x}') - h.c.} \right].
$$

(7.10)
This implies that, in order to preserve commutation relations, we have similar results to the one-dimensional case -

\[
\sum_{\mu=0}^{N} \sum_{\sigma=1}^{2} \Lambda_{\mu\sigma}(k) \Pi_{\mu\sigma}(k) = \frac{i\hbar}{2A(2\pi)^n} \tilde{\delta}_{ij}(k). \tag{7.11}
\]

For free fields, it is clear that \( \Pi = \mu \hat{A} \). Hence, we can also write \( \Pi_{\mu\sigma}(k) \) in the form of:

\[
\Pi_{\mu\sigma}(k) = -i\omega_{\mu\sigma}(k) \mu \hat{A}_{\mu\sigma}(k). \tag{7.12}
\]

The equation for the expansion coefficients \( \Lambda_{\mu\sigma}(k) \), is therefore:

\[
\sum_{\mu=0}^{N} \sum_{\sigma=1}^{2} \omega_{\mu\sigma}(k) \Lambda_{\mu\sigma}(k) \Lambda_{j\mu\sigma}(k) = \frac{\hbar}{2A\mu(2\pi)^n} \tilde{\delta}_{ij}(k). \tag{7.13}
\]

Next, we choose \( \Lambda_{\mu\sigma}(k) \) to be real, and as before, we can always choose an (unknown) function \( v_{\mu\sigma}(k) \) so that:

\[
\Lambda_{\mu\sigma}(k) = u_{\sigma}(k) \left[ \frac{h\nu_{\mu\sigma}(k)\epsilon_{\mu}(k)}{2A\mu(2\pi)^n} \right]^{1/2}. \tag{7.14}
\]

We will show later that \( v_{\mu\sigma}(k) \) must be interpreted as the electromagnetic component of group velocity, with non-electromagnetic phonon/exciton dispersion explicitly excluded. This is not completely identical to either the earlier narrow-band analysis [12] of this problem, or the simple one-dimensional theory. The difference can be attributed to the difference in the Hamiltonian energy that is introduced when the polarization fields are dispersive.

In order to demonstrate this, we first recall the standard identity that:

\[
\sum_{\sigma=1}^{2} u_{\sigma}(k)u_{j\sigma}(k) = \tilde{\delta}_{ij}(k). \tag{7.15}
\]

Next, substituting the above expression for \( \Lambda_{\mu\sigma}(k) \) into the equation for the field and mode commutators leads to:

\[
(I) \quad \sum_{\mu=0}^{N} \frac{k v_{\mu\sigma}(k)}{\omega_{\mu\sigma}(k)} = 1. \tag{7.16}
\]

As explained above, we have to determine a function \( v_{\mu\sigma}(k) \) which satisfies this condition, and we intend to demonstrate that the choice of \( v_{\mu\sigma}(k) \) as the (isotropic) electromagnetic component of group-velocity of the relevant polariton branch is sufficient - no other correction factors are needed. At this point, we notice an important fact; apart from the change in the resonance frequencies \( \Omega_{\nu}(k) \) due to their k-dependence, the above summation over \( \omega_{\mu\sigma}(k) \) is identical in analytic form to our one-dimensional expression. However, the k-dependence plays no role in obtaining the Cauchy theorem summation results, provided we define \( v_{\mu\sigma}(k) \) to be the same function of \( \Omega_{\nu}(k) \) and \( \omega_{\mu\sigma}(k) \) as before. In other words, the k-dependence of the phonon-exciton dispersion relation simply renormalizes the effective resonance frequency at each k-value, in the above summation over the roots of the dispersion relation. Since this renormalization is the same for each eigenvalue, the summation can be carried out using identical techniques to those used previously. Thus, we define, for \( \sigma = 1, 2 \):

\[
v_{\mu\sigma}(k) = \frac{\Omega_{\mu}(k)}{k} \left( 1 + \sum_{\nu} \frac{k^2 c^2 g_{\nu}}{(\Omega_{\nu}^2(k) - \omega_{\mu\sigma}(k)k^2)^2} \right)^{-1}. \tag{7.17}
\]

It should be noted that this \( v_{\mu\sigma}(k) \) is not the same as \( v_{\mu\nu}(k) \) in the one-dimensional case, although it has the same functional form. The difference is that it is a now a function of \( \Omega_{\nu}(k) \) and \( \omega_{\mu\sigma}(k) \), which include phonon/exciton dispersion effects. However, if we simply differentiate the slope of the function \( \omega_{\mu\sigma}(k) \), we do not obtain \( v_{\mu\sigma} \) as defined here - there are additional terms involving \( \partial \Omega_{\nu}^2(k)/\partial k \). For this reason, we refer to \( v_{\mu\sigma}(k) \) as the electromagnetic component of group velocity, which excludes additional transport terms.

