Hamiltonian treatment of the electromagnetic field in dispersive and absorptive structured media

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(Received 18 February 2006; published 7 June 2006)

We introduce a Hamiltonian formulation of electromagnetic fields in dispersive and absorptive structured media of arbitrary dimensionality; the Kramers-Kronig relations are satisfied by construction. Our method is based on an identification of the photonic component of the polariton modes of the system. Although the medium degrees of freedom are introduced in an oscillator model, only the susceptibility of the medium appears in the derived eigenvalue equation for the polaritons; the theory is applicable to both classical and quantum optics. A discrete polariton spectrum is obtained in the transparent regime below the absorption cutoff frequency, and the normalization condition contains the material dispersion in a simple way. In the absorptive regime, a continuous polariton spectrum is obtained. The expressions for the full electromagnetic field of the system can be written in terms of the modes of a limiting, non dispersive, nonabsorptive system, so the theory is well suited to studying the effect of dispersion or absorption on photonic dispersion relations and mode structure. Available codes for dispersive photonic modes can easily be leveraged to obtain polariton modes in both the transparent and absorptive regimes.

DOI: 10.1103/PhysRevA.73.063808 PACS number(s): 42.50.Nn, 42.70.Qs, 71.36.+c, 42.50.Ct

I. INTRODUCTION

Dispersion and absorption have become increasingly important in the study of structured media. Many applications, including those involving photovoltaic systems, ultrahigh-efficiency incandescent lighting systems [1,2], few-photon switches [3], low threshold microlasers [4], and infrared emission suppression [5] specifically leverage the absorptive and dispersive properties of host media.

For other proposed applications, absorption and dispersion are unavoidable effects that complicate an idealized system. Many of these proposals need to be evaluated and refined in light of realistic models of material absorption and dispersion. Some such proposed applications involve the classical and quantum states of light in high $Q$ defect cavities [6–9], and the propagation and generation of entangled photon states in dielectric systems [10–14]. It has been argued that a better understanding of the dynamics of few-photon wave packets in dispersive media [15] is essential to advances in certain practical quantum information systems, and it has been shown through numerical [16] and analytical [17] investigations that photonic band gaps can be significantly altered or even closed by very small material absorption.

From a materials standpoint, a significant class of interesting systems cannot even be described qualitatively without explicitly accounting for absorption and dispersion. For example, metallo-dielectric (plasmonic) nanophotonic media [18] are inherently absorptive and highly dispersive, as are media doped with atoms with excitation energies near photon energies of interest. The same holds for photonic crystals in which the photonic and electronic band gaps are in close proximity. Inherently dispersive and absorptive structured dielectric systems have also shown the promise of rich physics, both in exciton-polaritonic [19,20] and phonon-polaritonic phononic crystals [21], although most analyses presented to date have relied on simple, model dielectric response functions.

In each of the above-mentioned cases, the underlying physical description at some level must involve a Hamiltonian formulation of the electromagnetic field in the presence of dispersive and absorptive materials satisfying the Kramers-Kronig relations.

While the problem of quantization in nondispersive, nonabsorptive dielectrics has been well studied since the early work of Jauch and Watson [22], for dispersive and absorptive Kramers-Kronig dielectrics or metals the problem is significantly more complicated. The medium is introduced through a susceptibility $\chi^{(1)}(r,t)$, which is known either from experimental measurements or from ab initio calculations. The fundamental difficulty is as follows. Any Hamiltonian, being a state functional, involves dynamical variables at a single given time; however, dispersive response functions imply the medium polarization involves the electromagnetic fields at all previous times, so any naïve attempt to introduce the medium polarization into the Hamiltonian is doomed to failure. The response function simply aggregates too much information about the underlying medium to allow such a Hamiltonian formalism. The solution is the reintroduction of dynamical variables representing the material degrees of freedom. This has been accomplished by various methods [23], all with their own strengths and limitations. Huttnner and Barnett [24,25] introduced the medium as a Hopfield model [26], and were able to construct a Hamiltonian formulation for uniform dielectrics with dispersive and absorptive response. In a Langevin method pioneered by Matloob and coworkers, and furthered by Gruner and Welsch [27,28], the medium is introduced as phenomenological noise currents in the Maxwell equations, and the canonical operators of the theory are noise current operators. However, the electromagnetic fields are obtained only indirectly from the noise current operators via the Green function. More recently, Tip [29–31] introduced an auxiliary field method, in which two fields $F_2$ and $F_4$ are coupled to the usual electromagnetic fields of the Maxwell equations; the drawback is that the coupling and free-field dynamics of the auxiliary fields are postulated ad hoc, so intuition into the resulting physics is
more difficult. Both the Langevin and auxiliary field methods have been applied to inhomogeneous media [32,29]. The Huttner-Barnett approach has been applied to quantization of a dielectric half space [33], and it has been suggested that the scheme would become quite cumbersome to apply to non-uniform media [34].

However, here we present just such a scheme, and demonstrate an approach with an end result that is both elegant and easy to use. Instead of relying on a Green-function representation of light in the system, we focus directly on the dispersion relation (photonic band structure) and electromagnetic modes. Since the majority of studies of artificially structured photonic media are formulated in terms of mode and band-structure concepts, the approach here connects more readily with that literature than do others.

Our strategy is as follows. The medium response is introduced via a Hopfield-type model [26] as an oscillator field, which is well justified in the linear excitation regime [35]. The full dispersive and absorptive dielectric response of the system, \( \chi^{(1)}(r,\tau) \), is generated implicitly by the dynamics of the coupled electromagnetic and model medium fields. The dielectric function is easily understood in terms of the model, with dispersion being due to virtual coupling and absorption due to direct coupling. This system is Hamiltonian, and is quantized in a straightforward manner. The polaritons, which are the dressed modes of the system, are obtained through a Fano-type diagonalization procedure and have a natural interpretation. Our theory is applicable to inhomogeneous media in general, and we formulate it in terms of a “nominal” nondispersive and nonabsorptive system with index of refraction \( n(r) \). While this intermediate step is not necessary—the nominal system can always be taken to be vacuum—it should make our approach particularly useful in the design and analysis of dispersive and absorptive metamaterials for which the practitioner has good intuition for the mode structure of the nominal system.

In fact, since our method is based on describing the dynamics in terms of modes, it has practical applicability to any medium or artificially structured material that is best understood in the nondispersive, nonabsorptive limit in terms of modes; this includes, for example, waveguides, multilayer systems, photonic crystals, coupled microring resonators [36], other resonator systems [37,38], and plasmon nanowire structures [39,40].

The possible applications of this theory lie both in the classical and quantum domains. To this end, we have written everything so as to apply to both. We formally write commutators instead of Poisson brackets, but recall the correspondence

\[
\frac{1}{i\hbar} [\ldots, \ldots] \rightarrow \{\ldots, \ldots\},
\]

which yields the associated classical Poisson brackets and classical equations. Similarly, we use \( \dagger \) to denote the adjoint or complex conjugate. Since no commutators appear in any of the final equations, the results are equally applicable to the classical and quantum domains, requiring only that the variables be interpreted appropriately as amplitudes or Heisenberg operators.

The paper is structured as follows. In Sec. II we introduce our canonical Hamiltonian model for dispersive and absorptive dielectrics in terms of electromagnetic fields coupled to model oscillator fields. In Sec. III we describe the dielectric response of the model. In Sec. IV we first diagonalize the Hamiltonian for the dispersive and absorptive case, obtaining a generalized Hermitian eigenvalue problem for the polaritons, which exist at all frequencies in the absorption band. Subsequently, we perform the diagonalization for the nonabsorptive but dispersive frequency regime, in which there is a discrete spectrum of polaritons; these polaritons consist of both an electromagnetic component, which is just the usual dispersive electromagnetic mode, and a medium component, which is slaved to the electromagnetic component. In Sec. V we give the equations for systems with continuously labeled modes, choosing photonic crystals as a specific example. Section VI gives a summary and user’s guide, and provides an overview of how to apply the end results of this paper to straightforwardly calculate polariton modes within a Hamiltonian framework for systems of interest; examples are presented for both the transparent and absorptive frequency regimes. The reader may wish to turn immediately to Sec. VI for sense of what we accomplish here, and of the nature of the formalism we produce. In Sec. VII we give a discussion of our main results and conclusions. The appendixes give technical results and protracted calculations which would otherwise impede the flow of the text.

II. HAMILTONIAN FOR DISPERSIVE AND ABSORPTIVE MEDIA

Maxwell’s curl equations for dielectric media in the absence of free charges and currents are

\[
\dot{\mathbf{D}} = \nabla \times \mathbf{H}, \quad \dot{\mathbf{B}} = -\nabla \times \mathbf{E}.
\]

(1)

The divergence equations,

\[
\nabla \cdot \mathbf{D} = 0, \quad \nabla \cdot \mathbf{B} = 0,
\]

(2)

are taken as initial conditions; if the initial fields satisfy Eq. (2), then evolution according to Eq. (1) ensures the fields will satisfy Eq. [2] for all time. However, it is important to note that approximations of Eq. (1) often do not maintain the conditions (2) upon evolution. To ensure the divergencelessness of \( \mathbf{D} \) and \( \mathbf{B} \) even after making approximations, we take \( \mathbf{D} \) and \( \mathbf{B} \) fields as our primary fields, and consider \( \mathbf{E} \) and \( \mathbf{H} \) as derived fields. This allows us to restrict the possible space of \( \mathbf{D} \) and \( \mathbf{B} \) fields to those that satisfy Eq. (2). In practice this can be simply done, for example, by using a mode expansion in divergenceless fields. The choice of \( \mathbf{D} \) and \( \mathbf{B} \) as fundamental fields has its roots in the work of Born and Infeld [41]; Bialynicki-Birula and Bialynicka-Birula discuss it in some detail in their text [42].

The Maxwell equations require constitutive relations. Neglecting magnetic effects we have
\[ \mathbf{B}(\mathbf{r}, t) = \mu_0 \mathbf{H}(\mathbf{r}, t), \]
\[ \mathbf{D}(\mathbf{r}, t) = \varepsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}(\mathbf{r}, t), \quad (3) \]

where the dielectric response is typically specified by a relation between the polarization and the electric field. However, since we take \( \mathbf{D} \) and \( \mathbf{B} \) as our primary fields, we write \( \mathbf{P} \) in terms of (in general a functional of) \( \mathbf{D} \). There is clearly no loss of generality here, since any relation of the first type can be converted into a relation of the second type by the use of the second of Eq. (3).

Sometimes dispersive and absorptive effects are negligible, and over the frequency range of interest the optical response can be characterized by a nondispersive, nonabsorptive response of the form
\[ \mathbf{P}(\mathbf{r}, t) = \Gamma_{\mathbf{D}}^0(\mathbf{r}) \mathbf{D}(\mathbf{r}, t), \quad (4) \]

which, nonetheless, allows for material inhomogeneities. Superscript Roman indices indicate Cartesian components, which, nonetheless, allows for material inhomogeneities. The special case of isotropic media, the response function \( \Gamma_{\mathbf{D}}^0(\mathbf{r}) \) is related to the usual (local) index of refraction \( n(\mathbf{r}) \) by
\[ \Gamma_{\mathbf{D}}^0(\mathbf{r}) = \left( 1 - \frac{1}{n^2(\mathbf{r})} \right) \delta_{ij}, \quad (5) \]

where \( \delta_{ij} \) is the Kronecker delta. Equation (5) can be confirmed by using Eqs. (3) and (4) with Eq. (5) in Eq. (1), and comparing with the usual equations in this description of the optical response.

To include the effects of absorption and dispersion we need a treatment of the medium response more general than Eq. (4). Our goal here is to model a very general linear response of the form
\[ \mathbf{P}(\mathbf{r}, t) = \int ds \Gamma^0(\mathbf{r}, t-s) \mathbf{D}(\mathbf{r}, s), \quad (6) \]

where the constraint of causality requires that \( \Gamma^0(\mathbf{r}, \tau)=0 \) for all \( \tau<0 \). The response (6) is still of course local in space (i.e., we neglect spatial dispersion effects [43]), and we neglect any magnetic aspects of the response by assuming
\[ \Gamma^0(\mathbf{r}, t) = \Gamma^0(\mathbf{r}, t). \quad (7) \]

Other than these constraints, we allow for a \( \Gamma^0(\mathbf{r}, t) \) that is freely specified, either from experimental data or on the basis of a microscopic calculation of the material response. It is the dynamics described by the system of equations (1)–(3), (6), and (7) that we wish to generate from a Hamiltonian.

In many treatments, complications arise because the route to canonical formulation begins with a Lagrangian. However, this is not necessary. Even in the canonical formulation of a classical system for ultimate use in quantization, one is only required to provide a set of commutators (or Poisson brackets) and a Hamiltonian such that their use leads in the usual way to the desired dynamical equations, and such that the numerical value of the Hamiltonian in the classical theory is equal to the energy [44]. Earlier we introduced such an approach for nondispersive and nonabsorptive media [45].

However, the kind of very general response (6) we consider here requires for its description the inclusion of medium degrees of freedom. We take as our Hamiltonian
\[ \mathbf{H} = \mathbf{H}_{\text{em}} + \mathbf{H}_{\text{med}} + \mathbf{H}_{\text{int}}, \quad (8) \]

where \( \mathbf{H}_{\text{em}} \) is the Hamiltonian of the electromagnetic field in vacuum, \( \mathbf{H}_{\text{med}} \) is the Hamiltonian of the medium, and \( \mathbf{H}_{\text{int}} \) describes the interaction between the vacuum electromagnetic field and the medium. One route to determining \( \mathbf{H}_{\text{med}} \) and \( \mathbf{H}_{\text{int}} \) would be to rely on a microscopic calculation of the actual material medium of interest. Our approach here adopts a different philosophy. For a \( \Gamma^0(\mathbf{r}, t) \) assumed given to us, we construct effective Hamiltonians \( \mathbf{H}_{\text{med}} \) and \( \mathbf{H}_{\text{int}} \) that lead to the response (6) and are convenient for later calculations. Thus we provide formal Hamiltonians \( \mathbf{H}_{\text{med}} \) and \( \mathbf{H}_{\text{int}} \) rather than the actual ones.

### A. Vacuum electromagnetic Hamiltonian

For the vacuum electromagnetic Hamiltonian we take
\[ \mathbf{H}_{\text{em}} = \frac{1}{2\mu_0} \int d\mathbf{r} \mathbf{B}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}) + \frac{1}{2\varepsilon_0} \int d\mathbf{r} \mathbf{D}(\mathbf{r}) \cdot \mathbf{D}(\mathbf{r}). \quad (9) \]

In conjunction with the equal-time commutators (or alternatively Poisson brackets)
\[ [\mathbf{D}(\mathbf{r}), \mathbf{D}(\mathbf{r}')] = [\mathbf{B}(\mathbf{r}), \mathbf{B}(\mathbf{r}')] = 0, \]
\[ [\mathbf{D}(\mathbf{r}), \mathbf{B}(\mathbf{r}')] = i\hbar \varepsilon^{ij} \frac{\partial}{\partial \mathbf{r}^j} [\delta(\mathbf{r} - \mathbf{r}')] \quad (10) \]
where \( \varepsilon^{ij} \) is the Levi-Civita symbol (\( \varepsilon^{123} = -\varepsilon^{231} = 1 \), etc.), Hamilton’s equations
\[ i\hbar \frac{\partial \mathbf{D}}{\partial t} = [\mathbf{D}, \mathbf{H}], \]
\[ i\hbar \frac{\partial \mathbf{B}}{\partial t} = [\mathbf{B}, \mathbf{H}], \quad (11) \]

with \( \mathbf{H} = \mathbf{H}_{\text{em}} \) yield the Maxwell equations (1) subject to the constitutive relations (3), with \( P(\mathbf{r}, t)=0 \), i.e., the dynamics for the electromagnetic field in vacuum.

### B. Medium and coupling Hamiltonians

We introduce dynamical variables for the medium, and a medium Hamiltonian, in such a way that the addition of coupling to the electromagnetic field results in equations that lead to the desired dispersive and absorptive response. As many workers have realized, harmonic oscillator fields are appropriate for the description of linear excitations of a medium [26,35]. Following the Hopfield model [26], Huttner and Barnett [24] considered optical absorption as resulting from coupling of the electromagnetic field to a discrete oscillator field, with the discrete oscillator field coupled to continuum reservoir fields. Diagonalization of their medium Hamiltonian alone yields a set of dressed continuum fields.
that are coupled to the vacuum electromagnetic field. Thus there is a mathematical equivalence between the Hopfield model and the modified model excluding the discrete oscillator as an intermediary between the vacuum electromagnetic and reservoir fields. We are more concerned here with developing a general approach for use in problems in optics, rather than describing any purported model of actual material response by a discrete oscillator field, or the details of the two-step diagonalization that was the focus of earlier workers. Thus we take a simpler model, involving the direct coupling of vacuum electromagnetic modes to a continuum of oscillator fields. We emphasize that while the two-step coupling is often adopted, it is certainly not needed to model the response of physical systems. Given the dielectric response function \( \Gamma^{ij}(\mathbf{r}, t) \) [see, e.g., Eq. (6)], we can immediately produce a model medium of our type that generates it. This is detailed in Sec. III.

