Canonical formalism and quantization for a class of classical fields with application to radiative atomic decay in dielectrics

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For the description of spectral and radiative decay properties of atoms or molecules, placed in a photonic material, the electromagnetic field in the material must be quantized and active research is taking place in this area. Here a unified account is given of such quantization procedures. Led by the Maxwell example, we consider the canonical formalism and its quantization for a class of linear evolution equations $\partial_t F = NF - G$, obeying a conservation law for $G = 0$. If N has a nonempty null space (zero is an eigenvalue with associated nonpropagating solutions), an abstract form of the gauge concept makes its appearance and generalizations of the familiar Coulomb and Lorentz gauges are obtained. A canonical formalism is set up and quantized. The application to spatially inhomogeneous nonconducting electrodynamical systems is immediate, including the interaction with matter. Next atomic decay in a medium is considered, in particular in the presence of band gaps. For a simple two-level model with transition frequency in the gap, single-photon decay is inhibited and also a different stable eigenvalue of the combined system is found. An open problem in connection with random dielectrics, showing Anderson localization, is discussed. Finally, a mass renormalization, by means of a Kramers transformation, is presented. In general, the renormalized mass is no longer a scalar quantity. [S1050-2947(97)00412-5]

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I. INTRODUCTION

A. Physical background

The prime example, motivating the present investigation, consists of Maxwell's equations in a nonconducting, spatially inhomogeneous medium. The latter have recently become a subject of active research in view of the emergence of photonic crystals [1]. These are classical dielectric media, characterized by a periodic dielectric permeability, the periodicity giving rise to a band structure analogous to that found for electrons in solid-state physics. Such systems have become of interest in view of the possibility to alter the radiative decay properties of embedded atoms relative to their vacuum values. Possible technological applications are in the fields of semiconductor lasers, photovoltaic elements, and quantum computers.

Indeed, if a band gap is present and an embedded atom has a transition frequency in the gap, one expects that radiative decay does not take place by emission of a single photon (the emitted radiation cannot propagate away). However, even if no gap develops, decay rates may change appreciably from their free values due to alterations in the photonic density of states. As shown by Sprik *et al.* [2], for a scalar field model, the former can be obtained through an application of Fermi's golden rule after quantization of the field and turn out to be proportional to the local density of states $N_f(\mathbf{X})$ of the field modes, a quantity that in essence measures the magnitude of the field modes at the atomic position $\mathbf{X}$. In a band gap $N_f(\mathbf{X})$ vanishes and hence no decay takes place for transition frequencies in the gap.

Radiative decay of atoms placed in *random* dielectrics has hardly been studied (for random electromagnetic wave propagation see [3]). This may be an interesting matter since, if the dielectric shows (Anderson) localization, decay with transition frequency in a localization interval is once more expected to be inhibited. Here we can think of Lifshitz tails in the gap of a randomized photonic crystal. An obvious question is the dependence of the decay parameter on the size of a finite, randomized crystal that would exhibit localization in the infinite limit.

The emerging technique of making photonic crystals of cool atoms in crossed laser beams [4] is particularly interesting. Since atomic diameters are much smaller than the lattice distance, the atoms can be considered as point scatterers and recent theoretical work by van Coevorden *et al.* [5] shows the existence of a band gap for such systems. At the moment the fraction of occupied lattice sites in experimental setups is still 10% or less (with a random distribution over the sites, in its simplest form of Bernouilli type, but the existing experimental evidence does not rule out correlated behavior), but as this increases an interesting random situation develops.

Another promising approach is the use of colloid techniques to manufacture photonic crystals [6]. In this case there is no severe limitation on the packing fraction and the method allows the deposition of a variety of dielectric scatterers on the lattice sites. The embedding of molecules for the study of decay rates and randomization of the crystal are possible as well.

A second type of photonic structure is encountered within the context of transition radiation [7], the emission of electromagnetic radiation caused by the passage of fast electrons moving through layered dielectrics. At present this mechanism is studied as a tool to produce x-ray radiation for diagnostic purposes [8]. So far theoretical work on this phenomenon has been mainly classical. A second quantized approach can give significant simplifications since now we are dealing with a scattering process (from an electron with

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zero photons in to an electron with one photon out) and a first order calculation suffices.

The calculation of radiative decay rates for atoms or molecules placed in dielectrics requires quantization of the dielectric and active research is taking place in this field [9–12]. Thus an appropriate Lagrange and Hamilton formalism is set up, which is then quantized by replacing Poisson brackets by commutators according to Dirac's recipe. The precise procedure varies from one author to the other and usually the magnetic permeability is set equal to its vacuum value. Compared to the situation for free (constant permeabilities) fields, there is a snag. The point is that in a general dielectric the longitudinal fields and the nonpropagating fields differ. Among other matters, this makes the usual Coulomb gauge less suitable.

B. General canonical formalism and its quantization

The above situation has led us to construct a general approach to the quantization of linear evolution equations through the Lagrange-Hamilton procedure. If they possess a conservation law (energy conservation in the electromagnetic case), a suitable inner product can be defined, leading to a unitary time evolution in the corresponding Hilbert space. In Sec. II we give a few examples. Thus we consider

$$\partial_t F(t) = NF(t) - G(t), \quad N^* = -N, \tag{1.1}$$

in a separable, real Hilbert space. Here the anti-self-adjointness of N reflects the conservation property in the absence of $G(t)$. At this point a canonical setup looks straightforward, but the Hamiltonian does not necessarily correspond to the above conserved quantity. Being of minor importance in a ''stand alone'' system, this is not satisfactory if a coupling with other systems is contemplated. However, amends can be made and at the same time it becomes clear that the existence of nonpropagating modes (a nonempty null space of N) gives rise to further subtleties. In fact, the latter lead to a generalization of the usual scalar and vector potentials and associated gauge transformations. In Sec. XI we briefly point out how to proceed with convolutive time evolutions of the type

$$\partial_t F(t) = NF(t) + \int_0^t ds\ M(t-s)F(s), \tag{1.2}$$

which are relevant in the case in which the permeabilities are frequency dependent.

C. Application to Maxwell's equations

The final result obtained above is then applied to Maxwell's equations for a linear, nonhomogeneous, nonconducting material medium, including the situation with external currents or the coupling with a Schrödinger quantum particle system. In particular, we obtain the generalizations of the familiar Coulomb and Lorentz gauges. For atoms in dielectrics we not only find a (by now well-known) change in the gauge condition for the vector potential (caused by modified field modes as compared to the vacuum case), but also the atomic Coulomb potential is affected. Using an effective Hamiltonian setup, we then give a simple calculation to leading order in the atom-field interaction of an atomic decay rate. The scalar results of Sprik *et al.*, mentioned above, are confirmed for the Maxwell case.

Our next application concerns the situation of a two-level atom with transition frequency in a band gap of a classical photonic system. Making a two-layer approximation to the quantized photonic system (i.e., only one-photon processes are considered), this model can be treated quite rigorously and we find that a bound state of the coupled system in the gap occurs, originating from the atomic excited-state eigenvalue. In addition, a new bound state is found. The situation of a randomized band-gap system showing Anderson localization is briefly discussed.

As in other situations, there are divergencies in the formalism. In order to remove the latter, a Kramers transformation is made. The outcome is intriguing since the renormalized mass is no longer a scalar. In principle, this could be checked by measurements of spectral properties (Lamb shifts) of atoms or molecules embedded in dielectrics. In Sec. XI we comment upon our results and make a comparison with related work.

Below, the case $\varepsilon = \mu = 1$ is referred to as the vacuum case, everything else remaining the same. With the free case we mean a free electromagnetic field with arbitrary ε and μ and a finite dielectric is a system with ε and μ constant outside a bounded region (for the sake of brevity, a medium where both ε and μ may deviate from their vacuum value is referred to as a dielectric). Inner products are denoted as $(f,g) = \langle g|f\rangle$, adjoint operators by an asterisk, and complex conjugates by an overbar. A centered dot between two tensors indicates a contraction over the last index of the first and the first of the second. For the classical canonical formalism we mention [13] and for its quantization [14].

II. EXAMPLES

Below we give some examples of field equations with a conservation law. They all lead to a coupled set of first-order differential equations. Thus $\boldsymbol{f} = \boldsymbol{f}(\mathbf{x},t)$, $\mathbb{R}^d \times \mathbb{R} \to \mathbb{R}^n$, is an n-component field over $\mathbb{R}^d \times \mathbb{R}$ with value in $\mathbb{R}^n$, satisfying

$$\partial_t\, \boldsymbol{f}(\mathbf{x},t) = \mathbf{M}(\mathbf{x},\partial_{\mathbf{x}}) \cdot \boldsymbol{f}(\mathbf{x},t), \tag{2.1}$$

where $\mathbf{M}$ is an $n \times n$ matrix with real partial differential operators with, in general, nonconstant coefficients as entries. In addition, there exists a conserved quantity, referred to as the energy, given by

$$\mathcal{E} = \frac{1}{2} \int d\mathbf{x} [\boldsymbol{\rho}(\mathbf{x}) \cdot \boldsymbol{f}(\mathbf{x},t)]^2, \tag{2.2}$$

where $\boldsymbol{\rho}(\mathbf{x})$, $\mathbb{R}^d \to \mathbb{R}^n \times \mathbb{R}^n$, is a bounded, real, invertible $n \times n$ matrix with bounded inverse. Then

$$\boldsymbol{F}(\mathbf{x},t) = \boldsymbol{\rho}(\mathbf{x}) \cdot \boldsymbol{f}(\mathbf{x},t) \tag{2.3}$$

is real, square integrable for each t, $\boldsymbol{F}(\mathbf{x},t) \in \mathcal{H}_r = L^2(\mathbb{R}^d, d\mathbf{x}; \mathbb{R}^n)$ with conserved norm $\|\boldsymbol{F}(\mathbf{x},t)\|^2 = 2\mathcal{E}$, satisfying

$$\partial_t \boldsymbol{F}(\mathbf{x},t) = \mathbf{N}(\mathbf{x},\partial_{\mathbf{x}}) \cdot \boldsymbol{F}(\mathbf{x},t), \quad \mathbf{N} = \boldsymbol{\rho} \cdot \mathbf{M} \cdot \boldsymbol{\rho}^{-1}. \tag{2.4}$$

It follows that $\mathbf{N}$ must be antisymmetric

$$\mathbf{N}=-\mathbf{N}^*. \tag{2.5}$$

Supposing that $\mathbf{N}$ has a unique anti-self-adjoint extension (this is essential for the time evolution to exist), again denoted by $\mathbf{N}$, it generates a unitary time evolution on $\mathcal{H}_r$:

$$\boldsymbol{F}(t)=\exp[\mathbf{N}t]\cdot\boldsymbol{F}(0). \tag{2.6}$$

It is common practice to embed $\mathcal{H}_r$ in the corresponding complex Hilbert space $\mathcal{H}=L^2(\mathbb{R}^d,d\mathbf{x};\mathbb{C}^n)$ in which $\mathbf{K}=i\mathbf{N}$ is self-adjoint. Initially we shall not do so since we want to introduce a (real) Lagrange-Hamilton formalism. We now give a few examples. Other classical equations such as the massive Klein-Gordon equation and oscillator chains (phonons) provide further ones.

A. Classical scalar waves

We start from the classical scalar wave equation in the form (f: $\mathbb{R}^d\to\mathbb{R}$)

$$\partial_t^2 f(\mathbf{x},t)-\partial_{\mathbf{x}}\cdot c(\mathbf{x})^2\partial_{\mathbf{x}}f(\mathbf{x},t)=0. \tag{2.7}$$

Then, with $F_1=\partial_t f$ and $\boldsymbol{F}_2=c(\mathbf{x})\partial_{\mathbf{x}}f$, we obtain Eq. (2.4) with

$$\boldsymbol{F}=\begin{pmatrix}F_1\\ \boldsymbol{F}_2\end{pmatrix},\quad \mathbf{N}=\begin{pmatrix}0 & \partial_{\mathbf{x}}\cdot c\\ c\,\partial_{\mathbf{x}} & 0\end{pmatrix} \tag{2.8}$$

and

$$\mathcal{E}=\frac{1}{2}\int d\mathbf{x}\boldsymbol{F}(\mathbf{x},t)^2 \tag{2.9}$$

is conserved. In this case $\mathcal{H}_r=L^2(\mathbb{R}^d,d\mathbf{x};\mathbb{R}^4)$.

B. Schrödinger's equation

Although not a classical wave equation, Schrödinger's equation in $L^2(\mathbb{R}^d)$,

$$\partial_t\psi(\mathbf{x},t)=-i[-\partial_{\mathbf{x}}^2+V(\mathbf{x})]\psi(\mathbf{x},t)=-iH\psi(\mathbf{x},t), \tag{2.10}$$

can be rewritten in the form (2.4). Set $F_1=\mathrm{Re}\psi$ and $F_2=\mathrm{Im}\psi$. Now (note that H is a real operator)

$$\boldsymbol{F}=\begin{pmatrix}F_1\\ F_2\end{pmatrix},\quad \mathbf{N}=\begin{pmatrix}0 & H\\ -H & 0\end{pmatrix}, \tag{2.11}$$

the conserved quantity is the total probability ($=1$), and $\mathcal{H}_r=L^2(\mathbb{R}^d,d\mathbf{x};\mathbb{R}^2)$.

C. Maxwell's equations for a nonconducting material medium

We write Maxwell's equations for a nonconducting medium with external current density $\boldsymbol{J}$ as

$$\begin{aligned}&\partial_t\boldsymbol{D}=\partial_{\mathbf{x}}\times\boldsymbol{H}-\boldsymbol{J},\quad \boldsymbol{D}=\varepsilon\boldsymbol{E},\\ &\partial_t\boldsymbol{B}=-\partial_{\mathbf{x}}\times\boldsymbol{E},\quad \partial_{\mathbf{x}}\cdot\boldsymbol{B}|_{t=0}=0,\quad \boldsymbol{H}=\mu^{-1}\boldsymbol{B}.\end{aligned} \tag{2.12}$$

Here we assume $\varepsilon(\mathbf{x})$ and $\mu(\mathbf{x})$ to be ''nice,'' i.e., real, smooth, bounded [elements of C^2 $(\mathbb{R})$] scalar functions of $\mathbf{x}$, bounded from below and above by positive constants (hence invertible with bounded inverse). Discontinuities can be handled as a limiting case; see [15]. For $\boldsymbol{J}=0$ the conserved energy is

$$\begin{aligned}\mathcal{E}&=\frac{1}{2}\int d\mathbf{x}\{\varepsilon(\mathbf{x})\boldsymbol{E}^2+\mu(\mathbf{x})^{-1}\boldsymbol{B}^2\}=\frac{1}{2}\int d\mathbf{x}|\boldsymbol{F}(\mathbf{x},t)|^2\\ &=\frac{1}{2}\|\boldsymbol{F}(t)\|^2,\end{aligned} \tag{2.13}$$

where $\boldsymbol{F}(\mathbf{x},t)$ is the six-dimensional vector field

$$\boldsymbol{F}(\mathbf{x},t)=\begin{pmatrix}\varepsilon^{1/2}\boldsymbol{E}(\mathbf{x},t)\\ \mu^{-1/2}\boldsymbol{B}(\mathbf{x},t)\end{pmatrix}=\begin{pmatrix}\boldsymbol{F}_1(\mathbf{x},t)\\ \boldsymbol{F}_2(\mathbf{x},t)\end{pmatrix} \tag{2.14}$$

in $\mathcal{H}_r=L^2(\mathbb{R}^3,d\mathbf{x};\mathbb{R}^6)$, the Hilbert space of square integrable functions with value in $\mathbb{R}^6$ [norm $\|\ \|$ and inner product $(\ ,\ )$]. From Eq. (2.12) we obtain Eq. (2.4), where now

$$\begin{aligned}\mathbf{N}=\begin{pmatrix}0 & \mathbf{N}_{12}\\ \mathbf{N}_{21} & 0\end{pmatrix}&=\begin{pmatrix}0 & -\varepsilon^{-1/2}(\boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}})\mu^{-1/2}\\ \mu^{-1/2}(\boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}})\varepsilon^{-1/2} & 0\end{pmatrix}=\mathbf{W}\cdot\mathbf{N}_0\cdot\mathbf{W},\\ \mathbf{N}_0&=\begin{pmatrix}0 & -\boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}}\\ \boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}} & 0\end{pmatrix},\quad W=\begin{pmatrix}\varepsilon^{-1/2} & 0\\ 0 & \mu^{-1/2}\end{pmatrix}.\end{aligned} \tag{2.15}$$

The matrix entries are operator-valued 3×3 blocks. Thus $\varepsilon^{-1/2}$ represents $\varepsilon^{-1/2}\mathsf{U}$, where U is the 3×3 unit matrix, and $\boldsymbol{\epsilon}$ is the Levi-Civita pseudotensor ($\boldsymbol{\epsilon}_{123}=1$ and $\boldsymbol{\epsilon}$ is antisymmetric in all three subscripts). As is readily verified, $\mathbf{N}^*=-\mathbf{N}$. Full details for the complex case are given in a paper by Dorren and Tip [15].

