

Hamiltonian formalism for charged-particle systems interacting with absorptive dielectrics

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We use the auxiliary field method to obtain a quantized Hamilton formalism for charged particles interacting with absorptive dielectrics. We give various equivalent representations. For absorptive dielectrics extending over all space, such as absorptive photonic crystals, our formalism is equivalent to that of Dung *et al.* [Phys. Rev. A **65**, 043813 (2002)]. We also show that, as for nonabsorptive photonic crystals, to leading order the excited states of embedded two-level atoms do not decay radiatively if their transition frequencies are in a band gap.

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I. INTRODUCTION**A. General**

Interactions between atoms or molecules are usually divided into two categories, Coulomb and radiative ones. In many situations only one of the two has to be taken into account. In scattering situations, Coulomb interactions (supplemented with spin interactions if needed) suffice, whereas in an opposite case, such as superradiant systems, Coulomb interactions are usually neglected and only the interaction through the radiation field is considered. But there exist situations where both can be important. This is the case with the transfer of energy between particles such as atoms, molecules, or nanostructures at fixed positions in a background filled with an, in general absorptive, dielectric, the subject studied here.

For vacuum or other homogeneous nonabsorptive dielectrics and using the Coulomb gauge, the transfer processes can be separated into radiative ones (photon emission and reabsorption) and electrostatic ones, determined by Coulomb interactions. The latter are often subdivided into Dexter [1] processes, relevant for particles at small distances, and Förster processes [2,3], where the particles are at a larger distance. See Ref. [1] for a discussion of the various nonradiative processes encountered in practice.

Consider a typical situation with identical atoms at fixed positions in a homogeneous background with one initially excited and the others in their ground states. Then the excited atom can decay radiatively or transfer its energy to another one (resonant energy transfer). If only a finite number is present, we are dealing with a large molecule with fixed nuclei. Generically this molecule is not in an (excited) eigenstate. In this situation radiative and Förster processes are of a different nature. Radiative decay is possible and a definite decay rate exists. But if only Förster processes take place the state of the system shows an oscillating behavior in time and a rate does not exist. This changes for an infinite number of particles, randomly distributed in an otherwise spatially homogeneous medium. Then a hopping (random walk) model for the energy transfer is often employed. For a discussion of its properties, see Refs. [4,5]. But, as far as the author is aware, it has never been derived starting from a Schrödinger

or Liouville–von Neumann description.

Note that the separation into radiative and nonradiative processes is rather arbitrary. If one changes the gauge for the electromagnetic potentials, the division becomes different. In the extreme case of the Weyl or temporal gauge, all interactions become radiative in the sense that they are all determined by the vector potential. Neglecting contact interactions this is also true for the multipolar representation.

Recently Dung *et al.* considered the situation for an absorptive dielectric [6–8]. Their formalism is initially setup for dielectrics extending over all space. Different configurations are handled by a limiting procedure at the end of a calculation. Among other matters these authors investigated resonant energy transfer between two embedded atoms.

Here we take a somewhat different point of view, using the auxiliary field formalism developed by the author [9]. It does not require the dielectric to extend over all space and involves the use of a generalized Coulomb gauge, resulting in a splitting into nonradiative and radiative interactions that is close to the conventional one in the Coulomb gauge. Then the scalar potential obeys a Poisson equation featuring the static permeability $\varepsilon_{stat}(\mathbf{x})$. This may not be a convenient starting point for high-frequency processes such as resonant energy transfer. Thus we consider different, unitarily equivalent, Hamiltonians, which are better suited. In particular, we retrieve the Hamiltonian used by Dung *et al.* [6] for dielectrics extending over all space. For an example involving resonant energy transfer considered earlier by the above authors we confirm their result, which indicates that their limiting procedure is compatible with the present setup.

We recall that photonic crystals are special dielectrics with a spatial periodicity in $\varepsilon(\mathbf{x}, \omega)$,

$$\varepsilon(\mathbf{x}, \omega) = \varepsilon(\mathbf{x} + n_k \mathbf{a}_k, \omega), \quad n_k \in \mathbb{Z}, \quad (1.1)$$

where the \mathbf{a}_k 's, the lattice vectors, are linearly independent. Depending on their number, we are dealing with one-, two-, or three-dimensional photonic crystals. In the conservative case, where $\varepsilon = \varepsilon(\mathbf{x})$ is frequency independent, the radiative mode spectrum is given by the spectrum of the Helmholtz operator which can have gaps, the photonic band gaps. Excited embedded atoms with transition frequency ω_0 in the gap no longer decay radiatively, simply because there are no field modes present to carry away the excitation energy. In fact their excited states remain stable bound states. Numeri-

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cal work [10] shows that gaps can still exist for dispersive, nonabsorptive media, but if absorption is present, the situation changes, gaps do not exist in frequency intervals with $\text{Im } \varepsilon(\mathbf{x}, \omega) \neq 0$. For a detailed analysis, based upon the auxiliary field formalism, we refer to Ref. [11]. But in intervals on which $\text{Im } \varepsilon(\mathbf{x}, \omega) = 0$ gaps can still exist, again inhibiting radiative decay. But, as discussed in the sequel, it is fairly complicated to show that excited atomic states remain stable.

In setting up a tractable theoretical description for a general absorptive dielectric with embedded particles a number of initial assumptions must be made.

Assumption 1. The system contains particles at fixed positions (Born-Oppenheimer approximation) such as atoms or molecules, which we assume optically small, i.e., for radiative processes we can evaluate the vector potential at their nuclear or center-of-mass coordinates (long-wavelength approximation).

Assumption 2. The background consists of a linear, in general absorptive and spatially inhomogeneous, dielectric, where the real and imaginary parts of the frequency-dependent electric permeability $\varepsilon(\mathbf{x}, \omega)$ are connected by a Kramers-Kronig relation (causality). The macroscopic Maxwell's equations are assumed to be valid, thus $\varepsilon(\mathbf{x}, \omega)$ is a given quantity.

Assumption 3. A perennial matter is the choice of the field in which the particles are placed. Does the dielectric extend into a particle or not? We assume that it does not. Given the macroscopic description, this seems the only logical choice, the alternative breaking down on an atomic scale. Keeping the particles isolated from the dielectric resolves this dilemma. For rare-earth atoms (often used in optoelectronic devices) with an active inner shell electron this seems reasonable. Note that the associated excluded volume effects are automatically taken care of by the corresponding boundary conditions in the determination of the eigenmodes of the Helmholtz equation. No separate calculations of fields in empty spheres surrounded by a dielectric are required.

B. Maxwell's equations

A proper treatment requires a Hamiltonian formalism in which both particles and electromagnetic fields are quantized. It is only recently that a general quantization method was developed for absorptive dielectrics. At present two general methods are available. The first consists of adding an appropriate quantum Langevin noise term to the vacuum quantized Maxwell's equations [12,13], whereas the second, developed by the author [9], starts on a classical level by introducing auxiliary fields, thus restoring energy conservation and making quantization possible [14]. The two methods have been shown to be equivalent [15]. Here we use the second method and shall consider a number of equivalent Hamiltonians.

Let us first introduce Maxwell's equations for the case at hand. Starting point is the set of macroscopic, classical, Maxwell's equations for an isotropic, linear, absorptive dielectric. Thus, setting $\varepsilon_0 = \mu_0 = c = 1$ and $\mathbf{H} = \mathbf{B}$, we have

$$\partial_t \mathbf{D}(\mathbf{x}, t) = \partial_{\mathbf{x}} \times \mathbf{B}(\mathbf{x}, t) - \mathbf{J}(\mathbf{x}, t),$$

$$\partial_t \mathbf{B}(\mathbf{x}, t) = -\partial_{\mathbf{x}} \times \mathbf{E}(\mathbf{x}, t),$$

$$\partial_{\mathbf{x}} \cdot \mathbf{D}(\mathbf{x}, t) = \rho(\mathbf{x}, t), \quad \partial_{\mathbf{x}} \cdot \mathbf{B}(\mathbf{x}, t_0) = 0,$$

$$\mathbf{D}(\mathbf{x}, t) = \mathbf{E}(\mathbf{x}, t) + \mathbf{P}(\mathbf{x}, t),$$

$$\mathbf{P}(\mathbf{x}, t) = \int_{t_0}^t ds \chi(\mathbf{x}, t-s) \mathbf{E}(\mathbf{x}, s), \quad (1.2)$$

with t_0 some initial time which can be taken as $t_0 = -\infty$. We also assume $\chi(\mathbf{x}, 0) = 0$ (no initial current surges). The charge and current densities $\rho(\mathbf{x}, t)$ and $\mathbf{J}(\mathbf{x}, t)$ are due to the charges (mass m_n , charge e_n , and position \mathbf{x}_n) constituting embedded atoms or molecules,

$$\rho(\mathbf{x}, t) = \sum_n e_n \delta(\mathbf{x} - \mathbf{x}_n(t)),$$

$$\mathbf{J}(\mathbf{x}, t) = \sum_n e_n \dot{\mathbf{x}}_n \delta(\mathbf{x} - \mathbf{x}_n(t)). \quad (1.3)$$

As usual we now express the fields in terms of the vector and scalar potentials

$$\mathbf{E}(\mathbf{x}, t) = -\partial_t \mathbf{A}(\mathbf{x}, t) - \partial_{\mathbf{x}} \Phi(\mathbf{x}, t),$$

$$\mathbf{B}(\mathbf{x}, t) = \partial_{\mathbf{x}} \times \mathbf{A}(\mathbf{x}, t). \quad (1.4)$$

In view of the convolutive relation between \mathbf{P} and \mathbf{E} , it is convenient to switch to their Fourier transforms. With the space and frequency dependent, complex electric permeability (permittivity, dielectric function) $\varepsilon(\mathbf{x}, z)$ given by

$$\varepsilon(\mathbf{x}, z) = 1 + \hat{\chi}(\mathbf{x}, z),$$

$$\hat{\chi}(\mathbf{x}, z) = \int_0^{\infty} dt \exp[izt] \chi(\mathbf{x}, t), \quad \text{Im } z \geq 0, \quad (1.5)$$

we obtain, denoting Fourier transforms by a tilde,

$$\{\omega^2 \varepsilon(\mathbf{x}, \omega) - \mathbf{h}_0\} \tilde{\mathbf{A}}(\mathbf{x}, \omega) + i\omega \varepsilon(\mathbf{x}, \omega) \partial_{\mathbf{x}} \tilde{\Phi}(\mathbf{x}, \omega) = -\tilde{\mathbf{J}}(\mathbf{x}, \omega),$$

$$\mathbf{p} \cdot \varepsilon(\mathbf{x}, \omega) \mathbf{p} \tilde{\Phi}(\mathbf{x}, \omega) + i\omega \partial_{\mathbf{x}} \{\varepsilon(\mathbf{x}, \omega) \tilde{\mathbf{A}}(\mathbf{x}, \omega)\} = \tilde{\rho}(\mathbf{x}, \omega). \quad (1.6)$$

Here, with \mathbf{U} the unit 3×3 matrix and $\mathbf{e}_a = \mathbf{a}/a$, $a = |\mathbf{a}|$,

$$\mathbf{h}_0 = \mathbf{p}^2 \mathbf{U} - \mathbf{p} \mathbf{p} = \mathbf{p}^2 \mathbf{\Delta}_p = \partial_{\mathbf{x}} \times (\partial_{\mathbf{x}} \times \dots),$$

$$\mathbf{p} = -i \partial_{\mathbf{x}}, \quad \mathbf{\Delta}_p = \mathbf{U} - \mathbf{e}_p \mathbf{e}_p. \quad (1.7)$$

Note that $\mathbf{P}^{\parallel} = \mathbf{e}_p \mathbf{e}_p$ projects upon the longitudinal component \mathbf{f}^{\parallel} and $\mathbf{P}^{\perp} = \mathbf{\Delta}_p$ upon the transverse component \mathbf{f}^{\perp} of a three-dimensional vector field \mathbf{f} . In the sequel we denote by p^{-2} the inverse of $-\partial_{\mathbf{x}}^2$, so $-\partial_{\mathbf{x}}^2 p^{-2} = 1$.