To describe a large class of media, we require medium harmonic oscillators at (possibly) all frequencies. Since we neglect spatial dispersion effects, the harmonic oscillator modes can be identified with individual points in space. In our approach, the coupling between a material excitation at \( \mathbf{r} \) and the electromagnetic field is encapsulated in a position- and frequency-dependent coupling tensor \( \Lambda^{ij}(\mathbf{r}, \Omega) \), which is real and satisfies \( \Lambda^{ij}(\mathbf{r}, \Omega) = \Lambda^{ji}(\mathbf{r}, \Omega) \). Furthermore, at each \( (\mathbf{r}, \Omega) \), the coupling tensor \( \Lambda^{ij}(\mathbf{r}, \Omega) \) we introduce shares the same set of principal axes as the frequency-domain dielectric response tensor \( \Gamma^{ij}(\mathbf{r}, \Omega) \). Any material dielectric response can be described by the oscillator model introduced here, with a flat density of oscillator states above a minimum cut-off frequency \( \Omega_{\text{c}} \), which may be zero; effects of the density of states in the actual material system can be described through the frequency dependence of the effective coupling constant \( \Lambda^{ij}(\mathbf{r}, \Omega) \), as is shown in Sec. III.

We describe the dynamics of our model medium by a coordinate field \( \mathbf{X}_{\Omega}(\mathbf{r}, t) \) and its conjugate momentum field \( \Pi_{\Omega}(\mathbf{r}, t) \). To avoid introducing superfluous components, two steps are required. For a large class of structured media at frequencies of interest, there is absorption at some positions but not at others; to avoid introducing uncoupled oscillators, we define a (possibly multiply connected) region of space \( \mathcal{V}_0 \) that identifies the region over which the medium has nonzero absorption at frequency \( \Omega \). Then oscillators are only introduced for \( (\mathbf{r}, \Omega) \) such that \( \mathbf{r} \in \mathcal{V}_0 \).

Furthermore, even in regions with absorption, all three vector components of an oscillator are not necessarily required, because at any given position \( \mathbf{r} \) the dielectric response \( \Gamma^{ij}(\mathbf{r}, \Omega) \)—and therefore the real, symmetric coupling tensor \( \Lambda^{ij}(\mathbf{r}, \Omega) \)—may have one or more vanishing eigenvalues at a given frequency \( \Omega \). For each position \( \mathbf{r} \) and frequency \( \Omega \) we determine the local principal axes \( \{ \mathbf{\xi}_{\mathbf{r}}(\mathbf{r}, \Omega), \mathbf{\zeta}_{\mathbf{r}}(\mathbf{r}, \Omega), \mathbf{\zeta}_{\mathbf{r}}(\mathbf{r}, \Omega) \} \) of the tensor \( \Gamma^{ij}(\mathbf{r}, \Omega) \), and then write the coupling tensor \( \Lambda^{ij}(\mathbf{r}, \Omega) \) in that position- and frequency-dependent basis. Throughout the paper, we use \( \alpha \) and \( \alpha' \) to refer to the components in the basis of the local principal axes, reserving \( i \) and \( j \) to refer to Cartesian components. Then

\[
\Lambda^{\alpha\alpha'}(\mathbf{r}, \Omega) = \Lambda^{\alpha\alpha'}(\mathbf{r}, \Omega) \delta^{\alpha\alpha'} = \Lambda^{\alpha}(\mathbf{r}, \Omega) \delta^{\alpha\alpha'},
\]

where here no summation is taken over repeated indices. The medium fields are then introduced in reference to the local principal axes. For all \( \lambda \), if \( \Lambda^{\alpha}(\mathbf{r}, \Omega) \neq 0 \), we introduce a local oscillator polarized along \( \mathbf{\xi}_{\mathbf{r}}(\mathbf{r}, \Omega) \), and write the associated coordinate field component as \( X_{\Omega}^{\alpha}(\mathbf{r}, t) \). Of course, the tensors written in the basis of the local principal axes can be rewritten in a global Cartesian basis via an appropriate transformation at each \( (\mathbf{r}, \Omega) \), and vice versa. Throughout the paper, we simply write \( \mathbf{X}_{\Omega}(\mathbf{r}, t) \), and recall that \( \mathbf{X}_{\Omega}(\mathbf{r}, t) \) has 0, 1, 2, or 3 nonvanishing components as required; we revert to the more detailed notation when necessary. Similarly, we write the coupling tensor as \( \Lambda(\mathbf{r}, \Omega) \). For notational simplicity, we also typically refrain from indicating the time-dependence of the medium fields, and instead write \( \mathbf{X}_{\Omega}(\mathbf{r}) \) and \( \Pi_{\Omega}(\mathbf{r}) \).

The Hamiltonian of the model medium is then

\[
H_{\text{med}} = \int_{\Omega_{\text{c}}}^{\infty} d\Omega \int_{\mathcal{V}_0} d\mathbf{r} \frac{1}{2} \left[ \Pi_{\Omega}(\mathbf{r}) \cdot \Pi_{\Omega}(\mathbf{r}) + \Omega^2 \mathbf{X}_{\Omega}(\mathbf{r}) \cdot \mathbf{X}_{\Omega}(\mathbf{r}) \right],
\]

which yields harmonic oscillator field dynamics when we impose the equal-time commutator

\[
[X_{\Omega}^{\alpha}(\mathbf{r}), \Pi_{\Omega}^{\alpha'}(\mathbf{r'})] = i\hbar \delta_{\alpha\alpha'} \delta(\mathbf{r} - \mathbf{r'}) \delta(\Omega - \Omega'),
\]

with all other commutators (Poisson brackets) vanishing. The association of each medium oscillator with a point in space is apparent in the Hamiltonian (13); were there no coupling with the electromagnetic field, the dynamics of oscillators at different field points \( \mathbf{r} \) would be completely independent of each other. Making the change of variables

\[
\psi_{\Omega}(\mathbf{r}) = \sqrt{\frac{1}{2\hbar\Omega}} \left[ \Omega \mathbf{X}_{\Omega}(\mathbf{r}) + i\Pi_{\Omega}(\mathbf{r}) \right],
\]

and neglecting the zero point energy in the quantum case, we obtain the form

\[
H_{\text{med}} = \int_{\Omega_{\text{c}}}^{\infty} d\Omega \int_{\mathcal{V}_0} d\mathbf{r} \hbar \Omega \psi_{\Omega}(\mathbf{r}) \cdot \psi_{\Omega}(\mathbf{r}),
\]

and the equal-time commutation relation

\[
[\psi_{\Omega}^{\dagger}(\mathbf{r}), \psi_{\Omega}^{\dagger}(\mathbf{r'})] = \delta_{\mathbf{r}\mathbf{r'}} \delta(\Omega - \Omega'),
\]

with all other equal-time commutators vanishing.

We now turn to the coupling Hamiltonian, which we take to be

\[
H_{\text{int}} = -\frac{1}{\sqrt{2\hbar\Omega}} \int_{\Omega_{\text{c}}}^{\infty} d\Omega \int_{\mathcal{V}_0} d\mathbf{r} \mathbf{D}(\mathbf{r}) \cdot \Lambda(\mathbf{r}, \Omega) \cdot \mathbf{X}_{\Omega}(\mathbf{r}).
\]

Using this Hamiltonian with \( H_{\text{en}} \) (9) and \( H_{\text{med}} \) (16) to form the total Hamiltonian \( H \) (8), Hamilton’s equations (11), evaluated with the commutators (10), yield the Maxwell equations (1) subject to the constitutive relations (3), with
\[ P(r, t) = \sqrt{\varepsilon_0} \int_{\Omega_x}^{\infty} d\Omega \lambda(r, \Omega) \cdot X_{\Omega}(r, t). \]  
(19)

Thus we identify the polarization due to the medium fields. Whereas the dispersive constitutive relation involves the displacement fields at all previous times, Eq. (19) involves the medium fields only at a single time, as is required to build a Hamiltonian formulation.

### III. DIELECTRIC RESPONSE OF THE MODEL

Nonetheless, we can now identify the dielectric response function \( \Gamma^\mu(r, t) \) generated by this model. To do this we use Hamilton’s equation with the full Hamiltonian (8), together with the medium field commutation relations (14), to find the dynamical equations for the medium fields:

\[ \dot{X}_{\Omega}(r, t) = \Pi_{\Omega}(r, t), \]  
(20)
\[ \Pi_{\Omega}(r, t) = -\Omega^2 X_{\Omega}(r, t) + \varepsilon_0 \partial^2 \lambda(r, \Omega) \cdot D(r, t). \]  
(21)

Combining these equations yields a second-order equation for \( X_{\Omega}(r, t) \), driven by the displacement field. To allow for Fourier decomposition even on resonance, we allow the free oscillation of the medium fields to be infinitesimally damped in the usual way [46], and with the understanding that at the end of the calculation we take \( \eta \to 0^+ \), we modify the second-order equation resulting from Eqs. (20) and (21) to read

\[ \left[ \partial_t^2 + 2 \partial_t + (\partial^2 + \eta^2) \right] X_{\Omega}(r, t) = \varepsilon_0 \partial^2 \lambda(r, \Omega) \cdot D(r, t). \]  
(22)

Then inverting the differential operator on the left-hand side [48] yields the solution

\[ X_{\Omega}(r, t) = \frac{1}{\sqrt{\varepsilon_0}} \int_{\Omega_x} d\Omega \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega(t-s)} \frac{\Lambda(r, \Omega) \cdot D(r, s)}{\omega^2 - 2i\omega \eta + \Omega^2 + \eta^2} + \left[ X_{\Omega,\text{free}}(r) e^{-i\Omega t} + \text{c.c.} \right], \]  
(23)

where we have identified the solution of Eqs. (20) and (21) involving the time independent \( X_{\Omega,\text{free}}(r) \) describes the free (nondriven) oscillation of the medium fields, which we note has 0, 1, 2, or 3 nonvanishing components, as per the discussion concerning \( X_0(r) \) after Eq. (12). Substitution of Eq. (23) into the expression (19) for the polarization in terms of the medium fields gives

\[ P(r, t) = \int ds \Gamma(r, t-s) \cdot D(r, s) \]  
\[ + \int_{\Omega_x} d\Omega \varepsilon_0 \Lambda(r, \Omega) \cdot \left[ X_{\Omega,\text{free}}(r) e^{-i\Omega t} + \text{c.c.} \right], \]  
(24)

where we write the dielectric response of the model as

\[ \Gamma(r, t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma(r, \omega) e^{-i\omega t}, \]  

with

\[ \Gamma(r, \omega) = \int_{\Omega_x} d\Omega \frac{\Lambda(r, \Omega) \cdot \Lambda(r, \Omega)}{(\Omega - \omega - i\eta)(\Omega + \omega + i\eta)}. \]  
(25)

The second term on the right-hand side of Eq. (24) is a fluctuation term familiar from reservoir theory [47], and is associated with the fluctuation-dissipation theorem. Note that our model satisfies Eq. (7), as is expected in the absence of magnetic response effects. We have

\[ \Gamma(r, \omega)^\ast = \Gamma(r, -\omega), \]  
(26)

and it is clear from the form of our expression (25) for \( \Gamma(r, \omega) \) that the dielectric response is purely causal, since the poles of \( \Gamma(r, \omega) \) are at \( \omega = \pm \Omega - i\eta \) for all \( \Omega \in [\Omega_x, \infty] \), and are all in the lower half plane. Then using the well-known identity

\[ \lim_{\eta \to 0^+} \frac{1}{x - x_0 \pm i\eta} = \frac{P}{x - x_0} + i\pi \delta(x - x_0), \]  
(27)

and identifying real and imaginary parts of \( \Gamma(r, \omega) \), we find, for \( \omega > 0 \),

\[ \Re \Gamma(r, \omega) = P \int_{\Omega_x} d\Omega \frac{\Lambda(r, \Omega) \cdot \Lambda(r, \Omega)}{(\Omega - \omega)(\Omega + \omega)}, \]  
(28)
\[ \Im \Gamma^{\lambda\lambda'}(r, \omega) = \frac{\pi}{2\omega} \Lambda^{\lambda\lambda'}(r, \omega) \Lambda^{\lambda'\lambda''}(r, \omega). \]  
(29)

For \( \omega < 0 \), \( \Gamma^{\lambda\lambda'}(r, \omega) \) can be determined using Eq. (26).

A set of Kramers-Kronig relations follow from Eq. (25), and can be written in the form

\[ \Re \Gamma(r, \omega) = -\frac{2}{\pi} P \int_{\Omega_x} d\Omega \frac{\Omega \Im \Gamma(r, \Omega)}}{(\omega - \Omega)(\omega + \Omega)}, \]  
(30)
\[ \Im \Gamma(r, \omega) = \frac{2}{\pi} P \int_{\Omega_x} d\Omega \frac{\Omega \Re \Gamma(r, \Omega)}}{(\omega - \Omega)(\omega + \Omega)}. \]  
(31)

It is now apparent how any specified causal \( \Gamma^\mu(r, t) \) can be described by our model. From the Fourier transform of \( \Gamma^\mu(r, t) \) expressed in the basis of its local principal axes, and recalling that the \( \Lambda^{\lambda\lambda'}(r, \omega) \) are chosen to share the same set of principal axes as \( \Gamma^{\lambda\lambda'}(r, \omega) \), we can identify a set of coupling constants \( \Lambda^{\lambda\lambda'}(r, \omega) \). With \( \Im \Gamma(r, \omega) \) correctly described, causality guarantees that the corresponding \( \Re \Gamma(r, \omega) \) follows from Eq. (30).

### IV. POLARITON MODES

#### A. Nominal system

We now identify a useful decomposition of the total Hamiltonian. If dispersive and absorptive effects are small, the simple model (4) and (5) can often provide a good starting point. Whether or not this is the case, we introduce a nominal index profile \( n(r) \) and write the full Hamiltonian (8) as
In Eq. (32), where we have decomposed the vacuum electromagnetic field Hamiltonian as \( H_{\text{em}} = H_0 + H_{\Delta} \), with
\[
H_0 = \frac{1}{2\mu_0} \int d^3r \mathbf{B}(r) \cdot \mathbf{B}(r) + \frac{1}{2\varepsilon_0} \int d^3r \frac{\mathbf{D}(r) \cdot \mathbf{D}(r)}{n^2(r)},
\] (33)
\[
H_{\Delta} = \frac{1}{2}\varepsilon_0 \int d^3r \mathbf{D}(r) \Gamma^{ij}_0(r) \mathbf{D}(r).\] (34)

Hamilton’s equations using \( H_0 \) alone as the Hamiltonian, in conjunction with the commutators (10), yield the Maxwell equations (1) with the constitutive relation
\[
\mathbf{D}(r,t) = \varepsilon_0 \mathbf{E}(r,t),
\] (35)
which identifies \( H_0 \) as the Hamiltonian for a nominal, nonabsorptive and nondispersive system with refractive index \( n(r) \). For the nominal system, \( H_0 \) is numerically equal to the total energy in the electromagnetic fields in the presence of the dielectric [45]. The “counterterm” \( H_{\Delta} \) in the Hamiltonian \( H \) then guarantees that Eq. (32) provides the correct sum (8).

We emphasize that we do not rely on perturbative results in \( (H-H_0) \); in general we exhibit equations whose solutions describe the dynamics that follow from the full \( H \). Yet we see below that having a reference, “nominal system” is useful both in understanding the physics of those exact solutions, and in constructing them. This is in part because there are numerous freely available codes and algorithms for calculating mode fields and dispersion relations of nondispersive, nonabsorptive artificially structured media [49–52]. Of course, it is always possible to set \( n(r)=1 \), in which case the vacuum itself serves as the nominal system, and \( H_{\Delta}=0 \).

B. Nominal system modes

To construct the polariton modes, we first characterize the solutions of the dynamical equations where only \( H_0 \) is used as the Hamiltonian. We begin by identifying stationary solutions,
\[
\mathbf{D}(r,t) = \mathbf{D}_m(r)e^{-i\omega_m t} + \text{c.c.},
\]
\[
\mathbf{B}(r,t) = \mathbf{B}_m(r)e^{-i\omega_m t} + \text{c.c.},
\] (36)
of the classical equations, where c.c. stands for complex conjugate. Using the nominal Hamiltonian \( H_0 \) and the form (36) in Eq. (11), we obtain the time-independent Maxwell equations for the nominal system:
\[-i\omega_m H_0 \mathbf{D}_m(r) = \nabla \times \mathbf{B}_m(r),\]
\[i\omega_m \varepsilon_0 \mathbf{B}_m(r) = \nabla \times \left[ \frac{\mathbf{D}_m(r)}{n^2(r)} \right].\] (37)

In practice, normal modes are found by combining Eqs. (37) into a second-order Hermitian eigenvalue problem for \( \mathbf{B}_m(r) \):
\[
\nabla \times \left[ \frac{\nabla \times \mathbf{B}_m(r)}{n^2(r)} \right] = \frac{\omega_m^2}{c^2} \mathbf{B}_m(r).\] (38)

Solutions of Eq. (38) for \( \omega_m \neq 0 \) are sought, subject to the divergence condition
\[
\nabla \cdot \mathbf{B}_m(r) = 0.
\] (39)

It is also possible to construct a second-order eigenvalue problem for \( \mathbf{D}_m(r) \),
\[
\nabla \times \nabla \times \mathbf{D}_m(r) = \frac{\omega_m^2}{c^2} \mathbf{D}_m(r),\] (40)
although the associated operator is not Hermitian. Then solutions of Eq. (40) for \( \omega_m \neq 0 \) are subject to the divergence condition \( \nabla \cdot \mathbf{D}_m(r)=0 \).