In the following we shall say that a three-dimensional vector field $\boldsymbol{f}$ is transverse ($\perp$) if $\partial_{\mathbf{x}}\cdot\boldsymbol{f}=0$ and longitudinal ($\|$) if $\partial_{\mathbf{x}}\times\boldsymbol{f}=0$. Thus the Fourier transform $\tilde{\boldsymbol{f}}(\mathbf{k})$ of a transverse $\boldsymbol{f}$ is orthogonal to $\mathbf{k}$ and of a longitudinal $\boldsymbol{f}$ along $\mathbf{k}$. The associated projectors are denoted by $P^\perp$ and $P^\|$. A six-dimensional vector field is transverse (longitudinal) if both three-dimensional components have this property. The associated projectors are

$$\Pi^\perp=\begin{pmatrix}P^\perp & 0\\ 0 & P^\perp\end{pmatrix},\quad \Pi^\|=\begin{pmatrix}P^\| & 0\\ 0 & P^\|\end{pmatrix}. \tag{2.16}$$

The orthogonal eigenprojector of $\mathbf{N}$ at the eigenvalue 0 is

$$\mathbf{P}_0=\begin{pmatrix}\mathbf{P}_1 & 0\\ 0 & \mathbf{P}_2\end{pmatrix}=\begin{pmatrix}\sqrt{\varepsilon}\,\partial_{\mathbf{x}}[\partial_{\mathbf{x}}\cdot\varepsilon\,\partial_{\mathbf{x}}]^{-1}\partial_{\mathbf{x}}\sqrt{\varepsilon} & 0\\ 0 & \sqrt{\mu}\,\partial_{\mathbf{x}}[\partial_{\mathbf{x}}\cdot\mu\,\partial_{\mathbf{x}}]^{-1}\partial_{\mathbf{x}}\sqrt{\mu}\end{pmatrix}. \tag{2.17}$$

In the vacuum case ($\varepsilon=\mu=1$)

$$\begin{aligned}(\mathbf{P}_1\cdot\boldsymbol{X})(\mathbf{x})&=\partial_{\mathbf{x}}[\partial_{\mathbf{x}}^2]^{-1}\partial_{\mathbf{x}}\cdot\boldsymbol{X}\\ &=-\partial_{\mathbf{x}}\int d\mathbf{x}'[4\pi|\mathbf{x}-\mathbf{x}'|]^{-1}\partial_{\mathbf{x}'}\cdot\boldsymbol{X}(\mathbf{x}')\end{aligned} \tag{2.18}$$

and the same for $\mathbf{P}_2\cdot\boldsymbol{X}$. In Fourier (momentum) space the corresponding expression is simply $(\mathbf{P}_1\cdot\boldsymbol{X})(\mathbf{k})=\mathbf{e}_{\mathbf{k}}\mathbf{e}_{\mathbf{k}}\cdot\boldsymbol{X}(\mathbf{k})$, where $\mathbf{e}_{\mathbf{a}}$ is the unit vector along $\mathbf{a}$.

Remark. Note that in the vacuum case ($\varepsilon=\mu=1$) the nonpropagating fields (i.e., fields in the eigenspace of $\mathbf{N}_0$ at the eigenvalue 0) are precisely the longitudinal ones. In general, this is no longer true. Thus we have to distinguish between longitudinal and nonpropagating fields (and also between transverse and propagating ones).This would not be the case if we had considered the time evolution of $\binom{\boldsymbol{D}}{\boldsymbol{B}}$. Then, however, the time evolution would not be unitary unless the inner product is changed (and $\mathbf{N}_0$ no longer anti-self-adjoint).

Next we mention the eigenvalues and eigenvectors associated with the electric and magnetic Helmholtz operators. They come into play in the field quantization below. Differentiation of the two components of Eq. (2.4) and mutual substitution give the wave equations

$$\partial_t^2\boldsymbol{F}_j(t)=\mathbf{N}_j^2\cdot\boldsymbol{F}_j(t)=-\mathbf{H}_j\cdot\boldsymbol{F}_j(t), \tag{2.19}$$

with the non-negative electric and magnetic Helmholtz operators

$$\mathbf{H}_1=-\mathbf{N}_{12}\mathbf{N}_{21}=\mathbf{N}_{21}^*\mathbf{N}_{12}=\varepsilon^{-1/2}(\boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}})\cdot\mu^{-1}(\boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}})\varepsilon^{-1/2}, \tag{2.20}$$

$$\mathbf{H}_2=-\mathbf{N}_{21}\mathbf{N}_{12}=\mathbf{N}_{12}^*\mathbf{N}_{12}=\mu^{-1/2}(\boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}})\cdot\varepsilon^{-1}(\boldsymbol{\epsilon}\cdot\partial_{\mathbf{x}})\mu^{-1/2}. \tag{2.21}$$

Since $\mathbf{N}_{12}\cdot\mathbf{H}_2=\mathbf{H}_1\cdot\mathbf{N}_{12}$ and $\mathbf{N}_{21}\cdot\mathbf{H}_1=\mathbf{H}_2\cdot\mathbf{N}_{21}$ the eigenvectors of the $\mathbf{H}_j$'s are related. Thus, denoting the eigenvectors of $\mathbf{H}_1$ by $\mathbf{u}_{\lambda\alpha}$,

$$\mathbf{H}_1\cdot\mathbf{u}_{\lambda\alpha}=\lambda^2\mathbf{u}_{\lambda\alpha}, \tag{2.22}$$

where $\lambda\geqslant 0$ and α labels the degeneracy, $\mathbf{H}_2\cdot\mathbf{N}_{21}\cdot\mathbf{u}_{\lambda\alpha}=\lambda^2\mathbf{N}_{21}\cdot\mathbf{u}_{\lambda\alpha}$ and normalization is preserved. Note further that since $\mathbf{H}_1$ is a real operator, $\overline{\mathbf{u}_{\lambda\alpha}}$ is also an eigenvector at the same eigenvalue, so we can always use real eigenvectors by taking linear combinations.

Depending on the actual situation the spectrum $\sigma(\mathbf{H}_j)$ of $\mathbf{H}_j$ may not be the full positive real axis $\mathbb{R}^+$ (a band gap in a periodic dielectric) or may be partly a point spectrum (a localization interval in a realization of a random dielectric). However, for a finite dielectric, where the permeabilities only differ from their vacuum value in a bounded region, we encounter a scattering situation. Initially free electromagnetic wave packets scatter from the dielectric and again propagate freely as time $t\to\infty$. Now the Møller (wave) operators between the classical fields in the medium and classical free fields exist and $\sigma(\mathbf{H}_j)=\mathbb{R}^+$. In this case a connection with the usual plane-wave expansion can be made (see also [9]). Thus, in $L^2(\mathbb{R}^3,d\mathbf{x};\mathbb{C}^3)$, denoting

$$\mathbf{H}_0=-\mathbf{N}_0^2=-(\boldsymbol{\epsilon}\cdot\mathbf{p})^2=\mathbf{p}^2\boldsymbol{\Delta}_{\mathbf{p}}=\mathbf{p}^2\mathsf{U}-\mathbf{pp},\quad \mathbf{p}=-i\partial_{\mathbf{x}}, \tag{2.23}$$

we introduce the wave operators (note that $\boldsymbol{\Delta}_{\mathbf{p}}$ projects away the nonscattering longitudinal states)

$$\boldsymbol{\Omega}_{\pm}^{(h)}=s-\lim_{t\to\pm\infty}\exp[i\mathbf{H}_h t]\cdot\exp[-i\mathbf{H}_0 t]\cdot\boldsymbol{\Delta}_{\mathbf{p}}. \tag{2.24}$$

Their existence is readily verified by standard techniques. The eigenvectors of $\mathbf{H}_0$ at the eigenvalue k^2 are the ''plane waves'' [the $\mathbf{e}_j(\mathbf{k})$'s are two orthogonal polarization unit vectors $\perp\mathbf{k}$]

$$\mathbf{u}_{\mathbf{k}j}^{(0)}(\mathbf{x})=\langle\mathbf{x}|\mathbf{k}\rangle\mathbf{e}_j(\mathbf{k})=(2\pi)^{-3/2}\exp[i\mathbf{k}\cdot\mathbf{x}]\mathbf{e}_j(\mathbf{k}), \tag{2.25}$$

$$\langle\mathbf{u}_{\mathbf{k}j}^{(0)}|\mathbf{u}_{\mathbf{k}'j'}^{(0)}\rangle=\delta(\mathbf{k}-\mathbf{k}')\,\delta_{jj'}. \tag{2.26}$$

Then, arbitrarily choosing $\boldsymbol{\Omega}_+^{(h)}$,

$$\mathbf{u}_{\mathbf{k}j}^{(h)}=\boldsymbol{\Omega}_+^{(h)}\cdot\mathbf{u}_{\mathbf{k}j}^{(0)} \tag{2.27}$$

are eigenvectors of $\mathbf{H}_h$ at the same eigenvalue and, since $(\boldsymbol{\Omega}_+^{(h)})^*\cdot\boldsymbol{\Omega}_+^{(h)}=\boldsymbol{\Delta}_{\mathbf{p}}$, the normalization (2.26) is preserved. Also [see Eq. (2.22)]

$$\mathbf{u}_{\mathbf{k}j}^{(1)}(\mathbf{x})=\mathrm{k}^{-1}\varepsilon^{-1/2}(\mathbf{x})(\boldsymbol{\epsilon}\cdot\mathbf{p})\mu^{-1/2}(\mathbf{x})\cdot\mathbf{u}_{\mathbf{k}j}^{(2)}(\mathbf{x}). \tag{2.28}$$

In later sections $\hat{\mathbf{u}}_{\mathbf{k}j}(\mathbf{x})=\mathbf{u}_{\mathbf{k}j}^{(1)}(\mathbf{x})=\langle\mathbf{x}|\boldsymbol{\Omega}_+^{(1)}\cdot\mathbf{u}_{\mathbf{k}j}^{(0)}\rangle$. Note that here we can label the eigenvectors of $\mathbf{H}_1$ by $\mathbf{k}\in\mathbb{R}^3$ and $j=1,2$. This is also the case in periodic systems.

For later reference we observe that the potentials $\boldsymbol{A}$ and Φ in

$$\boldsymbol{E}=-\partial_t\boldsymbol{A}-\partial_{\mathbf{x}}\Phi,\quad \boldsymbol{B}=\partial_{\mathbf{x}}\times\boldsymbol{A} \tag{2.29}$$

satisfy

$$\begin{aligned}&\partial_t\partial_{\mathbf{x}}\cdot\varepsilon\boldsymbol{A}+\partial_{\mathbf{x}}\cdot\varepsilon\,\partial_{\mathbf{x}}\Phi=-\partial_{\mathbf{x}}\cdot\boldsymbol{D}=-\rho,\\ &\partial_t^2\varepsilon\boldsymbol{A}+\partial_{\mathbf{x}}\times\mu^{-1}(\partial_{\mathbf{x}}\times\boldsymbol{A})+\varepsilon\,\partial_t\partial_{\mathbf{x}}\Phi=\boldsymbol{J}.\end{aligned} \tag{2.30}$$

Finally, the vector potential in the Coulomb gauge and vacuum field Hamiltonian in second quantization are [14]

$$\boldsymbol{A}(\mathbf{x})=\sum_j\int d\mathbf{k}(2k)^{-1/2}\{a_{\mathbf{k}j}^*\overline{\mathbf{u}_{\mathbf{k}j}^{(0)}}(\mathbf{x})+a_{\mathbf{k}j}\mathbf{u}_{\mathbf{k}j}^{(0)}(\mathbf{x})\}, \tag{2.31}$$

$$H_f=\sum_j \int d\mathbf{k}\; k a^*_{\mathbf{k}j} a_{\mathbf{k}j}, \tag{2.32}$$

where the creation and annihilation operators are those for the states $\mathbf{u}^{(0)}_{\mathbf{k}j}$.

III. LAGRANGE FORMALISM

A. A naive approach with a deficiency

We now turn back to general equations of the type (2.4), i.e., we consider

$$\partial_t F(t)=NF(t), \quad N=-N^*, \tag{3.1}$$

in a separable, real Hilbert space $\mathcal{H}_r$ (we suppress the boldface vector notation). We want to obtain this equation from Hamilton's principle [13]

$$\delta\int_{t_1}^{t_2} dt L(\dot\xi,\xi,t)=0, \tag{3.2}$$

where L is the Lagrangian.. Taking $\xi=F$ for the coordinate field and

$$L=\frac{1}{2}(\dot\xi,\dot\xi)-\frac{1}{2}(N\xi,N\xi), \tag{3.3}$$

we have, using $\delta(A\xi,B\xi)=(A\xi,B\delta\xi)+(A\delta\xi,B\xi)$ $=2(B^*A\xi,\delta\xi)$ and $N^*=-N$,

$$\delta\int_{t_1}^{t_2} dt L(\dot\xi,\xi,t)=\int_{t_1}^{t_2} dt(-\partial_t^2\xi+N^2\xi,\delta\xi)=0,$$

leading to

$$\partial_t^2\xi=N^2\xi, \tag{3.4}$$

which is compatible with Eq. (3.1). Note that we refrained from introducing a Lagrangian density. Indeed, $\mathcal{H}$ can be a general Hilbert space, not necessarily a function space over $\mathbb{R}^d$. Still the momentum field can be introduced as a variational derivative of L:

$$\pi=\frac{\delta L}{\delta\dot\xi}=\dot\xi. \tag{3.5}$$

The Hamiltonian is now

$$H=(\pi,\dot\xi)-L=\frac{1}{2}(\pi,\pi)+\frac{1}{2}(N\xi,N\xi). \tag{3.6}$$

Insertion of the actual equations of motion now gives

$$H=(NF,NF), \tag{3.7}$$

which is not proportional to $\mathcal{E}=\frac{1}{2}\|F\|^2$.

The present procedure has a second deficiency in the case

$$\mathcal{H}=\mathcal{H}_1\oplus\mathcal{H}_2, \quad N=\begin{pmatrix} 0 & N_{12} \\ N_{21} & 0 \end{pmatrix}, \quad N_{21}=-N_{12}^*, \tag{3.8}$$

as in the examples. Then the Lagrangian equations of motion result in separate second-order equations for the two components of $F=\binom{F_1}{F_2}$ (the electric and magnetic Helmholtz equations in the electromagnetic case) and their connection is lost. (The situation is analogous to that of the electromagnetic potentials in the Lorentz gauge. They are the solutions of separate wave equations, the connection being provided by the Lorentz gauge condition.)

Remark. Below we restrict ourselves to the case (3.8). A structure similar to Eq. (3.8) can be obtained in the general situation as is detailed in Appendix A.

B. An improved approach

From Eq. (3.7) we note that the factors N in front of F are the cause of the inequality $H\neq\mathcal{E}$. Thus it makes sense to try $\xi=N^{-1}F$ as coordinate field. This works fine in the scalar wave case, where, switching back to vector notation,

$$\mathbf{N}^*\cdot\mathbf{N}=-\partial_{\mathbf{x}}c(\mathbf{x})^2\partial_{\mathbf{x}}\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \tag{3.9}$$

is strictly positive (and hence $\mathbf{N}$ invertible) for $c(\mathbf{x})>c_0>0$. The same is true in the phonon case not considered here. In the Maxwell case the situation is different due to the presence of nonpropagating modes. Obviously, we have to project these away before applying the inverse. As will soon become clear, this has important consequences in further developments: It gives rise to a generalization of the gauge concept.

Before investigating the general case, let us first consider how things work out for the vacuum Maxwell situation with external charges and currents. Here the null space of $\mathbf{N}_0$ consists of the longitudinal field modes, so we must project upon the transverse ones before taking the inverse. Since $\boldsymbol{B}$ is already transverse $\binom{\boldsymbol{\xi}_1}{\boldsymbol{\xi}_2}=\mathbf{N}_0^{-1}\binom{E^\perp}{\boldsymbol{B}}$ exists. Hence $\boldsymbol{E}^\perp=\partial_{\mathbf{x}}\times\boldsymbol{\xi}_2$ and $\boldsymbol{B}=-\partial_{\mathbf{x}}\times\boldsymbol{\xi}_1$, so $-\boldsymbol{\xi}_1$ is precisely the vector potential A in the Coulomb gauge (it is transverse, so $\partial_{\mathbf{x}}\cdot\boldsymbol{A}=0$). Maxwell's equations give

$$\partial_t\begin{pmatrix}\boldsymbol{E}^\perp\\ \boldsymbol{B}\end{pmatrix}=\mathbf{N}_0\cdot\begin{pmatrix}\boldsymbol{E}^\perp\\ \boldsymbol{B}\end{pmatrix}-\begin{pmatrix}\boldsymbol{J}^\perp\\ 0\end{pmatrix},$$

so

$$\partial_t\begin{pmatrix}\boldsymbol{\xi}_1\\ \boldsymbol{\xi}_2\end{pmatrix}=\begin{pmatrix}\boldsymbol{E}^\perp\\ \boldsymbol{B}\end{pmatrix}-\mathbf{N}_0^{-1}\cdot\begin{pmatrix}\boldsymbol{J}^\perp\\ 0\end{pmatrix}.$$

Due to the skew symmetry of $\mathbf{N}_0$, the first component of $\mathbf{N}_0^{-1}\cdot\binom{\boldsymbol{J}^\perp}{0}$ vanishes, leaving

$$\boldsymbol{E}^\perp=\partial_t\boldsymbol{\xi}_1=-\partial_t\boldsymbol{A}, \quad \boldsymbol{B}=-\partial_{\mathbf{x}}\times\boldsymbol{\xi}_1=\partial_{\mathbf{x}}\times\boldsymbol{A}.$$

Using Eq. (2.18), we have $\boldsymbol{E}^\parallel=-\partial_{\mathbf{x}}\Phi$, with $\Phi(\mathbf{x})=\int d\mathbf{x}'\rho(\mathbf{x}')/[4\pi|\mathbf{x}-\mathbf{x}'|]$, where $\rho(\mathbf{x})=\partial_{\mathbf{x}}\cdot\boldsymbol{E}(\mathbf{x})$ is the external charge density. Thus we have expressed the fields in terms of the potentials in the Coulomb gauge by means of a procedure that easily generalizes to more complex situations.