In the vacuum case, $\varepsilon(\mathbf{x}, \omega) = \varepsilon_0 = 1$, and in the Coulomb gauge, $\partial_{\mathbf{x}} \mathbf{A} = 0$, the first equation leads to an Helmholtz equation for $\tilde{\mathbf{A}}$, not involving $\tilde{\Phi}(\mathbf{x}, \omega)$, whereas the second equation reduces to the Poisson equation. But in our situa-

tion, adopting the Coulomb gauge, the vector and scalar potentials remain coupled. Instead of the Coulomb gauge we can set

$$\partial_{\mathbf{x}} \cdot \{\varepsilon(\mathbf{x}, \omega) \tilde{\mathbf{A}}(\mathbf{x}, \omega)\} = 0, \quad (1.8)$$

thus obtaining the Poisson equation

$$-\partial_{\mathbf{x}} \cdot \{\varepsilon(\mathbf{x}, \omega) \partial_{\mathbf{x}} \tilde{\Phi}(\mathbf{x}, \omega)\} = \mathbf{p} \cdot \varepsilon(\mathbf{x}, \omega) \mathbf{p} \tilde{\Phi}(\mathbf{x}, \omega) = \tilde{\rho}(\mathbf{x}, \omega). \quad (1.9)$$

Eliminating $\tilde{\Phi}$ we then obtain

$$\begin{aligned} & \{\omega^2 \varepsilon(\mathbf{x}, \omega) - \hbar_0 \tilde{\mathbf{A}}(\mathbf{x}, \omega)\} \\ & = -\tilde{\mathbf{J}}(\mathbf{x}, \omega) + \omega \varepsilon(\mathbf{x}, \omega) \mathbf{p} [\mathbf{p} \cdot \varepsilon(\mathbf{x}, \omega) \mathbf{p}]^{-1} \tilde{\rho}(\mathbf{x}, \omega), \end{aligned} \quad (1.10)$$

which is still quite complicated. Even accepting this, it is not clear that a Lagrange-Hamilton formalism can be found where the vacuum Coulomb potentials are replaced by the above $\tilde{\Phi}$, obeying Eq. (1.9). But in the auxiliary field formalism [9] these problems are overcome. After quantization various unitary transformations lead to different representations, the main subject of this work. Inner products are denoted as $(f, g) = \langle g | f \rangle$, infinite self-energies appearing in various summations are dismissed.

II. THE AUXILIARY FIELD FORMALISM

In this section we briefly present the essentials of the auxiliary field formalism, at the same time introducing the notation employed in the sequel. Using $\chi(\mathbf{x}, 0) = 0$ and introducing the Fourier transform $\nu(\mathbf{x}, \lambda)$ of $\chi'(\mathbf{x}, t)$ we have

$$\chi(\mathbf{x}, t) = \int d\lambda \nu(\mathbf{x}, \lambda) \lambda^{-1} \sin \lambda t, \quad \chi(\mathbf{x}, 0) = 0,$$

$$\chi'(\mathbf{x}, t) = \int d\lambda \nu(\mathbf{x}, \lambda) \cos \lambda t, \quad \chi'(\mathbf{x}, 0) = \int d\lambda \nu(\mathbf{x}, \lambda),$$

$$\begin{aligned} \hat{\chi}(\mathbf{x}, z) &= z^{-1} \int d\lambda [\lambda - z]^{-1} \nu(\mathbf{x}, \lambda) \\ &= \int d\lambda [\lambda^2 - z^2]^{-1} \nu(\mathbf{x}, \lambda), \end{aligned}$$

$$\hat{\chi}(\mathbf{x}, 0) = \int d\lambda \lambda^{-2} \nu(\mathbf{x}, \lambda),$$

$$\varepsilon(\mathbf{x}, \lambda) = 1 + \hat{\chi}(\mathbf{x}, \lambda + i0),$$

$$\text{Im } \varepsilon(\mathbf{x}, \lambda) = \text{Im } \hat{\chi}(\mathbf{x}, \lambda + i0) = \frac{\pi}{\lambda} \nu(\mathbf{x}, \lambda),$$

$$\begin{aligned} \varepsilon_{stat}(\mathbf{x}) &\equiv \varepsilon(\mathbf{x}, 0) = 1 + \hat{\chi}_{stat}(\mathbf{x}) = 1 + \hat{\chi}(\mathbf{x}, 0) \\ &= 1 + \int d\lambda \lambda^{-2} \nu(\mathbf{x}, \lambda), \end{aligned}$$

$$\nu(\mathbf{x}, \lambda) = \pi^{-1} \int_0^\infty dt \cos \lambda t \chi'(\mathbf{x}, t), \quad \nu(\mathbf{x}, 0) = 0. \quad (2.1)$$

We now set $\mathbf{F}_1(\mathbf{x}, t) = \mathbf{E}(\mathbf{x}, t)$, $\mathbf{F}_3(\mathbf{x}, t) = \mathbf{B}(\mathbf{x}, t)$, $\nu(\mathbf{x}, \lambda) = \sigma(\mathbf{x}, \lambda)^2$, $\sigma \geq 0$, and introduce

$$\mathbf{F}_2(\mathbf{x}, \lambda, t) = -\sigma(\mathbf{x}, \lambda) \int_{t_0}^t ds \sin \lambda(t-s) \mathbf{F}_1(\mathbf{x}, s),$$

$$\mathbf{F}_4(\mathbf{x}, \lambda, t) = -\sigma(\mathbf{x}, \lambda) \int_{t_0}^t ds \cos \lambda(t-s) \mathbf{F}_1(\mathbf{x}, s). \quad (2.2)$$

Note that $\sigma(\mathbf{x}, \lambda)$ and $\mathbf{F}_{2,4}(\mathbf{x}, \lambda, t)$ vanish outside the dielectric material and that $\mathbf{E}(\mathbf{x}, t)$ is transverse in this region. Now

$$\partial_t \mathbf{F}_1(\mathbf{x}, t) = \partial_{\mathbf{x}} \times \mathbf{F}_3(\mathbf{x}, t) + \int d\lambda \sigma(\mathbf{x}, \lambda) \mathbf{F}_4(\mathbf{x}, \lambda, t) - \mathbf{J}(\mathbf{x}, t),$$

$$\partial_t \mathbf{F}_2(\mathbf{x}, \lambda, t) = \lambda \mathbf{F}_4(\mathbf{x}, \lambda, t),$$

$$\partial_t \mathbf{F}_3(\mathbf{x}, t) = -\partial_{\mathbf{x}} \times \mathbf{F}_1(\mathbf{x}, t),$$

$$\partial_t \mathbf{F}_4(\mathbf{x}, \lambda, t) = -\sigma(\mathbf{x}, \lambda) \mathbf{F}_1(\mathbf{x}, t) - \lambda \mathbf{F}_2(\mathbf{x}, \lambda, t), \quad (2.3)$$

or, in compact form,

$$\partial_t \mathbf{F}(\mathbf{x}, t) = \mathbf{N} \mathbf{F}(\mathbf{x}, t) - \mathbf{G}(\mathbf{x}, t), \quad (2.4)$$

where

$$\mathbf{G}_1(\mathbf{x}, t) = \mathbf{J}(\mathbf{x}, t), \quad \mathbf{G}_{2,3,4}(\mathbf{x}, t) = 0. \quad (2.5)$$

As discussed earlier [9,11], Eq. (2.4) with the initial conditions

$$\mathbf{F}_2(\mathbf{x}, \lambda, t_0) = \mathbf{F}_4(\mathbf{x}, \lambda, t_0) = 0, \quad (2.6)$$

is equivalent to the original set of Maxwell's equations. The operator

$$\begin{aligned} \mathbf{N} &= \begin{pmatrix} 0 & \mathbf{N}_{em} \\ \mathbf{N}_{me} & 0 \end{pmatrix}, \\ \mathbf{N}_{em} &= \begin{pmatrix} \partial_{\mathbf{x}} \times & \langle \sigma(\mathbf{x}, \lambda) | \\ 0 & \lambda \end{pmatrix}, \\ \mathbf{N}_{me} &= \begin{pmatrix} -\partial_{\mathbf{x}} \times & 0 \\ -\sigma(\mathbf{x}, \lambda) & 0 \end{pmatrix}, \end{aligned} \quad (2.7)$$

where $\langle \sigma(\mathbf{x}, \lambda) | = \int d\lambda \sigma(\mathbf{x}, \lambda) \dots$, has block structure and

$$\mathbf{N}^2 = - \begin{pmatrix} \mathbf{H}_e & 0 \\ 0 & \mathbf{H}_m \end{pmatrix}, \quad (2.8)$$

where

$$\mathbf{H}_e = \begin{pmatrix} \hbar_0 + \chi'(\mathbf{x}, 0) & \langle \lambda \sigma(\mathbf{x}, \lambda) | \\ \lambda \sigma(\mathbf{x}, \lambda) & \lambda^2 \end{pmatrix}. \quad (2.9)$$

We note that \mathbf{N} is anti-self-adjoint, $\mathbf{N}^* = -\mathbf{N}$, in the real Hilbert space

$$\mathcal{H} = \oplus_{j=1}^4 \mathcal{H}_j, \quad \mathcal{H}_1 = \mathcal{H}_3 = L^2(\mathbb{R}^3, d\mathbf{x}; \mathbb{R}^3),$$

$$\mathcal{H}_2 = \mathcal{H}_4 = L^2(\mathbb{R}^3, d\mathbf{x}; \mathbb{R}^3) \otimes L^2(\mathbb{R}, d\lambda), \quad (2.10)$$

with inner product

$$(\mathbf{f}, \mathbf{g}) = \sum_{j=1}^4 (\mathbf{f}_j, \mathbf{g}_j)_j. \quad (2.11)$$

For details, see Refs. [9,11]. We shall occasionally write for $\mathbf{f} \in \mathcal{H}$,

$$\mathbf{f} = \begin{pmatrix} \mathbf{f}_e \\ \mathbf{f}_m \end{pmatrix}, \quad (2.12)$$

where $\mathbf{f}_e \in \mathcal{H}_e = \mathcal{H}_1 \oplus \mathcal{H}_2$ with inner product $(\mathbf{f}_e, \mathbf{g}_e)_e$ and $\mathbf{f}_m \in \mathcal{H}_m = \mathcal{H}_3 \oplus \mathcal{H}_4$ with inner product $(\mathbf{f}_m, \mathbf{g}_m)_m$. Next we determine the projector \mathbf{P} upon the null space $\mathcal{N}(\mathbf{N})$ of \mathbf{N} . The condition $\mathbf{N}^* \mathbf{f} = 0$ gives

$$\mathbf{f} = \mathbf{M}\phi, \quad \mathbf{M} = \begin{pmatrix} \mathbf{M}_e \\ \mathbf{M}_m \end{pmatrix}, \quad \phi = \begin{pmatrix} \varphi_e(\mathbf{x}) \\ \varphi_m(\mathbf{x}) \end{pmatrix},$$

$$\mathbf{M}_e = \begin{pmatrix} \partial_{\mathbf{x}} & 0 \\ -\frac{\sigma(\mathbf{x}, \lambda)}{\lambda} \partial_{\mathbf{x}} & 0 \end{pmatrix}, \quad \mathbf{M}_m = \begin{pmatrix} 0 & \partial_{\mathbf{x}} \\ 0 & 0 \end{pmatrix}, \quad (2.13)$$

where $\varphi_{e,m}(\mathbf{x})$ are scalar functions. Now

$$\mathbf{M}^* = \begin{pmatrix} -\partial_{\mathbf{x}} & \partial_{\mathbf{x}} \left\langle \frac{\sigma(\mathbf{x}, \lambda)}{\lambda} \right\rangle & 0 & 0 \\ 0 & 0 & -\partial_{\mathbf{x}} & 0 \end{pmatrix} \quad (2.14)$$

and

$$\mathbf{M}^* \mathbf{M} = \begin{pmatrix} \mathbf{P} \varepsilon_{stat}(\mathbf{x}) \mathbf{P} & 0 \\ 0 & \mathbf{P}^2 \end{pmatrix}. \quad (2.15)$$