It is clear that the mode function expansion of \( \hat{p}_{\nu}, \hat{\sigma}_{\nu} \) is also needed, for a complete demonstration of consistency. Using Maxwell's equations, if we expand \( \hat{p}_{\nu} \) as:
\[ \hat{p}_\nu = \sum_{\mu=0}^{N} \sum_{\sigma=0}^{2} \int d^3k \left[ p_{\mu\sigma}^{\nu}(k) \delta_{\mu\sigma}(k)e^{ik \cdot x - i \omega_{\mu\sigma}(k)t} + h.c. \right] , \quad (7.18) \]

then it follows that the expansion coefficient for the transverse component of \( p_{\mu\sigma} \) in the \( \mu \)-th frequency band must be:

\[ p_{\mu\sigma}^{\nu}(k) = \frac{ig_{\nu} k \times A_{\mu\sigma}(k)}{(\Omega_{\nu}^2(k) - \omega_{\mu\sigma}^2(k))} . \quad (7.19) \]

Similarly, if the canonical momentum for the atomic polarization field is:

\[ \hat{\pi}_\nu(t, x) = \sum_{\mu=0}^{N} \sum_{\sigma=0}^{2} \int d^3k \left[ \pi_{\mu\sigma}^{\nu}(k) \delta_{\mu\sigma}(k)e^{ik \cdot x - i \omega_{\mu\sigma}(k)t} + h.c. \right] , \quad (7.20) \]

then the corresponding transverse expansion coefficient is:

\[ \pi_{\mu\sigma}^{\nu}(k) = \frac{\omega_{\mu\sigma}(k) k \times A_{\mu\sigma}(k)}{\epsilon_0(\Omega_{\nu}^2(k) - \omega_{\mu\sigma}^2(k))} . \quad (7.21) \]

For these operators to have the correct equal-time commutators, the different oscillator position operators \( \hat{x} \) must commute amongst themselves at equal times, as must the different momentum operators \( \hat{p} \). This is trivial from the form of the mode operator expansion. However, the commutation relations (II) between the position and momentum operators are non-trivial. The relevant commutation conditions are therefore:

\[ [\hat{p}_{\mu}(x), \hat{\pi}_{\nu}(x')] = i\hbar \delta_{\mu\nu} \delta_{ij} \delta(x - x')/A = \sum_{\mu=0}^{N} \sum_{\sigma=0}^{2} \int d^3k \left[ p_{\mu\sigma}^{\nu}(k) \pi_{\nu\sigma}^{\mu}(k)e^{ik \cdot (x - x') - h.c.} \right] . \quad (7.22) \]

This in turn implies that:

\[ \sum_{\mu=0}^{N} \sum_{\sigma=0}^{2} p_{\mu\sigma}^{\nu}(k) \pi_{\nu\sigma}^{\mu}(k) = i\hbar \delta_{\mu\nu} \delta_{ij} \frac{e^{ik \cdot x}}{2A(2\pi)^n} . \quad (7.23) \]

Defining \( \delta_{ij}^{\parallel}(k) = k_i k_j / k^2 = \delta_{ij} - \delta_{ij}^{\perp}(k) \), and expanding the coefficients, gives two new equations. The transverse case is:

\[ \sum_{\mu=0}^{N} \omega_{\mu\sigma}(k)(\omega_{\nu\sigma}^2(k) - \Omega_{\nu\sigma}(k)) = \delta_{\nu\nu'} , \quad (7.24) \]

while the longitudinal equation is easily solved on defining \( u_{\mu}(k) = k/k \):

\[ p_{\mu0}^{\nu}(k) = u_{\mu}(k) \delta_{\mu\nu} \left[ \frac{1}{2A(2\pi)^n} \right]^{1/2} . \quad (7.25) \]

Finally, to ensure that there are correct field-atom commutators, we must satisfy the commutators III-VI. For these cross-terms between the oscillators and field variables, we find that conditions (III) and (IV), involving commutators between the field and the particle position (or the field momentum and particle momentum) are automatically satisfied. This occurs for the same reason that commutators like \([\hat{A}(x), \hat{A}(x')]\) or \([\hat{\pi}(x), \hat{\pi}(x')]\) must equal zero in our mode expansion. In all these cases involving pairs of canonical position-type operators or pairs of momentum-type operators, the commutator reduces to an odd integral over \( k \), which vanishes when integrated over all positive and negative \( k \)-values.