Returning to the general case, let P be the (nonzero) projector upon the null space $\mathcal{N}=\mathcal{N}(N)$ of N and $Q=1-P$. Since $\mathcal{N}(N)=\mathcal{N}(N^*N)=\mathcal{N}(-N^2)$ and, from Eq. (3.8),

$$N^*N=-\begin{pmatrix} N_{12}N_{21} & 0 \\ 0 & N_{21}N_{12} \end{pmatrix}=-\begin{pmatrix} N_1^2 & 0 \\ 0 & N_2^2 \end{pmatrix}, \quad (3.10)$$

it follows that P and Q decompose according to [cf. Eq. (2.17) for the Maxwell case]

$$P=\begin{pmatrix} P_1 & 0 \\ 0 & P_2 \end{pmatrix}, \quad Q=\begin{pmatrix} Q_1 & 0 \\ 0 & Q_2 \end{pmatrix}, \quad Q_j=1-P_j, \quad (3.11)$$

with P_j acting in $\mathcal{H}_j$. We also allow an inhomogeneous term $G(t)=\binom{\xi_1(t)}{\xi_2(t)}$ in Eq. (3.1):

$$\partial_t F(t)=NF(t)-G(t). \quad (3.12)$$

Since $\partial_t QF=QNF-QG=NQF-QG$ we have

$$\partial_t N^{-1}QF=QF-N^{-1}QG. \quad (3.13)$$

Splitting F into its components in the $\mathcal{H}_j$'s, $F=\binom{F_1}{F_2}$, we have

$$Q_1F_1=\partial_t(N^{-1}QF)_1+(N^{-1}QG)_1, \quad (3.14)$$

$$Q_2F_2=(NN^{-1}QF)_2=N_{21}(N^{-1}QF)_1. \quad (3.15)$$

Thus, with

$$\hat{\xi}=-(N^{-1}QF)_1, \quad (3.16)$$

we have

$$F_1=-\partial_t\hat{\xi}+(N^{-1}QG)_1+P_1F_1, \quad (3.17)$$

$$F_2=-N_{21}\hat{\xi}+P_2F_2, \quad (3.18)$$

and also

$$P_1\hat{\xi}=0. \quad (3.19)$$

Comparing Eqs. (3.17) and (3.18) with the vacuum Maxwell example earlier in this section, we see that a similar structure emerges and that Eq. (3.19) is the generalization of the Coulomb gauge condition.

C. Gauges and Lagrangians

As noted above, Eq. (3.19) fixes a particular gauge. We now change to a more general situation by setting

$$\xi=\hat{\xi}+P_1\eta, \quad \eta\in\mathcal{H}_1. \quad (3.20)$$

In the vacuum Maxwell case, discussed above, $\hat{\xi}$ is the vector potential in the Coulomb gauge and, using Eq. (2.18), $P_1\eta$ can be written as the gradient $\partial_{\mathbf{x}}\chi$ of a scalar χ, so we are dealing with a generalization of a gauge transformation. Now, noting that $N_{21}P_1\eta=0$,

$$F_1=-\partial_t\xi+P_1(F_1+\partial_t\eta)+(N^{-1}QG)_1, \quad (3.21)$$

$$F_2=-N_{21}\xi+P_2F_2. \quad (3.22)$$

Although it is possible to continue within this general setting, there is no immediate application and so we set, as in the Maxwell case,

$$G=\begin{pmatrix} G_1 \\ 0 \end{pmatrix}, \quad P_2F_2|_{t=0}=0. \quad (3.23)$$

Then $P_2F_2=0$ for all t and also $(N^{-1}QG)_1=N_{12}(N^{-2}QG)_2=0$, so

$$F_1=-\partial_t\xi+P_1(F_1+\partial_t\eta), \quad (3.24)$$

$$F_2=-N_{21}\xi. \quad (3.25)$$

Next we introduce a generalization ξ_0 of the scalar potential Φ of Maxwell theory: We assume that there exists a third real Hilbert space $\mathcal{H}_3$ and a (closed, densely defined) invertible operator M from $\mathcal{H}_3$ into $P_1\mathcal{H}_1$ (M: $\Phi\rightarrow-\partial_{\mathbf{x}}\Phi$ in the vacuum Maxwell case). Thus, for each $f\in\mathcal{H}_1$ there is a $g\in\mathcal{H}_3$ with

$$P_1f=-Mg. \quad (3.26)$$

Now ξ_0 is defined according to

$$P_1(F_1+\partial_t\eta)=-M\xi_0, \quad (3.27)$$

so

$$F_1=-\partial_t\xi-M\xi_0. \quad (3.28)$$

Note that Eqs. (3.28) and (3.25) are the generalizations of the expressions for the fields in terms of the potentials of the vacuum Maxwell case. Since $\partial_tF_1=N_{12}F_2-G_1$, $\partial_t(-\partial_t\xi-M\xi_0)=N_{12}\{-N_{21}\xi\}-G_1$ or

$$\partial_t^2\xi-N_{12}N_{21}\xi+\partial_tM\xi_0=G_1, \quad (3.29)$$

and defining the quantity ρ through

$$M^*P_1F_1=-\rho, \quad (3.30)$$

we arrive at the ''continuity equation''

$$\partial_t\rho-M^*P_1G=0. \quad (3.31)$$

Note the analogy with the vacuum Maxwell case. There P_1F_1 corresponds to $-\partial_{\mathbf{x}}\Phi$ and M^* with $\partial_{\mathbf{x}}$, so $M^*P_1F_1\rightarrow-\partial_{\mathbf{x}}^2\Phi=\rho$, the charge density. Since $P_1F_1(t)$ only depends on $P_1F(0)$ and $P_1G(t)$, so does $\rho(t)$ and we can consider it to be a given function of t, not depending on the dynamics generated by N. Insertion of Eq. (3.28) into Eq. (3.30) now gives

$$\partial_tM^*P_1\xi+M^*M\xi_0=\rho. \quad (3.32)$$

Equations (3.29) and (3.32) are abstract versions of Eq. (2.30) and can be derived through Hamilton's principle from the Lagrangian [$(\ ,\ )_j$ is the inner product in $\mathcal{H}_j$]

$$L=\frac{1}{2}(\partial_t\xi+M\xi_0,\partial_t\xi+M\xi_0)_1-\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2 +(G_1,\xi)_1-(\rho,\xi_0)_3. \tag{3.33}$$

IV. SPECIFIC GAUGES AND HAMILTONIANS

In Eq. (3.33) we encounter a phenomenon familiar from the Maxwell case, i.e., the absence of $\partial_t\xi_0$, thus preventing the construction of a Hamilton formalism for all dynamical variables. The way out follows the same route: We have to fix a suitable gauge. Here we consider three cases (C, L, and T), which can be viewed as abstractions of the Coulomb, Lorentz, and temporal gauges.

A. The *C* gauge

In this case we eliminate ξ_0 from the formalism by expressing it in terms of ρ. Thus we set

$$P_1\xi=0 \tag{4.1}$$

(as we have seen above the Coulomb gauge condition in the vacuum Maxwell case). Then $\xi=\hat{\xi}\in Q_1\mathcal{H}_1$ (i.e., $\eta=0$) and Eq. (3.32) reduces to $M^*M\xi_0=\rho$ (a generalization of the Poisson equation relating potential and charge density) or

$$\xi_0=(M^*M)^{-1}\rho, \tag{4.2}$$

so ($\partial_t\xi\perp M\xi_0=-P_1F_1$ and $\partial_tM\xi_0=P_1G_1$ in the present case)

$$\partial_t^2\xi-N_{12}N_{21}\xi=Q_1G_1 \tag{4.3}$$

and

$$L=\frac{1}{2}(\partial_t\xi,\partial_t\xi)_1-\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2+(G_1,\xi)_1 -\frac{1}{2}(M\xi_0,M\xi_0)_1, \tag{4.4}$$

with ξ_0 given by Eq. (4.2). The canonical momentum field associated with ξ and the Hamiltonian are

$$\pi=\dot{\xi}, \tag{4.5}$$

$$H=\frac{1}{2}(\pi,\pi)_1+\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2-(G_1,\xi)_1+\frac{1}{2}(M\xi_0,M\xi_0)_1 \tag{4.6}$$

and substitution of the equations of motion now results in

$$H=\frac{1}{2}\|F\|^2-(G_1,\xi)_1, \tag{4.7}$$

which equals $\mathcal{E}$ for vanishing G.

B. The *L* gauge

Here we encounter the generalization of the Lorentz gauge condition $\partial_t\Phi-\partial_{\mathbf{x}}\cdot\boldsymbol{A}=0$ of the vacuum Maxwell case:

$$\partial_t\xi_0-M^*P_1\xi=0, \tag{4.8}$$

resulting in

$$\partial_t^2\xi_0+M^*M\xi_0=\rho, \tag{4.9}$$

$$\partial_t^2\xi-N_{12}N_{21}\xi+MM^*\mathrm{P}_1\xi=G_1. \tag{4.10}$$

These equations can be derived from

$$L=\frac{1}{2}(\partial_t\xi,\partial_t\xi)_1+(M\xi_0,M\xi_0)_1-\frac{1}{2}(\partial_t\xi_0,\partial_t\xi_0)_3 -\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2\frac{1}{2}(M^*\xi,M^*\xi)_1+(G_1,\xi)_1-(\rho,\xi_0)_3. \tag{4.11}$$

Since the equations obtained for ξ_0 and ξ are decoupled, the gauge condition (4.8) is needed as a subsidiary condition to select the proper solutions. The momentum fields are $\pi_0=\dot{\xi}_0$ and $\pi=\dot{\xi}$, whereas

$$H=\frac{1}{2}(\pi,\pi)_1+\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2+\frac{1}{2}(M^*\xi,M^*\xi)_1 -\frac{1}{2}(\pi_0,\pi_0)_3+(M\xi_0,M\xi_0)_1-(G_1,\xi)_1+(\rho,\xi_0)_3. \tag{4.12}$$

In this case η satisfies

$$\partial_t^2\eta+MM^*\eta=P_1G_1. \tag{4.13}$$

C. The *T* gauge

In the vacuum Maxwell case the temporal gauge is fixed by $\Phi=0$, which now becomes

$$\xi_0=0, \tag{4.14}$$

so $\partial_t\eta=-P_1F_1$. Thus

$$F_1=-\partial_t\xi \tag{4.15}$$

and

$$\partial_t^2\xi+N_{12}N_{21}\xi=G_1. \tag{4.16}$$

Now

$$L=\frac{1}{2}(\partial_t\xi,\partial_t\xi)_1-\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2+(G_1,\xi)_1, \tag{4.17}$$

the momentum field is $\pi=\dot{\xi}$, and

$$H=\frac{1}{2}(\pi,\pi)_1+\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2-(G_1,\xi)_1. \tag{4.18}$$

V. QUANTIZATION

We are now in a position to quantize the Hamiltonian systems obtained in the preceding section. Our procedure starts with expanding the coordinate and momentum fields in

terms of an orthonormal basis. The expansion coefficients are ordinary generalized coordinates and momenta and H is expressed in the latter. Since these canonical variables satisfy the usual Poisson bracket relations, we can now apply Dirac's recipe of replacing Poisson brackets (PB's) by equal time commutators (we set $\hbar=1$). At this point we have obtained an abstract operator setting and our next step is to construct a Fock-space representation. Then all objects of interest can be expressed in terms of Fock-space creation and annihilation operators and finally, by making a special choice for the orthonormal basis above (the eigenvectors of $N_{21}^{*}N_{21}$, i.e., the Helmholtz operator $\mathbf{H}_1$ in the Maxwell case), we end up with expressions having a close resemblance to those common in nonrelativistic quantized electromagnetic field theory. We discuss the C-gauge case in full detail and give a few comments about the L-gauge situation.

A. The *C* gauge

In the C gauge $\xi\in Q_1\mathcal{H}_1$. Now let $\{u_j\}$ be an orthonormal basis for this subspace and set $\xi_j=\xi(u_j)=(u_j,\xi)_1$ and $\pi_j=\pi(u_j)=(u_j,\pi)_1$ [in general $\xi(f)=(f,\xi)_1$ and $\pi(f)=(f,\pi)_1$]. The latter form an infinite set of canonical pairs in terms of which

$$
\begin{aligned}
H&=\frac{1}{2}(\pi,\pi)_1+\frac{1}{2}(N_{21}\xi,N_{21}\xi)_2-(G_1,\xi)_1+\frac{1}{2}(M\xi_0,M\xi_0)_1=\frac{1}{2}(\pi,\pi)_1+\frac{1}{2}(-N_{12}N_{21}\xi,\xi)_1-(G_1,\xi)_1+\frac{1}{2}(M\xi_0,M\xi_0)_1\\
&=\frac{1}{2}\sum_j\,(\pi,u_j)_1^2+\frac{1}{2}\sum_j\,(-N_{12}N_{21}\xi,u_j)_1(u_j,\xi)_1-\sum_j\,(G_1,u_j)_1(u_j,\xi)_1+\frac{1}{2}(M\xi_0,M\xi_0)_1\\
&=\frac{1}{2}\sum_j\,\pi_j^2+\frac{1}{2}\sum_{j,h}\,(\xi,u_h)_1(u_h,-N_{12}N_{21}u_j)_1(u_j,\xi)_1-\sum_j\,(G_1,u_j)_1\xi_j+\frac{1}{2}(M\xi_0,M\xi_0)_1\\
&=\frac{1}{2}\sum_j\,\pi_j^2+\frac{1}{2}\sum_{j,h}\,(N_{21}u_h,N_{21}u_j)_2\xi_j\xi_h-\sum_j\,(G_1,u_j)\xi_j+\frac{1}{2}(M\xi_0,M\xi_0)_1
\end{aligned}
\tag{5.1}
$$

and the PB's can be defined in the usual way, leading to

$$\{\xi_j,\pi_h\}=\delta_{jh}. \tag{5.2}$$

Quantization is accomplished by replacing the PB's by the commutators

$$[\xi(u_j),\pi(u_h)]=i\delta_{jh}. \tag{5.3}$$

We give a representation of the operators $\xi(u_j),\pi(u_h)$ in terms of boson creation and annihilation operators acting in a Fock space in the usual way. So far we have dealt with real Hilbert spaces, but at this point we need their complex counterpart. This is a trivial matter for the examples in Sec. II. In the general case $\mathcal{H}$, the complexification of $\mathcal{H}_1$, is defined as follows: For $f,g\in\mathcal{H}_1$ and $\lambda,\mu\in\mathbb{C}$ we define $(\lambda f,\mu g)=\lambda\overline{\mu}(f,g)$ and for operators T, $T\lambda f=\lambda Tf$. Furthermore, let $\mathcal{F}=F(Q_1\mathcal{H})$ be the symmetric (boson) Fock space over $Q_1\mathcal{H}$ and $a^{*}(f)$ and $a(g)$ creation and annihilation operators acting in $\mathcal{F}$ (see Appendix B for details). Thus a^{*} is linear in its argument, $a^{*}(\mu f+g)=\mu a^{*}(f)+a^{*}(g)$, and we have

$$[a(f),a^{*}(g)]=(g,f),\quad f,g\in Q_1\mathcal{H}. \tag{5.4}$$

We now take, with $N_1=(N_{21}^{*}N_{21})^{1/2}$,

$$\xi(f)=a^{*}\left(\frac{1}{\sqrt{2N_1}}f\right)+a\left(\frac{1}{\sqrt{2N_1}}f\right), \tag{5.5}$$

$$\pi(f)=a^{*}\left(i\sqrt{\frac{N_1}{2}}f\right)+a\left(i\sqrt{\frac{N_1}{2}}f\right)=\xi(iN_1f).$$