Then

$$\mathbf{P} = \mathbf{M}(\mathbf{M}^* \mathbf{M})^{-1} \mathbf{M}^* = \begin{pmatrix} \mathbf{P}_e & 0 \\ 0 & \mathbf{P}_m \end{pmatrix} = \mathbf{1} - \mathbf{Q}, \quad (2.16)$$

where

$$\mathbf{P}_e = \mathbf{1} - \mathbf{Q}_e = \begin{pmatrix} \frac{1}{\mathbf{P} \cdot \varepsilon_{stat}(\mathbf{x}) \mathbf{P}} & \frac{1}{\mathbf{P} \cdot \varepsilon_{stat}(\mathbf{x}) \mathbf{P}} \left\langle \frac{\sigma(\mathbf{x}, \lambda)}{\lambda} \right\rangle \\ \frac{\sigma(\mathbf{x}, \lambda)}{\lambda} \frac{1}{\mathbf{P} \cdot \varepsilon_{stat}(\mathbf{x}) \mathbf{P}} & \frac{\sigma(\mathbf{x}, \lambda)}{\lambda} \frac{1}{\mathbf{P} \cdot \varepsilon_{stat}(\mathbf{x}) \mathbf{P}} \left\langle \frac{\sigma(\mathbf{x}, \lambda)}{\lambda} \right\rangle \end{pmatrix},$$

$$\mathbf{P}_m = \begin{pmatrix} \mathbf{P}^{\parallel} & 0 \\ 0 & 0 \end{pmatrix}. \quad (2.17)$$

III. LAGRANGE-HAMILTON FORMALISM IN THE C GAUGE

We assume that the charge and current densities are due to charged particles (mass m_n and charge e_n),

$$\rho(\mathbf{x}) = \sum_n e_n \delta(\mathbf{x} - \mathbf{x}_n), \quad \mathbf{J}(\mathbf{x}) = \sum_n e_n \dot{\mathbf{x}}_n \delta(\mathbf{x} - \mathbf{x}_n). \quad (3.1)$$

Next we introduce potentials [9,14]

$$\mathbf{F}_e = -\partial_t \boldsymbol{\xi} - M \boldsymbol{\zeta}, \quad \mathbf{F}_m = -\mathbf{N}_{me} \boldsymbol{\xi}, \quad (3.2)$$

which is a generalization of the situation in the vacuum case. Clearly

$$\mathbf{F}_1 = -\partial_t \boldsymbol{\xi}_1 - \partial_{\mathbf{x}} \zeta, \quad \mathbf{F}_3 = \partial_{\mathbf{x}} \times \boldsymbol{\xi}_1, \quad (3.3)$$

so we identify $\boldsymbol{\xi}_1$ with the vector potential \mathbf{A} and ζ with the scalar potential Φ . From Eq. (2.4),

$$\partial_t^2 \boldsymbol{\xi} + M \partial_t \boldsymbol{\zeta} = -\mathbf{H}_e \boldsymbol{\xi} + \mathbf{G}_e. \quad (3.4)$$

This equation of motion and that for the particles can be retrieved by Hamilton's principle from the Lagrangian

$$L = \frac{1}{2} (\partial_t \boldsymbol{\xi} + M \boldsymbol{\zeta}, \partial_t \boldsymbol{\xi} + M \boldsymbol{\zeta}) - \frac{1}{2} (\mathbf{H}_e \boldsymbol{\xi}, \boldsymbol{\xi}) + \sum_n \frac{1}{2} m_n \dot{\mathbf{x}}_n^2$$

$$+ (\mathbf{G}_e, \boldsymbol{\xi}) - (\rho, \zeta)_0, \quad (3.5)$$

where we dropped the subscript e in $(\mathbf{f}, \mathbf{g})_e$ and $(\phi, \psi)_0$ the inner product on $L^2(\mathbb{R}^3, d\mathbf{x})$. As in the vacuum case L does not contain $\partial_t \zeta$ and we have to fix a specific gauge in order to arrive at a Hamilton formalism. Thus we set $\mathbf{P}_e \boldsymbol{\xi} = 0$, or

$$\partial_{\mathbf{x}} \cdot \left\{ \boldsymbol{\xi}_1(\mathbf{x}) - \int d\lambda \frac{\sigma(\mathbf{x}, \lambda)}{\lambda} \boldsymbol{\xi}_2(\mathbf{x}, \lambda) \right\} = 0. \quad (3.6)$$

Note that $A(\mathbf{x}) = \xi_1(\mathbf{x})$ is transverse for \mathbf{x} outside the dielectric material. Applying $M^* = M^* P_e$ to Eq. (3.4) and noting that $P_e H_e = 0$, we obtain

$$M^* M \partial_t \zeta = M^* \mathbf{G} = -\partial_{\mathbf{x}} \mathbf{J} = \partial_t \rho, \quad (3.7)$$

so

$$-\partial_{\mathbf{x}} \varepsilon_{stat}(\mathbf{x}) \partial_{\mathbf{x}} \zeta = \rho, \quad (3.8)$$

a Poisson equation featuring $\varepsilon_{stat}(\mathbf{x})$. Since now $\xi \perp M \zeta$ we have

$$L = \frac{1}{2} (\partial_t \xi, \partial_t \xi) + \frac{1}{2} (M^* M \zeta, \zeta) - \frac{1}{2} (H_e \xi, \xi) + \sum_n \frac{1}{2} m_n \dot{\mathbf{x}}_n^2 + (\mathbf{G}_e, \xi) - (\rho, \zeta)_0. \quad (3.9)$$

The canonical momenta are

$$\boldsymbol{\pi} = \frac{\partial L}{\partial \dot{\xi}} = \partial_t \xi, \quad \mathbf{p}_n = \frac{\partial L}{\partial \dot{\mathbf{x}}_n} = m_n \dot{\mathbf{x}}_n + e_n \xi_1(\mathbf{x}_n), \quad (3.10)$$

and the Hamiltonian becomes (note that both $\boldsymbol{\pi}$ and ξ are in $\mathcal{Q}_e \mathcal{H}_e$)

$$\begin{aligned} H &= (\boldsymbol{\pi}, \partial_t \xi) + \sum_n \mathbf{p}_n \dot{\mathbf{x}}_n - L \\ &= \frac{1}{2} (\boldsymbol{\pi}, \boldsymbol{\pi}) + \frac{1}{2} (H_e \xi, \xi) + \sum_n \frac{1}{2m_n} [\mathbf{p}_n - e_n \xi_1(\mathbf{x}_n)]^2 + V_{stat} \\ &= \frac{1}{2} (\mathcal{Q}_e \boldsymbol{\pi}, \boldsymbol{\pi}) + \frac{1}{2} (H_e \xi, \xi) + \sum_n \frac{1}{2m_n} [\mathbf{p}_n - e_n \xi_1(\mathbf{x}_n)]^2 \\ &\quad + V_{stat}, \end{aligned} \quad (3.11)$$

where

$$V_{stat} = \frac{1}{2} (\rho, \zeta) \quad (3.12)$$

with ζ satisfying Eq. (3.8).

IV. QUANTIZATION

We discussed the quantization procedure earlier in Ref. [9] and here we only give a brief summary. From now onwards we use the complexified forms of the various Hilbert spaces introduced earlier. With \mathbf{f} and \mathbf{g} test functions from $\mathcal{Q}_e \mathcal{H}_e$ the basic commutation relations are

$$[\xi(\mathbf{f}), \boldsymbol{\pi}(\mathbf{g})] = i \text{Re}(\mathbf{f}, \mathbf{g}), \quad \mathbf{f}, \mathbf{g} \in \mathcal{Q}_e \mathcal{H}_e. \quad (4.1)$$

Next let $\{\mathbf{u}_{\mu\beta} | \mu \geq 0\}$ be the set of eigenfunctions of $\mathcal{Q}_e H_e$,

$$H_e \mathbf{u}_{\mu\beta} = \mu^2 \mathbf{u}_{\mu\beta}. \quad (4.2)$$

Then, in terms of the boson creation and annihilation operators $a(\mathbf{u}_{\mu\beta})$ and $a^*(\mathbf{u}_{\mu\beta})$, acting in the Fock space $\mathcal{F} = \mathcal{F}(\mathcal{H}_e)$, which satisfy, see Ref. [9],

$$[a(\mathbf{g}), a^*(\mathbf{f})] = \text{Re}(\mathbf{f}, \mathbf{g}), \quad \mathbf{f}, \mathbf{g} \in \mathcal{Q}_e \mathcal{H}_e, \quad (4.3)$$

$$\begin{aligned} \xi(\mathbf{f}) &= \sum_{\beta} \int d\mu [2\mu]^{-1/2} \{a^*(\mathbf{u}_{\mu\beta})(\mathbf{f}, \mathbf{u}_{\mu\beta}) + a(\mathbf{u}_{\mu\beta}) \\ &\quad \times (\mathbf{u}_{\mu\beta}, \mathbf{f})\}, \end{aligned}$$

$$\begin{aligned} \boldsymbol{\pi}(\mathbf{f}) &= i \sum_{\beta} \int d\mu [\mu/2]^{1/2} \{a^*(\mathbf{u}_{\mu\beta})(\mathbf{f}, \mathbf{u}_{\mu\beta}) - a(\mathbf{u}_{\mu\beta}) \\ &\quad \times (\mathbf{u}_{\mu\beta}, \mathbf{f})\}, \end{aligned} \quad (4.4)$$

and, since now

$$H_f = \sum_{\beta} \int d\mu \mu a^*(\mathbf{u}_{\mu\beta}) a(\mathbf{u}_{\mu\beta}), \quad (4.5)$$

we have, writing $\xi_1 = A$,

$$H = H_f + \sum_n \frac{1}{2m_n} [\mathbf{p}_n - e_n A(\mathbf{x}_n)]^2 + V_{stat}. \quad (4.6)$$

However, the static potential does not always exist. Indeed, for the Drude model $\varepsilon(\omega) = 1 - \Omega^2 \omega^{-1} (\omega + i\gamma)^{-1}$, leading to an infinite V_{stat} . This can be remedied by switching to a different Hamiltonian H' , using an appropriate unitary transformation U . Hence

$$H' = U H U^{-1}, \quad (4.7)$$

where we take

$$U = \exp[-i \xi(\mathbf{W})] \quad (4.8)$$

with

$$\mathbf{W} = \begin{pmatrix} \mathbf{W}_1 \\ 0 \end{pmatrix}, \quad \mathbf{W}_1(\mathbf{x}) = \partial_{\mathbf{x}} \zeta_0(\mathbf{x}),$$

$$\zeta_0(\mathbf{x}) = \sum_n \frac{e_n}{4\pi |\mathbf{x} - \mathbf{x}_n|}, \quad -\partial_{\mathbf{x}}^2 \zeta_0(\mathbf{x}) = \rho(\mathbf{x}). \quad (4.9)$$

Thus $\mathbf{W}_1 = P^{\parallel} \mathbf{W}_1$ and $U = \exp[-i(\mathbf{W}_1, A)_1]$. The coordinates are not affected by the transformation, but

$$\begin{aligned} \mathbf{p}'_n &= U \mathbf{p}_n U^{-1} = \mathbf{p}_n + e_n A^{\parallel}(\mathbf{x}_n), \\ \boldsymbol{\pi}' &= U \boldsymbol{\pi} U^{-1} = \boldsymbol{\pi} + \mathcal{Q}_e \mathbf{W}. \end{aligned} \quad (4.10)$$

Noting that $\boldsymbol{\pi} = \mathcal{Q}_e \boldsymbol{\pi}$ we have

$$\begin{aligned} U(\boldsymbol{\pi}, \boldsymbol{\pi}) U^{-1} &= (\mathcal{Q}_e \{\boldsymbol{\pi} + \mathbf{W}\}, \boldsymbol{\pi} + \mathbf{W}) \\ &= (\mathcal{Q}_e \boldsymbol{\pi}, \boldsymbol{\pi}) + (\mathcal{Q}_e \mathbf{W}, \mathbf{W}) + 2(\mathbf{W}, \mathcal{Q}_e \boldsymbol{\pi}) \\ &= (\boldsymbol{\pi}, \boldsymbol{\pi}) + 2V_{Coul} - 2V_{stat} + 2(\mathbf{W}, \boldsymbol{\pi}). \end{aligned} \quad (4.11)$$

Here we used

$$\begin{aligned}
(\mathbf{Q}_e \mathbf{W}, \mathbf{W}) &= (\mathbf{W}, \mathbf{W}) - (\mathbf{P}_e \mathbf{W}, \mathbf{W}) = (\mathbf{W}_1, \mathbf{W}_1)_1 \\
&\quad - (\mathbf{p}[\mathbf{p} \cdot \boldsymbol{\varepsilon}_{stat} \mathbf{p}]^{-1} \mathbf{p} \cdot \mathbf{W}_1, \mathbf{W}_1)_1 \\
&= (\partial_{\mathbf{x}} \zeta_0, \partial_{\mathbf{x}} \zeta_0)_1 - ([\mathbf{p} \cdot \boldsymbol{\varepsilon}_{stat} \mathbf{p}]^{-1} \partial_{\mathbf{x}}^2 \zeta_0, \partial_{\mathbf{x}}^2 \zeta_0)_1 \\
&= (\rho, \zeta_0)_1 - ([\mathbf{p} \cdot \boldsymbol{\varepsilon}_{stat} \mathbf{p}]^{-1} \rho, \rho)_1 \\
&= 2V_{Coul} - 2V_{stat}, \tag{4.12}
\end{aligned}$$

where

$$V_{Coul} = \frac{1}{2} (\rho, \zeta_0)_1 = \frac{1}{2} \sum_{m \neq n} \frac{e_m e_n}{4\pi |\mathbf{x}_m - \mathbf{x}_n|}. \tag{4.13}$$

Thus

$$H' = H_f + (\mathbf{W}, \boldsymbol{\pi}) + \sum_n \frac{1}{2m_n} [\mathbf{p}_n - e_n \mathbf{A}^\perp(\mathbf{x}_n)]^2 + V_{Coul}. \tag{4.14}$$

Note that $(\mathbf{W}, \boldsymbol{\pi})$ depends on

$$\boldsymbol{\pi}_1^\parallel = -\mathbf{E}^\parallel - \partial_{\mathbf{x}} \zeta, \tag{4.15}$$

which does not equal \mathbf{D}^\parallel as is the case in simpler situations. In changing to the primed system we also have to transform the states ψ according to

$$\psi' = U \psi. \tag{4.16}$$

Thus, if ψ is of the form $\psi = \varphi_m \otimes \varphi_{vac}$, where φ_m is a state of the material subsystem and φ_{vac} the field vacuum state, then

$$\psi' = U \varphi_m \otimes \varphi_{vac} = \varphi_m \otimes U \varphi_{vac}, \tag{4.17}$$

which now contains contributions from all Fock layers and *vice versa*.