Either Eq. (38) or Eq. (40) suffices as a master equation for the electromagnetic modes. For example, having obtained \( \mathbf{B}_m(r) \) satisfying the master equation (38), \( \mathbf{D}_m(r) \) is obtained from the first of Eqs. (37). Then the pair of fields \( (\mathbf{D}_m(r),\mathbf{B}_m(r)) \) identifies a stationary solution of the classical Maxwell equations (1) and (2); the square of the frequency is obtained from Eq. (38); we take the positive root, associating a real positive frequency \( \omega_m \) with each normal-mode solution. Taking the complex conjugate of Eqs. (37), we note that for every mode \( (\mathbf{D}_m(r),\mathbf{B}_m(r)) \) with frequency \( \omega_m \) there is another related mode
\[
(\mathbf{D}_m(r),\mathbf{B}_m(r)) = (\mathbf{D}_m^*(r),-\mathbf{B}_m^*(r))
\] (41)
with frequency
\[
\omega_m = \omega_m.
\] (42)

Throughout the paper, an overbar refers to the physical conjugate in this sense. In problems of interest such pairs of modes are physically easy to identify and pair, so in practice Eqs. (41) and (42) can be used to define the mode \( m \) in terms of the mode \( m \), and to thereby obtain half of the modes without resorting directly to the eigenvalue problem (38).

Since Eq. (38) defines a Hermitian eigenvalue problem, solutions associated with different eigenvalues are orthogonal, and we can choose the following normalization conditions for \( \mathbf{D}_m(r) \) and \( \mathbf{B}_m(r) \):
\[
\int_V d^3r \frac{\mathbf{B}_m^*(r) \cdot \mathbf{B}_m(r)}{\mu_0} = \frac{\hbar \omega_m}{2} \delta_{mm'},
\]
\[
\int_V d^3r \frac{\mathbf{D}_m^*(r) \cdot \mathbf{D}_m(r)}{\varepsilon_0 \mu_0 n^2(r)} = \frac{\hbar \omega_m}{2} \delta_{mm'},\] (43)
where we employ box normalization, the spatial integral is over the very large normalization region \( V \) of volume \( V \), and the mode fields \( \mathbf{D}_m(r) \) and \( \mathbf{B}_m(r) \) are taken to have periodic boundary conditions. Throughout most of this paper we use discrete modes for the electromagnetic field in this fashion. The exception is Sec. V, where we give the transition to systems with continuous or mixed labeling.

For the nominal system, arbitrary electromagnetic field solutions can be expanded in terms of the normal modes as
\[
\mathbf{D}(r,t) = \sum_m c_m^{(1)}(t) \mathbf{D}_m(r),
\]

\[
\mathbf{B}(r,t) = \sum_m c_m^{(2)}(t) \mathbf{B}_m(r),
\]

where we guarantee the reality, or Hermiticity in the quantum case, of \(\mathbf{B}(r,t)\) and \(\mathbf{D}(r,t)\) by imposing the conditions

\[C_m^{(1)} = (c_m^{(1)})^\dagger\]

\[C_m^{(2)} = -(C_m^{(2)})^\dagger.\]

These conditions can be satisfied identically by introducing new unrestricted mode amplitudes \(a_m\) such that \(C_m^{(1)} = a_m^\dagger a_m^\dagger\) and \(C_m^{(2)} = a_m - a_m^\dagger\). In conjunction with Eq. (41), this yields

\[
\mathbf{D}(r,t) = \sum_m [a_m(t) \mathbf{D}_m(r) + a_m^\dagger(t) \mathbf{D}_m^\dagger(r)],
\]

\[
\mathbf{B}(r,t) = \sum_m [a_m(t) \mathbf{B}_m(r) + a_m^\dagger(t) \mathbf{B}_m^\dagger(r)],
\]

where the sum is over all modes. From the commutators (Poisson brackets) Eqs. (10) then follow the equal-time commutators

\[
[a_m, a_m^\dagger] = 0,
\]

\[
[a_m, a_m^\dagger] = \delta_{mn^\prime}.
\]

Using Eqs. (45) in Eq. (33) we then find that the nominal system Hamiltonian is given by

\[
\mathcal{H}_0 = \sum_m \hbar \omega_m a_m^\dagger a_m,
\]

with neglect of the zero point energy in the quantum case, and Hamilton’s equation

\[
i\hbar \dot{a}_m = [a_m, \mathcal{H}_0]
\]

indeed generates the Maxwell curl equations (1) with the constitutive relation (35); the Maxwell divergence equations (2) are manifested satisfied due to the form of the expansion (45) in terms of the divergencelessness normal modes.

We can now use the expansion (45) of the electromagnetic fields to write the full Hamiltonian in terms of raising and lowering operators of the nominal system modes. First, the counterterm is given by

\[
\mathcal{H}_3 = \frac{1}{2} \sum_{m,m^\prime} (a_{m^\prime}^\dagger + a_{m^\prime}^\dagger) \Gamma_{m,m^\prime}^{(0)}(a_m + a_m^\dagger),
\]

where we define

\[
\Gamma_{m,m^\prime}^{(0)} = \frac{1}{\varepsilon_0} \int d\mathbf{r} \mathcal{D}_{mm^\prime}(\mathbf{r}) \Gamma_{m,m^\prime}^{(0)}(\mathbf{r}),
\]

We note that using Eq. (41) and the permutation symmetry of \(\Gamma_{m,m^\prime}^{(0)}(\mathbf{r})\) yields the properties

\[
\Gamma_{m,m^\prime}^{(0)} = \Gamma_{m^\prime,m}^{(0)} = \Gamma_{m,m^\prime}^{(0)}.
\]

The full Hamiltonian then becomes

\[
\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_3 + \mathcal{H}_3
\]

\[
= \sum_m \mathcal{H}_m + \sum_{m,m^\prime} \Gamma_{m,m^\prime}^{(0)}(a_m + a_m^\dagger)
\]

\[
+ \int d\Omega \int d\mathbf{r} \hbar \Omega \psi^\dagger_\Omega(\mathbf{r}) \psi_\Omega(\mathbf{r}) + \int d\Omega \int d\mathbf{r} \Delta L_m(r, \Omega) \cdot [\psi_\Omega(\mathbf{r}) + \psi^\dagger_\Omega(\mathbf{r})],
\]

where again we have neglected the zero-point energy in the quantum case, and where we have defined a mode coupling constant

\[
\Lambda_m^\lambda(r, \Omega) = -\sqrt{\frac{\hbar}{2\varepsilon_0 \Omega}} D_{m}^{\lambda^\dagger}(r) \Lambda^\lambda_m(r, \Omega).
\]

Recall that the overbar always refers to physical conjugation of a mode as described after Eq. (42), and that sums over modes in Eq. (54) are over all modes.

C. Polariton problem

It is the Hamiltonian (54) we seek to diagonalize, and find the full, or polariton, modes of the system. Generally, some of the nominal electromagnetic modes lie below the cutoff frequency \(\Omega_c\) for absorption, and for our calculation with periodic boundary conditions a discrete spectrum of modes can be found there. Above the cutoff frequency for absorption the polariton spectrum forms a continuum. At these frequencies, the problem of finding the polariton modes amounts to a Fano diagonalization with many discrete modes coupled to many continuous modes—a “many-many” Fano problem—with counter-rotating wave terms, and an extra coupling between the bare discrete modes. The single-single, single-many, and many-many rotating-wave approximation (RWA) cases were solved by Fano [54]; the many-many RWA case was subsequently solved by Mies [55] in a way specific to Fano’s configuration interaction problem; that solution does not apply here. The single-single non-RWA problem was solved by Hutten and Barnett in their seminal work [56]; here we must address an augmented many-many non-RWA case, which has not been previously solved. In addition, virtual coupling to the medium modes shifts the nominal system modes in the discrete spectrum below the cutoff frequency \(\Omega_c\).

We begin with an important classification of the polariton modes. Below we identify two types of modes, which we call longitudinal polarization (LP) polaritons and transverse polarization (TP) polaritons. For the first type, \(\mathcal{H}_3\) does not contribute to the dynamics, and the modes consist solely of contributions from the medium oscillators. We formally separate the Hamiltonian of these uncoupled modes from the Hamiltonian of the TP modes as

\[
\mathcal{H} = \mathcal{H}_LP + \mathcal{H}_TP.
\]
Fig. 1. (Color online) Schematic diagram of the frequency domains of the various modes of the system before (left) and after (right) the canonical transformation. The nominal electromagnetic mode operators \(a_m\) and medium modes \(\varphi_\Omega\) are plotted on the left, while the polariton mode operators \(c_m\) and \(c^\dagger_\Omega m\) are plotted on the right.

verse polarization modes; then we seek polariton operators that diagonalize the Hamiltonian in the form

\[
H_{LP} = \sum_m \int_{\Omega_g}^\infty A_m \Omega s^\dagger_\Omega m s^\da_\Omega m, \tag{57}
\]

\[
H_{TP} = \sum_m \hbar \omega_m c^\dagger_m c_m + \sum_m \int_{\Omega_g}^\infty \hbar \Omega c^\dagger_\Omega m c_\Omega m, \tag{58}
\]

where the \(c_\Omega m\) are the TP operators for the continuous polariton spectrum, and the \(c_m\) are the TP operators of the discrete polariton spectrum; we will find, for all \(m\), \(\omega_m < \Omega_g\). The \(s_\Omega m\) are the LP operators, and by definition they satisfy

\[
[s_\Omega m, H_{int}] = 0. \tag{59}
\]

The various modes are plotted schematically in Fig. 1. The polariton operators are required to satisfy harmonicity conditions

\[
[s_\Omega m, H] = \hbar \Omega s_\Omega m, \tag{60}
\]

\[
[c_m, H] = \hbar \omega_m c_m, \tag{61}
\]

\[
[c_\Omega m, H] = \hbar \Omega c_\Omega m, \tag{62}
\]

and the (equal-time) canonical commutation relations that apply are

\[
[s_\Omega m s^\dagger_\Omega m'], = \delta_{mm'} \delta(\Omega - \Omega'), \tag{63}
\]

\[
[c_m c^\dagger_m] = \delta_{mm'}, \tag{64}
\]

\[
[c_\Omega m c^\dagger_\Omega m'] = \delta_{mm'} \delta(\Omega - \Omega'), \tag{65}
\]

with all other equal-time commutators between the various \(c\) and \(s\) operators vanishing. The number of discrete modes is not known at the outset, although for sufficiently small absorption and dispersion it is expected to approach the number of nominal modes below the cutoff frequency in typical systems.

In summary, there are three types of polaritons. In the absorption regime \((\Omega \gg \Omega_g)\) we have LP and TP polaritons, which form continua, and in the purely dispersive regime \((\Omega < \Omega_g)\) we have discrete TP polaritons.

We mention now, and prove in Sec. IV H, that the dispersion and magnetic inducment fields can be written entirely in terms of the TP polariton operators and their corresponding mode fields as

\[
D(r, t) = \sum_m [c_m D^m_m(r) + c^\dagger_m D^{\dagger m}_m(r)]
\]

\[
+ \sum_m \int_{\Omega_g}^\infty d\Omega [c^\dagger_\Omega m \tilde{D}_\Omega m(r) + c_\Omega m \tilde{D}^{\dagger}_\Omega m(r)], \tag{66}
\]

\[
B(r, t) = \sum_m [c_m B^m_m(r) + c^\dagger_m B^{\dagger m}_m(r)]
\]

\[
+ \sum_m \int_{\Omega_g}^\infty d\Omega [c^\dagger_\Omega m \tilde{B}_\Omega m(r) + c_\Omega m \tilde{B}^{\dagger}_\Omega m(r)], \tag{67}
\]

where the \(\tilde{D}_m(r)\) and \(\tilde{B}_m(r)\) are the mode fields for discrete TP mode \(m\), and the \(\tilde{D}_\Omega m(r)\) and \(\tilde{B}_\Omega m(r)\) are the mode fields for the continuum TP mode \(\Omega m\). We now turn to the task of identifying three polariton types.

D. Longitudinal polarization (LP) polariton modes

We first identify the LP polariton modes. These are modes in the absorption regime composed entirely of medium fields which, although not necessarily longitudinal themselves, produce a longitudinal polarization. Thus they do not couple through \(H_{int}\) with the displacement field, which is purely transverse. It is possible to formulate the medium fields in \(k\) space so as to avoid inclusion of longitudinal polarizations from the outset. However, this approach leads to more complicated expressions elsewhere, especially if \(V_\Omega\) is not all of space. The real-space formulation of the medium fields we have used here, in contrast, respects the spatial locality of the dielectric response of the model.

Writing the lowering operators of the LP polaritons as \(s_\Omega m\) (57), we expand them in terms of the medium fields as

\[
s_\Omega m = \int_{\Omega_\Omega} dr \varphi_\Omega m(r) \cdot \varphi_\Omega (r), \tag{68}
\]

where the condition (59) requires that the expansion coefficients \(\varphi_\Omega m(r)\) obey

\[
\int_{\Omega_\Omega} dr \varphi^\dagger_\Omega m'(r) \cdot \varphi_\Omega m(r) = \delta_{mm'}, \tag{69}
\]

for all \(m\); we normalize them according to

\[
\int_{\Omega_\Omega} dr \varphi^\dagger_\Omega m'(r) \cdot \varphi_\Omega m(r) = \delta_{mm'}. \tag{70}
\]

In conjunction with the Hamiltonian (54) and the medium field commutators (17), it follows from Eqs. (68)–(70) re-
spective that the LP lowering operators $s_{\Omega m}$ have harmonic time dependence (60), and satisfy canonical commutation relations (63).

To identify the $\rho^{\Omega m}(r)$, we first note that we can extend the integrals in Eqs. (68)–(70) to all space, by extending the functions $\rho_{l}^{\Omega m}(r)$ to vanish for $r$ outside $V_\Omega$. Now the $D_{m'}(r)$ span the space of transverse vector fields, so since the integral over all space of the dot product of a longitudinal field with any transverse field vanishes, and since no transverse field has a vanishing overlap in this sense with every transverse field, $\Lambda(r,\Omega)\cdot \rho^{\Omega m}(r)$ must be a longitudinal vector field. Recalling the periodic boundary conditions employed, and neglecting any uniform fields over the volume, we must have

$$\Lambda(r,\Omega)\cdot \rho^{\Omega m}(r) = \nabla \phi^{\Omega m}(r)$$

(71)

for some scalar field $\phi^{\Omega m}(r)$. Since the left-hand side of Eq. (71) must be finite, we require that $\phi^{\Omega m}(r)$ be continuous and once differentiable. Taking components along the principal axis direction $\lambda$, it then follows from Eq. (71) and from the requirement that $\rho^{\Omega m}(r)$ vanish outside $V_\Omega$ that (no summation)

$$\rho^{\Omega m}_{\lambda}(r) = f^{\Omega}_{\lambda}(r) \partial_{\lambda} \phi^{\Omega m}(r),$$

(72)

with

$$f^{\Omega}_{\lambda}(r) = \frac{1}{\Lambda^{\lambda}(r,\Omega)}$$

if $\Lambda^{\lambda}(r,\Omega) \neq 0$. (73)

We need not specify $f^{\Omega}_{\lambda}(r)$ for $r$ such that $\Lambda^{\lambda}(r,\Omega)=0$, since

$$\partial_{\lambda} \phi^{\Omega m}(r) = 0$$

if $\Lambda^{\lambda}(r,\Omega) = 0$, (74)

which follows immediately from Eq. (71). Equations (72)–(74) describe the full set of LP polariton solutions.

It follows from Eq. (74) that $\phi^{\Omega m}$ is constant within any connected region outside of $V_\Omega$. Furthermore, given a local unit vector $\tilde{n}$ normal to the boundary of $V_\Omega$, which we denote $S_\Omega$, we have the boundary condition $\tilde{n} \times \nabla \phi^{\Omega m}(r) = 0$ on $S_\Omega$. Note that $V_\Omega$ may be multiply connected, and that we make no assumptions about the number of connected components. This is illustrated schematically in Fig. 2.