This leaves the requirements (V) and (VI), which are that \( \hat{\Delta} \) and \( \hat{\pi}_\nu \) must commute at equal times, as well as \( \hat{\pi} \) and \( \hat{p}_\nu \). These two requirements both imply the same restriction on the expansion coefficients, and hence on \( v_{\mu\sigma}(k) \), which is that for all \( k \) and \( \nu \) we must have the condition:

\[ \sum_{\mu=0}^{N} \frac{k v_{\mu\sigma}(k)}{\omega_{\mu\sigma}^2(k)(\omega_{\nu\sigma}^2(k) - \Omega_{\nu\sigma}^2(k))} = 0 . \quad (7.26) \]
Despite the complex nature of each of these conditions - which involve sums over all the roots of the dispersion equation, and must be satisfied for all the resonant frequencies $\omega_{\mu\sigma}$, as well as all momenta $k$ - we will show that each of these sums can be analytically evaluated without requiring an algebraic solution for the roots, just as before. For all of the commutation relation identities it is preferable to use techniques from complex function theory, which transform the sums over roots of the dispersion relation to complex contour integrals of related meromorphic functions. However, the transverse dispersion relations considered here have an identical analytic structure for a fixed $k$-value, with those in the one-dimensional case, so the previous analytic results follow without any further calculation. The main point here is that it is necessary for the ‘group-velocity’ coefficient to have the algebraic form given in Eq.(7.17) - which implies that it includes only part of the slope of the dispersion relation.

VIII. HAMILTONIAN

We now wish to show that when the Hamiltonian is expressed in terms of the operators $a_{\mu\sigma}(k)$ and $a_{\mu\sigma}^\dagger(k)$, $\mu = 0, \ldots, N$ and $\sigma = 0, 1, 2$, where the combination $\mu = \sigma = 0$ is omitted, it is of diagonal form. In order to prove this we shall study further the classical modes, in particular their orthogonality and normalization properties. Once this has been done, we shall be able to show that the Hamiltonian takes the form given in Eq. (7.6). Let us begin by restating the classical mode equations, Eqs. (10), in matrix form. Define the $3N + 3$ component vector $\vec{\lambda}$ by

$$\vec{\lambda} = \left( \begin{array}{c} \hat{A} \\ \hat{p}_{\nu_1} \\ \vdots \\ \hat{p}_{\nu_N} \end{array} \right), \quad (8.1)$$

or $\lambda_0 = \hat{A}$ and $\lambda_s = \hat{p}_{\nu_s}$ for $s \geq 1$, and the $3(N + 1) \times 3(N + 1)$ matrix $M(k)$ by

$$M(k) = \left( \begin{array}{cccc} k^2 c^2 I_3 & -ic^2 K & -ic^2 K & \ldots \\ -ig_{\nu_1} K & \Omega_{\nu_1}^2(k) I_3 & 0 & \ldots \\ -ig_{\nu_2} K & 0 & \Omega_{\nu_2}^2(k) I_3 & \ldots \\ \vdots & \vdots & \vdots & \ddots \end{array} \right). \quad (8.2)$$

Here $I_3$ is the $3 \times 3$ identity matrix and $K(k)$ is the anti-hermitian matrix given by

$$K = \left( \begin{array}{ccc} 0 & -k_3 & k_2 \\ k_3 & 0 & -k_1 \\ -k_2 & k_1 & 0 \end{array} \right), \quad (8.3)$$

which has the action on an arbitrary vector $A$

$$KA = k \times A. \quad (8.4)$$

The equations for the modes can now be expressed as (for each value of $k$)

$$M\vec{\lambda} = \omega^2 \vec{\lambda}. \quad (8.5)$$

The matrix $M$ is not hermitian, but if it is multiplied by the positive, diagonal, $3(N + 1) \times 3(N + 1)$ matrix $G$, 

$$G = \left( \begin{array}{cccc} I_3 & 0 & 0 & \ldots \\ 0 & \frac{c^2}{g_{\nu_1}} I_3 & 0 & \ldots \\ 0 & 0 & \frac{c^2}{g_{\nu_2}} I_3 & \ldots \\ \vdots & \vdots & \vdots & \ddots \end{array} \right), \quad (8.6)$$

then the combination $GM$ is hermitian. Therefore, if

$$M\vec{\lambda}^{(1)} = \omega_1^2 \vec{\lambda}^{(1)}, \quad M\vec{\lambda}^{(2)} = \omega_2^2 \vec{\lambda}^{(2)}, \quad (8.7)$$