Then

$$[\xi(f),\pi(g)]=i\ \mathrm{Re}(f,g), \tag{5.6}$$

from which Eq. (5.3) follows. With $\{u_j\}$ an orthonormal basis for $Q_1\mathcal{H}$ we have

$$
\begin{aligned}
(N_1\xi,N_1\xi)&=\sum_j\,(N_1\xi,u_j)(u_j,N_1\xi)\\
&=\sum_j\,(\xi,N_1u_j)(N_1u_j,\xi)\\
&=\sum_j\,\xi(N_1u_j)\xi(N_1u_j)^{*}\\
&=\sum_j\,\left[a^{*}\left(\sqrt{\frac{N_1}{2}}u_j\right)+a\left(\sqrt{\frac{N_1}{2}}u_j\right)\right]^2
\end{aligned}
$$

and, skipping the zero-point energies arising from commuting through a^{*}'s and a's, noting that $(G_1,\overline{u_j})=(u_j,G_1)$ since G_1 is an element of the real space $\mathcal{H}_1$,

$$
\begin{aligned}
H=\sum_j\,a^{*}(\sqrt{N_1}u_j)a(\sqrt{N_1}u_j)-\sum_j\,\Bigg\{&(G_1,u_j)a^{*}\left(\frac{1}{\sqrt{2N_1}}u_j\right)\\
&+(u_j,G_1)a\left(\frac{1}{\sqrt{2N_1}}u_j\right)\Bigg\}+\frac{1}{2}(M\xi_0,M\xi_0).
\end{aligned}
\tag{5.7}
$$

In particular, if N_1 has a pure point spectrum and $\{u_j\}$ is a set of eigenvectors with associated eigenvalues $\{\lambda_j\}$ ($\{u_j\}$ is

complete, since N_1 is invertible, but not necessarily unique since degeneracies are allowed), then, since for suitable functions F, $F(N_1)u_j = F(\lambda_j)u_j$,

$$H = \sum_j \lambda_j a^*(u_j)a(u_j) - \sum_j \frac{1}{\sqrt{2\lambda_j}}\{(G_1,u_j)a^*(u_j) + (u_j,G_1)a(u_j)\} + \frac{1}{2}(M\xi_0, M\xi_0). \tag{5.8}$$

If the spectrum of N_1^2 is continuous and N_1^2 has an eigenfunction expansion $N_1^2 = \Sigma_\alpha \int_0^\infty d\lambda\ \lambda |u_{\lambda\alpha}\rangle\langle u_{\lambda\alpha}|$, where α labels the degeneracy, we have in Eq. (5.7)

$$\begin{aligned} a^*(\sqrt{N_1}u_j) &= \sum_\alpha \int d\lambda\ a^*[(\sqrt{N_1}u_j, u_{\lambda\alpha})u_{\lambda\alpha}] \\ &= \sum_\alpha \int d\lambda\ a^*[(u_j, \sqrt{N_1}u_{\lambda\alpha})u_{\lambda\alpha}] \\ &= \sum_\alpha \int d\lambda \sqrt{\lambda}\ (u_j, u_{\lambda\alpha})a^*(u_{\lambda\alpha}), \end{aligned}$$

$$a(\sqrt{N_1}u_j) = \sum_\alpha \int d\lambda \sqrt{\lambda}\ (u_{\lambda\alpha}, u_j)a(u_{\lambda\alpha}), \tag{5.9}$$

etc., resulting in

$$H = \sum_\alpha \int d\lambda\ \lambda a^*(u_{\lambda\alpha})a(u_{\lambda\alpha}) - \sum_\alpha \int d\lambda \frac{1}{\sqrt{2\lambda}}\{(G_1,u_{\lambda\alpha})a^*(u_{\lambda\alpha}) + (u_{\lambda\alpha},G_1)a(u_{\lambda\alpha})\} + \frac{1}{2}(M\xi_0, M\xi_0). \tag{5.10}$$

Note that the above expressions are similar to those encountered in the vacuum Maxwell case (2.32). There the $u_{\lambda\alpha}$'s are the plane-wave states $\mathbf{u}^{(0)}_{\mathbf{k}j}$ [Eq. (2.25)], $\lambda = k = |\mathbf{k}|$, and $\Sigma_\alpha \int d\lambda$ is replaced by $\Sigma_j \int d\mathbf{k}$.

The following remarks are in order.

(i) The spectrum of N_1^2 outside zero need not always be pure, either point or continuous. In the free electromagnetic case and also for finite dielectrics its spectrum is purely continuous. If, in a realization of a random system, there is a localization interval, we encounter a mixed spectrum.

(ii) In the electromagnetic case a common procedure consists of confining the fields to a box and imposing suitable boundary conditions, thus rendering the spectrum of N_1^2 pure point, whereupon the size of the box is made infinite. Here our (separable) Hilbert space is still general, but nevertheless, as is less well known, the spectrum of N_1^2 can be made pure point by adding a suitable Schmidt class perturbation with arbitrarily small Schmidt norm (see [16], p. 525, theorem 2.1 and p. 527, theorem 2.3), which can later be made to vanish.

B. The L gauge

Here ξ and π are no longer confined to $Q_1\mathcal{H}_1$ but things proceed as before except that N_1^2 is replaced by $T_1 = (N_1^2 + MM^*)^{1/2} = N_1 \oplus M_1$ and $M_1 = (MM^*)^{1/2}$ (the operators act in the orthogonal subspaces $Q_1\mathcal{H}$ and $P_1\mathcal{H}$). Let $f \in \mathcal{H}$, $f = g + h$, $g \in Q_1\mathcal{H}$, and $h \in P_1\mathcal{H}$. Then

$$\begin{aligned} \xi(f) &= a^*\left(\frac{1}{\sqrt{2T_1}}f\right) + a\left(\frac{1}{\sqrt{2T_1}}f\right) = a^*\left(\frac{1}{\sqrt{2N_1}}g\right) \\ &\quad + a\left(\frac{1}{\sqrt{2N_1}}g\right) + a^*\left(\frac{1}{\sqrt{2M_1}}h\right) + a\left(\frac{1}{\sqrt{2M_1}}h\right), \\ \pi(f) &= \xi(iT_1 f). \end{aligned} \tag{5.11}$$

With the pair (ξ_0, π_0) we proceed similarly. Let $\mathcal{H}'$ be the complexification of $\mathcal{H}_3$ [inner product $(\ ,\ )'$] and $b^*(f)$ and $b(g)$ creation and annihilation operators, respectively, acting in $\mathcal{F}(\mathcal{H}')$, satisfying

$$[b(f), b^*(g)] = -(g,f)', \quad f,g \in \mathcal{H}'. \tag{5.12}$$

Now, with $M_2 = (M^*M)^{1/2}$,

$$\xi_0(f) = b^*\left(\frac{1}{\sqrt{2M_2}}f\right) + b\left(\frac{1}{\sqrt{2M_2}}f\right), \quad \pi_0(f) = \xi(iM_2 f). \tag{5.13}$$

Then, if $\{y_j\}$ is an orthonormal basis for $\mathcal{H}$ and $\{w_j\}$ for $\mathcal{H}'$,

$$\begin{aligned} H &= \sum_j a^*(\sqrt{T_1}y_j)a(\sqrt{T_1}y_j) - \sum_j b^*(\sqrt{M_2}w_j)b(\sqrt{M_2}w_j) \\ &\quad - \sum_j \left\{(G_1,y_j)a^*\left(\frac{1}{\sqrt{2T_1}}y_j\right) + (y_j,G_1)a\left(\frac{1}{\sqrt{2T_1}}y_j\right)\right\} \\ &\quad + \sum_j \left\{(\rho,v_j)b^*\left(\frac{1}{\sqrt{2M_2}}w_j\right) + (v_j,\rho)b\left(\frac{1}{\sqrt{2M_2}}w_j\right)\right\}. \end{aligned} \tag{5.14}$$

In the case where N_1, M_1, and M_2 have a pure point spectrum we take $\{u_j\}, \{\lambda_j\}$ as before and for $\{w_j\}$ a (complete) set of eigenvectors of M_2 with $\{\mu_j\}$ the associated eigenvalues. Then $\{v_j = Mw_j\}$ is an orthonormal basis for $P_1\mathcal{H}$ and $T_1 v_j = \mu_j v_j$. Thus

$$\begin{aligned} H &= \sum_j \lambda_j a^*(u_j)a(u_j) + \sum_j \mu_j\{a^*(v_j)a(v_j) \\ &\quad - b^*(w_j)b(w_j)\} - \sum_j \frac{1}{\sqrt{2\lambda_j}}\{(G_1,u_j)a^*(u_j) \\ &\quad + (u_j,G_1)a(u_j)\} - \sum_j \frac{1}{\sqrt{2\mu_j}}\{(G_1,v_j)a^*(v_j) \\ &\quad + (v_j,G_1)a(v_j)\} + \sum_j \frac{1}{\sqrt{2\mu_j}}\{(\rho,v_j)b^*(w_j) \\ &\quad + (v_j,\rho)b(w_j)\}. \end{aligned} \tag{5.15}$$

Remark. Note that we defined $b^*(v_j)$ and $b(v_j)$ such that a minus sign appears on the right-hand side of Eq. (5.12). This is common practice in the indefinite metric formulation in relativistic field theory [14,17]. Here we encounter essentially the same situation and the same methods can be used with appropriate modifications. In particular the quantum version of Eq. (4.8) can be used to single out the set of physical states. Thus we see that this is a general feature, due to the presence of a nonempty null space of the classical dynamics generator.

VI. APPLICATION TO MAXWELL'S EQUATIONS

We apply the formalism developed in Sec. IV to Maxwell's equations as given in Sec. II. Noting that $\mathbf{N}_{21}\cdot\mathbf{g}=-\mu^{-1/2}\partial_{\mathbf{x}}\times\varepsilon^{-1/2}\mathbf{g}$,

$$\boldsymbol{F}_1=\varepsilon^{1/2}\boldsymbol{E}=-\partial_t\varepsilon^{1/2}\boldsymbol{A}-\varepsilon^{1/2}\partial_{\mathbf{x}}\Phi=-\partial_t\boldsymbol{\xi}-\mathbf{M}\xi_0,$$

$$\boldsymbol{F}_2=\mu^{-1/2}\boldsymbol{B}=\mu^{-1/2}\partial_{\mathbf{x}}\times\boldsymbol{A}=-\mathbf{N}_{21}\cdot\boldsymbol{\xi}. \tag{6.1}$$

Also, since $\mathbf{P}_1\cdot\varepsilon^{1/2}\partial_{\mathbf{x}}\Phi=\varepsilon^{1/2}\partial_{\mathbf{x}}\Phi$, we can identify $\mathcal{H}_1$ as $L^2(\mathbb{R}^3,d\mathbf{x};\mathbb{R}^3)$, $\mathcal{H}_0$ as the space of real scalar functions f for which $\varepsilon^{1/2}\partial_{\mathbf{x}}f\in\mathcal{H}_1$, and

$$\boldsymbol{\xi}=-\varepsilon^{1/2}\boldsymbol{A},\quad \boldsymbol{\pi}=-\varepsilon^{1/2}\partial_t\boldsymbol{A},\quad \xi_0=\Phi,\quad \mathbf{M}=\varepsilon^{1/2}\partial_{\mathbf{x}}. \tag{6.2}$$

Moreover, $(\mathbf{M}f,\boldsymbol{g})_1=(f,\mathbf{M}^*\cdot\boldsymbol{g})_0=(f,-\partial_{\mathbf{x}}\cdot\varepsilon^{1/2}\boldsymbol{g})_0$, so $\mathbf{M}^*\cdot\boldsymbol{g}=-\partial_{\mathbf{x}}\cdot\varepsilon^{1/2}\boldsymbol{g}$, $\mathbf{M}^*\cdot\mathbf{M}=-\partial_{\mathbf{x}}\cdot\varepsilon\cdot\partial_{\mathbf{x}}$, and $\mathbf{M}\mathbf{M}^*\cdot\boldsymbol{g}=-\varepsilon^{1/2}\partial_{\mathbf{x}}\partial_{\mathbf{x}}\cdot\varepsilon^{1/2}\boldsymbol{g}$.

A. The *C* gauge

In the C gauge we have

$$\partial_{\mathbf{x}}\cdot\varepsilon\boldsymbol{A}=0, \tag{6.3}$$

leading to [see Eq. (2.30)]

$$\partial_t^2\varepsilon\boldsymbol{A}+\partial_{\mathbf{x}}\times\mu^{-1}(\partial_{\mathbf{x}}\times\boldsymbol{A})=\mathbf{Q}_1\boldsymbol{J}, \tag{6.4}$$

$$\partial_{\mathbf{x}}\cdot\varepsilon\cdot\partial_{\mathbf{x}}\Phi=-\rho, \tag{6.5}$$

and ($\boldsymbol{G}_1=\varepsilon^{-1/2}\boldsymbol{J}$)

$$L=\int d\mathbf{x}\left\{\frac{1}{2}\varepsilon(\partial_t\boldsymbol{A})^2-\frac{1}{2}\mu^{-1}(\partial_{\mathbf{x}}\times\boldsymbol{A})^2+\boldsymbol{J}\cdot\boldsymbol{A}-\frac{1}{2}\rho\Phi\right\}. \tag{6.6}$$

Thus $\boldsymbol{\pi}=-\varepsilon^{1/2}\partial_t\boldsymbol{A}$ and

$$H=\int d\mathbf{x}\left\{\frac{1}{2}\varepsilon(\partial_t\boldsymbol{A})^2+\frac{1}{2}\mu^{-1}(\partial_{\mathbf{x}}\times\boldsymbol{A})^2-\boldsymbol{J}\cdot\boldsymbol{A}+\frac{1}{2}\rho\Phi\right\}. \tag{6.7}$$

The PB's now become

$$\{\boldsymbol{\xi}(\mathbf{x}),\boldsymbol{\pi}(\mathbf{y})\}=\{\varepsilon^{1/2}(\mathbf{x})\boldsymbol{A}(\mathbf{x}),\varepsilon^{1/2}(\mathbf{y})\partial_t\boldsymbol{A}(\mathbf{y})\}=\mathbf{Q}_1(\mathbf{x},\mathbf{y}), \tag{6.8}$$

where $\mathbf{Q}_1(\mathbf{x},\mathbf{y})=\langle\mathbf{x}|\mathbf{Q}_1|\mathbf{y}\rangle$ is the kernel associated with the projector upon the propagating modes $\mathbf{Q}_1$ (the projector upon the transverse states in the vacuum case).

B. The *L* gauge

In the L gauge

$$\partial_t\Phi+\partial_{\mathbf{x}}\cdot\varepsilon\boldsymbol{A}=0, \tag{6.9}$$

leading to the equations of motion

$$\partial_t^2\Phi-\partial_{\mathbf{x}}\cdot\varepsilon\partial_{\mathbf{x}}\Phi=\rho, \tag{6.10}$$

$$\partial_t^2\boldsymbol{A}+\varepsilon^{-1}\partial_{\mathbf{x}}\times\mu^{-1}(\partial_{\mathbf{x}}\times\boldsymbol{A})-\partial_{\mathbf{x}}^2\varepsilon\boldsymbol{A}=\varepsilon^{-1}\boldsymbol{J}. \tag{6.11}$$

In particular,

$$\begin{aligned}\{-\mathbf{N}_{12}\cdot\mathbf{N}_{12}+\mathbf{M}\mathbf{M}^*\}\cdot\boldsymbol{f}&=\varepsilon^{-1/2}\partial_{\mathbf{x}}\times\mu^{-1}(\partial_{\mathbf{x}}\times\varepsilon^{-1/2}\boldsymbol{f})\\&\quad-\varepsilon^{1/2}\partial_{\mathbf{x}}\partial_{\mathbf{x}}\cdot\varepsilon^{1/2}\boldsymbol{f}\\&=-\varepsilon^{-1/2}(\partial_{\mathbf{x}}\cdot\mu^{-1}\partial_{\mathbf{x}}\varepsilon^{-1/2})\boldsymbol{f}\\&\quad+\varepsilon^{-1/2}\partial_{\mathbf{x}}\mu^{-1}\partial_{\mathbf{x}}\varepsilon^{-1/2}\cdot\boldsymbol{f}\\&\quad-\varepsilon^{1/2}\partial_{\mathbf{x}}\partial_{\mathbf{x}}\cdot\varepsilon^{1/2}\boldsymbol{f}.\end{aligned} \tag{6.12}$$