The procedure for constructing the full set of LP polariton modes is as follows. Find a set of continuous scalar fields $\phi^{\Omega m}(r)$ that span the space of continuous once-differentiable functions on the normalization volume up to addition of an overall constant, subject to the condition (74). Obtain the corresponding set of $\rho^{\Omega m}(r)$ from Eq. (72). Then the $\rho^{\Omega m}(r)$ can be constructed from the $\phi^{\Omega m}(r)$ by Gram-Schmidt orthogonalization of the $\rho^{\Omega m}(r)$ with respect to the normalization condition (70).

There is a formal mathematical analogy here to electrostatics with perfect conductors and free charges. The field $\phi^{\Omega m}(r)$ corresponds to the electromagnetic scalar potential, and the quantity $-\Lambda(r,\Omega)\cdot \rho^{\Omega m}(r)$ corresponds to the electric field. The local charge density is proportional to $-\nabla^2 \phi^{\Omega m}(r)$. And the region of space outside of $V_\Omega$, where there is no coupling, plays the role of a perfect conductor. The complete set of fields $\phi^{\Omega m}(r)$ then corresponds to those potentials that can be generated by arbitrary charge distributions in the region $V_\Omega$.

While a route to constructing the LP polariton modes has been identified here for completeness, the explicit construction of these modes is unnecessary for most applications. This is because the displacement and magnetic induction fields only involve the transverse polarization polariton modes, as mentioned before Eq. (66), and we now turn to the task of finding them.

E. Transverse polarization (TP) polariton modes

We first consider the TP polaritons in the absorptive regime. To find them, we begin with a general expansion of the $c_{\Omega m}$ in terms of nominal system modes and medium oscillator modes that are coupled together in the Hamiltonian (54). We write

$$c_{\Omega m} = \sum_{n} (a_{n}^{\Omega m} a_{n} + \beta_{n}^{\Omega m} a_{n}^{\dagger})$$

$$+ \int_{\xi_\Omega} d\Omega' \int_{V_\Omega} d\mathbf{r} \left[ \alpha_{\Omega m}^{\Omega m}(\mathbf{r}) \cdot \psi_{\Omega'}(\mathbf{r}) + \beta_{\Omega m}^{\Omega m}(\mathbf{r}) \cdot \psi_{\Omega'}^{\dagger}(\mathbf{r}) \right],$$

(75)

where recall that the sums include all nominal system modes in the sum, regardless of whether they lie in the frequency range of the discrete or continuous polariton band. We refer to the terms containing $\alpha$'s and $\beta$'s as respectively as corotating and counterrotating terms.

The harmonicity condition (62), in conjunction with Eqs. (75) and (54), yields an equation that we do not reproduce here, which provides the conditions on the expansion coefficients $\alpha_{n}^{\Omega m}$, $\alpha_{\Omega m}^{\Omega m}(r)$, $\beta_{n}^{\Omega m}$, and $\beta_{\Omega m}^{\Omega m}(r)$. We refer to it as the "harmonic" equation." Taking commutators of the harmonic equation with $a_{\Omega m}$, $a_{m}^{\dagger}$, $\psi_{\Omega m}(r)$, and $\psi_{\Omega m}^{\dagger}(r)$, and relabeling the primes, we obtain the following system of equations:

FIG. 2. (Color online) Schematic illustration of the scalar potential $\phi^{\Omega m}$ for the longitudinal polarization modes. The hatched regions represent $V_\Omega$, the region in which absorption is present at frequency $\Omega$. The scalar potential is constant within connected non-absorbing regions.
\[ (\hbar \Omega - \hbar \omega_n) \beta_{n'}^{\Omega m} - \sum_n \Gamma_n^{(0)} (\alpha_{n}^{\Omega m} - \rho_{n}^{\Omega m}) \]
\[ - \int_{\Omega}^\infty d\Omega' \int \mathcal{V}_{\Omega'} \, d\mathbf{r} \, \mathbf{A}_n^*(\mathbf{r}, \Omega') \cdot [\alpha_{\Omega m}^* (\mathbf{r}) - \beta_{\Omega m}^*(\mathbf{r})] = 0, \]

(76)

\[ (\hbar \Omega + \hbar \omega_n) \beta_{n'}^{\Omega m} - \sum_n \Gamma_n^{(0)} (\alpha_{n}^{\Omega m} - \rho_{n}^{\Omega m}) \]
\[ - \int_{\Omega}^\infty d\Omega' \int \mathcal{V}_{\Omega'} \, d\mathbf{r} \, \mathbf{A}_n^*(\mathbf{r}, \Omega') \cdot [\alpha_{\Omega m}^* (\mathbf{r}) - \beta_{\Omega m}^*(\mathbf{r})] = 0, \]

(77)

\[ (\hbar \Omega - \hbar \Omega') \beta_{\Omega m}^{\Omega m}(\mathbf{r}) - \sum_n \mathbf{A}_n(\mathbf{r}, \Omega') (\alpha_{n}^{\Omega m} - \rho_{n}^{\Omega m}) = 0, \]

(78)

\[ (\hbar \Omega + \hbar \Omega') \beta_{\Omega m}^{\Omega m}(\mathbf{r}) - \sum_n \mathbf{A}_n(\mathbf{r}, \Omega') (\alpha_{n}^{\Omega m} - \rho_{n}^{\Omega m}) = 0, \]

(79)

where we recall that \( \mathbf{A}_n(\mathbf{r}, \Omega) \) is defined in Eq. (55). We note that the harmonicity equation can be reconstructed from Eqs. (76)–(79) by multiplying each of these equations by an appropriate operator and then composing the results, so Eqs. (76)–(79) are necessary and sufficient for the harmonicity equation to be satisfied, and thus for harmonicity condition (62) to hold. For every solution to Eqs. (76)–(79) labeled by \( m \), i.e., \( (\alpha_n^{\Omega m}, \beta_n^{\Omega m}, \alpha_n^{\Omega m}, \beta_n^{\Omega m}(\mathbf{r}), \beta_n^{\Omega m}(\mathbf{r})) \), there is another solution, which we label \( \bar{m} \), given by

\[ (\alpha_n^{\Omega m} = \bar{\alpha}_n^{\Omega m} + \alpha_n^{\Omega m}), \beta_n^{\Omega m} = \bar{\beta}_n^{\Omega m} + \beta_n^{\Omega m}, \alpha_n^{\Omega m} = \bar{\alpha}_n^{\Omega m}, \beta_n^{\Omega m}(\mathbf{r}) = \bar{\beta}_n^{\Omega m}(\mathbf{r}), \]

(80)

where \( \alpha_n^{\Omega m} = (\alpha_n^{\Omega m})^* \), etc. To confirm Eq. (80), let \( m \to \bar{m} \) and \( n' \to \bar{n}' \) in Eqs. (76)–(79), take the complex conjugate of these equations, use properties (42) and (51), and reorganize the sums. This yields the original set of equations, i.e., Eqs. (76)–(79), but with the replacements \( (\alpha_n^{\Omega m}, \beta_n^{\Omega m}, \alpha_n^{\Omega m}, \beta_n^{\Omega m}, \alpha_n^{\Omega m}, \beta_n^{\Omega m}(\mathbf{r}) \to \bar{\alpha}_n^{\Omega m}, \beta_n^{\Omega m}(\mathbf{r}) \to \bar{\beta}_n^{\Omega m}(\mathbf{r}) \), and Eq. (80) results.

We now solve the system (76)–(79). Comparing Eqs. (76) and (77) immediately gives

\[ \sum_n \left[ (\hbar \Omega)^2 - (\hbar \omega_n)^2 \right] \sum_n \left[ \frac{1}{2} \left( \mathbf{A}_n^{\Omega m}(\mathbf{r}, \Omega') \cdot \mathbf{A}_n(\mathbf{r}, \Omega') \right) \gamma_n^{\Omega m} \right] = \frac{Z_{\Omega m}}{\hbar} \int \mathcal{V}_{\Omega} \, d\mathbf{r} \, \mathbf{A}_n^{\Omega m}(\mathbf{r}, \Omega') \cdot \mathbf{A}_n(\mathbf{r}, \Omega') \gamma_n^{\Omega m}. \]

(87)
Equation (87) is a generalized Hermitian eigenvalue problem with the eigenvalue-eigenvector pair \((Z^{\Omega m}, \gamma^{\Omega m}_n)\). To our knowledge, such an equation has never appeared in a previous treatment of a multichannel Fano-diagonalization problem [55,58]. Since \(Z^{\Omega m}\) is the eigenvalue in a generalized Hermitian eigenvalue problem, it is guaranteed to be real.

Making use of the expressions (28) and (29) for the medium response, Eq. (87) becomes

\[
\sum_n \left[ \frac{(\hbar \Omega)^2 - (\hbar \omega_n)^2}{2 \hbar \omega_n} \delta_{n', n} + (\Delta \Re \Gamma^{\Omega}_{m n'}) \right] \gamma^{\Omega m}_n = \frac{Z^{\Omega m}}{\pi} \sum_n (\Im \Gamma^{\Omega}_{m n'}) \gamma^{\Omega m}_n,
\]

where

\[
(\Delta \Re \Gamma^{\Omega}_{m n'}) = \frac{1}{\varepsilon_0} \int \, d\mathbf{r} \mathbf{D}^*_n(\mathbf{r}) \cdot [\{\Re \Gamma(\mathbf{r}, \Omega)\] \nonumber
- \Gamma_0(\mathbf{r})] \cdot \mathbf{D}_m(\mathbf{r}),
\]

\[
(\Im \Gamma^{\Omega}_{m n'}) = \frac{1}{\varepsilon_0} \int \, d\mathbf{r} \mathbf{D}^*_n(\mathbf{r}) \cdot [\Im \Gamma(\mathbf{r}, \Omega)] \cdot \mathbf{D}_m(\mathbf{r}).
\]

Physical quantities appear in Eq. (88) in a very natural way. Here the information about the model coupling constant \(\Lambda(\mathbf{r}, \Omega)\) has completely given way to the aggregate real and imaginary parts of the dielectric response; for the real part of the response, the quantity that appears is in fact just the dispersive part of the response beyond the nominal value. The eigenvalue equation for the underlying polaritons is now expressed in terms of the physically important quantities, and the dispersive and absorptive contributions to the dielectric response appear in separate roles here, rather than as real and imaginary parts of a single quantity, reflecting their physical difference. Furthermore, the real response appears as its difference from the nominal response, so there is a simple connection to the nominal system. From a practical standpoint, Eq. (88) can be applied straightforwardly; one takes matrix elements consisting of overlap integrals between nominal system modes and the medium response; the generalized Hermitian eigenvalue problem can be solved using standard routines to obtain \(Z^{\Omega m}\) and \(\gamma^{\Omega m}_n\), and then the medium contributions are obtained from Eqs. (82) and (84). We note that Eqs. (88)–(90) hold for any choice of nominal index profile \(n(\mathbf{r})\); no perturbative analysis has been used. We can also isolate the effect of inclusion of the counterrotating terms in the expansion (75), since performing the above derivation in the rotating wave approximation, i.e., with all \(\beta^{\Omega m}_n=0\), yields the eigenvalue equation (88), but with the replacement \(\gamma^{\Omega m}_n \rightarrow \gamma^{\Omega m}_n - \alpha^{\Omega m}_n\) and with the term multiplying the \(\delta_{n', n}\) replaced by \((\hbar \Omega - \hbar \omega_n)\). Furthermore, although it is not immediately apparent from Eq. (88), the eigenvalues \(Z^{\Omega m}\) depend only on \(\Gamma(\mathbf{r}, \omega)\), and are independent of the choice of nominal index profile \(n(\mathbf{r})\). This is verified in Sec. IV G.

Finally, imposing the equal-time commutation relation (65) yields the normalization condition for the \(\gamma^{\Omega m}_n\). We defer the details to Appendix A, where the normalization condition is determined to be

\[
\sum_{n, n'} \gamma^{\Omega m*}_n (\Im \Gamma^{\Omega}_{m n'}) \gamma^{\Omega m}_n = \frac{\pi \hbar}{\pi^2 + (\hbar \Omega)^2} \delta_{n n'}.
\]

F. Polaritons in the transparent regime

Below the absorption cutoff frequency \(\Omega_c\), the polariton spectrum is discrete, and the TP polariton operators there are the \(c_m\) introduced in Eq. (58). We begin with a general expansion of the \(c_m\) in terms of nominal system modes and medium modes that are coupled together in the Hamiltonian (54):

\[
c_m = \sum_n (\alpha^m_n c_n + \beta^m_n c^*_{n^*}) + \int_{\Omega_g} d\Omega' \int_{\Omega_g'} \, d\mathbf{r} \left( \mathbf{\alpha}^m_{\Omega_g}(\mathbf{r}) \cdot \psi_{\Omega_g'}(\mathbf{r}) \right) + \mathbf{\beta}^m_{\Omega_g}(\mathbf{r}) \cdot \psi_{\Omega_g'}(\mathbf{r}) \right].
\]

Then, following the approach used in the absorptive regime, the harmonicity condition (61) leads to the following system of equations:

\[
(h \tilde{\omega}_m - h \omega_n) \alpha^m_n = \sum_n \left(1^{(0)}_{nn} (\alpha^m_n - \beta^m_n) - \int_{\Omega_g} d\Omega' \int_{\Omega_g'} \, d\mathbf{r} \mathbf{\alpha}^m_{\Omega_g}(\mathbf{r}) \cdot \mathbf{\alpha}^m_{\Omega_g'}(\mathbf{r}) \cdot \left[ \mathbf{\alpha}^m_{\Omega_g}(\mathbf{r}) - \mathbf{\beta}^m_{\Omega_g}(\mathbf{r}) \right] = 0,
\]

\[
(h \tilde{\omega}_m + h \omega_n) \beta^m_n = \sum_n \left(1^{(0)}_{nn} (\alpha^m_n - \beta^m_n) - \int_{\Omega_g} d\Omega' \int_{\Omega_g'} \, d\mathbf{r} \mathbf{\alpha}^m_{\Omega_g}(\mathbf{r}) \cdot \mathbf{\beta}^m_{\Omega_g'}(\mathbf{r}) \cdot \left[ \mathbf{\alpha}^m_{\Omega_g}(\mathbf{r}) - \mathbf{\beta}^m_{\Omega_g}(\mathbf{r}) \right] = 0,
\]

\[
(h \tilde{\omega}_m - h \Omega) \alpha^m_{\Omega_g}(\mathbf{r}) = \sum_n \Lambda_n(\mathbf{r}, \Omega) (\alpha^m_n - \beta^m_n) = 0, \tag{95}
\]

\[
(h \tilde{\omega}_m + h \Omega) \beta^m_{\Omega_g}(\mathbf{r}) = \sum_n \Lambda_n(\mathbf{r}, \Omega) (\alpha^m_n - \beta^m_n) = 0. \tag{96}
\]

Physical conjugate solutions are given by

\[
\alpha^m_{\Omega_g*} = \alpha^m_n, \beta^m_{\Omega_g*} = \beta^m_n, \alpha^m_{\Omega_g*}(\mathbf{r}) = \alpha^m_{\Omega_g}(\mathbf{r}), \beta^m_{\Omega_g*}(\mathbf{r}) = \beta^m_{\Omega_g}(\mathbf{r}). \tag{97}
\]

As with Eqs. (76) and (77), comparing Eqs. (93) and (94) immediately gives

\[
\beta^m_n = \frac{h \tilde{\omega}_m - h \omega_n}{h \tilde{\omega}_m + h \omega_n} \alpha^m_n. \tag{98}
\]

Making use of Eq. (98), Eqs. (95) and (96) have the following solutions in terms of the \(\alpha^m_n\):
Here we define the coefficients $\gamma_n^m$ as

$$\gamma_n^m = \alpha_n^m - \beta_n^m = \frac{2\hbar \omega_n}{\hbar \omega_m + \hbar \omega_n} \alpha_n^m.$$  

(100)

The system (93)–(96) yields a null-space equation, and as before it is possible to combine factors involving the model coupling tensor into a term involving only the dielectric response:

$$\sum_n G'_{n,n} (\bar{\omega}_m) \gamma_n^m = 0,$$  

(101)

where

$$G'_{n,n} (\bar{\omega}_m) = \frac{(\hbar \bar{\omega}_m)^2 - (\hbar \omega_n)^2}{2\hbar \omega_n} \delta_{n,n} + (\Delta \Gamma)^{m'}_{mn'},$$  

(102)

(\Delta \Gamma)^{m'}_{mn'} = \frac{1}{\epsilon_0} \int dV \mathbf{D}_n (r) \cdot [\Gamma (r, \bar{\omega}_m) - \Gamma_0 (r)] \cdot \mathbf{D}_{n'} (r).$$  

(103)

We note that (\Delta \Gamma)^{m'}_{mn'} = (\Delta \Gamma)^{m'}_{n'n}, due to the permutation symmetry of $\Gamma_{ij}$. Equation (101) plays the role for discrete modes that Eq. (88) plays for the modes at absorbing frequencies. And the homogeneous system of equations (101) plays the role here for the transparent regime that the generalized Hermitian eigenvalue problem (88) plays for the absorptive regime. Solutions to Eq. (101) exist for the frequencies $\bar{\omega}_m, \bar{\omega}_n \neq 0, \Omega_{mn}$, such that $\det G (\bar{\omega}_m) = 0$. The $\gamma_n^m$ are then obtained by solving the system (101), which is linear once $\bar{\omega}_m$ has been found. Note that it is not clear a priori how many solutions there are, and this number is generally not the same as the number of eigenvalues below the absorption cutoff frequency in the nominally system.