V. A REPRESENTATION IN TERMS OF FREE FIELDS AND WAVE OPERATORS

We now turn to a formulation which features the free electromagnetic and auxiliary fields. In \mathcal{H}_e we define

$$\mathbf{H}_0 = \begin{pmatrix} \hbar_0 & 0 \\ 0 & \lambda^2 \end{pmatrix}, \quad \mathbf{Q}_0 = 1 - \mathbf{P}_0 = \begin{pmatrix} \mathbf{P}^\perp & 0 \\ 0 & 1 \end{pmatrix}. \tag{5.1}$$

Then the wave operators

$$\Omega_\pm = \lim_{t \rightarrow \pm\infty} \exp[iH_e t] \exp[-iH_0 t] \mathbf{Q}_0 \tag{5.2}$$

exist under certain conditions, see Appendix A. Special cases are a finite dielectric in vacuum and certain absorptive dielectrics extending over all space, in particular photonic crystals. The wave operators have the properties

$$\mathbf{H}_e = \Omega_\pm \mathbf{H}_0 \Omega_\pm^*, \quad \Omega_\pm \Omega_\pm^* = \mathbf{Q}_e, \quad \Omega_\pm^* \Omega_\pm = \mathbf{Q}_0, \tag{5.3}$$

and the eigenvectors of \mathbf{H}_e and \mathbf{H}_0 are related according to

$$\mathbf{u}_{\mu\beta} = \Omega \mathbf{u}_{\mu\beta}^{(0)}, \quad \mathbf{u}_{\mu\beta}^{(0)} = \Omega^* \mathbf{u}_{\mu\beta}. \tag{5.4}$$

In the following we choose $\Omega = \Omega_-$. Next we define Ω_F on \mathcal{F} through

$$\Omega_F \varphi = \varphi^{(0)} \oplus \Omega \varphi^{(1)} \oplus \{\Omega \varphi_1^{(2)} \otimes \Omega \varphi_2^{(2)}\}_{sym} \oplus \dots, \tag{5.5}$$

for $\varphi \in \mathcal{F}$ of the form

$$\varphi = \varphi^{(0)} \oplus \varphi^{(1)} \oplus \{\varphi_1^{(2)} \otimes \varphi_2^{(2)}\}_{sym} \oplus \dots. \tag{5.6}$$

Let now

$$\begin{aligned}
\hat{H} &= \Omega_F^* H' \Omega_F, \quad \hat{a}(\mathbf{f}) = \Omega_F^* a(\mathbf{f}) \Omega_F, \\
\hat{a}^*(\mathbf{f}) &= \Omega_F^* a^*(\mathbf{f}) \Omega_F. \tag{5.7}
\end{aligned}$$

Then, from its action in Fock space,

$$\Omega_F^* a^*(\mathbf{u}_{\mu\beta}) \Omega_F = a^*(\Omega^* \mathbf{u}_{\mu\beta}) = a^*(\mathbf{u}_{\mu\beta}^{(0)}), \tag{5.8}$$

leading to

$$\Omega_F^* \boldsymbol{\xi}(\mathbf{f}) \Omega_F = \boldsymbol{\xi}^{(0)}(\Omega^* \mathbf{f}), \quad \Omega_F^* \boldsymbol{\pi}(\mathbf{f}) \Omega_F = \boldsymbol{\pi}^{(0)}(\Omega^* \mathbf{f}), \tag{5.9}$$

where

$$\begin{aligned}
\boldsymbol{\xi}^{(0)}(\mathbf{f}) &= \sum_\beta \int d\mu [2\mu]^{-1/2} \\
&\quad \times \{a^*(\mathbf{u}_{\mu\beta}^{(0)})(\mathbf{f}, \mathbf{u}_{\mu\beta}^{(0)}) + a(\mathbf{u}_{\mu\beta}^{(0)})(\mathbf{u}_{\mu\beta}^{(0)}, \mathbf{f})\}, \\
\boldsymbol{\pi}^{(0)}(\mathbf{f}) &= i \sum_\beta \int d\mu [\mu/2]^{1/2} \\
&\quad \times \{a^*(\mathbf{u}_{\mu\beta}^{(0)})(\mathbf{f}, \mathbf{u}_{\mu\beta}^{(0)}) - a(\mathbf{u}_{\mu\beta}^{(0)})(\mathbf{u}_{\mu\beta}^{(0)}, \mathbf{f})\}. \tag{5.10}
\end{aligned}$$

Thus we obtain

$$\begin{aligned}
\hat{H}_f &= \sum_\beta \int d\mu \mu a^*(\mathbf{u}_{\mu\beta}^{(0)}) a(\mathbf{u}_{\mu\beta}^{(0)}) \\
&= H_f^{(0)} = H_{em}^{(0)} + H_{aux}^{(0)}, \\
\Omega_F^* \boldsymbol{\pi}(\mathbf{W}) \Omega_F &= \boldsymbol{\pi}^{(0)}(\Omega^* \mathbf{W}), \\
\hat{A}^\perp(\mathbf{x}_n) &= \Omega_F^* A^\perp(\mathbf{x}_n) \Omega_F \\
&= \sum_\beta \int d\mu [2\mu]^{-1/2} \\
&\quad \times \{a^*(\mathbf{u}_{\mu\beta}^{(0)}) \bar{\mathbf{u}}_{\mu\beta 1}^\perp(\mathbf{x}_n) + a(\mathbf{u}_{\mu\beta}^{(0)}) \mathbf{u}_{\mu\beta 1}^\perp(\mathbf{x}_n)\}, \tag{5.11}
\end{aligned}$$

$$\hat{H} = H_{em}^{(0)} + H_{aux}^{(0)} + \boldsymbol{\pi}^{(0)}(\Omega^* \mathbf{W}) + \sum_n \frac{1}{2m_n} [\mathbf{p}_n - e_n \hat{A}^\perp(\mathbf{x}_n)]^2 + V_{Coul}. \quad (5.12)$$

We note that all interactions with the dielectric are stored in the wave operator Ω . If we set Ω equal to 1, then $\boldsymbol{\pi}^{(0)}(\Omega^* \mathbf{W}) \rightarrow \boldsymbol{\pi}^{(0)}(\mathbf{W}) = 0$ since $\mathbf{W}_1 = \partial_x \zeta_0$ is longitudinal and the $\mathbf{u}_{\mu\beta 1}^{(0)}$'s are transverse. Then \hat{H} reduces to the sum of its vacuum counterpart and $H_{aux}^{(0)}$. Finally we note that the states transform according to $\hat{\psi} = \Omega_F^* \psi'$ and in particular

$$\hat{\psi} = \Omega_F^* \varphi_m \otimes \varphi_{vac} = \varphi_m \otimes \varphi_{vac} = \psi', \quad (5.13)$$

since the vacuum state is not affected by Ω_F^* .

VI. A SIMPLE MODEL FOR TWO-LEVEL ATOMS INTERACTING WITH DIELECTRICS

A. An approximate Hamiltonian

In applications the charged particle system often consists of neutral atoms or molecules with fixed, infinitely heavy, nuclei. Here we consider atoms. Let atom n have N_n electrons. We denote by m and $-e$ the electronic mass and charge. Then atom n has nuclear charge $N_n e$. Denoting by \mathbf{X}_n its nuclear coordinate and $\mathbf{r}_{\alpha n}$ the electronic coordinate of the α th electron in atom n relative to \mathbf{X}_n , we have

$$H' = H_f + (\mathbf{W}, \boldsymbol{\pi}) + \sum_{n, \alpha_n} \frac{1}{2m} [\mathbf{p}_{\alpha_n} + eA^\perp(\mathbf{X}_n + \mathbf{r}_{\alpha_n})]^2 + V_{Coul}. \quad (6.1)$$

Assuming $\mathbf{r}_{\alpha n}$ to be small relative to \mathbf{X}_n , we make the long-wavelength approximation, $A^\perp(\mathbf{X}_n + \mathbf{r}_{\alpha n}) \rightarrow A^\perp(\mathbf{X}_n)$. We also neglect the $(A^\perp)^2$ term. This approximation is usually made but there are situations, for instance in multiphoton processes, where this is not allowed. In addition we assume that the atoms are well separated, so that we can make the dipole-coupling approximation,

$$\begin{aligned} \zeta_0(\mathbf{x}) &\rightarrow \sum_{n, \alpha_n} e \mathbf{r}_{\alpha_n} \cdot \partial_x \frac{1}{4\pi|\mathbf{x} - \mathbf{X}_n|} = - \sum_n \mathbf{d}_n \partial_x \frac{1}{4\pi|\mathbf{x} - \mathbf{X}_n|}, \\ \mathbf{W}_1(\mathbf{x}) &\rightarrow - \sum_n \mathbf{d}_n \cdot \partial_x \partial_x \frac{1}{4\pi|\mathbf{x} - \mathbf{X}_n|} = \sum_n \mathbf{d}_n \cdot \delta^\parallel(\mathbf{x} - \mathbf{X}_n), \end{aligned} \quad (6.2)$$

where

$$\mathbf{d}_n = -e \sum_{\alpha_n} \mathbf{r}_{\alpha_n} \quad (6.3)$$

is the dipole-moment operator for atom n and $\delta^\parallel(\mathbf{x}) = \langle \mathbf{x} | \mathbf{e}_p \mathbf{e}_p | \mathbf{0} \rangle = (1/4\pi) \{ \mathbf{U} - 3\mathbf{e}_x \mathbf{e}_x \} (1/|\mathbf{x}|^3)$ is the longitudinal δ function. Also,

$$V_{Coul} \rightarrow \sum_n V_n + \frac{1}{2} \sum_{m \neq n} V_{mn},$$

$$V_{mn} = \mathbf{d}_m \mathbf{d}_n \cdot \delta^\parallel(\mathbf{X}_m - \mathbf{X}_n) = \mathbf{d}_m \mathbf{d}_n \cdot \langle \mathbf{X}_m | \mathbf{e}_p \mathbf{e}_p | \mathbf{X}_n \rangle, \quad (6.4)$$

where V_n is the sum of Coulomb potentials for atom n ,

$$V_n = \frac{1}{2} \sum_{\alpha_n \neq \beta_n} \frac{e^2}{4\pi|\mathbf{r}_{\alpha_n} - \mathbf{r}_{\beta_n}|} - \sum_{\alpha_n} \frac{Ne^2}{4\pi|\mathbf{r}_{\alpha_n}|}, \quad (6.5)$$

and, for later reference, we note that

$$\begin{aligned} (V_{stat})_{mn} &\rightarrow \mathbf{d}_m \mathbf{d}_n \cdot \langle \mathbf{X}_m | (\mathbf{P}_e)_{11} | \mathbf{X}_n \rangle \\ &= \mathbf{d}_m \mathbf{d}_n \cdot \langle \mathbf{X}_m | \mathbf{p} [\mathbf{p} \cdot \boldsymbol{\varepsilon}_{stat}(\mathbf{x}) \mathbf{p}]^{-1} \mathbf{p} | \mathbf{X}_n \rangle. \end{aligned} \quad (6.6)$$