Remark. Note that the two operators in the middle expression in Eq. (6.12) act in orthogonal subspaces, whereas in the vacuum case the two last terms in the third expression cancel, leaving $-\mathbf{N}_{12}\cdot\mathbf{N}_{12}+\mathbf{M}\mathbf{M}^*=-\partial_{\mathbf{x}}^2$, the well-known free field result. The coordinate fields are $\{\xi_0=\Phi,\boldsymbol{\xi}=-\varepsilon^{1/2}\boldsymbol{A}\}$, whereas

$$L=\int d\mathbf{x}\left\{\frac{1}{2}\varepsilon(\partial_t\boldsymbol{A})^2-\frac{1}{2}(\partial_t\Phi)^2-\frac{1}{2}\mu^{-1}(\partial_{\mathbf{x}}\times\boldsymbol{A})^2+\varepsilon(\partial_{\mathbf{x}}\Phi)^2+\boldsymbol{J}\cdot\boldsymbol{A}-\rho\Phi\right\}, \tag{6.13}$$

so the momentum fields are $\{\pi_0=\partial_t\Phi,\boldsymbol{\pi}=-\varepsilon^{1/2}\partial_t\boldsymbol{A}\}$. Thus

$$H=\int d\mathbf{x}\left\{\frac{1}{2}\varepsilon(\partial_t\boldsymbol{A})^2+\frac{1}{2}\mu^{-1}(\partial_{\mathbf{x}}\times\boldsymbol{A})^2-\frac{1}{2}(\partial_t\Phi)^2-\varepsilon(\partial_{\mathbf{x}}\Phi)^2-\boldsymbol{J}\cdot\boldsymbol{A}+\rho\Phi\right\} \tag{6.14}$$

and the PB's become (U is the unit 3×3 matrix)

$$\{\Phi(\mathbf{x}),\Phi(\mathbf{y})\}=\delta(\mathbf{x}-\mathbf{y}),$$

$$\{\varepsilon^{1/2}(\mathbf{x})\boldsymbol{A}](\mathbf{x}),\varepsilon^{1/2}(\mathbf{y})\partial_t\boldsymbol{A}](\mathbf{y})\}=\mathsf{U}\delta(\mathbf{x}-\mathbf{y}). \tag{6.15}$$

VII. QUANTIZATION OF MAXWELL'S EQUATIONS IN THE *C* GAUGE AND COUPLING TO MATERIAL PARTICLES

In applications the C gauge is the most convenient one to use and from now on we restrict ourselves to this case. The quantized version of Eq. (6.7) is directly obtained from Eq. (5.10):

$$H=H_f+H_{ext},$$

$$H_f=\sum_\alpha\int d\lambda\ \lambda\ a^*(\mathbf{u}_{\lambda\alpha})a(\mathbf{u}_{\lambda\alpha}),$$

$$H_{ext}=-\int d\mathbf{x}\ \boldsymbol{J}(\mathbf{x})\cdot\boldsymbol{A}(\mathbf{x})+\frac{1}{2}\int d\mathbf{x}\ \rho(\mathbf{x})\Phi(\mathbf{x}),$$

$$\boldsymbol{A}(\mathbf{x})=\varepsilon(\mathbf{x})^{-1/2}\sum_{\alpha}\int d\lambda(2\lambda)^{-1/2}\{a^*(\mathbf{u}_{\lambda\alpha})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{x})+a(\mathbf{u}_{\lambda\alpha})\mathbf{u}_{\lambda\alpha}(\mathbf{x})\},\tag{7.1}$$

where H_f is the field Hamiltonian in the absence of external charges and currents. Here we assumed that $\mathbf{H}_1$ has a purely continuous spectrum and possesses an eigenfunction expansion $\{\mathbf{u}_{\lambda\alpha}\}$ [see Eq. (2.22)]. As discussed in Sec. II, the eigenvectors can be related to their vacuum counterparts $\hat{\mathbf{u}}_{\mathbf{k}j}=\boldsymbol{\Omega}_{+}^{(1)}\mathbf{u}_{kj}^{(0)}$ for a finite dielectric and the labeling through $\mathbf{k}$ and j can also be used for periodic systems. Then $\Sigma_\alpha\int d\lambda$ is replaced by $\Sigma_j\int d\mathbf{k}$ in Eq. (7.1) and, with $\hat{a}_{\mathbf{k}j}=a(\hat{\mathbf{u}}_{\mathbf{k}j})$, etc.,

$$H_f=\sum_j\int d\mathbf{k}\ \mathrm{k}\hat{a}_{\mathbf{k}j}^*\hat{a}_{\mathbf{k}j},\tag{7.2}$$

$$\boldsymbol{A}(\mathbf{x})=\varepsilon(\mathbf{x})^{-1/2}\sum_j\int d\mathbf{k}(2k)^{-1/2}\{\hat{a}_{\mathbf{k}j}^*\overline{\mathbf{u}}_{\mathbf{k}j}(\mathbf{x})+\hat{a}_{\mathbf{k}j}\mathbf{u}_{\mathbf{k}j}(\mathbf{x})\}.\tag{7.3}$$

Finding the Lagrangian and Hamiltonian for a full, coupled matter-field system does not pose any further problems if we assume that the force $\mathbf{F}$ on a charged particle positioned in $\mathbf{x}$ in a material medium, due to an external field, is still given by the Lorentz force

$$\mathbf{F}=e\{\boldsymbol{E}(\mathbf{x},t)+\mathbf{v}\times\boldsymbol{B}(\mathbf{x},t)\}.\tag{7.4}$$

Then the Hamiltonian for a set of charged particles in an external field is

$$H=\sum_j\left\{\frac{1}{2m_j}[\mathbf{p}_j-e_j\boldsymbol{A}(\mathbf{x}_j,t)]^2+e_j\Phi(\mathbf{x}_j,t)\right\}\tag{7.5}$$

and for the full interacting system (charges e_j, $|e_j|=1$, masses m_j), dismissing self-energies,

$$H=H_m+H_f+H_{int},\tag{7.6}$$

$$H_m=\sum_j\frac{1}{2m_j}\mathbf{p}_j^2+\frac{1}{2}\sum_{j>h}e_je_h\Phi(\mathbf{x}_j,\mathbf{x}_h),\tag{7.7}$$

$$H_{int}=-\sum_j\frac{e_j}{2m_j}\{\mathbf{p}_j\cdot\boldsymbol{A}(\mathbf{x}_j)+\boldsymbol{A}(\mathbf{x}_j)\cdot\mathbf{p}_j\}+\sum_j\frac{1}{2m_j}e_j^2\boldsymbol{A}(\mathbf{x}_j)^2,\tag{7.8}$$

where

$$-\partial_{\mathbf{x}}\cdot\varepsilon(\mathbf{x})\partial_{\mathbf{x}}\Phi(\mathbf{x},\mathbf{y})=\delta(\mathbf{x}-\mathbf{y}).\tag{7.9}$$

The following remarks are in order.

(i) As noted earlier by Kweon and Lawandy [12], $\boldsymbol{A}(\mathbf{x})$ is not transverse in the C gauge so the operators $\mathbf{p}_j$ and $\boldsymbol{A}(\mathbf{x}_j)$ do not commute in general. In addition, if we solve Eq. (7.9) for Φ we do not obtain a Coulomb potential. In Eq. (7.9) $\varepsilon(\mathbf{x})$ is sandwiched between two space derivatives and a convolution is involved. These matters are rather irrelevant in practical cases of atoms in a medium with optical characteristic length scale, where ε hardly changes over atomic dimensions. Still, if such subtleties become important, one reaches the borders of a setup where the phenomenological approach through the permeabilities seems trustworthy and a more sophisticated treatment is preferable.

(ii) Quantization in the L gauge does not pose specific problems. The whole procedure can be patterned after [14]. The complications that occur (due to the indefinite Hamiltonian) in the vacuum case once more make their appearance.

VIII. ATOMIC RADIATIVE DECAY IN DIELECTRICS

In this section we study the radiative decay of an excited atom in a dielectric. For simplicity we consider a hydrogen atom with an infinitely heavy nucleus at the position $\mathbf{X}$ and make the long-wavelength approximation.

Remark. In the present situation the long-wavelength approximation involves the following: (i) The evaluation of the vector potential in the point $\mathbf{X}$. (ii) the replacement of $\varepsilon(\mathbf{x})$ by $\varepsilon(\mathbf{X})$ in (7.9), leading to atomic Coulomb potentials of the type $|4\pi\varepsilon(\mathbf{X})r|^{-1}$ thus the atomic eigenvalues depend on $\mathbf{X}$ if the atom is situated in a space region where ε deviates from unity; and (iii) in the vacuum case it is common practice to make the dipole approximation at this point: The symmetry-breaking properties caused by general $\mathbf{x}$-dependent permeabilities make this less obvious in the present situation.

Also we assume (as for a finite dielectric) that the classical field has, in addition to the eigenvalue zero for the nonpropagating modes, only a continuous spectrum covering the positive real axis. The interaction with the field will turn the excited atomic states into resonances, their imaginary parts giving the radiative decay rates of the now unstable states. We calculate the corresponding complex eigenvalues to lowest nonvanishing order in the interaction, using an effective Hamiltonian formalism. We have

$$H=H_{at}+H_f+H_{int},\quad H_{at}=\frac{\mathbf{p}^2}{2m}-\frac{1}{4\pi\varepsilon(\mathbf{X})r},$$

$$\mathbf{p}=-i\partial_{\mathbf{r}},\quad r=|\mathbf{r}|,$$

$$H_{int}=-\frac{e}{m}\mathbf{p}\cdot\boldsymbol{A}(\mathbf{X})+\frac{e^2}{2m}\boldsymbol{A}(\mathbf{X})^2,$$

$$\boldsymbol{A}(\mathbf{X})=\varepsilon(\mathbf{X})^{-1/2}\sum_{\alpha}\int d\lambda\ \sigma(\lambda)(2\lambda)^{-1/2}\times\{a^*(\mathbf{u}_{\lambda\alpha})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})+a(\mathbf{u}_{\lambda\alpha})\mathbf{u}_{\lambda\alpha}(\mathbf{X})\}.\tag{8.1}$$

Here we introduced a cutoff function $\sigma(\lambda)$ in the definition of $\boldsymbol{A}(\mathbf{X})$ in order to avoid divergencies. In the following we neglect the $\boldsymbol{A}(\mathbf{X})^2$ term in the interaction. Further approximations will be made along the way, our main aim being an investigation of the leading deviations from the vacuum case. We write

$$H=H_0+H_{int},\quad H_0=H_{at}+H_f.\tag{8.2}$$

The atomic eigenvalues λ_n have associated eigenvectors φ_{nlm}, $H_{at}\varphi_{nlm}=\lambda_n\varphi_{nlm}$, and eigenprojectors $P_n^{at}=\Sigma_l P_{nl}^{at}=\Sigma_{l,m}|\varphi_{nlm}\rangle\langle\varphi_{nlm}|$. We set $\varphi_{100}=\varphi_1$ for the ground state. For later use it is convenient to write the three orthogonal $2p$ states as the components of a three-dimensional vector $\boldsymbol{\varphi}_2=\mathbf{p}\varphi_2$, where φ_2 is a scalar quantity. Hence $P_{21}^{at}=|\boldsymbol{\varphi}_2\rangle\cdot\langle\boldsymbol{\varphi}_2|=\mathbf{p}|\varphi_2\rangle\cdot\langle\varphi_2|\mathbf{p}$. Let φ_{vac} be the vacuum state for the field, $P_0^f=|\varphi_{vac}\rangle\langle\varphi_{vac}|$, and let P_n^f be the projector upon the nth Fock layer. In particular, $P_1^f=\Sigma_\alpha\int d\lambda|\mathbf{u}_{\alpha\lambda}\rangle\langle\mathbf{u}_{\alpha\lambda}|$. The eigenstates of H_0 are $\psi_{nlm}=\varphi_{nlm}\otimes\varphi_{vac}$ and we write $P_n=\Sigma_{l,m}|\psi_{nlm}\rangle\langle\psi_{nlm}|=P_n^{at}\otimes P_0^f$.

By the Feshbach projection formula (P is a projector, $Q=1-P$, $\mathrm{Im}z\neq 0$, taken positive from now on, $H_P=PHP$, $H_{PQ}=PHQ$, etc.)

$$[z-H]^{-1}=[z-H_Q]^{-1}Q+\{P+[z-H_Q]^{-1}H_{QP}\}\mathcal{G}_P(z)\times\{P+H_{PQ}[z-H_Q]^{-1}\},$$

$$\mathcal{G}_P(z)=[z-H_P-H_{PQ}[z-H_Q]^{-1}H_{QP}]^{-1}=[z-H^{(eff)}(z)]^{-1}. \tag{8.3}$$

With $P=P_n$ and $Q=1-P_n$ and noting that they commute with H_0, we have

$$P_n[z-H]^{-1}P_n=[z-H_n^{(eff)}(z)]^{-1}P_n,$$

$$H_n^{(eff)}(z)=\lambda_nP_n+P_nH_{int}P_n+P_nH_{int}Q_n[z-H_{Q_n}]^{-1}\times Q_nH_{int}P_n=\lambda_nP_n+P_nH_{int}[z-H_{Q_n}]^{-1}H_{int}P_n. \tag{8.4}$$

Since $\langle\varphi_{vac}|\boldsymbol{A}(\mathbf{X})|\varphi_{vac}\rangle=0$, we have $P_nH_{int}P_n=0$, $P_nH_{int}Q_n=P_nH_{int}$, etc., which were used to arrive at the final expression. We determine the resonance poles of $[z-H]^{-1}$ originating from the atomic eigenvalues, due to the perturbation H_{int}, by approximately solving

$$zP=H_n^{(eff)}(z)P \tag{8.5}$$

for complex z. In the solution z is the perturbed eigenvalue and P the associated projector. In essentially the same way as in the vacuum case one calculates

$$\langle\varphi_{vac}|\boldsymbol{A}(\mathbf{X})Q[z-H_0]^{-1}\boldsymbol{A}(\mathbf{X})|\varphi_{vac}\rangle=[2\varepsilon(\mathbf{X})]^{-1}\sum_\alpha\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}\mathbf{u}_{\lambda\alpha}(\mathbf{X})\times[z-H_{at}-\lambda]^{-1}\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X}). \tag{8.6}$$

Then, to leading (second) order in the interaction,

$$H_n^{(eff)}(z)=\lambda_nP_n+P_nH_{int}[z-H_0]^{-1}H_{int}P_n=\left\{\lambda_nP_n^{at}+\left(\frac{e}{m}\right)^2[2\varepsilon(\mathbf{X})]^{-1}\times\sum_\alpha\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}P_n^{at}\mathbf{p}\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})\times[z-H_{at}-\lambda]^{-1}\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\mathbf{p}P_n^{at}\right\}\otimes P_0^f. \tag{8.7}$$

As an example we consider the truncated system where only the $1s$ ($n=1$) and $2p$ ($n=2$, $l=1$) states are retained. As far as the field is concerned, everything is in the vacuum subspace and we take this for granted. Thus

$$H^{(eff)}(z)=\lambda_2P_{21}^{at}+\left(\frac{e}{m}\right)^2[2\varepsilon(\mathbf{X})]^{-1}\sum_\alpha\times\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}P_{21}^{at}\mathbf{p}\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})\times[z-H_{at}-\lambda]^{-1}\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\mathbf{p}P_{21}^{at}=\lambda_2P_{21}^{at}+\left(\frac{e}{m}\right)^2[2\varepsilon(\mathbf{X})]^{-1}\sum_\alpha\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}\times[z-\lambda_1-\lambda]^{-1}P_{21}^{at}\mathbf{p}\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})P_1^{at}\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\mathbf{p}P_{21}^{at}. \tag{8.8}$$

Using the spherical symmetry of φ_1 and φ_2 we have

$$P_{21}^{at}\mathbf{p}\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})P_1^{at}\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\mathbf{p}P_{21}^{at}=|\boldsymbol{\varphi}_2\rangle\cdot\langle\varphi_2|\mathbf{p}\mathbf{p}\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})|\varphi_1\rangle\langle\varphi_1|\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\mathbf{p}\mathbf{p}|\varphi_2\rangle\langle\boldsymbol{\varphi}_2|=\frac{1}{9}|\langle\varphi_1|\mathbf{p}\cdot\boldsymbol{\varphi}_2\rangle|^2|\boldsymbol{\varphi}_2\rangle\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\langle\boldsymbol{\varphi}_2|, \tag{8.9}$$

so

$$H^{(eff)}(z)=\lambda_2P_{21}^{at}+\kappa(\mathbf{X})\sum_\alpha\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}\times[z-\lambda_1-\lambda]^{-1}|\boldsymbol{\varphi}_2\rangle\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\langle\boldsymbol{\varphi}_2|,$$

$$\kappa(\mathbf{X})=\left(\frac{e}{3m}\right)^2[2\varepsilon(\mathbf{X})]^{-1}|\langle\varphi_1|\mathbf{p}\cdot\boldsymbol{\varphi}_2\rangle|^2. \tag{8.10}$$

We now iterate Eq. (8.10). To zeroth order $z_0=\lambda_2$, $P_0=P_{21}^{at}$, and to first order

$$z_1P_1=\lambda_2P_{21}^{at}+\kappa(\mathbf{X})\sum_\alpha\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}\times[\lambda_2+i0-\lambda_1-\lambda]^{-1}|\boldsymbol{\varphi}_2\rangle\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\langle\boldsymbol{\varphi}_2|=\lambda_2P_{21}^{at}+|\boldsymbol{\varphi}_2\rangle\cdot\mathbf{M}(\mathbf{X})\cdot\langle\boldsymbol{\varphi}_2|. \tag{8.11}$$