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then
\[
\langle \lambda^{(2)} | G M \lambda^{(1)} \rangle = \omega_1^2 \langle \lambda^{(2)} | G \lambda^{(1)} \rangle
\]  
(8.8)
\[
= \langle G M \lambda^{(2)} | \lambda^{(1)} \rangle = \omega_2^2 \langle \lambda^{(2)} | G \lambda^{(1)} \rangle.
\]  
(8.9)

This implies that if \(\omega_1^2 \neq \omega_2^2\), then
\[
\langle \lambda^{(2)} | G \lambda^{(1)} \rangle = 0,
\]  
(8.10)
and we have part of the desired orthogonality relation.

In order to learn more we must examine the \(3N + 3\) eigenvectors in more detail. Define the projection operator, which projects each component of \(\lambda\) onto its longitudinal component,
\[
P(\hat{k}) = \left( \begin{array}{ccc} |\hat{k}\rangle \langle \hat{k}| & 0 & \cdots \\ 0 & |\hat{k}\rangle \langle \hat{k}| & \cdots \\ \vdots & \vdots & \ddots \end{array} \right),
\]  
(8.11)
where \(|\hat{k}\rangle \langle \hat{k}|\) is the projection onto the vector \(\hat{k}\). A short calculation shows that \([P(\hat{k}), M] = 0\) which implies that the eigenvectors \(\lambda\) can be taken to lie in either the subspace projected out by \(P(\hat{k})\) (longitudinal modes), or in the orthogonal subspace (transverse modes). The longitudinal modes can be found by taking the inner product of Eqs. (7.2) with \(\hat{\lambda}\) and we have part of the desired orthogonality relation.

We are now left with \(2N + 2\) transverse solutions. We first note that each transverse eigenvector is two-fold degenerate. This follows from the fact that if \(\lambda^{(2)}\) with \(\hat{\lambda}\) giving \(\langle \lambda^{(2)} | \Omega_\mu(\hat{k}) - \omega^2 \rangle \hat{k} \cdot \hat{p}_\nu = 0\).

There are \(N\) physical solutions to these equations given by \(\hat{\lambda} = 0\), \(\hat{p}_{\nu_m} \times \hat{k}\), and \(\hat{p}_\nu = 0\) for \(\nu \neq \nu_m\) with eigenvalue \(\Omega_\mu^{(2)}(k)\) for \(\mu = 1, \ldots, N\). There is also one unphysical solution (it violates the gauge condition) given by \(\hat{\lambda} \propto \hat{k}\) and all of the \(\hat{p}_{\nu}\) being equal to zero.

We are now left with \(2N + 2\) transverse solutions. We first note that each transverse eigenvalue is two-fold degenerate. This follows from the fact that if \(M \lambda = \omega^2 \lambda\), then \(M(K_{3N} \lambda) = \omega^2(K_{3N} \lambda)\), where
\[
K_{3N} = \left( \begin{array}{ccc} K & 0 & \cdots \\ 0 & K & \cdots \\ \vdots & \vdots & \ddots \end{array} \right),
\]  
(8.13)
which can be verified by noting that if \(\hat{\lambda}\) and \(\hat{p}_\nu\) satisfy Eqs. (7.2), so do \(\hat{k} \times \hat{\lambda}\) and \(\hat{k} \times \hat{p}_\nu\). We choose the two eigenvectors \(\lambda^{(1)}\) and \(\lambda^{(2)}\), which correspond to the eigenvalue \(\omega_\mu^2\), to be orthogonal in the sense that
\[
\langle \lambda^{(1)} | G \lambda^{(2)} \rangle = 0.
\]  
(8.14)
This, along with Eq. (8.10) implies that
\[
\langle \lambda^{(\mu\sigma)} | G \lambda^{(\mu'\sigma')} \rangle = \delta_{\mu\mu'} \delta_{\sigma\sigma'},
\]  
(8.15)
which is our final orthonormality relation. Here, due to isotropy, the mode frequency \(\omega_{\mu\sigma}\) does not depend on the polarization index, \(\sigma\), for the transverse modes.