Then

$$H' = H_f + \sum_n H_n + \frac{1}{2} \sum_{m \neq n} V_{mn} + (\mathbf{W}, \boldsymbol{\pi}) + (\mathbf{I}, \boldsymbol{\xi}), \quad (6.7)$$

where

$$H_n = \sum_{\alpha_n} \frac{1}{2m} \mathbf{p}_{\alpha_n}^2 + V_n \quad (6.8)$$

is the Hamiltonian for atom n and

$$\mathbf{I} = \begin{pmatrix} \mathbf{I}_1 \\ 0 \end{pmatrix}, \quad \mathbf{I}_1(\mathbf{x}) = \frac{e}{m} \sum_n \mathbf{p}_n \delta^\perp(\mathbf{x} - \mathbf{X}_n) \quad (6.9)$$

with $\mathbf{p}_n = \sum_{\alpha_n} \mathbf{p}_{\alpha_n}$ the total electronic momentum of atom n and $\delta^\perp(\mathbf{x} - \mathbf{y})$ is the transverse δ function, $\delta^\perp(\mathbf{x}) = \langle \mathbf{x} | \boldsymbol{\Delta}_p | \mathbf{0} \rangle$. Finally we restrict the field Hamiltonian to the first two (vacuum and single photon) Fock layers,

$$H_f \rightarrow H_f^{(0)} + H_f^{(1)}, \quad (6.10)$$

with associated projectors $P^{(0)}$ and $P^{(1)}$. Since $(\mathbf{W}, \boldsymbol{\pi}) + (\mathbf{I}, \boldsymbol{\xi})$ ladders one layer up and down, we then have, with

$$|\varphi\rangle = \{ (\mathbf{W}, \boldsymbol{\pi}) + (\mathbf{I}, \boldsymbol{\xi}) \} |\varphi_{vac}\rangle, \quad (6.11)$$

that

$$\begin{aligned} H' &= H_f^{(0)} + H_f^{(1)} + \sum_n H_n + \frac{1}{2} \sum_{m \neq n} V_{mn} \\ &\quad + |\varphi\rangle \langle \varphi_{vac}| + |\varphi_{vac}\rangle \langle \varphi| \\ &= H_f^{(0)} + H_f^{(1)} + \sum_n H_n + V^{(int)} + |\varphi\rangle \langle \varphi_{vac}| + |\varphi_{vac}\rangle \langle \varphi|. \end{aligned} \quad (6.12)$$

Note that φ still contains atomic operators. Additional structure is present in the case of two two-level atoms a and b with coinciding levels, the ground states (projectors P_{a1} and P_{b1}) being nondegenerate and excited states (which may be degenerate) with opposite parity (projectors P_{a2} and P_{b2}), as is the case with atomic s and p states. Then the projectors

$$\begin{aligned} P_E &= (P_{a1} \otimes P_{b2} + P_{a2} \otimes P_{b1}) \otimes P^{(0)} \\ &\quad + (P_{a1} \otimes P_{b1} + P_{a2} \otimes P_{b2}) \otimes P^{(1)}, \end{aligned}$$

$$P_F = 1 - P_E = (P_{a1} \otimes P_{b1} + P_{a2} \otimes P_{b2}) \otimes P^{(0)} \\ + (P_{a1} \otimes P_{b2} + P_{a2} \otimes P_{b1}) \otimes P^{(1)}, \quad (6.13)$$

commute with H' . Thus, if we consider a situation with initial state $\psi = \varphi_{a2} \otimes \varphi_{b1} \otimes \varphi_{vac}$, we have $P_F \psi = 0$ and the problem reduces to one with Hamiltonian $H'_E = H'E$ in the corresponding subspace.

B. The multipolar representation

Using the same notation as above we can obtain, starting from the original Hamiltonian H , the multipolar representation by applying the unitary Power-Zienau-Woolley transformation [16]

$$H_{MP} = U H U^{-1}, \quad (6.14)$$

where

$$U = \exp[-iG], \quad G = (\mathbf{P}_1, \mathbf{A}) = (\mathbf{P}, \xi), \quad (6.15)$$

with

$$\mathbf{P} = \begin{pmatrix} \mathbf{P}_1 \\ 0 \end{pmatrix},$$

$$\mathbf{P}_1(\mathbf{x}) = -e \sum_{n, \alpha_N} \int_0^1 du \mathbf{r}_{\alpha_n} \delta(\mathbf{x} - \mathbf{X}_n - u \mathbf{r}_{\alpha_n}). \quad (6.16)$$

Since the calculations closely follow those of the vacuum case, we only state the result, where we made the long-wavelength and dipole-coupling approximations. Thus, also skipping magnetic field terms and contact terms,

$$H_{MP} = H_f - \sum_n \mathbf{d}_n E(\mathbf{X}_n) + \frac{1}{2} \sum_n (\mathbf{P}_{1n}, \mathbf{P}_{1n})_1 + \sum_{n, \alpha_N} \frac{\mathbf{p}_{\alpha_n}^2}{2m}. \quad (6.17)$$

Note that skipping the contact terms is consistent with the assumption of well-separated atoms, but is no longer justified if their wave functions start to overlap. This is often the case in actual situations, where atoms are close to sensitizers, large molecules that are needed for the efficient excitation of atomic states.

VII. DIELECTRICS EXTENDING OVER ALL SPACE

There are situations for dielectrics extending over all of \mathbb{R}^3 , including photonic crystals, where Ω_{\pm} exist and moreover

$$\Omega_{11} = 0, \quad (7.1)$$

in which case

$$\mathbf{u}_{\mu\beta 1} = \Omega_{12} \mathbf{u}_{\mu\beta 2}^{(0)}. \quad (7.2)$$

Hence in the hatted representation (H.c. denotes the Hermitian conjugate),

$$\hat{A}^{\perp}(\mathbf{x}) = \sum_{\beta} \int d\mu (2\mu)^{-1/2} a(\mathbf{u}_{\mu\beta 2}^{(0)}) \langle \mathbf{x} | \mathbf{P}^{\perp} \Omega_{12} \mathbf{u}_{\mu\beta 2}^{(0)} \rangle + \text{H.c.}, \\ \boldsymbol{\pi}^{(0)}(\Omega^* \mathbf{W}) = \sum_{\beta} \int d\mu (\mu/2)^{1/2} a(\mathbf{u}_{\mu\beta 2}^{(0)}) (\mathbf{W}_1, \mathbf{P}^{\parallel} \Omega_{12} \mathbf{u}_{\mu\beta 2}^{(0)}) \\ + \text{H.c.}, \quad (7.3)$$

which only involve the creation and annihilation operators for the auxiliary fields. Thus $H_{em}^{(0)}$ is completely decoupled from the remainder of \hat{H} . Setting

$$\boldsymbol{\pi}^{(0)}(\Omega^* \mathbf{W}) = \int d\mathbf{x} \rho(\mathbf{x}) \vartheta(\mathbf{x}) \\ = - \int d\mathbf{x} \partial_{\mathbf{x}} \cdot \mathbf{W}_1(\mathbf{x}) \vartheta(\mathbf{x}) \\ = \int d\mathbf{x} \mathbf{W}_1(\mathbf{x}) \cdot \partial_{\mathbf{x}} \vartheta(\mathbf{x}), \quad (7.4)$$

we have

$$\partial_{\mathbf{x}} \vartheta(\mathbf{x}) = \sum_{\beta} \int d\mu (\mu/2)^{1/2} a(\mathbf{u}_{\mu\beta 2}^{(0)}) \langle \mathbf{x} | \mathbf{P}^{\parallel} \Omega_{12} \mathbf{u}_{\mu\beta 2}^{(0)} \rangle + \text{H.c.} \quad (7.5)$$

In Appendix B it is shown how the above expressions can be recast as

$$\hat{A}^{\perp}(\mathbf{x}) = - \int d\mathbf{y} \int d\lambda \langle \mathbf{x} | \mathbf{P}^{\perp} \mathbf{R}_e(\lambda) | \mathbf{y} \rangle (\lambda/2)^{1/2} \sigma(\mathbf{y}, \lambda) \mathbf{f}(\mathbf{y}, \lambda) \\ + \text{H.c.}, \\ \partial_{\mathbf{x}} \vartheta(\mathbf{x}) = i \int d\mathbf{y} \int d\lambda \langle \mathbf{x} | \mathbf{P}^{\parallel} \mathbf{R}_e(\lambda) | \mathbf{y} \rangle (\lambda/\sqrt{2}) \sigma(\mathbf{y}, \lambda) \mathbf{f}(\mathbf{y}, \lambda) \\ + \text{H.c.}, \quad (7.6)$$

where

$$[\mathbf{f}(\mathbf{y}, \lambda), \mathbf{f}^*(\mathbf{y}', \lambda')] = \mathbf{U} \delta(\mathbf{y} - \mathbf{y}') \delta(\lambda - \lambda') \quad (7.7)$$

and

$$\mathbf{R}_e(z) = [z^2 \boldsymbol{\varepsilon}(\mathbf{x}, z) - \mathbf{h}_0]^{-1} \quad (7.8)$$

In terms of the \mathbf{f} 's,

$$H_{aux} = \int d\mathbf{y} \int d\lambda \lambda \mathbf{f}^*(\mathbf{y}, \lambda) \cdot \mathbf{f}(\mathbf{y}, \lambda). \quad (7.9)$$

Apart from the term $H_{em}^{(0)}$, which is decoupled from the remainder of \hat{H} , the Hamiltonian coincides with the one used by Dung *et al.* [6], since in our case $\sigma(\mathbf{y}, \lambda) = \sqrt{2} \nu(\mathbf{y}, \lambda)$, $\text{Im} \boldsymbol{\varepsilon}(\mathbf{y}, \lambda) = (\pi/\lambda) \nu(\mathbf{y}, \lambda)$ and $\varepsilon_0 = c = 1$. Although the formalism of Ref. [6] requires the medium to extend over all space, these authors could also treat different situations by first assuming a small amount of absorption, then expressing the quantity of interest in terms of the Helmholtz Green's

function and finally replace the latter by the actual one for the case that certain space regions are nonabsorbing such as vacuum regions. In the following section we show that our formalism, where no such limiting procedure is required, gives precisely the same result.

VIII. AN EXAMPLE CONCERNING RESONANT ENERGY TRANSFER

Using the model of Sec. VI we consider two two-level atoms a and b with coinciding levels and nondegenerate ground states (labeled 1) embedded in an absorptive dielectric. Thus

$$H_n = \lambda_1 P_{n1} + \lambda_2 P_{n2},$$

$$H_m^{(0)} = H_a + H_b = 2\lambda_1 P_{a1} \otimes P_{a2} + (\lambda_1 + \lambda_2) \{P_{a1} \otimes P_{b2} + P_{a2} \otimes P_{b1}\} + 2\lambda_2 P_{a2} \otimes P_{b2}. \quad (8.1)$$

The eigenstates of $H_m^{(0)}$ are zeroth approximations ($V_{ab} = 0$) of corresponding states of $H_m = H_m^{(0)} + V_{ab}$, where $V_{ab} = V^{(int)}$. We assume that the states corresponding to $\lambda_1 + \lambda_2$ are embedded in the field continuum and coupled to the latter. Then, generically, these states turn into resonances once the matter-field interaction is turned on. Now radiative decay becomes possible but also meaningful transition amplitudes between two different eigenstates of H_m , originating from those of $H_m^{(0)}$ at $\lambda_1 + \lambda_2$, exist. In fact the first atom is coupled to a continuum involving the second atom interacting with the radiation field. Treating V_{ab} as a perturbation we are then dealing with resonant energy transfer. This case was studied by Dung *et al.* [6]. We shall use the primed representation, reduced to the model of Sec. VI (the same result follows in the hatted one).

$$\begin{aligned} H' &= H_f^{(0)} + H_f^{(1)} + H_a + H_b + V_{ab} + |\varphi\rangle\langle\varphi_{vac}| + |\varphi_{vac}\rangle\langle\varphi| \\ &= H_f^{(0)} + H_f^{(1)} + H_m^{(0)} + V_{ab} + |\varphi\rangle\langle\varphi_{vac}| + |\varphi_{vac}\rangle\langle\varphi| \\ &= H_f^{(0)} + H_f^{(1)} + H_m^{(0)} + V_{ab} + H_{mf}^{(int)} \\ &= H_f^{(0)} + H_f^{(1)} + H_m^{(0)} + H^{(int)} = H^{(0)} + H^{(int)}. \end{aligned} \quad (8.2)$$