This expression closely resembles that of the vacuum case except that there $\Sigma_\alpha \mathbf{u}_{\lambda\alpha}(\mathbf{X})\overline{\mathbf{u}_{\lambda\alpha}}(\mathbf{X})$ can be replaced by $\frac{2}{3}\mathsf{U}$, leading to $P_1 = P_{21}^{at}$. In general, this isotropy is lost and λ_2 splits up into three different values; see below. Making the isotropy approximation $\Sigma_\alpha \mathbf{u}_{\lambda\alpha}(\mathbf{X})\overline{\mathbf{u}_{\lambda\alpha}}(\mathbf{X}) \to 1/3\Sigma_\alpha |\mathbf{u}_{\lambda\alpha}(\mathbf{X})|^2 \mathsf{U}$ we have $P_1 = P_{21}^{at}$ and

$$z_1 = \lambda_2 + \frac{1}{3}\kappa(\mathbf{X}) \sum_\alpha \int d\lambda\, \sigma(\lambda)^2 \lambda^{-1} \times [\lambda_2 + i0 - \lambda_1 - \lambda]^{-1} |\mathbf{u}_{\lambda\alpha}(\mathbf{X})|^2. \tag{8.12}$$

Hence, setting $\sigma(\lambda) = 1$,

$$\begin{aligned}\Gamma(\mathbf{X}) &= \mathrm{Im} z_1 \\ &= -\frac{\pi}{3}\kappa(\mathbf{X}) \sum_\alpha \int d\lambda\, \lambda^{-1} \delta(\lambda_2 - \lambda_1 - \lambda) |u_{\lambda\alpha}(\mathbf{X})|^2 \\ &= -\frac{\pi}{3}\kappa(\mathbf{X}) N_f(\mathbf{X}),\end{aligned} \tag{8.13}$$

where $N_f(\mathbf{X})$ is the so-called local density of states for the classical (i.e., not quantized) field, its integral over $\mathbf{X}$ being the field density of states N_f itself. The above result, valid for general ε and μ, generalizes the scalar case result by Sprik *et al.* [2]. For its numerical evaluation the mode functions $\mathbf{u}_{\lambda\alpha}(\mathbf{X})$ must be calculated. The translation symmetry present in a photonic crystal case makes a further Bloch decomposition possible, but actual calculations do not yet seem to exist. In other situations it is sometimes convenient to recast Eq. (8.13) into a different form, featuring the electric classical Helmholtz Green's function. With $\omega_0 = \lambda_2 - \lambda_1$ we have

$$\begin{aligned}\Gamma(\mathbf{X}) &= -\frac{2\pi}{3}\kappa(\mathbf{X}) \sum_\alpha \int d\lambda\, \delta(\omega_0 - \lambda^2) \langle \mathbf{X}|\mathbf{u}_{\lambda\alpha}\rangle \cdot \langle \mathbf{u}_{\lambda\alpha}|\mathbf{X}\rangle \\ &= \mathrm{Im}\frac{2}{3}\kappa(\mathbf{X}) \mathrm{tr} \langle \mathbf{X}|\mathbf{Q}_1[\omega_0^2 + i0 - \mathbf{H}_1]^{-1}\mathbf{Q}_1|\mathbf{X}\rangle \\ &= \mathrm{Im}\frac{2}{3}\kappa(\mathbf{X}) \mathrm{tr} \langle \mathbf{X}|[\omega_0^2 + i0 - \mathbf{H}_1]^{-1}|\mathbf{X}\rangle \\ &= \mathrm{Im}\frac{2}{3}\kappa(\mathbf{X}) \mathrm{tr} \langle \mathbf{X}|\mathbf{R}(\omega_0^2 + i0)|\mathbf{X}\rangle \\ &= \mathrm{Im}\frac{2}{3}\kappa(\mathbf{X}) \mathrm{tr}\mathbf{G}(\mathbf{X}, \mathbf{X}, \omega_0^2 + i0),\end{aligned} \tag{8.14}$$

where [see Eq. (2.17)] $\mathbf{Q}_1 = 1 - \mathbf{P}_1$ is the projector upon the propagating states associated with $\mathbf{H}_1$ and tr stands for the trace over a 3×3 matrix. Since the imaginary part is taken in the above expression we could replace $\mathbf{Q}_1$ by the unit operator, the contribution of $\mathbf{P}_1$ [$\sim \delta(\omega_0^2)$] being zero. Thus the calculation of $\Gamma(\mathbf{X})$ amounts to the evaluation of the Green's function $\mathbf{G}(\mathbf{X}, \mathbf{X}, \omega_0^2 + i0) = \langle \mathbf{X}|\mathbf{R}(\omega_0^2 + i0)|\mathbf{X}\rangle$. In general, this is a complicated matter. Even for dielectric (Mie) spheres this has only recently been achieved in full generality by the author [18]. In that reference explicit formulas in terms of Bessel functions are given for this and related objects.

In the case where the isotropy approximation is not justified we solve Eq. (8.11) by writing $\mathbf{M} = \Sigma_{j=1}^3 m_j \mathbf{a}_j \overline{\mathbf{b}_j}$, where $\{\mathbf{a}_j, \mathbf{b}_j\}$ form a biorthogonal set, $\mathbf{a}_j \cdot \overline{\mathbf{b}_h} = \delta_{jh}$, $\Sigma_{j=1}^3 \mathbf{a}_j \overline{\mathbf{b}_j} = \mathsf{U}$. Then

$$z_1 \mathrm{P}_1 = \sum_{j=1}^3 (\lambda_2 + m_j) |\mathbf{a}_j \cdot \boldsymbol{\varphi}_2\rangle\langle \mathbf{b}_j \cdot \boldsymbol{\varphi}_2|, \tag{8.15}$$

from which we conclude that the eigenvalues are $\lambda_2 + m_j$ with associated, in general, nonorthogonal projectors $|\mathbf{a}_j \cdot \boldsymbol{\varphi}_2\rangle\langle \mathbf{b}_j \cdot \boldsymbol{\varphi}_2|$.

The following remarks are in order.

(i) Other cases can be handled in a similar way. For photonic crystals with a band gap the gap is excluded in the λ integral so $\Gamma(\mathbf{X})$ vanishes for ω_0 in the gap. Note that above $\Gamma(\mathbf{X})$ is only calculated to leading order. At this point the possibility cannot be excluded that an eigenvalue situated in a gap actually disappears. The vanishing of $\Gamma(\mathbf{X})$ does not imply that the atom does not decay. It may do so through the simultaneous emission of three or more photons provided the photon energies are not in a gap.

(ii) Above we calculated the imaginary part of a perturbed eigenvalue. If we also want to determine the shift in its real part the cutoff function $\sigma(\lambda)$ is necessary in order to avoid divergencies. Divergencies also appear if one tries to include degenerate states at a given eigenvalue (the $2s$ and $2p$ states in the above example). See Sec. X for a renormalization procedure.

IX. BAND-GAP SYSTEMS AND RANDOMNESS

A. Atoms in band-gap dielectrics

In the preceding section we obtained results for the behavior of the excited states of a ''two-level'' atom coupled to the quantized electromagnetic field in a dielectric. These results were obtained to leading order in the atom-field interaction, which amounts to a truncation of the photon Fock space to the first two layers (vacuum and single particle).

For a better understanding of the situation where a band gap is present, it is useful to consider first the spectrum of the two-level system and field Hamiltonian restricted to the first few Fock layers with the interaction deleted. We assume that the Helmholtz operator $\mathbf{H}_1$ has, apart from the eigenvalue zero, associated with the nonpropagating modes an (absolutely) continuous spectrum covering the positive real axis except for a finite gap (λ_a, λ_b), which starts at some positive value, and that the difference of the two atomic eigenvalues $\omega_0 = \lambda_2 - \lambda_1$ is in this interval.

Using the fact that the spectrum of $H_\alpha \otimes I_\beta + I_\alpha \otimes H_\beta$ is the set of points $\{\lambda_\alpha + \lambda_\beta\}$, where λ_α runs through the spectrum of H_α and λ_β through that of H_β, we obtain the results displayed in Fig. 1. In Fig. 1(a) the spectrum of the field Hamiltonian, restricted to the first two Fock layers, is presented (see Appendix B for the layer Hamiltonians). Note the eigenvalue zero, associated with the vacuum state, at the bottom of the spectrum. In Figs. 1(b) and 1(c) the next layers are added. Adding further layers removes the gap completely in

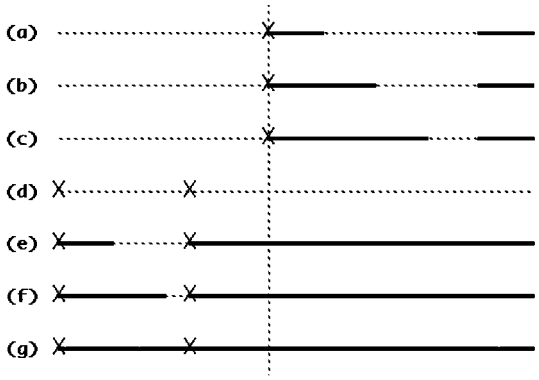

FIG. 1. Spectrum of a band-gap system (crosses are eigenvalues, lines continuous spectrum). In (a) the spectrum of the field Hamiltonian restricted to the two first Fock layers is given. Note the vacuum eigenvalue in the origin and the band gap. In (b) and (c) the third and fourth layers are added, respectively. In (d) the eigenvalues of the two-level system are presented. In (e)–(g) the combinations of (a)–(d), (b)–(d), and (c)–(d) are given, respectively. Note the complete embedding of the excited state in the last case.

this example (in general, if $\lambda_a>0$, there is no gap left in the full H_f). The two eigenvalues $\lambda_1$and λ_2 of the two-level system are pictured in Fig. 1(d) and in Figs 1(e)–1(g) the combined atom–truncated-field spectrum is given. Now λ_1 is associated with the product of atomic ground state and field vacuum state and λ_2 with the atomic excited state and vacuum state. With two and three layers present λ_2 is at the bottom of the second piece of continuous spectrum with a gap to its left but with three layers [Fig. 1(g)] it has become continuum embedded.

The next step is to add the interaction $H_{int}=-(e/m)\mathbf{p}\cdot\boldsymbol{A}(\mathbf{X})$, again suitably truncated. We discuss the situation for the two-level, two-layer case, relevant for single-photon processes. Now H_{int} reduces to a finite-rank operator

$$H_{int}=\eta\{|\varphi_1\otimes\boldsymbol{\psi}\rangle\cdot\langle\boldsymbol{\varphi}_2\otimes\varphi_{vac}|+|\boldsymbol{\varphi}_2\otimes\varphi_{vac}\rangle\cdot\langle\varphi_1\otimes\boldsymbol{\psi}|$$
$$+|\varphi_1\otimes\varphi_{vac}\rangle\langle\boldsymbol{\varphi}_2\otimes\cdot\,\boldsymbol{\psi}|+|\boldsymbol{\varphi}_2\otimes\cdot\,\boldsymbol{\psi}\rangle\langle\varphi_1\otimes\varphi_{vac}|\},$$

$$\boldsymbol{\psi}=\boldsymbol{A}(\mathbf{X})|\varphi_{vac}\rangle$$
$$=[2\varepsilon(\mathbf{X})]^{-1/2}\sum_\alpha\int d\lambda\ \sigma(\lambda)\lambda^{-1/2}\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})|\mathbf{u}_{\lambda\alpha}\rangle,$$

$$\eta=-(e/3m)\langle\varphi_1|\mathbf{p}\cdot\boldsymbol{\varphi}_2\rangle. \tag{9.1}$$

Here φ_1 and $\boldsymbol{\varphi}_2$ are chosen such that $\langle\varphi_1|\mathbf{p}\cdot\boldsymbol{\varphi}_2\rangle$ is positive and $\otimes\cdot$ indicates a tensor product over vector components, followed by a contraction. Set $E=P_1^{at}\otimes P_0^f+P_2^{at}\otimes P_1^f$ and $F=P_2^{at}\otimes P_0^f+P_1^{at}\otimes P_1^f$. Then $E+F=1$ and $(H_1^f=\Sigma_\alpha\int d\lambda\ \lambda|\mathbf{u}_{\lambda\alpha}\rangle\langle\mathbf{u}_{\lambda\alpha}|$ is the field Hamiltonian restricted to layer one)

$$H_{int}E=EH_{int},$$

$$H_0E=EH_0=H_{0E}=\lambda_1P_1^{at}\otimes P_0^f+(\lambda_2+H_1^f)P_2^{at}\otimes P_1^f,$$

$$H_{int}F=FH_{int},$$

$$H_0F=FH_0=H_{0F}=\lambda_2P_2^{at}\otimes P_0^f+(\lambda_1+H_1^f)P_1^{at}\otimes P_1^f. \tag{9.2}$$

Thus E and F reduce H: It breaks up into two parts H_E and H_F, acting in the orthogonal subspaces $\mathcal{H}_E$ and $\mathcal{H}_F$ (superselection sectors). Note that H_{0E} has the isolated eigenvalue λ_1 and the pieces of an absolutely continuous spectrum $[\lambda_2,\lambda_2+\lambda_a]$ and $[\lambda_2+\lambda_b,\infty]$, whereas λ_2 is an isolated eigenvalue of H_{0F}, which has the intervals $[\lambda_1,\lambda_1+\lambda_a]$ and $[\lambda_1+\lambda_b,\infty]$ as a continuous spectrum. H_{int} has finite rank and so have its restrictions to $\mathcal{H}_E$ and $\mathcal{H}_F$. In particular they are trace-class operators, implying that H_E and H_F have the same (absolutely) continuous spectrum. Further, since the interaction is compact, the spectra of these operators outside the above intervals can only consist of isolated eigenvalues with finite degeneracy. For these mathematical details, see [16,19]. In the present case this means that the isolated eigenvalues may move or disappear and new ones may appear. The situation is controlled by the Weinstein-Aronszajn theorem [16]. Alternatively, the isolated eigenvalues can be analyzed further by means of the Feshbach formula (8.3). Again we are led to the relation $zP=H^{(eff)}(z)P$, where on $\mathcal{H}_E$, taking $P=P_1^{at}\otimes\mathrm{P}_0^f$,

$$z=\lambda_1+\kappa(\mathbf{X})\sum_\alpha\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}[z-\lambda_2-\lambda]^{-1}|\mathbf{u}_{\lambda\alpha}(\mathbf{X})|^2, \tag{9.3}$$

and on $\mathcal{H}_F$, with $P=P_2^{at}\otimes\mathrm{P}_0^f$,

$$zP=\lambda_2P_2^{at}+\kappa(\mathbf{X})\sum_\alpha\int d\lambda\ \sigma(\lambda)\lambda^{-1}$$
$$\times[z-\lambda_1-\lambda]^{-1}|\boldsymbol{\varphi}_2\rangle\cdot\mathbf{u}_{\lambda\alpha}(\mathbf{X})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\cdot\langle\boldsymbol{\varphi}_2|. \tag{9.4}$$

Here κ is again given by Eq. (8.10). We write Eq. (9.3) as

$$z=\lambda_1+\kappa(\mathbf{X})\sum_\alpha\int_0^{\lambda_a}d\lambda\ \sigma(\lambda)^2\lambda^{-1}[z-\lambda_2-\lambda]^{-1}|\mathbf{u}_{\lambda\alpha}(\mathbf{X})|^2$$
$$+\kappa(\mathbf{X})\sum_\alpha\int_{\lambda_b}^{\infty}d\lambda\ \sigma(\lambda)^2\lambda^{-1}[z-\lambda_2-\lambda]^{-1}|\mathbf{u}_{\lambda\alpha}(\mathbf{X})|^2$$
$$=\lambda_1+X(z)+Y(z)=Z(z). \tag{9.5}$$

Now, considering $[z-\lambda_2-\lambda]^{-1}$, we note that $Z(z)<\lambda_1$ for $z<\lambda_1$, $Z(z)\to\lambda_1$ as $z\to-\infty$ and $Z(z)\to-\infty$ as $z\uparrow\lambda_1$. It follows that there exists a solution $z_1<\lambda_1$ and that $z_1\to\lambda_1$ as $\kappa\to0$. This remains true if there is no gap, i.e., $\lambda_a=\lambda_b$. Next consider the region $(\lambda_2+\lambda_a,\lambda_2+\lambda_b)$. Now $X(z)\to\infty$ as $z\downarrow\lambda_2+\lambda_a$ and $Y(z)\to-\infty$ as $z\uparrow\lambda_2+\lambda_b$. Thus $Z(z)$ ranges through the whole real axis as z runs through $(\lambda_2+\lambda_a,\lambda_2+\lambda_b)$, so there is a second solution z_3 in this interval. We have $z_3\to\lambda_2+\lambda_a$ as $\kappa\to0$, but for $\kappa=0$ the only solution is λ_1, i.e., $z_3(\kappa)$ is not analytic in $\kappa=0$.

The analysis of Eq. (9.4) is more complicated. In the isotropic approximation it takes the form

$$zP=\lambda_2P_2+\frac{1}{3}\kappa(\mathbf{X})\sum_\alpha\int d\lambda\ \sigma(\lambda)^2\lambda^{-1}$$
$$\times[z-\lambda_1-\lambda]^{-1}|\mathbf{u}_{\lambda\alpha}(\mathbf{X})|^2\mathrm{P}_2. \tag{9.6}$$

and, by a similar argument as above, there is a solution $z_2 \in (\lambda_1+\lambda_a, \lambda_1+\lambda_b)$, which can be considered as the perturbed λ_2. A more involved argument leads to a corresponding result for the three solutions in the nonisotropic case.