Next, we impose canonical commutation relations on the $c_m$. As in the absorptive regime this calculation is lengthy, so we defer details to Appendix A, where it is found that the equal-time commutation relation (64) holds if the dispersive electromagnetic modes are normalized as specified there. It is also shown there that for equal times,

$$[c_m^*, s_{lm}^*] = 0,$$  

(104)

as required.

G. Master equations

We define

$$\vec{D}_m (r) = \sum_n \gamma_n^m \mathbf{D}_n (r),$$  

(105)

$$\vec{B}_m (r) = \sum_n \gamma_n^m \mathbf{B}_n (r),$$  

(106)

which are shown below to be the respective displacement and magnetic induction mode fields of polariton $m$ in the transparent regime.

Equation (101) can be reworked into a master equation in terms of $\mathbf{D}_n (r)$ or $\mathbf{B}_n (r).$ We multiply Eq. (101) by $\omega_n^2$, and make use of Eq. (43) and the nominal system master equation (40) for the displacement field. Employing standard vector identities, this then yields master equations for the $\mathbf{D}$ and $\mathbf{B}$ fields,

$$\frac{\omega_n^2}{c^2} \vec{D}_m (r) - \mathbf{\nabla} \times \mathbf{\nabla} \times [1 - \Gamma (r, \bar{\omega}_m)] \cdot \vec{D}_m (r) = 0,$$  

(107)

$$\frac{\omega_n^2}{c^2} \vec{B}_m (r) - \mathbf{\nabla} \times [1 - \Gamma (r, \bar{\omega}_m)] \cdot \mathbf{\nabla} \times \vec{B}_m (r) = 0.$$  

(108)

It is also possible to obtain equations analogous to Eq. (37). Substituting the expressions (66) and (67) into the first of Eqs. (1), making use of the harmonicity of the $c$ operators to take the time derivative explicitly, and taking the commutator of the resultant equation with $c_m^*$ yields an equation analogous to the first of Eqs. (37); the second can be obtained by from the first in conjunction with Eq. (108). Thus

$$-i \bar{\omega}_m \epsilon_0 \vec{D}_m (r) = \mathbf{\nabla} \times \vec{B}_m (r),$$

$$i \omega_m \epsilon_0 \vec{B}_m (r) = \mathbf{\nabla} \times [1 - \Gamma (r, \bar{\omega}_m)] \cdot \vec{D}_m (r),$$  

(109)

and analogous to Eq. (41), we see that for every mode $m$, there is another mode $\tilde{m}$ with mode fields and frequency given by

$$\vec{D}_{\tilde{m}} (r) = \vec{D}_m (r), \vec{B}_{\tilde{m}} (r) = \vec{B}_m (r),$$

(110)

Eqs. (109) are just the time independent Maxwell curl equations for dispersive media, which can be obtained in naive ways, without a Hamiltonian or reference to a medium. Thus the curl equations (109) and the master equations (107) and (108) are completely model independent, retaining no trace of our microscopic model whatsoever, but depending only on the response tensor $\Gamma$. Therefore the dispersive modes can be obtained by any method at all, for example using a finite difference time domain (FDTD) method [49], a local eigensolver [51], the cutting surface method [52], or an iterative scheme.

What is not obvious from such naive ways of finding the mode fields are their normalization conditions. In terms of the mode fields, the field normalization conditions, derived in Appendix A, become [53]
\[
\frac{1}{\varepsilon_0} \int V \, d\mathbf{D}^*_m(r) \cdot \left[ 1 - \Gamma(r, \omega_m) + \frac{\omega_m}{2} \Gamma''(r, \omega_m) \right] \cdot \mathbf{D}_m(r) = \frac{\hbar \omega_m}{2}, \\
= \frac{\hbar \omega_m}{2}, \quad (111)
\]
\[
\frac{1}{\mu_0} \int V \, d\mathbf{B}^*_m(r) \cdot \mathbf{B}_m(r) + \left( \frac{c}{\omega_m} \right)^2 \frac{1}{\mu_0} \int V \, d\mathbf{V} \times \mathbf{B}^*_m(r) \cdot \mathbf{V} \times \mathbf{B}_m(r) = \frac{\hbar \omega_m}{2}, \quad (112)
\]
where we have defined the material dispersion
\[
\Gamma''(r, \omega_m) = \frac{\partial}{\partial \omega} \Gamma(r, \omega) \bigg|_{\omega_m}. \quad (113)
\]
Note that Eq. (112) follows from Eq. (111) using Eq. (109). By comparison with Eq. (43) for the nondispersive case, we see that in the dispersive case we have obtained an extra term in each normalization condition which is proportional to \( \Gamma'' \), and which is due to the medium contribution. We note here that although only a single frequency derivative appears in Eqs. (111) and (112), the results are exact—they do not rely on perturbation theory. Furthermore, these normalization conditions are independent of the choice of nominal system and are straightforward for the practitioner to use. Having obtained the dispersive modes and dispersion relation using any preferred method, the modes can be normalized using only the known macroscopic response function \( \Gamma(r, \omega_m) \).

Having obtained the \( \gamma^m_n \) or \( \mathbf{D}_m(r) \), the coefficients for the medium component of the polariton mode field are given by Eq. (99), which using the definition of \( \mathbf{D}_m(r) \) can be written in the form
\[
\alpha^{m}_n(r) \quad \beta^{m}_n(r) = -\sqrt{\frac{\hbar}{2e_0 \varepsilon Y}} \left( \frac{1}{\hbar \omega_m + \hbar \Omega'} \right) \mathbf{D}^*_m(r) \cdot \Delta(r, \Omega'). \quad (114)
\]

At this point we can make an interesting connection with earlier work. In 1921, Brillouin [59] derived an expression for the cycle-averaged energy density of electromagnetic fields in dispersive dielectrics, which has been used to quantize the electromagnetic field in dispersive but completely nonabsorptive systems [60]. Brillouin’s derivation made use of slow-variation approximations, including the requirement of near monochromaticity and the rotating wave approximation; in our notation and system of units, and for nonmagnetic media, his expression applied to the cycle-averaged energy of a single excited transparent-regime transverse polariton mode is
\[
\tilde{H}_{BR} = \int V \, d\mathbf{r} \, \frac{1}{\varepsilon_0} \left( \frac{\partial (\omega_m^2(r, \omega))}{\partial \omega} \right) \frac{1}{n^2(r, \omega)} \mathbf{D}^*_m(r) \cdot \mathbf{D}_m(r) + \frac{1}{\mu_0} \mathbf{B}^*_m(r) \cdot \mathbf{B}_m(r), \quad (115)
\]
where \( n^2(r, \omega) = \varepsilon(r, \omega)/\varepsilon_0 \). Performing the integral and making use of the normalization conditions (111) and (112), we find agreement with our result for the energy of a single polariton as obtained from Eq. (58), although here we have not resorted to any approximations in the derivation, and our expressions are not cycle averaged.

Returning now to the absorptive regime, we define
\[
\mathbf{D}_{\Omega m}(r) = \sum_n \gamma^m_n \mathbf{D}_n(r), \quad (116)
\]
\[
\mathbf{B}_{\Omega m}(r) = \sum_n \gamma^m_n \mathbf{B}_n(r), \quad (117)
\]
which we demonstrate below to be the respective \( \mathbf{D} \) and \( \mathbf{B} \) field modes of the polariton \( \Omega_m \), although we note their units differ from those of the displacement and magnetic induction fields because of the continuous frequency index \( \Omega \).

We find that Eq. (88) can also be reworked into a master equation in terms of either \( \mathbf{D}_{\Omega m}(r) \) or \( \mathbf{B}_{\Omega m}(r) \). Following the procedure used to obtain Eqs. (107) and (108) from Eq. (101), Eq. (88) can be written as a master equation for \( \mathbf{D} \) or \( \mathbf{B} \):
\[
\frac{\Omega^2}{c^2} \mathbf{D}_{\Omega m}(r) - \nabla \times \nabla \times \left( [1 - \text{Re} \Gamma(r, \Omega)] \cdot \mathbf{D}_{\Omega m}(r) \right) = \frac{\eta^m_{\Omega m}}{2\pi} \nabla \times \left( [\text{Im} \Gamma(r, \Omega)] \cdot \mathbf{D}_{\Omega m}(r) \right), \quad (118)
\]
\[
\frac{\Omega^2}{c^2} \mathbf{B}_{\Omega m}(r) - \nabla \times \nabla \times \left( [1 - \text{Re} \Gamma(r, \Omega)] \cdot \mathbf{B}_{\Omega m}(r) \right) = \frac{\eta^m_{\Omega m}}{2\pi} \nabla \times \left( [\text{Im} \Gamma(r, \Omega)] \cdot \mathbf{B}_{\Omega m}(r) \right). \quad (119)
\]
Equations (118) and (119) have a number of salient features, which for brevity we consider only for the \( \mathbf{B} \) master equation (119) in what follows. First, since the nominal system mode amplitudes and index profile \( n(r) \) do not appear explicitly in Eq. (119), we see that the eigenvalue \( \eta^m_{\Omega m} \) is independent of the choice of nominal system. Furthermore, while we have introduced an underlying oscillator model to describe the optical response of the medium, in the eigenvalue equation for the polaritons all the details of the oscillator model have disappeared, except that it leads to the physically significant \( \Gamma'(r, \Omega) \).

Second, we note that Eq. (119) can be written in the form
\[
\frac{\Omega^2}{c^2} \mathbf{B}_{\Omega m}(r) - \nabla \times \left( 1 - \Gamma(r, \Omega; \eta^m_{\Omega m}) \right) \cdot \nabla \times \mathbf{B}_{\Omega m}(r) = 0, \quad (120)
\]
with
\[
\Gamma(r, \Omega; Z) = [\text{Re} \Gamma(r, \Omega)] - \frac{Z}{\pi} [\text{Im} \Gamma(r, \Omega)], \quad (121)
\]

where since \( Z \) is real, \( \Gamma(r, \Omega; Z) \) is also real. Equation (120) is just a master equation that would result in the artificial model of a dispersive, \textit{nonabsorptive} dielectric response, with an effective dispersive dielectric tensor given by \( \Gamma(r, \Omega; Z) \) for a given fixed \( Z \).

This means that we can find the modes of the physical absorptive system by solving for the modes and discrete band structure of the family of artificial but nonabsorptive systems characterized by \( \Gamma(r, \Omega; Z) \) for all \( Z \in (-\infty, \infty) \), and thereby scanning the \((Z, \Omega)\) plane (positive \( \Omega \)) for solutions. This is the inverse process of solving the eigenvalue equation (88) for eigenvalues \( Z \) at each \( \Omega \). Yet it yields the same image in the \((Z, \Omega)\) plane. For \( Z=0 \), Eq. (120) becomes just the master equation that arises if absorption is neglected. Thus the mode fields and dispersion relation obtained by making the replacement \( \Gamma(r, \Omega) \rightarrow \text{Re} \Gamma(r, \Omega) \) are solutions to the full absorptive problem with \( Z=0 \).

As in the transparent regime, we can identify effective Maxwell curl equations analogous to Eq. (37), even in the absorptive regime. The derivation is parallel to that which leads to Eq. (109). The equations are

\[
-i \Omega \mu_0 \bar{\mathbf{D}}_{\Omega m}(r) = \nabla \times \bar{\mathbf{B}}_{\Omega m}(r),
\]

\[
i \Omega \epsilon_0 \bar{\mathbf{B}}_{\Omega m}(r) = \nabla \times \left[ [1 - \Gamma(r, \Omega; Z_{\Omega m})] \cdot \bar{D}_{\Omega m}(r) \right],
\]

and for every mode \( \Omega m \), there is a physical conjugate mode \( \Omega \bar{m} \) with mode fields and eigenvalue \( Z_{\Omega \bar{m}} \) given by

\[
(\bar{D}_{\Omega \bar{m}}(r), \bar{B}_{\Omega \bar{m}}(r)) = (\bar{D}_{\Omega m}^*(r), -\bar{B}_{\Omega m}^*(r)),
\]

\[
Z_{\Omega \bar{m}} = Z_{\Omega m}. \quad (123)
\]

In terms of the mode fields \( \bar{D}_{\Omega m}(r) \) and \( \bar{B}_{\Omega m}(r) \), defined in Eqs. (116) and (117), the normalization condition equivalent to Eq. (91) is

\[
\frac{1}{\epsilon_0} \int_{V} dr \bar{D}_{\Omega m}^*(r) \cdot [\text{Im} \Gamma(r, \Omega)] \cdot \bar{D}_{\Omega m}(r) = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega m}^2)^2} \delta_{\Omega m},
\]

\[
\frac{c^2}{\Omega^2 \mu_0} \int_{V} dr (\nabla \times \bar{B}_{\Omega m}^*(r)) \cdot [\text{Im} \Gamma(r, \Omega)] \cdot (\nabla \times \bar{B}_{\Omega m}(r)) = \frac{\pi \hbar}{\pi^2 + (Z_{\Omega m}^2)^2} \delta_{\Omega m}, \quad (124)
\]

\[
\Rightarrow \frac{c^2}{\Omega^2 \mu_0} \int_{V} dr (\nabla \times \bar{B}_{\Omega m}^*(r)) \cdot [\text{Im} \Gamma(r, \Omega)] \cdot (\nabla \times \bar{B}_{\Omega m}(r))
\]

\[
= \frac{\pi \hbar}{\pi^2 + (Z_{\Omega m}^2)^2} \delta_{\Omega m}, \quad (125)
\]

as derived in Appendix A. Note that Eq. (125) follows from Eq. (124) using Eq. (122), and furthermore that like the eigenvalue equation (118), the normalization conditions are independent of the choice of nominal system.

The coefficients that identify the contribution from the medium fields to the TP polariton mode amplitudes \( c_{\Omega m} \) can be written in a simple form using the \( \mathbf{D}_{\Omega m}(r) \). Equations (82) and (84) reduce to

\[
\alpha_{\Omega m}(r) = - \sqrt{\frac{\hbar}{2 \epsilon_0 \Omega}} \left( \frac{1}{\hbar \Omega - \hbar \Omega'} + Z_{\Omega m} \delta(\hbar \Omega - \hbar \Omega') \right) \times \mathbf{D}_{\Omega m}(r) \cdot \Lambda(r, \Omega'),
\]

\[
\beta_{\Omega m}(r) = - \sqrt{\frac{\hbar}{2 \epsilon_0 \Omega'}} \left( \frac{1}{\hbar \Omega' + \hbar \Omega} \right) \mathbf{D}_{\Omega m}(r) \cdot \Lambda(r, \Omega'),
\]

so that given the electromagnetic component mode field \( \mathbf{D}_{\Omega m}(r) \) of a polariton, the medium component field follows immediately.

\section*{H. Inverse transformation}

Having obtained the canonical transformation to the polariton basis, we now determine the inverse transformation. The procedure is to expand the medium and nominal system operators in polariton operators, then calculate the various commutators of the nominal system and medium operators with the polariton operators two ways: once using the expansion (68), (75), and (92) of the polariton operators in terms of nominal system and medium operators, and once using the expansion of the nominal system and medium operators in terms of the polaritons. Comparing these two results, and then using the relations (80) and (97), yields the inverse transformation

\[
a_m = \sum_n \int_{\Omega g} d\Omega \left( (\alpha_{\Omega m}^*) c_n - (\beta_{\Omega m}^*) c_n^\dagger \right)
\]

\[
+ \sum_n \left\{ (\alpha_{m}^*) c_n - (\beta_{m}^* c_n^\dagger \right\},
\]

\[
\psi_{\Omega m}(r) = \sum_m \int_{\Omega g} d\Omega' \left( (\alpha_{\Omega m}^*) c_m - (\beta_{\Omega m}^*) c_m^\dagger \right)
\]

\[
+ \sum_m \left\{ (\alpha_{m}^*) c_m - (\beta_{m}^* c_m^\dagger \right\} s_{\Omega m},
\]

Making use of the inverse transformation (128) and the definitions (116) and (105) of \( \mathbf{D}_{\Omega m}(r) \) and \( \mathbf{B}_{\Omega m}(r) \) in the expression (45) for the displacement field \( \mathbf{D}(r,t) \) in terms of the nominal system operators and modes \( a_m \) and \( \mathbf{D}_m(r) \), we obtain the expression (66) for the displacement field in terms of the polariton operators and polariton displacement field modes. Similarly, we obtain the corresponding expression (67) for the field \( \mathbf{B}(r,t) \) in terms of polariton operators and polariton \( \mathbf{B} \) field modes. Clearly, \( \mathbf{D}_{\Omega m}(r) \) and \( \mathbf{B}_{\Omega m}(r) \) are the displacement and magnetic induction field modes for the polariton \( c_{\Omega m} \), and \( \mathbf{D}_m(r) \) and \( \mathbf{B}_m(r) \) are mode fields for the \( c_m \). We note that the dispersive (tilde) electromagnetic mode
fields provide a nonorthogonal basis for the physical electromagnetic fields.