We now consider

$$\begin{aligned} &\langle\varphi_{out}|T(z)|\varphi_{in}\rangle, \\ T(z) &= H^{(int)} + H^{(int)}[z - H]^{-1}H^{(int)}, \\ \varphi_{in} &= \varphi_{a2} \otimes \varphi_{b1} \otimes \varphi_{vac}, \\ \varphi_{out} &= \varphi_{a1} \otimes \varphi_{b2} \otimes \varphi_{vac} \end{aligned} \quad (8.3)$$

with $\varphi_{a2} \otimes \varphi_{b1}$ and $\varphi_{a1} \otimes \varphi_{b2}$ eigenstates of $H_m^{(0)}$ at the same eigenvalue $\lambda = \lambda_{a2} + \lambda_{b1} = \lambda_{a1} + \lambda_{b2}$ and $z = \lambda + i\delta$, where eventually δ is set equal to zero. Thus we consider the transfer of the excitation energy $\omega_0 = \lambda_{a2} - \lambda_{a1} > 0$ from atom a to atom b . We note that V_{ab} already contributes to $T(z)$ to first order in a Born expansion, but, since it only connects differ-

ent Fock layers, $H_{mf}^{(int)}$ starts contributing in the second order. Thus to leading order in both interactions

$$T(z) \rightarrow T^{(0)}(z) = V_{ab} + H_{mf}^{(int)}[z - H^{(0)}]^{-1}H_{mf}^{(int)}, \quad (8.4)$$

which is the quantity we shall consider. Since $H_{mf}^{(int)}$ maps the vacuum state into the next Fock layer,

$$\langle\varphi_{vac}|T^{(0)}(z)|\varphi_{vac}\rangle = V_{ab} + \langle\varphi|[z - H_m^{(0)} - H_f^{(1)}]^{-1}|\varphi\rangle. \quad (8.5)$$

Now

$$\begin{aligned} &\langle\varphi_{a1} \otimes \varphi_{b2}|V_{ab}|\varphi_{a2} \otimes \varphi_{b1}\rangle \\ &= \langle\varphi_{a1}|\mathbf{d}_A|\varphi_{a2}\rangle\langle\varphi_{b2}|\mathbf{d}_B|\varphi_{b1}\rangle : \delta^{\parallel}(\mathbf{X}_A - \mathbf{X}_B). \end{aligned} \quad (8.6)$$

Next we note that, see Eqs. (6.11) and (4.4),

$$|\varphi\rangle = \sum_{\beta} \int d\mu \left[i \sqrt{\frac{\mu}{2}} (\mathbf{W}, \mathbf{u}_{\mu\beta}) + \frac{1}{\sqrt{2\mu}} (\mathbf{I}, \mathbf{u}_{\mu\beta}) \right] |\mathbf{u}_{\mu\beta}\rangle. \quad (8.7)$$

Observing that $H^{(0)}|\mathbf{u}_{\mu\beta}\rangle = (H_m^{(0)} + \mu)|\mathbf{u}_{\mu\beta}\rangle = (H_m^{(0)} + H_e^{1/2})|\mathbf{u}_{\mu\beta}\rangle$ and using the orthogonality properties of the set $\{|\mathbf{u}_{\mu\beta}\rangle\}$, we obtain (for the actual calculation, see Appendix C)

$$\begin{aligned} \langle\varphi_{out}|T(z)|\varphi_{in}\rangle &= \omega_0^2 \langle\varphi_{a1}|\mathbf{d}_A|\varphi_{a2}\rangle \\ &\quad \times \langle\varphi_{b2}|\mathbf{d}_B|\varphi_{b1}\rangle : \langle\mathbf{X}_A|\mathbf{R}_e(\omega_0)|\mathbf{X}_B\rangle. \end{aligned} \quad (8.8)$$

This is precisely the expression, Eq. (31), obtained by Dung *et al.* in Ref. [6], since

$$\langle\mathbf{X}_A|\mathbf{R}_e(\omega_0)|\mathbf{X}_B\rangle = -\mathbf{G}(\mathbf{X}_A, \mathbf{X}_B, \omega_0), \quad (8.9)$$

with \mathbf{G} the Green's function used by these authors. Note that our derivation follows a different, maybe somewhat simpler, route. In addition, our expression applies to general absorptive dielectrics, vacuum regions are allowed, *a posteriori* validating the limiting procedure used by Dung *et al.*

IX. ATOMS IN PHOTONIC CRYSTALS WITH BAND GAPS

In Ref. [11] photonic band gaps for absorptive systems were defined as frequency intervals for which the Fourier transforms $\tilde{\mathbf{E}}$ and $\tilde{\mathbf{B}}$ vanish for any initial $\mathbf{E}(t_0)$ and $\mathbf{B}(t_0)$. This definition reduces to the usual one, featuring the local density of states, for nonabsorptive situations. In the above reference it was shown that photonic crystals cannot have band gaps in frequency intervals on which $\text{Im } \varepsilon(\mathbf{x}, \omega)$ is nonvanishing. But if $\nu(\mathbf{x}, \omega) = 0$ for ω contained in a finite interval J , the Kramers-Kronig relations are still obeyed and J can contain a band gap Δ . Let this be the case and consider $\mathbf{R}_e(\omega_0)$ for $\omega_0 \in \Delta$. Then

$$\langle\mathbf{X}_A|\mathbf{R}_e(\omega_0)|\mathbf{X}_B\rangle = \langle\mathbf{X}_A|[\omega_0^2 \varepsilon(\mathbf{x}, \omega_0 + i\delta) - \mathbf{h}_0]^{-1}|\mathbf{X}_B\rangle, \quad (9.1)$$

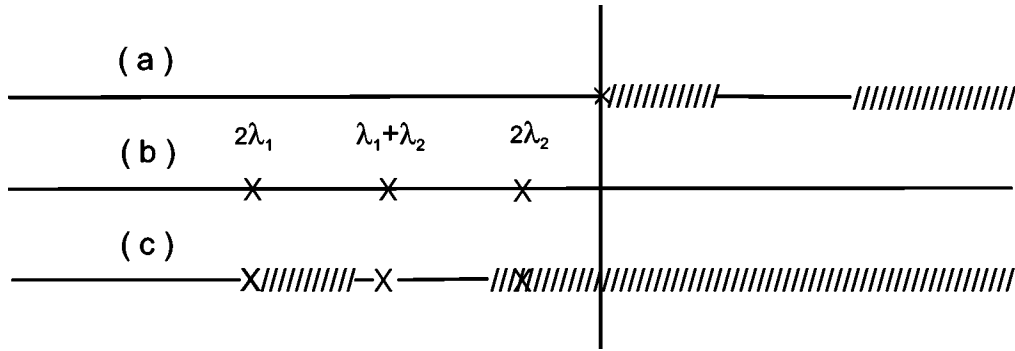


FIG. 1. Spectra of the field Hamiltonian with a band gap (a), of two two-level atoms with coinciding levels λ_1 and λ_2 (b) and their combination (c). Crosses are eigenvalues, dashed lines continuous spectrum. In (c) only the relevant part of the spectrum is shown.

where now $\varepsilon(\mathbf{x}, \omega_0)$ is real and in addition $\mathbf{R}_e(z)$ is analytic across the real axis for $\text{Re}z \in \Delta$ [11]. Thus we can set $\delta=0$ in Eq. (9.1),

$$\langle \mathbf{X}_A | \mathbf{R}_e(\omega_0) | \mathbf{X}_B \rangle = \langle \mathbf{X}_A | [\omega_0^2 \varepsilon(\mathbf{x}, \omega_0) - \mathbf{h}_0]^{-1} | \mathbf{X}_B \rangle, \quad (9.2)$$

where

$$\mathbf{R}_e(\omega_0) = \sum_{\beta} \int_{\mathbb{R} \setminus \Delta} d\mu [\omega_0^2 - \mu^2]^{-1} |\mathbf{u}_{\mu\beta 1}\rangle \langle \mathbf{u}_{\mu\beta 1}|, \quad (9.3)$$

is self-adjoint. Note that now a single atom with nuclear position \mathbf{X} cannot decay radiatively by one-photon emission since the decay constant is proportional to $\text{Im}\langle \mathbf{X} | \mathbf{R}_e(\omega_0) | \mathbf{X} \rangle = 0$ [9]. Note also that band gaps cannot directly be related to gaps in the spectrum of \mathbf{H}_e . In fact the spectrum of this operator does not have gaps. But if we discard its off-diagonal terms, i.e.,

$$\mathbf{H}_e \rightarrow \mathbf{H}_e^{(0)} = \begin{pmatrix} \mathbf{h}_0 + \chi'(0) & 0 \\ 0 & \lambda^2 \end{pmatrix}, \quad (9.4)$$

then the auxiliary field part completely decouples and does not contribute to $\mathbf{R}_e(z)$.

Consider the Drude model with dispersion but no absorption, i.e., $\varepsilon(\mathbf{x}, \omega) = 1 - f(\mathbf{x})(\Omega^2/\omega^2)$. Then $\chi'(0) = f(\mathbf{x})\Omega^2$, so the eigenvalue equation $[\omega^2 \varepsilon(\mathbf{x}, \omega) - \mathbf{h}_0]\phi = 0$ can be rewritten as $[\mathbf{h}_0 + f(\mathbf{x})\Omega^2]\phi = [\mathbf{h}_0 + \chi'(0)]\phi = \omega^2 \phi$. Numerical work [10,17] has shown that band gaps can occur in this situation. Suppose such a band gap is present and let the field be coupled to two atoms with coinciding levels λ_1 and λ_2 as in Sec. VIII. Without interactions the spectrum consists of the union of $2\lambda_1$, $\lambda_1 + \lambda_2$, and $2\lambda_2$ with the spectrum of the field Hamiltonian (vacuum eigenvalue $\lambda_{vac} = 0$ and pieces of continuum) and it can happen that in the model of Sec. VIII $\lambda_1 + \lambda_2$ is an isolated eigenvalue. Strictly speaking it is continuum embedded but we can use the decomposition given in Sec. VI, cf. Eq. (6.13), to remove this feature. For a picture of this situation, see Fig. 1. Then, generically, the interaction will shift and/or split this eigenvalue but it remains real, i.e., it remains a stable state (this changes if multi-photon processes come into play but the associated decay rates are of a completely different magnitude).