We did not check the details, but expect the situation to be similar for the three-layer case. With four or more layers λ_2 is already an embedded eigenvalue without interaction and it is expected to turn into a resonance once H_{int} is switched on.

The physical interpretation of the above is that if ω_0 is in the gap the atom cannot decay by the emission of one or two photons (in fact, the emission of an even number of photons is prohibited for other reasons), but *it can* by the emission of three or more. Of course, the precise situation depends on the value of the atomic eigenvalues, the properties of the gap, and the presence of symmetries (which can inhibit certain decay processes), but the general rule of thumb is that n-photon decay does not take place if the energies of the photons involved (they need not have the same energy) are in the gaps of the corresponding classical dielectric. The situation can be expected to be similar for more precise models for an atom or molecule.

B. Band-gap systems with randomness

Next we consider the same case but with some random part ε_ω added to the electric permeability: $\varepsilon(\mathbf{x}) \to \varepsilon(\mathbf{x}) + \varepsilon_\omega(\mathbf{x})$. As is customary in this field, the index ω labels the realization of the random process involved. Here we can think of a lattice of dielectric spheres where additional dielectric objects are placed between them in a random way, that some spheres are removed from the lattice in a random way, that the positions of the original lattice spheres are randomized, etc. Another, intensely studied case is the Anderson model, which in the present context amounts to random coupling constants λ_j multiplying the ε of each of the spheres on the lattice positions: $\varepsilon_j \to \lambda_j \varepsilon_j$. For a general introduction into random classical wave motion, see [3] and for the mathematical background [20].

In general, it is expected that a random perturbation gives rise to the occurrence of a spectrum in the gap of the classical system, the so-called Lifshitz tails, and that this spectrum is Anderson localized (the vanishing of the diffusion coefficient in this energy region). Recently, some mathematically precise results have been obtained in this direction. In [21] it is shown that for a periodic system with a gap with randomness of a specific type added there is an interval in the original gap containing a dense point spectrum with associated exponentially decaying eigenfunctions (the definition of localization adopted in mathematical work). Using a different method, a similar result was obtained in [22]. There the periodicity of the original band-gap system is not required.

Next consider a two-level atom in a randomized band-gap dielectric that contains a localization interval Δ in the gap such that the atomic transition frequency is contained in Δ. Then the situation of Fig. 1 changes. Suppose that Δ is at the left of the second field continuum; see Fig. 2(a). Thus, in the two-layer approximation, λ_2 becomes embedded in a dense set of square integrable field states and is at the bottom of a field continuum; see Fig. 2(c). Next, the interaction, which is now also a random quantity, is switched on. The decomposition above can still be made and the gap remains (we still have a finite-rank perturbation), but it is by no means clear as to what happens to λ_2. Present day perturbation theory cannot cope with eigenvalues embedded in a dense point spectrum. This raises the question whether single photon decay is still inhibited. From a physical point of view this is expected to be the case. There are no propagating classical field modes with energy in Δ, so if the transition frequency ω_0 is in Δ, radiation cannot propagate away.

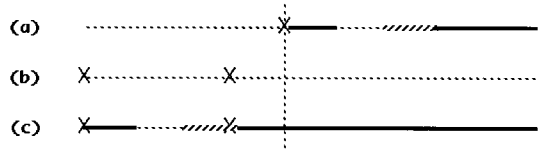

FIG. 2. Spectrum of a randomized band-gap system (crosses and lines are the same as in Fig. 1, hatched lines are localization regions). In (a) the spectrum of the first two Fock layers is displayed. Note the localized states to the left of the right part of the continuous spectrum. In (b) the eigenvalues of the two-level system are given, whereas (c) shows the combined spectrum. In this case the second eigenvalue is embedded in the dense point spectrum and at the bottom of the continuous spectrum.

The situation may change if we average over randomness. Then the problem can be formulated in a bigger space, a direct integral relative to the underlying probability measure (see [23] for details), and in this space the classical field Hamiltonian may once more have a continuous spectrum in Δ. Thus we are back to the situation of Sec. VIII. In this connection note that the eigenvalues in Δ strongly depend on the actual realization [20] and averaging leads to a smearing out. Further, if the random process is ergodic, the integrated density of states is deterministic [20]: It has the same value for almost every realization. However, this is not true for the local density of states, which determines the decay constant in Eq. (8.10). In a system of randomly placed dielectric scatterers the atom can be close to one of them in one realization but further away in another one, so the local density of states will be different in both cases.

X. DIVERGENCIES: KRAMERS TRANSFORMATION

Divergencies turn up if one attempts to calculate atomic properties such as the Lamb shift of atomic levels. In the vacuum case a Kramers transformation can be used to circumvent them [14,24,25]. Here we extend it to material media, once more making the restriction to the long-wavelength approximation. Our starting point is Eq. (8.1), where we introduce a cutoff $\sigma(\lambda)$ in $\boldsymbol{A}(\mathbf{X})$ (a smoothing in coordinate space) and replace m by the ''bare mass'' m_0. We write Eq. (8.1) as

$$H = \frac{1}{2m_0}[\mathbf{p} - e\boldsymbol{A}_\sigma(\mathbf{X})]^2 + V(\mathbf{x}) + H_f,$$

$$V(\mathbf{x}) = -|4\pi\varepsilon(\mathbf{X})x|^{-1},$$

$$\boldsymbol{A}_\sigma(\mathbf{X}) = \varepsilon(\mathbf{X})^{-1/2} \sum_\alpha \int d\lambda\ \sigma(\lambda)(2\lambda)^{-1/2}$$

$$\times\{a^*(\mathbf{u}_{\lambda\alpha})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})+a(\mathbf{u}_{\lambda\alpha})\mathbf{u}_{\lambda\alpha}(\mathbf{X})\}. \tag{10.1}$$

The idea is to transform away the offending term $\mathbf{p}\cdot\boldsymbol{A}(\mathbf{X})$ in favor of a more manageable change in V. For this purpose we introduce the Hertz vector

$$\boldsymbol{Z}_\sigma(\mathbf{X})=i\varepsilon(\mathbf{X})^{-1/2}\sum_\alpha\int d\lambda\ \sigma(\lambda)2^{-1/2}\lambda^{-3/2}$$
$$\times\{a^*(\mathbf{u}_{\lambda\alpha})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})-a(\mathbf{u}_{\lambda\alpha})\mathbf{u}_{\lambda\alpha}(\mathbf{X})\} \tag{10.2}$$

and note that

$$[\boldsymbol{Z}_\sigma(\mathbf{X}),H_f]=-i\boldsymbol{A}_\sigma(\mathbf{X}),$$
$$[\boldsymbol{Z}_\sigma(\mathbf{X}),\boldsymbol{A}_\sigma(\mathbf{X})]=-i\mathbf{F}_\sigma(\mathbf{X}),$$
$$[\boldsymbol{Z}_\sigma(\mathbf{X}),\mathbf{F}_\sigma(\mathbf{X})]=0, \tag{10.3}$$

where

$$\mathbf{F}_\sigma(\mathbf{X})=\varepsilon(\mathbf{X})^{-1}\sum_\alpha\int d\lambda\ \sigma(\lambda)^2(2\lambda^2)^{-1}$$
$$\times\{\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\mathbf{u}_{\lambda\alpha}(\mathbf{X})+\mathbf{u}_{\lambda\alpha}(\mathbf{X})\overline{\mathbf{u}}_{\lambda\alpha}(\mathbf{X})\}. \tag{10.4}$$

Now let $\mathbf{V}$ be a real, symmetric matrix and

$$U_a=\exp[ia\mathbf{p}\cdot\mathbf{V}\boldsymbol{Z}_\sigma(\mathbf{X})],\quad T_a=U_aTU_a^{-1}. \tag{10.5}$$

Since

$$T_a=T+ia[\mathbf{p}\cdot\mathbf{V}\cdot\boldsymbol{Z}_\sigma(\mathbf{X}),T]$$
$$-\frac{a^2}{2}[\mathbf{p}\cdot\mathbf{V}\cdot\boldsymbol{Z}_\sigma(\mathbf{X}),[\mathbf{p}\cdot\mathbf{V}\cdot\boldsymbol{Z}_\sigma(\mathbf{X}),T]]\cdots, \tag{10.6}$$

we obtain

$$H_{fa}=H_f+a\mathbf{p}\cdot\mathbf{V}\cdot\boldsymbol{A}_\sigma(\mathbf{X})+\frac{a^2}{2}\mathbf{p}\cdot\mathbf{V}\cdot\mathbf{F}_\sigma(\mathbf{X})\cdot\mathbf{V}\cdot\mathbf{p},$$
$$\boldsymbol{A}_\sigma(\mathbf{X})_a=\boldsymbol{A}_\sigma(\mathbf{X})+a\mathbf{p}\cdot\mathbf{V}\cdot\mathbf{F}_\sigma(\mathbf{X}),$$
$$V_a(\mathbf{x})=V[\mathbf{x}+a\mathbf{V}\cdot\boldsymbol{Z}_\sigma(\mathbf{X})]. \tag{10.7}$$

With these results we obtain H_a and equating the terms linear in $\boldsymbol{A}(\mathbf{X})_\sigma$ to zero we find

$$a\mathbf{V}=e[m_0+e^2\mathbf{F}_\sigma(\mathbf{X})]^{-1}=e\mathbf{m}_\sigma^{-1}, \tag{10.8}$$

leading to

$$H_a=\frac{1}{2}\mathbf{p}\cdot\mathbf{m}_\sigma^{-1}\cdot\mathbf{p}+V[\mathbf{x}+e\mathbf{m}_\sigma^{-1}\cdot\boldsymbol{Z}_\sigma(\mathbf{X})]+H_f+\frac{e^2}{2m_0}\boldsymbol{A}_\sigma(\mathbf{X})^2. \tag{10.9}$$

In the vacuum case $\mathbf{m}_\sigma$ reduces to a scalar, $\mathbf{m}_\sigma=m_\sigma\mathsf{U}=m\mathsf{U}$. In removing the cutoff, i.e., in the limit $\sigma\to1$, m is kept constant and identified as the observed mass. Here the situation is rather puzzling, hinting in the direction of a nonscalar renormalized mass $\mathbf{m}$. Note that the presence of V is not related to this problem. Next, once $\mathbf{m}$ is fixed, setting $\sigma=1$ in $V[\mathbf{x}+e\mathbf{m}^{-1}\cdot\boldsymbol{Z}_\sigma(\mathbf{X})]$ gives a pathological situation, due to the small-λ (infrared) behavior of the Hertz vector. Its vacuum expectation value, for instance, vanishes. As pointed out by van Kampen [25], this can be remedied by transforming back. Taking $U_a=\exp[-i\mathbf{p}\cdot\mathbf{m}^{-1}\cdot\boldsymbol{Z}_\tau(\mathbf{X})]$ in Eq. (10.5), we obtain

$$H=\frac{1}{2}\mathbf{p}\cdot\mathbf{m}_\tau^{-1}\cdot\mathbf{p}-\mathbf{p}\cdot\mathbf{m}^{-1}\cdot\boldsymbol{A}_\tau(\mathbf{X})+V[\mathbf{x}+e\mathbf{m}^{-1}\cdot\boldsymbol{Z}_{1-\tau}(\mathbf{X})]$$
$$+H_f+\frac{e^2}{2m_0}\boldsymbol{A}_\sigma(\mathbf{X})^2,$$
$$\mathbf{m}_\tau^{-1}=\mathbf{m}^{-1}+\mathbf{m}^{-1}\cdot\mathbf{F}_\tau\cdot\mathbf{m}^{-1}. \tag{10.10}$$

With $\tau(\lambda)$ equal to one for small λ and vanishing for large λ, $\boldsymbol{A}_\tau(\mathbf{X})$, $\mathbf{F}_\tau(\mathbf{X})$, and $\boldsymbol{Z}_{1-\tau}(\mathbf{X})$ are now well behaved. For further discussion (for the vacuum case) see [25], where also the further renormalization of $H_f+(1/2m_0)\boldsymbol{A}_\sigma(\mathbf{X})^2$ is discussed, leading to an expression featuring the renormalized mass. A similar procedure can be followed in the present case where now $\mathbf{m}$ enters. Thus Eq. (10.10) is a convenient starting point for Lamb shift calculations.

XI. DISCUSSION

A. Summary of results

The quantization of Maxwell's equations for material media has led us to develop a general approach to the canonical formalism and quantization of a given linear evolution equation $\partial_tF(t)=NF(t)-G(t)$ in a real Hilbert space. The gauge concept makes its appearance if N has a nonempty null space and we obtained generalizations of the familiar gauges of electrodynamics. The application to Maxwell's equations in a medium, characterized by permeabilities $\varepsilon(\mathbf{x})$ and $\mu(\mathbf{x})$, is immediate.

Next we considered atoms placed in dielectrics and found an expression for the spontaneous decay rate featuring the local density of states, thus generalizing an earlier result of Sprik *et al.* for a scalar model [2] to a dielectric with general ε and μ.

If the classical dielectric has a band gap and the (two-level) atom has its transition frequency in the gap, the atom is stable relative to single-photon decay. The original atomic eigenvalues are modified, but remain real. Also a new eigenvalue of the combined system appears. We make some remarks in the case where the medium shows Anderson localization. This can lead to an atomic eigenvalue embedded in a dense (electromagnetic) point spectrum.

A mass renormalization is necessary for the calculation of Lamb shifts. We gave a version of the nonrelativistic Kramers–van Kampen approach. Due to the nonisotropic nature of the medium, a nonscalar renormalized mass $\mathbf{m}$ emerged. In the vacuum case the renormalized mass is the experimentally observed electron mass. Here we can take a similar point of view, but experimentally this is problematic since, even if we are able to position an atom in the medium away from the dielectric scatterers (for instance, by means of laser or colloid techniques) and try to retrieve $\mathbf{m}$ from its spectral and decay properties, other, nondielectric effects will wash out the tiny changes caused by the medium.

The approach of starting from the (approximative) phenomenological equations and then quantizing leaves something to be desired. The usual classical and quantum derivations of the phenomenological Maxwell equations involve an averaging over volumes of atomic size [26–28]. At a more sophisticated level a starting point could be the Heisenberg equations of motion for the field operators of the interacting system, from which a Bethe-Salpeter equation can be constructed for the fields only (in fact, the latter originated within a field-theoretical context [29]). Here we are still dealing with field operators, but we can, if so desired, make use of the Glauber-Sudarshan p-representation concept (expansion in coherent states) [30] to replace them by c-number quantities. If we are only interested in absorptive processes, the complicated irreducible vertex term in the Bethe-Salpeter equation can be skipped, leaving a formalism containing only an effective Hamiltonian (mass operator). However, the way the matter part is ''integrated out'' will depend on the actual process one wants to describe. It is also far from obvious how spatial dependences, so conveniently stored in the classical permeabilities, are recovered. In certain scattering situations, for finite dielectrics, a simpler procedure suffices. In a channel with equal initial and final matter states the irreducible vertex term is not involved. Using the Feshbach projection formula, the transition operator can be written in terms of an effective Hamiltonian in which only the mass operator appears and nonlinear photonic processes are included.

In conclusion, we note that the whole quantization procedure as discussed above and elsewhere aims for the determination of a second quantized formalism and the actual equations of motion for the *fields* are rather irrelevant once the Hamiltonian has been obtained. On the other hand, their merit is the simple way in which the presence of matter is stored in the permeabilities.

B. Lagrange-Hamilton formalism: Connection with related work

In general, a given equation of motion can be formulated in terms of a Lagrange formalism in many ways, each featuring different generalized coordinates. Indeed the literature shows such a variation in the choice of the latter. Glauber and Lewenstein [9], for the case $\mu=1$ and, initially, $\boldsymbol{J}=\mathbf{0}$, take $\boldsymbol{A}$ as the coordinate, leading to the associated momentum $-\boldsymbol{D}$. The gauge condition (6.3) is also employed here. They then continue by making an eigenmode expansion of the various field operators. Actually, except that $\mu=1$, the expansion is the same as employed here, since not $\boldsymbol{A}$ but $\sqrt{\varepsilon}\boldsymbol{A}$ is expanded [see their Eq. (2.13) and below]. The reason for this is also the same, the self-adjointness of the Helmholtz differential operator, giving an orthogonal set of eigenvectors. (Here we note that, alternatively, the weight in the inner product can be modified, making the expansion of $\boldsymbol{A}$ itself orthogonal. This, however, has the drawback that the free differential operator acts in a different Hilbert space.) The authors then consider the relation between the above expansion and the eigenmode expansion in terms of vacuum modes (plane waves), the two being related through Møller wave operators. This requires the latter to exist, i.e., $\varepsilon(\mathbf{x})-1$ must tend to zero sufficiently rapidly for large $\mathbf{x}$, so this relation is not available for periodic dielectrics. Finally, the spontaneous decay of a two-level atom in a dielectric is studied. Now $\boldsymbol{J}$ no longer vanishes and indeed the relation (6.5) between charge density and scalar potential appears [their Eq. (7.3a)].