I. Other fields

Here we briefly describe the electric and polarization fields in terms of the polariton modes. Substituting Eqs. (15) and (129) into the expression (19) for the polarization field, then making use of Eqs. (28), (29), (71), (110), (114), (121), (123), (126), and (127) yields an expression for the polarization, separated into transverse and longitudinal pieces as

$$\mathbf{P}(r, t) = \mathbf{P}_T(r, t) + \mathbf{P}_L(r, t),$$

where

$$\mathbf{P}_T(r, t) = \sum_m c_m \mathbf{\tilde{P}}_m(r) + \sum_m \int_{\Omega} d\Omega c_{\Omega m} \mathbf{\tilde{P}}_{\Omega m}(r) + \text{c.c.},$$

$$\mathbf{P}_L(r, t) = \sum_m \int_{\Omega} d\Omega \xi_{\Omega m} \mathbf{\tilde{P}}^{(L)}_{\Omega m}(r) + \text{c.c.},$$

with the mode polarization fields given by

$$\mathbf{\tilde{P}}_m(r) = \Gamma(r, \omega_m) \cdot \mathbf{D}_m(r),$$

$$\mathbf{\tilde{P}}_{\Omega m}(r) = \Gamma(r, \Omega; Z^{\Omega m}) \cdot \mathbf{D}_{\Omega m}(r),$$

$$\mathbf{\tilde{P}}^{(L)}_{\Omega m}(r) = \sqrt{\frac{\hbar}{2\Omega}} (\nabla \phi^{\Omega m}(r))^*. \tag{135}$$

It then follows that

$$\mathbf{E}(r, t) = \frac{1}{\varepsilon_0} [\mathbf{D}(r, t) - \mathbf{P}(r, t)] = \mathbf{E}_T(r, t) + \mathbf{E}_L(r, t),$$

where

$$\mathbf{E}_T(r, t) = \sum_m c_m \mathbf{\tilde{E}}_m(r) + \sum_m \int_{\Omega} d\Omega c_{\Omega m} \mathbf{\tilde{E}}_{\Omega m}(r) + \text{c.c.},$$

$$\mathbf{E}_L(r, t) = \sum_m \int_{\Omega} d\Omega \xi_{\Omega m} \mathbf{\tilde{E}}^{(LP)}_{\Omega m}(r) + \text{c.c.},$$

with

$$\mathbf{\tilde{E}}_m(r) = \frac{1}{\varepsilon_0} \left\{ [1 - \Gamma(r, \omega_m)] \cdot \mathbf{D}_m(r) \right\},$$

$$\mathbf{\tilde{E}}_{\Omega m}(r) = \frac{1}{\varepsilon_0} \left\{ [1 - \Gamma(r, \Omega; Z^{\Omega m})] \cdot \mathbf{D}_{\Omega m}(r) \right\}, \tag{139}$$

$$\mathbf{\tilde{E}}^{(LP)}_{\Omega m}(r) = \sqrt{\frac{\hbar}{2\Omega\varepsilon_0}} (\nabla \phi^{\Omega m}(r))^*, \tag{140}$$

so that \(\mathbf{\tilde{E}}_{\Omega m}(r)\) is of the form expected from Eqs. (109) and (122).

Whereas the Hamiltonian (8) with commutators (10) and (14), yields dynamics of the Maxwell equations augmented by a fluctuation polarization, here we find that the polariton mode fields satisfy the time-independent Maxwell equations with an effective dielectric response \(\Gamma(r, \Omega; Z^{\Omega m})\), but without any additional fluctuation term.

Of course, \(\mathbf{H}\) field expressions are obtained from corresponding \(\mathbf{B}\) field expressions through division by \(\mu_0\).

V. SYSTEMS WITH CONTINUOUSLY LABELED MODES

For many systems of interest, the modes are labelled continuously or both continuously and discretely in combination. Examples include waveguides, Bragg gratings, photonic crystals, multilayer thin films, coupled microring structures, and periodic quantum well structures. Often the modes of interest are labeled by a discrete-continuous pair \((u, \mathbf{k})\), where \(\mathbf{k}\) is the crystal wave vector and \(u\) is a “band index.”

Then in the equations of Sec. IV, the discrete mode index \(m\) is replaced by \(m \rightarrow (u, \mathbf{k})\). As an example, we consider photonic crystals of arbitrary dimensionality; the equations involving other systems with discrete-continuous labeling can be derived analogously. Since a related discussion for non-dispersive, nonabsorptive systems was presented earlier [45], here we focus only on the full dispersive system (tilde) modes, which are solutions to the master equations described in Sec. IV.

Systems with continuously labeled modes arise from systems subject to periodic boundary conditions for which the normalization region \(V\) is extended infinitely in one or more orthogonal directions. The cross section of the original finite \(V\) is always invariant in any direction perpendicular to the direction(s) of extent. Then the extended \(V\) can be written as \(V = D_\infty \otimes F\), where \(F\) is a finite “cross section” in dimensions in which \(V\) is of finite extent, and \(D_\infty\) is the subspace of the dimensions of infinite extent, which has dimensionality \(d \in \{1, 2, 3\}\).

For these continuous systems, the derivation of Sec. IV holds, but subject to the extension of \(V\), and replacement of summations according to

$$\sum_m \rightarrow \int d^d k,$$ \tag{142}

where the integral is over the region of reciprocal space corresponding to the real space region \(D_\infty\). Making these replacements requires simultaneous modification of the other quantities introduced in Sec. IV, which involve a factor \(C_\infty\) which depends on \(D_\infty\) according to

$$D_\infty \quad C_\infty$$

$$i \text{ axis} \quad \frac{L_i}{2\pi}$$

$$ij \text{ plane} \quad \frac{A_{ij}}{(2\pi)^2}$$
\[ ij \text{ space } \frac{V}{(2\pi)^3} \]  

(143)

where \( L_i \) is the extent of the original finite \( V \) in the \( i \) direction, \( A_{ij} \) is the area of the original finite \( V \) in the \( ij \) plane, and recall that \( V \) is the volume of the original finite normalization region \( V \). In the interest of brevity, we quote only end results whenever possible, and in what follows we work out the continuous-\( k \) equations in the context of a photonic crystal.

A. Example: Photonic crystal

Photonic crystals [61] are a particularly interesting examples of inhomogeneous media, defined by the property

\[ \Gamma^{ij}(r + R, \Omega) = \Gamma^{ij}(r, \Omega) \]

(144)

where \( R \) is any lattice vector. Interesting systems exist for one-dimensional (1D), 2D, and 3D periodic lattices. We thus take the coupling constant and nominal index to have the periodicity of the lattice:

\[ \Lambda^{ij}(r + R, \Omega) = \Lambda^{ij}(r, \Omega), \]

\[ \Gamma^{ij}_{\Omega}(r + R) = \Gamma^{ij}_{\Omega}(r). \]

(145)

Numerous codes for the calculation of nondispersive, nonabsorptive band structures are freely available [50,51], so it is easy to obtain the nominal bands and nominal system modes.

As discussed above, we write \( m \rightarrow (u, k) \), where \( k \) is the wave vector which is restricted to the first Brillouin zone, and \( u \) is a discrete band index. Under the transition from discrete to continuous \( k \), the governing equations (109) and (122) and thus also the master equations (107) and (108) are unchanged. They retain their physical conjugate properties (110) and (123), and the physical conjugate to a nominal mode \((u, k)\) is another mode at \(-k\). Using this property to label the bands at \(-k\), we can write

\[ \bar{(u, k)} = (u, -k). \]

(146)

So the modes are first determined in one half of \( k \) space (e.g., in three dimensions, the first Brillouin zone for \( k_z \geq 0 \)), with the modes in the other half of \( k \) space given by

\[ \bar{(D}_{\bar{u}(-k)}(r), \bar{B}_{\bar{u}(-k)}(r)} = (\bar{D}_{u}(r), \bar{B}_{u}(r)) \]

(147)

with \( \bar{\omega}_{u(-k)} = \bar{\omega}_{u} \). The analogous relation holds between the \( \bar{D}_{\Omega}(r) \) and \( \bar{B}_{\bar{\Omega}}(r) \).

The electromagnetic fields are then expanded in terms of polariton operators and mode fields as

\[ \mathbf{D}(r, t) = \sum_{u} \int d^d k [c_{u k} \bar{D}_{u k}(r) + c_{u k}^\dagger \bar{D}_{u k}^*(r)] \]

\[ + \sum_{u} \int d^d k \int_{\Omega} \tilde{\Omega} [c_{\bar{\Omega} u k} \bar{D}_{\bar{\Omega} u k}(r) + c_{\bar{\Omega} u k}^\dagger \bar{D}_{\bar{\Omega} u k}^*(r)], \]

(148)

where the \( k \) integrals are over the \((d\)-dimensional\) first Brillouin zone, and where we have used Eq. (147). The (equal-time) canonical commutation relations are then

\[ [c_{u k}^\dagger, c_{u' k'}] = \delta_{uu'} \delta(k - k'), \]

\[ [c_{\bar{\Omega} u k}^\dagger, c_{\bar{\Omega} u' k'}] = \delta_{uu'} \delta(\bar{\Omega} - \bar{\Omega}') \delta(k - k'), \]

\[ [s_{\bar{\Omega} u k}, s_{\bar{\Omega} u' k'}] = \delta_{uu'} \delta(\bar{\Omega} - \bar{\Omega}') \delta(k - k'), \]

(149)

(150)

(151)

where \( \delta(k - k') \) is a delta function with dimensionality \( d \), and where all other equal-time commutators between the \( c \) and \( s \) operators vanish. The mode fields are normalized according to

\[ \frac{1}{\varepsilon_0} \int_V dr \bar{D}_{ak}^*(r) \cdot \left[ 1 - \Gamma(r, \bar{\omega}_{ak}) + \frac{\hbar}{2} \Gamma^\dagger(r, \bar{\omega}_{ak}) \right] \bar{D}_{ak}(r) \]

\[ = \frac{\hbar \bar{\omega}_{ak}}{2} \delta(k - k'), \]

(152)

\[ \frac{1}{\varepsilon_0} \int_V dr \bar{D}_{\bar{\Omega} u k'}^*(r) \cdot [\mathrm{Im} \Gamma(r, \bar{\Omega})] \bar{D}_{\bar{\Omega} u k}(r) \]

\[ = \frac{\pi \hbar}{\pi^2 + (Z_{\bar{\Omega} u k'})^2} \delta(\bar{\Omega} - \bar{\Omega}') \delta(k - k'). \]

(153)

The normalization of the \( \bar{B}_{ak} \) and \( \bar{B}_{\bar{\Omega} u k} \) fields follows from Eqs. (109) and (122). Since the full dielectric function \( \Gamma^{ij}(r, \Omega) \) is periodic, Bloch’s theorem applied to the master equations (107) and (108) guarantees the existence of mode fields in Bloch form. Choosing convenient scaling factors, we then write the \( \bar{D}_{ak}(r), \bar{B}_{ak}(r), \bar{D}_{\bar{\Omega} u k}(r), \) and \( \bar{B}_{\bar{\Omega} u k}(r) \) in Bloch form as

\[ \bar{D}_{ak}(r) = \sqrt{\frac{C_{ak}}{V}} \bar{d}_{ak}(r) e^{i k r}, \]

\[ \bar{B}_{ak}(r) = \sqrt{\frac{C_{ak}}{V}} \bar{b}_{ak}(r) e^{i k r}, \]

\[ \bar{D}_{\bar{\Omega} u k}(r) = \sqrt{\frac{C_{\bar{\Omega} u k}}{V}} \bar{d}_{\bar{\Omega} u k}(r) e^{i k r}, \]

\[ \bar{B}_{\bar{\Omega} u k}(r) = \sqrt{\frac{C_{\bar{\Omega} u k}}{V}} \bar{b}_{\bar{\Omega} u k}(r) e^{i k r}, \]

(154)

where lower case characters represent the periodic Bloch fields, which have the periodicity of the lattice; so for any lattice vector \( R \),

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\( \tilde{d}_{ak}(r) = -\tilde{d}_{ak}(r + R) \),

\( \tilde{b}_{ak}(r) = -\tilde{b}_{ak}(r + R) \),

and the same holds for \( \tilde{d}_{\Omega m k} \) and \( \tilde{b}_{\Omega m k} \). Normalization of these periodic Bloch fields involves a unit cell \( V_{\text{cell}} \), which can be chosen to be any region which tiles the full normalization region \( V \) via translations by lattice vectors.

In the transparent regime, the normalization condition for the \( \tilde{d}_{ak}(r) \) is

\[
\frac{1}{\varepsilon_0} \int_{V_{\text{cell}}} \frac{dr}{V_{\text{cell}}} \tilde{d}_{ak}(r) \cdot \left( 1 - \Gamma'(r, \tilde{\omega}_{ak}) + \frac{\tilde{\omega}_{ak}}{2} \Gamma'(r, \tilde{\omega}_{ak}) \right) \tilde{d}_{ak}(r) = \frac{\hbar \tilde{\omega}_{ak}}{2},
\]

where \( \Gamma' \) is defined in Eq. (113), and where the spatial integral is over the unit cell, which has volume \( V_{\text{cell}} \). In the absorptive regime, the normalization condition is

\[
\frac{1}{\varepsilon_0} \int_{V_{\text{cell}}} \frac{dr}{V_{\text{cell}}} \tilde{d}_{\Omega m k}(r) \cdot \left[ \text{Im} \Gamma'(r, \Omega) \right] \cdot \tilde{d}_{\Omega m k}(r) = \frac{\pi \hbar}{\pi + (Z_{\Omega m k})^2} \delta_{\Omega m k}.
\]

We note that under a transition from discrete to continuous wave vectors, the variables which appear in Secs. I–IV are slightly modified as follows. Below, we use \( \tilde{Q} \) to denote a generic quantity with discretely labeled wave vector and \( Q \) to denote the corresponding quantity with continuously labeled wave vector. Then the transition from discrete to continuous labeling is given by

\[
\tilde{Q} \rightarrow \left( \frac{1}{N^2} \right)^{\frac{1}{2}} Q
\]

for some \( N \); \( N=1 \) gives the transition for \( \tilde{Q} \) which is any of the variables \( \omega_{ak} \), \( \tilde{D}_{ak}(r) \), \( \tilde{B}_{ak}(r) \), \( \alpha_{ak}(r, \Omega) \), \( c_{ak} \), \( \tilde{c}_{ak} \), \( \gamma_{ak} \), \( \rho_{\Omega m k} \), \( \phi_{\Omega m k} \), \( \beta_{\Omega m k} \), \( \gamma_{\Omega m k} \), \( \alpha_{\Omega m k} \), \( \beta_{\Omega m k} \), \( \gamma_{\Omega m k} \), \( \alpha_{\Omega m k} \), \( \beta_{\Omega m k} \), and \( \gamma_{\Omega m k} \); and \( N=2 \) gives the transition for \( \alpha_{nk} \), \( \beta_{nk} \), \( \gamma_{nk} \), \( \rho_{nk} \), \( \phi_{nk} \), \( \beta_{nk} \), and \( \gamma_{nk} \).

VI. CALCULATING POLARITONS: SUMMARY AND USER’S GUIDE

We emphasize here that the effort in constructing the formalism has yielded a theory that is easy to use in the end. Calculating the polariton modes is in fact straightforward, and here we review the process of calculating polariton modes for artificially structured inhomogeneous media. Most importantly, in both the transparent and absorbing regimes, the photonic component of the polariton modes can be obtained from the dielectric response function, after which the corresponding medium field component modes are obtained trivially; together a photonic component and its associated medium field comprise the components of a polariton. For the sake of simplicity, we take the dielectric response to be isotropic in this section: \( \varepsilon_i(r, \omega) = \varepsilon(r, \omega) \delta_{ij} \). We employ box normalization, and seek first the photonic component of the polariton modes of the system. We label modes by \( m \); for example, for a photonic crystal we might have \( m = (u, k) \), where \( u \) is a band index and \( k \) denotes the wave vector, which until Sec. VI C we take to be a discrete label.