We now express the fields in terms of the eigenvectors, substitute them into the Hamiltonian density in Eq. (8.8), and integrate over the relevant \(n\)-dimensional volume. In particular, we have that
\[
\lambda^{(\mu\sigma)}_0 = \Lambda_{\mu\sigma}, \quad \lambda^{(\mu\sigma)}_i = p^{\nu}_{\mu\sigma},
\]  
(8.16)
\[
\Pi_{\mu\sigma} = -i \omega_{\mu\sigma} \Lambda_{\mu\sigma}, \quad \pi^{\nu}_{\mu\sigma} = -i \omega_{\mu\sigma} \lambda^{(\mu\sigma)}_i.
\]  
(8.17)
Adding the requirement that \(\lambda^{(\mu\sigma)}(k)^* = \lambda^{(\mu\sigma)}(-k)\) (it can be shown that \(M(-k)\lambda^{(\mu\sigma)}(k)^* = \omega^2(k)\lambda^{(\mu\sigma)}(k)^*\) which implies that \(\lambda^{(\mu\sigma)}(k)^*\) is in the two-dimensional subspace spanned by \(\lambda^{(\mu\sigma)}(-k)\) for \(\sigma = 1, 2\) and utilizing Eq. (8.15), we find that the terms of the form \(\tilde{a}_{\mu\sigma}(k)\tilde{a}_{\mu'\sigma'}(-k)\) vanish giving for the transverse modes
\[
H_{\text{trans}} = 2(2\pi)^n \mu A \sum_{\mu=0}^{N} \sum_{\sigma=1}^{2} \int d^3k \langle \chi(\mu\sigma) (k) | G \chi(\mu\sigma) (k) \rangle \omega^2_\mu (k) \hat{a}^\dagger_{\mu\sigma} (k) \hat{a}_{\mu\sigma} (k).
\] (8.18)

In order to show that the Hamiltonian assumes the form given in Eq. (7.6) we need to prove that
\[
2(2\pi)^n \mu A \langle \chi(\mu\sigma) (k) | G \chi(\mu\sigma) (k) \rangle \omega_\mu (k) = \hbar.
\] (8.19)

Making use of Eqs. (7.14) and (7.19) this condition becomes
\[
\left( 1 + \sum_\nu \frac{k^2 c^2 g_\nu}{(\Omega^2_\nu (k) - \omega^2_\mu)^2} \right) \frac{k v_{\mu\sigma}}{\Omega_\mu} = 1.
\] (8.20)

This agrees precisely with Eq. (7.17) and is true even including phonon dispersion \((\alpha_\nu \neq 0)\). However, as pointed out earlier, when there is phonon dispersion we cannot interpret \(v_{\mu\sigma}\) as the total group velocity - it only includes an electromagnetic contribution, i.e., it is no longer equal to \(\partial \omega / \partial k\).

In order to complete the diagonalization of the Hamiltonian we must consider the longitudinal modes. The fields \(\Lambda\) and \(\Pi\) have no longitudinal components so that this part of the diagonalization procedure involves only the fields \(p_\nu\) and \(\pi_\nu\). We find that
\[
H_{\text{long}} = (2\pi)^n \frac{2A}{\epsilon_0} \sum_{\mu=1}^{N} \int d^3k \frac{\omega^2_\mu (k)}{g_\mu} p^\mu_{\mu0} (k)^* \cdot p^\mu_{\mu0} (k) \hat{a}^\dagger_{\mu0} \hat{a}_{\mu0} (k),
\] (8.21)

where we have made use of the fact that \(p^\nu_{\mu0} (k) = \delta_{\mu\nu} p^\nu_{\mu0} (k)\), and we note that \(\omega_{\mu0} (k) = \Omega_\mu (k)\). The Hamiltonian assumes the expected form,
\[
H_{\text{long}} = \sum_{\mu=1}^{N} \int d^3k \hbar \omega_{\mu0} (k) \hat{a}^\dagger_{\mu0} \hat{a}_{\mu0} (k),
\] (8.22)

when explicit expressions for the vectors, \(p^\mu_{\mu0} (k)\) from Eq. (7.22) are used.