Kweon and Lawandy [12] considered the case $\varepsilon(\mathbf{x})$ periodic, $\mu(\mathbf{x})=1$. Again $\boldsymbol{A}$ is the coordinate field and Eq. (4.8) the gauge condition, but the expansion functions are chosen differently. Since here $\mu=1$, the magnetic Helmholtz operator $\mathbf{N}_2^2=-(\boldsymbol{\epsilon}\cdot\mathbf{p})\varepsilon^{-1}(\boldsymbol{\epsilon}\cdot\mathbf{p})$ is transverse, self-adjoint, and its eigenvectors are used in the expansions. These authors study the suppression of atomic radiative decay in a band gap and also give an expression for atomic decay rates (their Eq. 131). Their observation of a dependence on $\varepsilon(\mathbf{x})^{-2}$, instead $\varepsilon(\mathbf{x})^{-1}$ as in the present work, is tied to the different choice of expansion functions [see Eq. (2.21) and below].

Huttner and Barnett [10] consider the quantization problem in connection with dissipation for the special case that the material part of the system has harmonic behavior. They use the vector potential as the coordinate field and employ the Coulomb gauge. Since the full system is considered, there are no problems with field quantization. The specific nature of the Hamiltonian allows a transformation (in essence, a Bogoliubov transformation) to a new representation, featuring an $\mathbf{x}$-independent but frequency-dependent ε. More recently, Matloob *et al.* [11] also considered the case of an absorbing medium and specific dielectric media.

C. Outlook

We did not consider more general situations such as frequency-dependent permeabilities (lossy dielectrics). This case is often encountered, for instance, in our example of transition radiation [8]. Here we outline briefly how such situations can be treated by means of the present formalism. For details, see [32], where the electromagnetic case is discussed. Instead of Eq. (3.1) we now have in $\mathcal{H}_c$ (for notational shortness formulated for the complexified case)

$$\partial_t F(t) = -iKF(t) - i\int_0^t ds\; L(t-s)F(s). \tag{11.1}$$

Note that only $t\geqslant 0$ appears, so we can consider $L(|t|)$ without penalty. Suppose it has a Fourier transform

$$L(|t|) = \int d\omega\; \exp[-i\omega t]\widetilde{L}(\omega), \tag{11.2}$$

with $\widetilde{L}(\omega)\geqslant 0$ (this property holds in the Maxwell case with general linear-response expressions for the susceptibilities). Now let $F_1(t)=F(t)$ and $F_2(t,\omega)$ be a second field with the property $F_2(0,\omega)=0$. Consider the set

$$\partial_t F_1(t) = -iKF(t) - i\int d\omega\; \widetilde{L}(\omega)^{1/2}F_2(t,\omega), \tag{11.3}$$

$$\partial_t F_2(t,\omega) = -i\omega F_2(t,\omega) - i\widetilde{L}(\omega)^{1/2}F_1(t). \tag{11.4}$$

Solving the second and substituting into the first then gives Eq. (11.1). However, $F=\binom{F_1}{F_2}$ now has a unitary time evolu-

tion in $\mathcal{H}=\mathcal{H}_c\oplus\mathcal{H}'$, $\mathcal{H}'=\mathcal{H}_c\otimes L^2(\mathbb{R},d\omega)$, and a canonical formalism and its quantization can again be obtained.

Nonlinear evolutions, relevant if a description of parametric and other nonlinear processes is required, were not considered. Probably, some of these can be handled. The alternative, starting from a fully quantized matter-field system and integrating out the matter part, using an effective Hamiltonian approach, is likely to give a more accurate result, in particular, a microscopic description of the coefficients in the nonlinear terms.

The result (8.13) for the decay of atomic excited states coincides with what would have been obtained using Fermi's golden rule. The effective Hamiltonian approach employed is rather crude and it would be interesting to investigate the possibility of developing a complex scaling (dilatation analytic) method for this case. Such spectral deformation methods [31] have been quite successful for Schrödinger operators. They lead to a deformation of the continuous spectrum away from the real axis, which reduces the calculation of resonances to the perturbation theory of isolated eigenvalues of a non-self-adjoint operator. In particular this can be useful if multiphoton processes are studied. In the case of a finite dielectric a version of the exterior scaling method [31] for the field part seems to be indicated. In Eq. (8.13) the factor $\varepsilon(\mathbf{X})^{-1}$ is not always significant, for instance, if interstitial atoms in a dielectric are situated at positions where the permeabilities are unity. (In the point interaction model, studied by van Coevorden *et al.* [5], $\varepsilon=1$ everywhere outside the lattice points.) As emphasized by Sprik *et al.* [2], who obtained a similar result, the field quantities are purely classical. However, the quantum-electrodynamical formalism is needed to find out how these classical quantities enter into the expression of the perturbed eigenvalue.

In band-gap systems single-photon atomic decay can be inhibited. Still multiphoton decay may occur and this makes it worthwhile to investigate this situation. For this a higher-order approximation for the effective Hamiltonian of Sec. VIII can be used.

Atomic decay in random band-gap systems showing Anderson localization is intriguing. Here we encounter situations where the atomic eigenvalue is embedded in a dense (field) point spectrum and its perturbation theory seems to be an open question. As mentioned in Sec. IX, it is possible to consider the situation in a broader context where an averaged decay parameter may exist. If so, the next step would be the calculation of its fluctuations.

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APPENDIX A: LAGRANGE FORMALISM FOR THE GENERAL CASE

We return to Eq. (3.12), $\partial_t F(t)=NF(t)-G(t)$, $N^*=-N$, in the real Hilbert space $\mathcal{H}_r$, where we no longer assume the structure (3.8). N, being closed, densely defined, allows the decomposition

$$N=U|N|,\quad |N|=(N^*N)^{1/2}, \tag{A1}$$

where N is a partial isometry (for details, see [16], p. 334). Since $N^*=-N$, we have, with P the projector upon the null space of N and $Q=1-P$,

$$U^*U=Q,\quad U^*=-U,\quad |N|=NU=UN. \tag{A2}$$

We also note that if $A^*=-A$, $Af\perp f$ since $(Af,f)=(f,A^*f)=(f,-Af)=-(Af,f)=0$. Now let

$$V_\pm=\frac{1}{\sqrt{2}}(1\pm U)Q. \tag{A3}$$

Then $V_\pm$ are unitary on $Q\mathcal{H}_r$, $V_+^2=-V_-^2=U$, and their ranges are orthogonal, $V_+f\perp V_-f$. Now let

$$F_\pm(t)=V_\pm F(t),\quad G_\pm(t)=V_\pm G(t). \tag{A4}$$

Then

$$\begin{aligned}\partial_t F_+(t)&=\frac{1}{\sqrt{2}}(1+U)QU|N|F(t)-G_+(t)\\&=\frac{1}{\sqrt{2}}(U-1)|N|F(t)-G_+(t)\\&=-|N|\frac{1}{\sqrt{2}}(1-U)F(t)-G_+(t)\\&=-|N|F_-(t)-G_+(t),\end{aligned}$$

and similarly $\partial_t F_-(t)=+|N|F_+(t)-G_-(t)$, so

$$\partial_t\begin{pmatrix}F_+(t)\\F_-(t)\end{pmatrix}=\begin{pmatrix}0&-|N|\\|N|&0\end{pmatrix}\begin{pmatrix}F_+(t)\\F_-(t)\end{pmatrix}-\begin{pmatrix}G_+(t)\\G_-(t)\end{pmatrix}. \tag{A5}$$

Note that, although $F_+(t)\perp F_-(t)$, each of them is not confined to a fixed subspace of $\mathcal{H}_r$. Also, since

$$\exp[Nt]Q=\exp[U|N|t]Q=\{\cos(|N|t)+U\sin(|N|t)\}Q,$$

a generalization of de Moivre's formula, we have for vanishing $G(t)$

$$\begin{aligned}F(t)&=PF(t)+\cos(|N|t)QF(0)+\sin(|N|t)UQF(0)\\&=PF(t)+F_a(t)+F_b(t).\end{aligned} \tag{A6}$$

Here $F_a(t)\perp F_b(t)$ and each is confined to a fixed subspace of $\mathcal{H}_r$, but now their equations of motion are decoupled. Starting from Eq. (A5), we can follow a procedure similar to that used in Sec. III: With $\hat{\xi}=-|N|^{-1}F_-(t)$,

$$\begin{aligned}F_+(t)&=-\partial_t\hat{\xi}-|N|^{-1}G_-(t),\\F_-(t)&=-|N|\hat{\xi},\end{aligned} \tag{A7}$$

and

$$\partial_t^2\hat{\xi}+|N|^2\hat{\xi}=G_+(t)-|N|^{-1}\partial_t G_-(t), \tag{A8}$$

which can be obtained from Hamilton's principle with

$$L=\frac{1}{2}(\partial_t\hat{\xi},\partial_t\hat{\xi})-\frac{1}{2}(|N|\hat{\xi},|N|\hat{\xi}) + (G_+(t)-|N|^{-1}\partial_t G_-(t),\hat{\xi}). \tag{A9}$$

Note that here everything is restricted to $Q\mathcal{H}_r$. The remaining dynamics

$$\partial_t PF(t)=-PG(t) \tag{A10}$$

can also be incorporated through a new coordinate field ξ_0 and also gauge transformations in the spirit of Eq. (3.20) can be introduced.

APPENDIX B: FOCK SPACE

Given the Hilbert space $\mathcal{H}$, the symmetric Fock space $\mathcal{F}(\mathcal{H})$ over $\mathcal{H}$ is defined as

$$\mathcal{F}(\mathcal{H})=\mathbb{C}\oplus\mathcal{H}\oplus(\mathcal{H}\otimes\mathcal{H})_{sym}\oplus(\mathcal{H}\otimes\mathcal{H}\otimes\mathcal{H})_{sym}\oplus\cdots =\oplus_{n=0}^{\infty}\mathcal{F}_n, \tag{B1}$$

where $(\mathcal{H}\otimes\mathcal{H})_{sym}$ etc., are symmetrized tensor products. With $f=(f^{(0)},f^{(1)},f^{(2)},f^{(3)},\ldots)$, $f^n\in\mathcal{F}_n$ and, similarly for g, the inner product on $\mathcal{F}$ is defined as

$$(f,g)=\sum_{n=0}^{\infty}\frac{1}{n!}(f^{(n)},g^{(n)})_n, \tag{B2}$$

where $(\ ,\ )_n$ is the inner product on $\otimes_{k=1}^n\mathcal{H}$ [so $(\ ,\ )_1$ is the inner product on $\mathcal{H}$]. With $\varphi\in\mathcal{H}$ and $f^{(n)}=(f_{k_1}^{(n)}\otimes\cdots\otimes f_{k_n}^{(n)})_{sym}=\Sigma_{k_1,\ldots,k_n}f_{k_1}^{(n)}\otimes\cdots\otimes f_{k_n}^{(n)}$, where the sum is over all permutations of $1,\ldots,n$, we define $a(\varphi)$ and $a^*(\varphi)$ by

$$[a(\varphi)f]_{n-1}=\sum_{k_1,\ldots,k_n}f_{k_1}^{(n)}\otimes\cdots\otimes f_{k_{n-1}}^{(n)}(f_{k_n}^{(n)},\varphi)_1, \tag{B3}$$

$$[a^*(\varphi)f]_{n+1}=\sum_{k_1,\ldots,k_{n+1}}f_{k_1}^{(n)}\otimes\cdots\otimes f_{k_{n+1}}^{(n)},\quad f_{n+1}^{(n)}=\varphi.$$

Since a general element of $\mathcal{F}_n$ can be written as a linear combination of $f^{(n)}$'s of the above type, this fixes $a^*(\varphi)$ and $a(\varphi)$. Explicitly, for the case $\mathcal{H}=L^2(\mathbb{R}^d)$,

$$[a(\varphi)f]_{n-1}(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_{n-1})=\int d\mathbf{x}_n \times f^{(n)}(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_n)\overline{\varphi}(\mathbf{x}_n),$$

$$[a^*(\varphi)f]_{n+1}(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_{n+1}) =\sum_{j=1}^{n}\varphi(\mathbf{x}_j) \times f^{(n)}(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_{j-1},\mathbf{x}_{n+1},\mathbf{x}_{j+1},\ldots,\mathbf{x}_n) +\varphi(\mathbf{x}_{n+1})f^n(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_n). \tag{B4}$$

These definitions result in the equations used in the main text such as the commutation relations (5.4).

As discussed in the main text, the field Hamiltonian has the form

$$H_f=\sum_{\alpha}\int d\lambda\ \lambda a^*(u_{\lambda\alpha})a(u_{\lambda\alpha}). \tag{B5}$$

Since H_f leaves each Fock layer $\mathcal{F}_n$ invariant, we can decompose it as a direct sum of its components on the layers

$$H_f=\oplus_{n=0}^{\infty}H_f^{(n)}, \tag{B6}$$

where

$$H_f^{(0)}=0,\quad H_f^{(1)}=N_1,\quad H_f^{(2)}=I\otimes N_1+N_1\otimes I,\ldots. \tag{B7}$$

This expression is a convenient starting point for the study of spectral properties associated with atomic decay in a dielectric with a band gap.

[1] J. D. Joannopoulos, R. D. Meade, and J. N. Winn, *Photonic Crystals* (Princeton University Press, Princeton, 1995).

[2] R. Sprik, B. A. van Tiggelen, and A. Lagendijk, Europhys. Lett. **35**, 265 (1996).

[3] *Scattering and Localization of Classical Waves in Random Media*, edited by Ping Sheng (World Scientific, Singapore, 1990).

[4] M. Weidemüller, A. Hemmerich, A. Görlitz, T. Esslinger, and T. W. Hänsch, Phys. Rev. Lett. **75**, 4583 (1995); G. Birkl, M. Gatzke, I. H. Deutsch, S. L. Rolston, and W. D. Phillips, *ibid.* **75**, 2823 (1995); J. N. Tan, J. J. Bollinger, B. Jelenkovic, and D. J. Wineland, *ibid.* **75**, 4198 (1995).

[5] D. V. van Coevorden, R. Sprik, A. Tip, and A. Lagendijk, Phys. Rev. Lett. **77**, 2412 (1996).

[6] A. van Blaaderen, R. Ruel, and P. Wiltzius, Nature (London) **385**, 321 (1997).

[7] V. L. Ginzburg and V. N. Tsytovich, *Transition Radiation and Transition Scattering* (Hilger, Bristol, 1990).

[8] A. E. Kaplan, C. T. Law, and P. L. Shkolnikov, Phys. Rev. A **52**, 6795 (1995).

[9] R. J. Glauber and M. Lewenstein, Phys. Rev. A **43**, 467 (1991).

[10] B. Huttner and S. M. Barnett, Phys. Rev. A **46**, 4306 (1992).

[11] R. Matloob, R. Loudon, S. Barnett, and J. Jeffers, Phys. Rev. A **52**, 4823 (1995).

[12] G. Kweon and N. M. Lawandy, Opt. Commun. **118**, 388 (1995).

[13] E. C. G. Sudarshan and N. Mukunda, *Classical Dynamics: A Modern Perspective* (Wiley, New York, 1974).

[14] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Photons and Atoms* (Wiley, New York, 1989).

[15] H. J. S. Dorren and A. Tip, J. Math. Phys. **32**, 3060 (1991).
[16] T. Kato, *Perturbation Theory for Linear Operators*, 2nd ed. (Springer-Verlag, Berlin, 1984).
[17] C. Itzykson and J.-B. Zuber, *Quantum Field Theory* (McGraw-Hill, New York, 1980).
[18] A. Tip, J. Math. Phys. **38**, 3545 (1997).
[19] M. Reed and B. Simon, *Methods of Modern Mathematical Physics IV, Analysis of Operators* (Academic, New York, 1978).
[20] R. Carmona and J. Lacroix, *Spectral Theory of Random Schrödinger Operators* (Birkhäuser, Basel, 1990).
[21] A. Figotin and A. Klein, Commun. Math. Phys. **184**, 411 (1997).
[22] J-.M. Combes, P. Hislop, C. Shubin, and A. Tip (unpublished).
[23] A. Tip, J. Math. Phys. **35**, 113 (1994).
[24] H. A. Kramers, *Rapport du 8e conseil Solvay* (Solvay Foundation, Brussels, 1950), p. 241.
[25] N. G. van Kampen, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **26**, 41 (1951).
[26] L. Rosenfeld, *Théorie des Électrons* (Hermann, Paris, 1951) [*Theory of Electrons* (North-Holland, Amsterdam, 1951)].
[27] S. R. de Groot, *The Maxwell Equations* (North-Holland, Amsterdam, 1969).
[28] F. N. H. Robinson, Physica (Amsterdam) **54**, 329 (1971).
[29] E. E. Salpeter and H. A. Bethe, Phys. Rev. **84**, 1232 (1951).
[30] C. W. Gardiner, *Handbook of Stochastic Methods*, 2nd ed. (Springer, Berlin, 1985).
[31] P. D. Hislop and I. M. Sigal, *Introduction to Spectral Theory with Applications to Schrödinger Operators* (Springer, Berlin, 1996).
[32] A. Tip (unpublished).