We suppose the user is given a causal \( \varepsilon(r, \omega) \) that describes the system of interest. In this paper we work in terms of a response function \( \Gamma(r, \omega) \) defined in Eq. (6), which we take to be isotropic in this section, and which is constructed from \( \varepsilon(r, \omega) \) using

\[
\Gamma(r, \omega) = \left( 1 - \frac{\varepsilon_0}{\varepsilon(r, \omega)} \right) \delta_{ij}.
\]

The coupling tensor of the underlying model of the medium is chosen to satisfy Eq. (29), in which it is defined in terms of the dielectric response function; thus

\[
\Lambda(r, \omega) = \frac{2 \omega}{\pi} \text{Im} \Gamma(r, \omega).
\]

The electromagnetic fields are given in terms of the polariton basis by

\[
\begin{align*}
\mathbf{D}(r, t) &= \sum_m [c_m \tilde{D}_m(r) + c_m^* \tilde{D}_m^*(r)] \\
&\quad + \sum_m \int d\Omega [c_{\Omega m} \tilde{D}_{\Omega m}(r) + c_{\Omega m}^* \tilde{D}_{\Omega m}^*(r)], \\
\mathbf{B}(r, t) &= \sum_m [c_m \tilde{B}_m(r) + c_m^* \tilde{B}_m^*(r)] \\
&\quad + \sum_m \int d\Omega [c_{\Omega m} \tilde{B}_{\Omega m}(r) + c_{\Omega m}^* \tilde{B}_{\Omega m}^*(r)],
\end{align*}
\]

where the \( \tilde{D}_m(r) \) and \( \tilde{D}_{\Omega m}(r) \) are the respective mode fields of the polaritons in the transparent and absorbing frequency regimes, and the frequency integral is taken over the absorbing regime.

A. Transparent (dispersive) regime

First we consider calculating the mode fields and eigenfrequencies in the nonabsorbing regime, i.e., for frequencies at which \( \text{Im} \varepsilon(r, \omega) \) vanishes. The dispersion relation \( \tilde{\omega}_m \) and electromagnetic field modes \( \tilde{B}_m(r) \) of the system described by \( \varepsilon(r, \omega) \) are calculated according to the usual Maxwell master equation (108) using any preferred method, such as an FDSD method [49], a local eigensolver [51], or an iterative scheme. For iterative schemes, the modes of a nominal, nondispersive and nonabsorptive medium characterized by an index profile \( n_0(r) \) can be used to provide initial guesses for the modes. The \( \tilde{D}_m(r) \) are then obtained from the \( \tilde{B}_m(r) \) using Eq. (109). The normalization condition on the displacement fields (111) takes the form
The modes are found by scanning through values of the parameters of the system, and the eigenvalue is a quantity which represents the electromagnetic fields dressed by the medium, rather than the other way around. This differs from the nondispersive case by the appearance of the second term in the square brackets.

If so desired, the associated medium fields can be explicitly constructed with little effort from Eq. (114) using Eq. (160).

B. Absorbing regime

At frequencies at which the medium is absorbing, the polariton spectrum is continuous, with solutions at all frequencies—even for a single given wave vector, for example. At frequencies close to those of the bands of the limiting nonabsorptive but dispersive structure, the polariton modes typically have more electromagnetic character, while away from the dispersive bands, the modes have more medium field character to them. Since the spectrum is continuous, the frequency $\omega$ is no longer an eigenvalue, and instead the eigenvalue is a quantity $Z$, which is proportional to the contribution to the polariton from medium fields with natural frequency equal to that of the polariton.

Here are two methods for calculating polaritons:

Method (i). This is a method that can easily leverage codes for the photonic modes of dispersive systems. It is shown in Sec. IV that in the absorptive regime, the polariton mode displacement fields are solutions to the dispersive, nonabsorptive Maxwell master equation, but with an effective $Z$-dependent dielectric tensor $\epsilon(\mathbf{r}, \omega; Z)$. Thus

$$\nabla \times \frac{\epsilon_0}{\epsilon(\mathbf{r}, \omega; Z)} \cdot \nabla \times \mathbf{B}_{\text{m}}(\mathbf{r}) = \frac{\omega^2}{c^2} \mathbf{B}_{\text{m}}(\mathbf{r}),$$

with $\epsilon_0/\epsilon(\mathbf{r}, \omega; Z) = 1 - \Gamma(\mathbf{r}, \omega; Z)$, with

$$\Gamma(\mathbf{r}, \omega; Z) = [\text{Re} \Gamma(\mathbf{r}, \omega)] - \frac{Z}{\pi}[\text{Im} \Gamma(\mathbf{r}, \omega)].$$

The modes are found by scanning through values of the parameter $Z$ (which are real, $-\infty < Z < \infty$), and solving the dispersive Maxwell master equation (164) for eigenvalues $\omega$ and eigenmodes which give the displacement fields of the modes. Often, the modes with small $|Z|$ are the ones of most interest; they span the band width around the limiting dispersive but nonabsorptive band, and that is the band width which represents the electromagnetic fields dressed by the medium, rather than the other way around.

The displacement field modes are obtained from the $\mathbf{B}_{\text{m}}(\mathbf{r})$ using Eq. (122), and are then normalized using Eq. (124):

$$\frac{1}{\epsilon_0} \int_{\mathcal{V}} d\mathbf{r} \mathbf{D}'_{\text{m}}(\mathbf{r}) \cdot [\text{Im} \Gamma(\mathbf{r}, \omega)] \cdot \mathbf{D}_{\text{m}}(\mathbf{r}) = \frac{\pi \hbar}{\pi^2 + Z^2} \delta_{\text{m}m'},$$

where $Z$ is the eigenvalue $Z_{\text{m}m'}$.
gins with a calculation of the dispersive photonic component modes \( \mathbf{D}_{\mathbf{k}}(\mathbf{r}) \) and \( \mathbf{B}_{\mathbf{k}}(\mathbf{r}) \). Here is how it is done: given \( \Gamma(\mathbf{r},\omega) \), the dielectric constant \( \varepsilon(\mathbf{r},\omega) \) is calculated using Eq. (159). The photonic bands are then found using the MIT Photonic Bands Package (MPB) [50], a freely available frequency domain eigensolver for the photonic band structure and modes of nondispersive, nonabsorptive systems. An outer iterative loop over the MPB code is used to solve for the dispersive modes. If the code is thought of at each \( \mathbf{k} \) as a mapping from a dielectric function to a set of mode frequencies, i.e., MPB: \( \varepsilon(\mathbf{r},\omega) \rightarrow \{\tilde{\omega}_n\} \), then the iterative loop seeks fixed points at which for some \( u, \tilde{\omega}_u = \omega \). These give the dispersive mode frequencies, and their associated eigenvectors give the dispersive photonic modes, which are just the photonic component of the polariton modes. The nominal crystal frequencies are used as a starting point for the iteration at the first \( \mathbf{k} \) point, and the frequencies obtained at a given \( \mathbf{k} \) point are used as a starting point for the subsequent adjacent \( \mathbf{k} \) point. The modes are normalized according to Eq. (163). The TM (E parallel to rod axis) dispersive band structure is plotted in black in Fig. 4, along with the nominal crystal band structure in pink (online).

We demonstrate that the medium components of the polariton modes have reasonable behavior by calculating them from the photonic components of the mode fields and eigenvalues according to Eq. (114). To make a definite calculation, we assume the dielectric material of the photonic crystal is a semiconductor with an electronic band gap of \( \omega_c a/2\pi c = 0.85 \). Now consider any dispersive mode with wave vector \( \mathbf{k} \) and unitless frequency \( \tilde{\omega}_n a/2\pi c = 0.8 \) (dashed line in Fig. 5), which is just below the electronic band gap. In Fig. 5 we plot the magnitudes of the corotating and countercorotating medium coefficients \( |\alpha_{\omega}^{n\mathbf{k}}(\mathbf{r})| \) and \( |\beta_{\omega}^{n\mathbf{k}}(\mathbf{r})| \) as a function of unitless natural oscillator frequency \( \omega a/2\pi c \), in units of \( |(a/2\pi c)\sqrt{\text{Im} \Gamma(\mathbf{r},\omega)/(\hbar\varepsilon_n(\mathbf{r}))}\mathbf{D}_{\omega}^*(\mathbf{r})| \). The patterned area below \( \omega_n \) in Fig. 5 indicates the transparent regime, in which there are no medium oscillators. Other things being equal, the amplitudes of the medium coefficients are larger wherever the displacement mode field is concentrated and wherever the dielectric material is more absorptive at the medium oscillator’s natural frequency. The amplitudes also fall off with increasing frequency away from the polariton frequency.

2. GaAs slab waveguide near the electronic band gap

We next consider a single-polariton state in a dielectric slab waveguide. We take the structure to be composed of a 262-nm-thick GaAs layer, and to be of infinite extent in the \( xy \) plane and bounded by perfect conductors, as depicted in Fig. 6. We take as input the dielectric constant \( \varepsilon(\omega) \) of GaAs. This could come from experimental data, semiempirical formulas, or \textit{ab initio} calculations. Here we take data for \( \varepsilon(\omega) \) based on \textit{ab initio} calculations [62]. The real and imaginary parts of \( \varepsilon(\omega) \), which obey the Kramers-Kronig relations, are

![Fig. 4](image-url)  
**Fig. 4.** (Color online) In-plane polariton band structure for TM modes of 2D photonic crystal with lattice constant \( a \) (black lines with symbols) and the photonic bands of a corresponding nominal, nondispersive crystal (pink lines). The crystal is composed of a triangular lattice of air cylinders of radius \( r/a = 0.417 \) in a dispersive background medium with dielectric response \( \Gamma(\omega) \) shown inset (black curve), along with the nominal nondispersive response (pink), which corresponds to index \( n = 3.25 \). In the frequency range \( \omega a c \in [0,0.66] \) for which the bands are given, the medium exhibits normal dispersion and its index of refraction spans \( n \in [3.1,3.59] \).

![Fig. 5](image-url)  
**Fig. 5.** (Color online) For the dispersive photonic crystal with band structure given in Fig. 4, medium field amplitudes in a transparent-regime polariton with unitless frequency \( \omega a/2\pi c = 0.8 \). The medium contribution is given as \( |\alpha_{\omega}^{n\mathbf{k}}(\mathbf{r})| \) (left scale) and \( |\beta_{\omega}^{n\mathbf{k}}(\mathbf{r})| \) (right scale) in units of \( |(a/2\pi c)\sqrt{\text{Im} \Gamma(\mathbf{r},\omega)/(\hbar\varepsilon_n(\mathbf{r}))}\mathbf{D}_{\omega}^*(\mathbf{r})| \). The patterned area indicates the absence of medium oscillators, i.e., the transparent frequency regime, and the dashed line indicates the frequency of the polariton mode of interest. The electronic gap is at \( \omega_c a/2\pi c = 0.85 \).
given in Fig. 7(a). The corresponding \(\Gamma(\omega)\) is obtained from \(\varepsilon(\omega)\) using Eq. (159), and is plotted in Fig. 7(b). The absorption cutoff frequency \(\omega_c\) is taken to be 1.42 eV/\(h\), which corresponds to the electronic band gap of GaAs from Fig. 7. We consider here only the polaritons with an in-plane (continuous) wave vector \(k=\left(k_x, k_y\right)\), and perpendicular but in-plane displacement field polarization. The normalization region \(V\) comprises the slab region of height \(L_z\) in the \(z\) direction, and the (infinite) \(xy\) plane. The unit cell \(V_{\text{cell}}\) is then taken to be of height \(L_z\), and its extent in the \(xy\) plane can be freely chosen, as described after Eq. (156). Since the electromagnetic fields vanish on the bounding conductors, the periodic boundary condition on \(V\) is guaranteed to hold. Equation (164) is solved readily. We can neglect solutions with radiation incident from outside the waveguide region, since they do not interact with the fields inside the GaAs slab. The electromagnetic component modes inside the waveguide are characterized by \(m=(u,k)\), where \(u \in \{1,2,3,\ldots\}\) gives the \(z\) direction “quantum number” and \(k\) is the in-plane wave vector. Then the eigenvalues \(Z^{\text{vock}}\) are used to normalize the mode fields using Eq. (157).

Assuming for the moment no excitation in the transparent regime, an arbitrary one-polariton state in the Hilbert space is given by

\[
|\Phi\rangle = \sum_{u} \int_{\omega_{g}}^{\omega_{e}} d\omega \int d^{2}k A^{1}_{\text{vock}}(\omega, k)|0\rangle,
\]

where \(|0\rangle\) is the vacuum state, and

\[
\sum_{u} \int_{\omega_{g}}^{\omega_{e}} d\omega \int d^{2}k |A^{1}_{\text{vock}}|^2 = 1.
\]

We consider a single-band “windowed” state from band \(\bar{u}\) centered at the point \((\bar{\omega}, \bar{k})\), which we write as \(|\bar{\omega}, \bar{u}, \bar{k}\rangle\). That is, we take \(A^{1}_{\text{vock}}\) to have support on \(u=\bar{u}\), and on a small window around \((\bar{\omega}, \bar{k})\), with area \(Q\) in 2D \(k\) space and full width \(W\) in frequency space. We consider \(\sqrt{Q}\) and \(W\) to be much smaller than any other inverse length or inverse time scale in the problem, except for the inverse of the evolution time coordinate. Inside the window, \(A^{1}_{\text{vock}}\) is taken to have uniform value \((WQ)^{-1/2}\), as is required to satisfy Eq. (168).

The degree of electromagnetic character of this polariton state is given by the quantity \(V(t)\), which we define as the ratio of electromagnetic energy to total energy, per unit of window energy \(hW\), neglecting zero point energy. Defining \(\mathcal{H}_{\text{nonvac}}(H_{\text{em,nonvac}}(t))\) to be the total (electromagnetic) Hamiltonian neglecting zero-point energy, q.v. (8) and (9), we have

\[
V(t) = \frac{\langle \Phi | H_{\text{em,nonvac}}(t) | \Phi \rangle}{\langle \Phi | H_{\text{nonvac}} | \Phi \rangle} \cdot \frac{1}{hW}.
\]

For a window state \(|\bar{\omega}, \bar{u}, \bar{k}\rangle\) we write this ratio as \(V^{\text{vock}}(t)\).

We note here that the time evolution does not just exhibit loss in electromagnetic energy, \(V^{\text{vock}}(t)\) vanishes in the limit \(t \to -\infty\). Before \(t=0\) the evolution is such that energy flows into the electromagnetic fields. Then for \(t > 0\) the dynamics show a decay in \(V^{\text{vock}}(t)\) as expected. We note also that while \(V^{\text{vock}}(t)\) describes the electromagnetic character of the modes, it does not describe the state obtained by driving the electromagnetic fields with a source, which we will address in a subsequent publication.

In Fig. 8 we plot \(\max_{\bar{u}} V^{\text{vock}}(0)\) for \(\bar{u} \in \{1,2,3\}\), on the \((\bar{\omega}, \bar{k})\) plane \((\bar{\omega} \gg \omega_{c})\). Superimposed are the dotted curves giving \(\omega_{\text{vock}}(u)\) for \(u \in \{1,2,3\}\) for the formal dispersive but non-absorptive system obtained if the absorption is switched off, i.e., \(\Gamma(\omega, u) \to \Gamma^{\text{real}}(\omega, u)\). These curves also indicate points \((\omega, k)\) at which \(Z^{\text{vock}}\) vanishes for the appropriate band \(u\).

We note some other features of Fig. 8. First, polaritons close in frequency to dispersive-limit bands tend to be highly electromagnetic in character, and appear to define broadened bands. A cross section at \(k=25\ \mu\text{m}^{-1}\) is given in Fig. 9. Note also that as a broadened band approaches the electronic
Hamiltonian Treatment of the polarization constant $k/\hbar$ of the corresponding discrete band in the dispersive but nonabsorptive limit, i.e., $\Gamma(\omega) \rightarrow \text{Re} \Gamma(\omega)$.

FIG. 8. (Color online) The lowest three effective broadened polariton bands for a GaAs waveguide with 1D confinement. Brightness indicates the degree of electromagnetic character of the polariton modes in the absorption regime ($\omega \gg \omega_g$). Dotted lines give the transparent regime bands below $\omega_g$; in the absorption regime above $\omega_g$ they give the three lowest bands of the transparent system obtained if absorption is switched off: $\Gamma(\omega) \rightarrow \text{Re} \Gamma(\omega)$.

band-gap frequency, it tapers out and merges with the discrete transparent-regime polariton band, which is indicated by the dotted line at frequencies below the electronic band-gap frequency $\omega_g$.

We demonstrate that the medium components of the mode fields have reasonable behavior by calculating them from the electromagnetic mode fields and eigenvalues according to Eqs. (126) and (127). Consider the second polariton band $m=(u,k)=(2,25 \mu m^{-1})$, for which $\hbar \omega_{\text{disp}}=1.82 \text{ eV}$ is the energy of the corresponding discrete band in the dispersive but nonabsorptive limit, i.e., $\Gamma(\omega) \rightarrow \text{Re} \Gamma(\omega)$.