The final Hamiltonian, which is the sum of \(H_{\text{trans}}\) and \(H_{\text{long}}\), has \((3N+2)\) mode operators for each value of \(k\), and can be written in the form
\[
H = \sum_{\mu=0}^{N} \sum_{\sigma=0}^{2} \int \hbar \omega_{\mu\sigma} (k) \hat{a}^\dagger_{\mu\sigma} (k) \hat{a}_{\mu\sigma} (k) d^3k.
\] (8.23)

Here the lower limit \(\sigma = 0'\) excludes the combination of \(\mu = 0\) and \(\sigma = 0\), which would imply a longitudinal polariton. Also, there is a requirement of having \((N+1)\) distinct roots for this form to be valid. The corresponding field operators (in the full three-dimensional case) have the expansions:
\[
\hat{D} = i \sum_{\mu, \sigma=1, 2} \int d^3k \left[ \frac{\hbar k v^E_{\mu} (k) \varepsilon (\omega_{\mu} (k))}{4\pi} \right]^{1/2} e_\sigma (k) \hat{a}_{\mu\sigma} (k) e^{ik \cdot x} + hc
\]
\[
\hat{E}^+ = i \sum_{\mu, \sigma=1, 2} \int d^3k \left[ \frac{\hbar k v^E_{\mu} (k)}{4\pi \varepsilon (\omega_{\mu} (k))} \right]^{1/2} e_\sigma (k) \hat{a}_{\mu\sigma} (k) e^{ik \cdot x} + hc
\]
\[
\hat{B} = -i \sum_{\mu, \sigma=1, 2} \int d^3k \left[ \frac{\hbar k v^E_{\mu} (k)}{4\pi} \right]^{1/2} u_\sigma (k) \hat{a}_{\mu\sigma} (k) e^{ik \cdot x} + hc.
\] (8.24)

Here we have introduced the electric field mode \(e (k) = k \times u (k) / |k|\) to simplify the expansion. We note, as in the one-dimensional case, that the transverse field expansion for the electric field is simply derived from the displacement field by using the frequency-dependent permittivity. The main feature introduced by the dispersion is the replacement of a frequency term \(\omega\), that would normally appear in the expansion coefficients, by a new term with the same units, but equal to \(k v^E_{\mu} (k)\) instead. We do not give the expansion for the longitudinal part of the electric field here explicitly, except to point out that it is equal to \(-P^\parallel / \varepsilon_0\).
A simple theory of a one-dimensional, dispersive waveguide was introduced, including a polarizable model of the medium with \( N \) discrete localized resonances. This can be thought of as a limiting case of an ideal insulator, in which the polarization field is due to localized electrons at each atomic location. The theory is exactly equivalent to the usual classical theory of a dispersive dielectric medium, in the sense that it results in the Sellmeir equations for the refractive index. These are well-known to lead to an excellent fit to the classical dispersion properties of transparent media, and have the usual causality requirements automatically satisfied. The theory was quantized and a set of \( N + 1 \) mode operators introduced, for the polaritons in each branch of the dispersion relation, provided there were \( N + 1 \) distinct, positive roots. In this case, the mode-expansion has a universal and simple form, only depending on the group velocity.

This model is necessarily causal, and implements the causality requirements through band-gaps, rather than isolated poles. It does omit many important correction factors that occur in practice. In particular, our mode expansion neglects scattering off inhomogeneities. For this reason, transmission inside the transmission band is essentially lossless. It also omits nonlinearities due to phonon-phonon, photon-photon, and photon-phonon interactions, which are responsible for additional non-electromagnetic damping of the polaritons. However, these effects can certainly be added to the Hamiltonian once a mode expansion is established.

Next, a quantum theory of an isotropic \( n \)-dimensional dispersive waveguide was introduced, with \( n = 1, 2 \) or 3. Without any additional phonon/exciton dispersion, the theory is exactly equivalent to the classical Drude-Lorentz theory of a dispersive dielectric medium. The complete \( n \)-dimensional theory was quantized, and a set of \( (3N + 2) \) mode operators introduced, for each branch of the dispersion relation; again, with the restriction of distinct, positive roots. As in the one-dimensional case, the mode expansion depends on the permittivity and the electromagnetic group velocity, in the case of transverse polaritons. However, the group velocity factor in this case is modified to include only the electromagnetic component of the group velocity.

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