In the general case the analysis is more complicated. Considering the model of Sec. VIII again, we study the perturbation of $\lambda_1 + \lambda_2$, for instance by determining the corresponding pole of $[z - H']^{-1}$. Then this pole is also a pole of $P[z - H']^{-1}P$ for some suitable projector P . Taking $P = P_{vac} = |\varphi_{vac}\rangle \langle \varphi_{vac}|$ and $Q = 1 - P$, we are dealing with $\langle \varphi_{vac} | [z - H']^{-1} | \varphi_{vac} \rangle$, which, by the Feshbach formula, equals

$$\begin{aligned} & \langle \varphi_{vac} | [z - H']^{-1} | \varphi_{vac} \rangle \\ &= [z - H_m^{(0)} - V_{ab} \\ & \quad - \langle \varphi_{vac} | H^{(int)} Q [z - QH'Q]^{-1} | \varphi_{vac} \rangle]^{-1}. \end{aligned} \quad (9.5)$$

The idea is then to find solutions z_j , P_j of

$$z_j P_j = [H_m^{(0)} + V_{ab} + \langle \varphi_{vac} | H^{(int)} Q [z - QH'Q]^{-1} | \varphi_{vac} \rangle] P_j, \quad (9.6)$$

by iteration, starting from the unperturbed case

$$z_j P_j = H_m^{(0)} P_j. \quad (9.7)$$

To leading order we can replace $\langle \varphi_{vac} | H^{(int)} Q [z - QH'Q]^{-1} | \varphi_{vac} \rangle$ by $\langle \varphi_{vac} | H_{mf}^{(int)} Q [z - H^{(0)}]^{-1} | \varphi_{vac} \rangle = \langle \varphi_{vac} | T^{(0)}(z) | \varphi_{vac} \rangle$, with $T^{(0)}(z)$ given by Eq. (8.4). If we are only interested in $\lambda_1 + \lambda_2$, we can take $P = P_{12} \otimes P_{vac}$, $P_{12} = P_{a_1} \otimes P_{b_2} + P_{a_2} \otimes P_{b_1}$. We can now follow the pattern of Sec. VIII, except that different matrix elements of $\langle \varphi | [z - H^{(0)}]^{-1} | \varphi \rangle$ are involved. We refrain from presenting the actual calculations, which are quite lengthy. An essential point is that the final formula contains matrix elements of the atomic operator $\lim_{\delta \downarrow 0} \{ [\omega + i\delta - H_e^{1/2}]^{-1} Q_e \}_{11}$ for $\omega = \pm(\lambda_1 + \lambda_2)$, which is self-adjoint. This is trivial for negative ω , whereas for $\omega > 0$ we have $(z = \omega + i\delta)$ and the limit for $\delta \downarrow 0$ is introduced to remove the contribution from the eigenspace $Q_e \mathcal{H}_e$

$$[z - H_e^{1/2}]^{-1} Q_e = \lim_{\varepsilon \downarrow 0} \int_{\varepsilon}^{\infty} du 2u [z - u]^{-1} \delta(u^2 - H_e). \quad (9.8)$$

But

$$\begin{aligned}
 [\delta(u^2 - H_e)]_{11} &= \frac{1}{2\pi i} \{ [u^2 - i0 - H_e]^{-1} - [u^2 + i0 - H_e]^{-1} \}_{11} \\
 &= \frac{1}{2\pi i} \{ R_e^*(u) - R_e(u) \}, \quad (9.9)
 \end{aligned}$$

so

$$\begin{aligned}
 [z - H_e^{1/2}]^{-1} Q_e]_{11} &= \frac{1}{\pi i} \lim_{\varepsilon \downarrow 0} \int_{\varepsilon}^{\infty} du u [z - u]^{-1} \\
 &\quad \times \{ R_e^*(u) - R_e(u) \}. \quad (9.10)
 \end{aligned}$$

Then, if Δ is a band gap, the integrand vanishes for $u \in \Delta$ so the left-hand side has an analytic continuation across Δ , which is self-adjoint for $\omega \in \Delta$. This results in the perturbed eigenvalues (the original one may split up), originating from $\lambda_1 + \lambda_2$, being real if $\lambda_1 + \lambda_2$ is in a band gap.

If there are $N > 2$ identical atoms the same procedure leads to the result that if the unperturbed eigenvalue $(N - 1)\lambda_1 + \lambda_2$ (one excited atom and the others in their ground states) lays in a gap then the perturbed eigenvalues remain real. In this case the excitation energy can be transferred from one atom to another nonradiatively (Förster transitions) but not by photon emission and reabsorption, nor can the excited atom decay through photon emission.

X. DISCUSSION

A. Summary

We presented a quantum Hamilton formalism for a system of atoms embedded in absorptive dielectrics. Various unitarily equivalent versions were introduced. In the first case, Eq. (4.6), the presence of the dielectric is encountered in the field Hamiltonian and in the potentials, which depend on ε_{stat} rather than the vacuum potentials. In addition the full vector potential \mathbf{A} is present in the atom-field interaction term. In the second case, Eq. (4.14), the field Hamiltonian is the same but the particles interact through vacuum Coulomb potentials and only the transverse vector potential is present in the atom-field interaction term. The part containing its longitudinal component is now replaced by an interaction term containing the longitudinal electric field and the gradient of the scalar potential Eq. (4.15). If suitable wave operators exist there is a third form (5.12) in which the field Hamiltonian is simply the vacuum field Hamiltonian. Here all interactions between the atoms and the dielectric are stored in the wave operators. If the dielectric extends over all space this case reduces to that of Dung *et al.* [6]. For an example considered by these authors we verified that their limiting procedure results in the same expression as obtained here. We then turned to the behavior of a model of excited atoms in a photonic crystal with transition frequency in a band gap. We found that for a finite number of atoms with one initially excited and the remainder in their ground states, radiative decay is inhibited. This result was obtained to leading order in the relevant interactions. However, its extension with the full interaction present seems to be a formidable problem. This is due to the fact that gaps are not a spectral

property of a Hamiltonian but of the projected object $R_e(z)$. It should be realized that in practice there is always some absorption present. But in suitable frequency intervals it may be sufficiently small to be ignored in which case the above results are relevant.

B. Transformation of the initial and final states

In the previous sections we performed unitary transformations of the Hamiltonian. In general this leads to corresponding transformations of the states. But this is not always the case. The situation was considered by Aharonov and Au [18] for field gauge transformations and some generalizations thereof. Using their ideas it can be shown (see Appendix D) that, switching from the primed to the hatted frame, $\hat{X} = \Omega_F^* X' \Omega_F$, we have

$$\langle \varphi_{out} | \hat{X} | \varphi_{in} \rangle = \langle \varphi_{out} | X' | \varphi_{in} \rangle, \quad (10.1)$$

for φ_{in} and φ_{out} of the type $\varphi_m \otimes \varphi_f$, where the φ_m 's are matter states and the φ_f 's eigenstates of

$$H_f^{(0)} = H_{em}^{(0)} + H_{aux}^{(0)}, \quad (10.2)$$

at the same eigenvalue. But this is no longer true for field eigenstates at different eigenvalues. The above result no longer holds for the transformation from the original framework to the primed or multipolar ones. Indeed, if we calculate the transition amplitude (8.3) in the original representation of Sec. VIII, using the same states, the result is different. Indeed, if we start with the object $\langle \varphi_m \otimes \varphi_{vac} | X | \varphi_m \otimes \varphi_{vac} \rangle$ in the original representation, then

$$\langle \varphi_m \otimes \varphi_{vac} | X | \varphi_m \otimes \varphi_{vac} \rangle = \langle \varphi_m \otimes U \varphi_{vac} | X' | \varphi_m \otimes U \varphi_{vac} \rangle, \quad (10.3)$$

rather than the quantity $\langle \varphi_m \otimes \varphi_{vac} | X' | \varphi_m \otimes \varphi_{vac} \rangle$ that we considered earlier for specific X' . Now

$$U \varphi_{vac} = \exp[-i\xi(\mathbf{W})] \varphi_{vac} = \exp[-i\mathbf{A}(\partial_{\mathbf{x}} \zeta_0)] \varphi_{vac} \quad (10.4)$$

contains contributions from all Fock layers. This is troublesome since our formalism does not provide a prescription as to which initial or final state should be taken in a specific framework. On the other hand we left out the mechanism that produces the initial excited atomic state and taking this into account may change the picture. Suppose that an atom is excited by absorbing a photon (another situation is that where an electron causes the excitation). Then the initial state is the product of the photon state, say a plane wave with frequency ω_0 , and the atomic ground state (or a product of atomic ground states if there are more atoms). The final state has the same structure so we are dealing with elastic scattering of a photon. To leading order the scattering amplitude contains a direct contribution without excitation of an atom and indirect ones where an atom is excited, the excitation energy is transferred to a second atom, which then decays by photon emission. In such situations results like Eq. (10.1) may still hold. But even if this is the case, choosing the state $\varphi_m \otimes \varphi_{vac}$ is of an *ad hoc* nature. More information about the proper choice of initial and final states might be obtained

from a corresponding microscopic approach, starting from an interacting (second) quantized matter-field system with minimal coupling. But, as far as the author is aware, such a formalism is not available for general spatially inhomogeneous situations. This matter, especially if embedded atoms are present, needs further investigation.

C. Outlook

It sometimes happens that absorption can be neglected in a certain frequency interval but that dispersion has to be taken into account. This raises the question if such a dispersive system can directly be quantized. This is indeed the case for the Drude-Lorentz model as we intend to discuss on another occasion. A related subject is a perturbative approach for small absorption situations. Some results have been obtained for the Drude model [19] and more general cases are under investigation.

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APPENDIX A: THE WAVE OPERATORS

The wave operators

$$\Omega_{\pm} = \lim_{t \rightarrow \pm\infty} \Omega(t) = \lim_{t \rightarrow \pm\infty} \exp[iH_e t] \exp[-iH_0 t] Q^{(0)},$$

$$H_e = H_0 + V, \quad (\text{A1})$$

acting in \mathcal{H} , were discussed to some extent in Refs. [9] and [15]. If they exist and are complete, we have the properties

$$H_e = \Omega_{\pm} H_0 \Omega_{\pm}^*, \quad \Omega_{\pm} \Omega_{\pm}^* = Q_e, \quad \Omega_{\pm}^* \Omega_{\pm} = Q^{(0)}. \quad (\text{A2})$$

In particular the (absolutely) continuous spectra of H_e and $H^{(0)}$ then coincide. Differentiation and integration of $\Omega(t)$ gives

$$\Omega_{\pm} = Q^{(0)} + i \int_0^{\pm\infty} dt \exp[iHt] V \exp[-iH_0 t] Q^{(0)} \quad (\text{A3})$$

and existence follows if

$$\int_{-\infty}^{+\infty} dt \|V \exp[-iH_0 t] \mathbf{f}\| < \infty \quad (\text{A4})$$

for a dense set $\mathbf{f} \in Q^{(0)}\mathcal{H}$. Note that $\mathbf{h}_0 \cdot \mathbf{f}_1 = \mathbf{p}^2 \mathbf{f}_1$ for $\mathbf{f} \in Q^{(0)}\mathcal{H}$. Then, if $\chi'(\mathbf{x}, 0)$ and $\sigma(\mathbf{x}, \lambda)$ vanish for \mathbf{x} outside some finite region (or have sufficiently fast decay) the norms $\|\chi'(\mathbf{x}, 0) \exp[-i\mathbf{p}^2 t] \mathbf{f}_1\|_1$ and $\|\langle \lambda \sigma(\mathbf{x}, \lambda) \rangle \exp[-i\mathbf{p}^2 t] \mathbf{f}_1\|_2$ have the required time-decay property, whereas $\|\langle \lambda \sigma(\mathbf{x}, \lambda) \rangle \exp[-i\lambda^2 t] \mathbf{f}_2\|_2$ can be handled by a method used for rank one perturbations [20]. But if the medium extends

over all space this proof breaks down. Nevertheless it can happen that even then the Ω 's exist and that actually $P_1 \Omega P_1 = \Omega_{11} = 0$. We have the following.

Proposition. Suppose that $\varepsilon(\mathbf{x}, z)$ has the properties

$$\begin{aligned} \text{Im } \varepsilon(\mathbf{x}, z) &\geq c_1 > 0 \quad \forall \mathbf{x} \in \mathcal{A}, \\ \omega &= |\text{Re } z| \in [\omega_1, \omega_2], \quad \eta = \text{Im } z \in [0, \eta_0], \\ |\varepsilon(\mathbf{x}, z)| &< c_2 < \infty, \\ \lim_{|\omega| \rightarrow \infty} \varepsilon(\mathbf{x}, z) &= 1, \quad z = \omega + i\eta, \quad \eta \in [0, \eta_0], \\ |\varepsilon_{stat}(\mathbf{x})| &\geq c_3 > 0, \\ |\varepsilon(\mathbf{x}, z) - \varepsilon_{stat}(\mathbf{x})| &\leq c_4 |z|, \quad |z| \text{ sufficiently small,} \\ 0 &\leq c_4 < \infty, \\ |z^2 \hat{\chi}(\mathbf{x}, z)| &\leq c_5 < \infty, \end{aligned} \quad (\text{A5})$$

where \mathcal{A} is either \mathbb{R}^3 or the volume occupied by the absorptive material in the union of the unit cells of a photonic crystal. $[\omega_1, \omega_2]$, $0 < \omega_1 < \omega_2 < \infty$, is an arbitrary finite interval and $\eta_0 > 0$, which can be taken arbitrarily small. Then, with P_1^\perp the projector upon $P^\perp \mathcal{H}_1$, we have for all $\mathbf{f} \in \mathcal{H}$,

$$\lim_{t \rightarrow \infty} P_1 \exp[iH_e t] P_1^\perp \mathbf{f} = 0. \quad (\text{A6})$$

This result is physically reasonable since it states that for large times the electric field dies out as is to be expected in an infinite absorptive medium. We omit the rather lengthy proof. The basic idea is to express Eq. (A6) in terms of the Laplace transform ($\text{Im } z > 0$)

$$P_1 [z^2 - H_e]^{-1} P_1 = [z^2 \varepsilon(\mathbf{x}, z) - \mathbf{h}_0]^{-1} P_1 = R_e(z) P_1. \quad (\text{A7})$$

For photonic crystals it is known [11] that, except for $z = 0$, $R_e(z)$ is analytic across the real axis with singularities forming islands in the lower half plane away from the real axis (this was confirmed numerically for a two-dimensional case in Ref. [21]). From this the proposition immediately follows.