The medium field excitation is shown in Fig. 10, where $c \text{ max}_r |\alpha_{\omega}^{\text{em}}(r)|$ is plotted as a function of $\hbar \omega'$ for $m=(u,k)=(2,25 \mu m^{-1})$, for polaritons of energies $\hbar \omega =\{\hbar \omega_{\text{disp}}-0.1 \text{ eV}, \hbar \omega_{\text{disp}}, \hbar \omega_{\text{disp}}+0.1 \text{ eV}\}$.

FIG. 9. (Color online) Cross section through Fig. 8 at propagation constant $k=25 \mu m^{-1}$, showing $V_{\text{max}}(0)$ for the lowest three polariton modes as a function of mode energy $\hbar \omega$.

VII. DISCUSSION

We have presented a Hamiltonian formulation of dispersive and absorptive structured (inhomogeneous) media explicitly obeying the Kramers-Kronig relations. The main results for the practitioner are summarized in Sec. VI. Our method focuses on the photonic component of the polariton modes of the system. This has an advantage over noise-current methods in that existing codes and computational methods for photonic band structures can easily be leveraged to calculate the polariton modes while maintaining a Hamiltonian formulation. The medium components of the polariton modes are easily obtained once the photonic components have been determined. This is demonstrated in Sec. VI, in which we construct polaritons for an example 2D dispersive photonic crystal and for a dispersive and absorptive waveguide. Given an arbitrary space- and frequency-dependent dielectric response function, the model and canonical description of the modes of the coupled medium and electromagnetic field can immediately be found.

In our formulation the medium is represented using a natural, intuitive model. There is a clear connection between the coupling constants of the model medium and the dielectric response, and only the susceptibility of the medium appears in the final equations for the polariton modes. The numerical value of the Hamiltonian of the system is equal to its energy, so this formulation gives both quantum and classical results; furthermore, having a Hamiltonian allows conserved quantities to be identified and understood for dispersive and absorptive frequency regimes. Throughout, we have explicitly treated transparent and absorptive frequency regimes, which have qualitatively different polariton characteristics.

In the transparent regime below the (optionally vanishing) absorption cutoff frequency, we obtain a discrete polariton spectrum, and the polariton dispersion relation is exactly the dispersion relation of the electromagnetic modes of the dispersive system. Each polariton mode consists of its dispersive electromagnetic component augmented by the associ-
ated medium excitation component, and the electromagnetic components are obtained from the usual dispersive Maxwell master equation.

In the absorptive regime, we obtain a continuous spectrum of polaritons; however, it is possible to identify effective broadened polariton bands. Furthermore, the electromagnetic component of each polariton mode is found to obey the familiar nonabsorptive Maxwell master equation, but with an effective dielectric function parametrized by the eigenvalue. It is inevitable that a continuous spectrum will arise in the absorptive regime; absorption causes a temporal decay of electromagnetic fields, which implies a linewidth in the (real) polariton spectrum. In contrast with our real, continuous spectrum, it is possible to obtain a discrete spectrum of bands [17], but at the cost of the spectrum being complex, and therefore incompatible with a Hamiltonian formulation.

Since the method utilizes the full modes of the system, it is especially applicable to structured media for which these are a natural way of expressing the fields in the nondispersive, nonabsorptive limit.

Our approach is well-suited to considering the effect of material dispersion and absorption on photonic band gaps in particular systems of interest. Such a question is naturally posed in terms of the band gap of a nondispersive or nonabsorptive limiting photonic crystal being closed or altered by the material dispersion. Since our approach references a nominal crystal, the mechanism for closing and altering gaps can easily be explored within our framework. The method is also especially well suited to the problem of propagation in structured media, where earlier treatments (see, e.g., Ref. [63]) have relied on perturbative approaches. Important linear and nonlinear propagation problems with dispersion include the generation and control of entangled few-photon wave packets in structured dispersive media, second harmonic generation, and spontaneous parametric downconversion; we plan to turn to these issues in future communications.

The model of the medium described here can of course be improved. We do not include the effect of propagation of the medium fields, which would model a spatially dispersive material. However, since our method introduces a physically intuitive model Hamiltonian for these fields, they could be made to propagate by augmenting the Hamiltonian by a suitable kinetic term.

ACKNOWLEDGMENTS

This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) and Photonics Research Ontario. N. A. R. Bhat acknowledges financial support from the University of Toronto. We thank F. Nastos for providing the response function of GaAs on which one of the example calculations is based.

APPENDIX A: CANONICAL COMMUTATORS FOR THE POLARITONS

Here we impose canonical commutation relations on the polariton operators, and obtain normalization conditions on the photonic component mode fields.

1. Polaritons in the absorptive regime

Making use of the expansion (75) in the commutation relation (65), we obtain

\[
\left[ c_{\Omega m \omega}^\dagger c_{\Omega'm'\omega'} \right] = \sum_n \left( \alpha_n^{\Omega m} \alpha_n^{\Omega'm'\omega} - \beta_n^{\Omega m} \beta_n^{\Omega'm'\omega'} \right) + \int_{\Omega_x} d\Omega' \int_{\Omega_y} d\Omega^{\Omega m}(\mathbf{r}) \cdot \alpha_n^{\Omega'm'\omega}(\mathbf{r}) - \beta_n^{\Omega m}(\mathbf{r}) \cdot \beta_n^{\Omega'm'\omega'}(\mathbf{r}),
\]

(A1)

Substitution of Eqs. (81) and (83) in Eq. (A1) yields a useful intermediate result for the electromagnetic part:

\[
\sum_n \left( \alpha_n^{\Omega m} \alpha_n^{\Omega'm'\omega} - \beta_n^{\Omega m} \beta_n^{\Omega'm'\omega'} \right) = \sum_{n,\omega} \gamma_n^{\Omega m}(\frac{\hbar \Omega + \hbar \Omega'}{2 \hbar \omega_{\omega'}}) \delta_{n,\omega} \gamma_n^{\Omega m}.
\]

(A2)

Substitution of Eqs. (82) and (84) into Eq. (A1) gives

\[
\int_{\Omega_x} d\Omega' \int_{\Omega_y} d\Omega^{\Omega m}(\mathbf{r}) \cdot \alpha_n^{\Omega'm'\omega}(\mathbf{r}) - \beta_n^{\Omega m}(\mathbf{r}) \cdot \beta_n^{\Omega'm'\omega'}(\mathbf{r})
\]

\[
= \int_{\Omega_x} d\Omega' \int_{\Omega_y} d\Omega^{\Omega m}(\mathbf{r}) \sum_{n,\omega} \gamma_n^{\Omega m}(\mathbf{r}, \Omega') \Lambda_n^{\Omega m}(\mathbf{r}, \Omega')
\]

\[
\times \left( \frac{1}{\hbar \Omega - \hbar \Omega'} + Z^{\Omega m}(h \Omega - h \Omega') \right)
\]

\[
\times \left( \frac{1}{h \Omega - h \Omega'} + Z^{\Omega m}(h \Omega' - h \Omega') \right)
\]

\[
- \frac{1}{h \Omega + h \Omega'} \cdot \Lambda_n^{\Omega m}(\mathbf{r}, \Omega') \gamma_n^{\Omega m}.
\]

(A3)

Care must be taken in expanding the factors inside the square brackets. Combining the two principal parts involves an extra contribution from the double pole for $\Omega=\Omega'$. This is resolved with the identity [64]

\[
\left( \frac{P}{x-y} \right) \left( \frac{P}{x-y} \right) = \left( \frac{P}{x'-x} \right) \left( \frac{P}{x-y} \right) + \pi^2 \delta(x-y) \delta(x-y).
\]

(A5)

Combination of the two delta functions requires use of the identity

\[
\delta(x-y) \delta(x'-y) = \delta(x-x') \delta(y - \frac{1}{2} (x + x')).
\]

(A6)

The use of Eqs. (A5) and (A6) and more straightforward identities simplifies Eq. (A3); combining this result with Eqs. (A2) in (A1) leads to

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\[ [c_{\Omega m}, c_{\Omega' m'}^\dagger] = \sum_{n, n'} \gamma_{n n'} \left\{ \frac{1}{\hbar \Omega - \hbar \Omega'} \left[ \frac{(\hbar \Omega')^2 - (\hbar \Omega_n)^2}{2\hbar \Omega_n} \delta_{m n} - \frac{\Delta \text{Re} \Gamma}{\pi} (\text{Im} \Gamma)_{m n} + \frac{Z_{\Omega m} \Omega'}{\pi} (\text{Im} \Gamma'_{m n}) \right] + \frac{\pi^2 + Z_{\Omega m} \Omega'}{\pi} \right\} \gamma_{m n}' \] (A.7)

Application of the eigenvalue equation (88) twice gives

\[ [c_{\Omega m}, c_{\Omega' m'}^\dagger] = \delta(\hbar \Omega - \hbar \Omega') \times \left( \frac{\pi^2 + Z_{\Omega m} \Omega'}{\pi} \right) \sum_{n, n'} \gamma_{n n'} \Omega_{n, n'} (\text{Im} \Gamma_{n m}) \gamma_{m n} \] (A.8)

The summand can be simplified by using the orthogonality condition for the generalized Hermitian eigenvalue problem (88), which is

\[ \sum_{n, n'} \gamma_{n n'} \Omega_{n, n'} (\text{Im} \Gamma_{n m}) \gamma_{m n} = \delta_{m m} \] (A9)

for some normalization factor \( \delta_{m m} \). Consistency of Eq. (A8) with Eq. (65) then specifies \( \delta_{m m} \), so that the canonical commutation relation (65) is obtained if the amplitudes are normalized according to Eq. (91). In terms of the mode fields \( \tilde{D}_{\Omega m}(r) \) and \( \tilde{B}_{\Omega m}(r) \), defined in Eqs. (116) and (117), the normalization condition becomes Eqs. (124) and (125).

We note here that the absorptive regime TP polariton operators commute with the LP polariton operators:

\[ [c_{\Omega m}, c_{\Omega' m'}^\dagger] = 0. \] (A10)

This is determined by substitution of the expansions (68) and (75) into the left-hand side of Eq. (A10), use of the commutation relation (17), use of the expression (82) for the medium field coefficients in terms of the nominal system coefficients for the TP polaritons, and the longitudinality of the polarization of the LP modes expressed in Eq. (69). Recall that the expression (82) ensures suppression of the homogeneous solution to Eq. (79).

2. Polaritons in the transparent regime

Expanding \( c_m \) using Eq. (92) in the commutation relation (64) gives

\[ [c_m, c_m^\dagger] = T_{mn} + T_{mn}^{med}, \] (A11)

where

\[ T_{mn}^{med} = \sum_n (\alpha_m^a \alpha_{n m}^{a*} - \beta_m^b \beta_{n m}^{b*}). \] (A12)

The medium term \( T_{mn}^{med} \) gives the electromagnetic and medium contributions, respectively. The electromagnetic contribution can be expressed in terms of the dispersive electromagnetic mode fields as follows: we write the \( \alpha_m^a \) and \( \beta_m^b \) in terms of \( \gamma_n^a \) using Eq. (100); then factor the result and make use of the normalization condition for the nominal modes (43) and apply the system (101) as well, in the form

\[ \sum_{n, n'} \gamma_{n n'}^a G_{n n'}(\tilde{\omega}_n) \gamma_{m n}^a = 0, \] (A14)

where we recall the definition of \( G_{n n'}(\tilde{\omega}_n) \) in the text after Eq. (103). Identifying the dispersive modes defined in Eq. (105), the result is

\[ T_{mn}^{med} = \int_{\Omega}^\infty d\Omega' \int_{\Omega' \Omega} \tilde{D}_{m n}^*(r) \left( \frac{1}{\hbar \tilde{\omega}_m} \right) \left( \frac{1}{\hbar \tilde{\omega}_m} \right) \gamma_{m n} \]

\[ + \left( \frac{1}{\hbar \tilde{\omega}_m} \right) \left( \frac{1}{\hbar \tilde{\omega}_m} \right) \gamma_{m n} \gamma_{m n} \] (A15)

The medium contribution to the commutator in Eq. (A11) is related to the electromagnetic contribution by substituting Eq. (99) into Eq. (A13), yielding

\[ T_{mn}^{med} = \int_{\Omega}^\infty d\Omega' \int_{\Omega' \Omega} \frac{d\Omega' \sum_{m m'} \gamma_{m m'}^a A_{m m'}(r, \Omega')}{\tilde{\omega}_m + \tilde{\omega}_m + \tilde{\omega}_m + \tilde{\omega}_m} \cdot \gamma_{m n}. \] (A16)

For \( m \neq m' \), the rest of the calculation of the commutator (A11) mirrors that for the continuous polariton spectrum; the electromagnetic and medium contributions are combined, and the system (101) is identified twice, yielding [65]

\[ [c_m, c_{m'}^\dagger] = 0; \quad m \neq m'. \] (A17)

For \( m = m' \), the partial fraction expansion cannot be used due to the double pole. Instead, taking \( m = m' \), Eq. (A16) is simplified by using the frequency derivative of the Kramers-Kronig relation (30). Then the medium contribution is found to be

\[ T_{mn}^{med} = \int_{\Omega}^\infty d\Omega' \int \tilde{D}_m^*(r) \cdot \frac{1}{\hbar} \left( \Gamma'(r, \tilde{\omega}_m) \right) \tilde{D}_m(r), \] (A18)

where we recall the definition of the material dispersion \( \Gamma'(r, \tilde{\omega}_m) \) in Eq. (113). With the requirement (64), Eq. (A11), along with Eqs. (A18) and (A15), gives the normalization conditions for the electromagnetic mode fields as Eqs. (111) and (112). The normalization condition (112) for \( \tilde{B}_{m m}(r) \) was
obtained from the normalization condition (111) for $\tilde{D}_m(\mathbf{r})$ by using Eqs. (37) and (38) as well as partial integration. Equation (112) can be written in an alternative form that is sometimes easier to use:

$$\frac{1}{\mu_0 J} \int d\mathbf{r} \tilde{B}_m^*(\mathbf{r}) \cdot \tilde{B}_m(\mathbf{r})$$

$$= \frac{1}{\mu_0} \int d\mathbf{r} \tilde{D}_m(\mathbf{r}) \cdot \left[ 1 - \Gamma(\mathbf{r}, \tilde{\omega}_m) \right] \cdot \tilde{D}_m(\mathbf{r}). \quad (A19)$$

We note that the transparent regime TP polariton operators commute with the LP polariton operators, as stated in Eq. (104). This is determined by following the same line of reasoning as leads to Eq. (A10).

**APPENDIX B: CONSISTENCY CONDITIONS**

Two useful identities for the transformation coefficients can be obtained by substituting the expressions (128) and (129) into the commutation relations (46) and (17). Respectively, these give

$$\delta_{m'} = \sum_m \int d\Omega [\alpha_{n', \alpha}^{\Omega m} \beta_{n', \beta}^{\Omega m} - \beta_{n, \alpha}^{\Omega m} \alpha_{n, \beta}^{\Omega m}]$$

$$+ \sum_m (\alpha_{n', \alpha}^{\Omega m} \alpha_{n', \beta} - \beta_{n, \alpha}^{\Omega m} \beta_{n, \beta}), \quad (B1)$$

and

$$\delta_j \delta(\Omega - \Omega') \delta(\mathbf{r} - \mathbf{r}')$$

$$= \sum_m \left\{ \int_{\Omega}^\infty d\Omega' \alpha_{n', \alpha}^{\Omega m} \alpha_{n', \beta}^{\Omega m} \right\}$$

$$- \beta_{n', \alpha}^{\Omega m} \beta_{n', \beta}^{\Omega m}$$

$$- \beta_{n, \alpha}^{\Omega m} \alpha_{n, \beta}^{\Omega m} \right\}, \quad (B2)$$

where here we refer to components of $\alpha_{n', \alpha}^{\Omega m}$ by writing, e.g., $\alpha_{n', \alpha}^{\Omega m}(\mathbf{r})$, and similarly for $\beta_{n, \alpha}^{\Omega m}(\mathbf{r})$. 

[16] M. Floreascu (private communication).
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...治 by G. V. Eleftheriades and K. G. Balmain (Wiley, New York, 2005).


[53] An alternative form for Eq. (112) is given in Eq. (A19).


[56] We take this opportunity to correct a typographical error in Huttner and Barnett’s [24] Hamiltonian. In their Eq. (2.25b), the expression \( \hat{b}^{\dagger}_a(\lambda, -k, t) + \hat{b}_a(\lambda, k, t) \) should read \( \hat{b}^{\dagger}_a(\lambda, k, t) + \hat{b}_a(\lambda, -k, t) \). In their Eq. (A1), the expression \( \hat{b}^{\dagger}_a(-k) + \hat{b}_a(k) \) should read \( \hat{b}^{\dagger}_a(k) + \hat{b}_a(-k) \). We thank Stephen Barnett [57] for distilling this problem.

[57] S. M. Barnett (private communication).


[62] F. Nastos (private communication).


[65] A similar technique can be used to verify that all commutators between \( c_m \) and \( c_{\Omega m}^\dagger \), vanish.