APPENDIX B: RELATIONS USED IN SEC. VII

We write, using the spectral decomposition of $H^{(0)}$,

$$H^{(0)} = \int \mu E^{(0)}(d\mu),$$

$$E_j^{(0)}(d\mu) = \sum_{\beta} d\mu |\mathbf{u}_{\mu\beta j}^{(0)}\rangle \langle \mathbf{u}_{\mu\beta j}^{(0)}|, \quad (\text{B1})$$

$$\begin{aligned} P_1 \Omega \mathbf{f} &= \lim_{\delta \downarrow 0} i \delta \int R_e(z) P_1 E^{(0)}(d\mu) \mathbf{f} + \lim_{\delta \downarrow 0} \int R_e(z) \\ &\times \begin{pmatrix} 0 & \langle \lambda \sigma | \\ 0 & 0 \end{pmatrix} E^{(0)}(d\mu) \mathbf{f}, \end{aligned} \quad (\text{B2})$$

or

$$(\Omega \mathbf{f})_1 = \lim_{\delta \downarrow 0} i \delta \int \mathbf{R}_e(z) \mathbf{E}_1^{(0)}(d\mu) \mathbf{f}_1 + \lim_{\delta \downarrow 0} \int \mathbf{R}_e(z) \times \langle \lambda \sigma | \mathbf{E}_2^{(0)}(d\mu) \mathbf{f}_2. \quad (\text{B3})$$

Provided

$$\lim_{\delta \downarrow 0} i \delta \int \mathbf{R}_e(z) \mathbf{E}_1^{(0)}(d\mu) \mathbf{f}_1 = 0 \quad (\text{B4})$$

(true if the above proposition holds), then

$$(\Omega \mathbf{f})_1 = \Omega_{12} \mathbf{f}_2 = \int \mathbf{R}_e(\mu) \langle \lambda \sigma | \mathbf{E}_2^{(0)}(d\mu) \mathbf{f}_2, \quad (\text{B5})$$

i.e., $\Omega_{11} = 0$. For $\mathbf{f}_2 = \mathbf{u}_{\mu\beta 2}^{(0)}$ we have

$$\langle \mathbf{x} | \Omega_{12} | \mathbf{u}_{\mu\beta 2}^{(0)} \rangle = \int d\mathbf{y} \int d\lambda \langle \mathbf{x} | \mathbf{R}_e(\mu) | \mathbf{y} \rangle \lambda \sigma(\mathbf{y}, \lambda) \mathbf{u}_{\mu\beta 2}^{(0)}(\mathbf{y}, \lambda). \quad (\text{B6})$$

In Sec. VII we encountered

$$\begin{aligned} \hat{A}^\perp(\mathbf{x}) &= \sum_{\beta} \int d\mu (2\mu)^{-1/2} a(\mathbf{u}_{\mu\beta 2}^{(0)}) \langle \mathbf{x} | \mathbf{P}^\perp \Omega_{12} | \mathbf{u}_{\mu\beta 2}^{(0)} \rangle + \text{H.c.}, \\ \partial_{\mathbf{x}} \vartheta(\mathbf{x}) &= -i \sum_{\beta} \int d\mu (\mu/2)^{1/2} a(\mathbf{u}_{\mu\beta 2}^{(0)}) \langle \mathbf{x} | \mathbf{P}^\parallel \Omega_{12} | \mathbf{u}_{\mu\beta 2}^{(0)} \rangle + \text{H.c.} \end{aligned} \quad (\text{B7})$$

With \mathbf{X} is an arbitrary operator on \mathcal{H}_1 ,

$$\begin{aligned} &\sum_{\beta} \int d\mu g(\mu) a(\mathbf{u}_{\mu\beta 2}^{(0)}) \langle \mathbf{x} | \mathbf{X} \Omega_{12} | \mathbf{u}_{\mu\beta 2}^{(0)} \rangle \\ &= \sum_{\beta} \int d\mu g(\mu) \int d\mathbf{y} \int d\lambda a(\mathbf{u}_{\mu\beta 2}^{(0)}) \\ &\quad \times \langle \mathbf{x} | \mathbf{X} \mathbf{R}_e(\mu) | \mathbf{y} \rangle \lambda \sigma(\mathbf{y}, \lambda) \mathbf{u}_{\mu\beta 2}^{(0)}(\mathbf{y}, \lambda). \end{aligned} \quad (\text{B8})$$

Since $\lambda^2 \mathbf{u}_{\mu\beta 2}^{(0)}(\mathbf{y}, \lambda) = \mu^2 \mathbf{u}_{\mu\beta 2}^{(0)}(\mathbf{y}, \lambda)$, $\mathbf{u}_{\mu\beta 2}^{(0)}(\mathbf{y}, \lambda)$ vanishes unless $\mu = \lambda$, so

$$\begin{aligned} &\sum_{\beta} \int d\mu g(\mu) a(\mathbf{u}_{\mu\beta 2}^{(0)}) \langle \mathbf{x} | \mathbf{X} \Omega_{12} | \mathbf{u}_{\mu\beta 2}^{(0)} \rangle \\ &\quad \times \int d\mathbf{y} \int d\lambda \langle \mathbf{x} | \mathbf{X} \mathbf{R}_e(\lambda) | \mathbf{y} \rangle \lambda g(\lambda) \sigma(\mathbf{y}, \lambda) \\ &\quad \times \sum_{\beta} \int d\mu \langle \mathbf{y}, \lambda | \mathbf{u}_{\mu\beta 2}^{(0)} \rangle a(\mathbf{u}_{\mu\beta 2}^{(0)}) \\ &= - \int d\mathbf{y} \int d\lambda \langle \mathbf{x} | \mathbf{X} \mathbf{R}_e(\lambda) | \mathbf{y} \rangle \lambda g(\lambda) \sigma(\mathbf{y}, \lambda) \mathbf{f}(\mathbf{y}, \lambda), \end{aligned} \quad (\text{B9})$$

where

$$\mathbf{f}(\mathbf{y}, \lambda) = - \sum_{\beta} \int d\mu \mathbf{u}_{\mu\beta 2}^{(0)}(\mathbf{y}, \lambda) a(\mathbf{u}_{\mu\beta 2}^{(0)}), \quad (\text{B10})$$

and its adjoint satisfy the commutation relation

$$[\mathbf{f}(\mathbf{y}, \lambda), \mathbf{f}^*(\mathbf{y}', \lambda')] = \mathbf{U} \delta(\mathbf{y} - \mathbf{y}') \delta(\lambda - \lambda'). \quad (\text{B11})$$

Thus

$$\begin{aligned} \hat{A}^\perp(\mathbf{x}) &= - \int d\mathbf{y} \int d\lambda \langle \mathbf{x} | \mathbf{P}^\perp \mathbf{R}_e(\lambda) | \mathbf{y} \rangle \\ &\quad \times (\lambda/2)^{1/2} \sigma(\mathbf{y}, \lambda) \mathbf{f}(\mathbf{y}, \lambda) + \text{H.c.}, \\ \partial_{\mathbf{x}} \hat{\theta}(\mathbf{x}) &= i \int d\mathbf{y} \int d\lambda \langle \mathbf{x} | \mathbf{P}^\parallel \mathbf{R}_e(\lambda) | \mathbf{y} \rangle \\ &\quad \times \lambda (\lambda/2)^{1/2} \sigma(\mathbf{y}, \lambda) \mathbf{f}(\mathbf{y}, \lambda) + \text{H.c.}, \end{aligned} \quad (\text{B12})$$

and also

$$H_{aux} = \int d\mathbf{y} \int d\lambda \lambda \mathbf{f}^*(\mathbf{y}, \lambda) \cdot \mathbf{f}(\mathbf{y}, \lambda). \quad (\text{B13})$$

APPENDIX C: CALCULATION OF EQ. (8.8)

Using Eq. (8.7) we obtain

$$\begin{aligned} \langle \varphi | [z - H^{(0)}]^{-1} | \varphi \rangle &= \sum_{\beta} \int d\mu \left\{ -i \sqrt{\frac{\mu}{2}} (\mathbf{u}_{\mu\beta}, \mathbf{W}) + \frac{1}{\sqrt{2\mu}} (\mathbf{u}_{\mu\beta}, \mathbf{I}) \right\} [z - H_m^{(0)} - \mu]^{-1} \left\{ i \sqrt{\frac{\mu}{2}} (\mathbf{W}, \mathbf{u}_{\mu\beta}) + \frac{1}{\sqrt{2\mu}} (\mathbf{I}, \mathbf{u}_{\mu\beta}) \right\} \\ &= \frac{1}{2} \sum_{\beta} \int d\mu \{ (\mathbf{u}_{\mu\beta}, \mathbf{W}) \mu [z - H_m^{(0)} - \mu]^{-1} (\mathbf{W}, \mathbf{u}_{\mu\beta}) + (\mathbf{u}_{\mu\beta}, \mathbf{I}) \mu^{-1} [z - H_m^{(0)} - \mu]^{-1} (\mathbf{I}, \mathbf{u}_{\mu\beta}) \\ &\quad + (\mathbf{u}_{\mu\beta}, \mathbf{W}) [z - H_m^{(0)} - \mu]^{-1} (\mathbf{W}, \mathbf{u}_{\mu\beta}) + (\mathbf{u}_{\mu\beta}, \mathbf{I}) [z - H_m^{(0)} - \mu]^{-1} (\mathbf{I}, \mathbf{u}_{\mu\beta}) \} \\ &= \frac{1}{2} \{ \langle \mathbf{W} | \mathbf{H}_e^{1/2} [z - H_m^{(0)} - \mathbf{H}_e^{1/2}]^{-1} \mathbf{Q}_e | \mathbf{W} \rangle + \langle \mathbf{I} | \mathbf{H}_e^{-1/2} [z - H_m^{(0)} - \mathbf{H}_e^{1/2}]^{-1} \mathbf{Q}_e | \mathbf{I} \rangle - i \langle \mathbf{W} | [z - H_m^{(0)} - \mathbf{H}_e^{1/2}]^{-1} \\ &\quad \times \mathbf{Q}_e | \mathbf{I} \rangle + i \langle \mathbf{I} | [z - H_m^{(0)} - \mathbf{H}_e^{1/2}]^{-1} \mathbf{Q}_e | \mathbf{W} \rangle \} \\ &= - \frac{1}{2} \langle \mathbf{W} | \mathbf{Q}_e | \mathbf{W} \rangle + \sum_{j=1}^4 X_j = -V_{Coul} + V_{stat} + \sum_{j=1}^4 X_j. \end{aligned} \quad (\text{C1})$$

Since $-\langle \varphi_{a_1} \otimes \varphi_{b_2} | V_{Coul} | \varphi_{a_2} \otimes \varphi_{b_1} \rangle = -\langle \varphi_{a_1} \otimes \varphi_{b_2} | V_{ab} | \varphi_{a_2} \otimes \varphi_{b_1} \rangle$, this term cancels Eq. (8.6). For X_1 we obtain (terms containing \mathbf{d}_a or \mathbf{d}_b twice vanish due to the orthogonality of φ_{b_2} and φ_{b_1} , respectively, φ_{a_1} and φ_{a_2})

$$\begin{aligned}
\langle \varphi_{a_1} \otimes \varphi_{b_2} | X_1 | \varphi_{a_2} \otimes \varphi_{b_1} \rangle &= \frac{1}{2} \langle \varphi_{a_1} \otimes \varphi_{b_2} | \mathbf{d}_a \cdot ([z - H_m^{(0)}] \langle \mathbf{X}_a | \mathbf{P}^{\parallel} ([z - H_m^{(0)} - H_e^{1/2}]^{-1} \mathbf{Q}_e)_{11} \mathbf{P}^{\parallel} | \mathbf{X}_b \rangle \mathbf{d}_b | \varphi_{a_2} \otimes \varphi_{b_1} \rangle \\
&\quad + \frac{1}{2} \langle \varphi_{a_1} \otimes \varphi_{b_2} | \mathbf{d}_b \cdot ([z - H_m^{(0)}] \langle \mathbf{X}_b | \mathbf{P}^{\parallel} ([z - H_m^{(0)} - H_e^{1/2}]^{-1} \mathbf{Q}_e)_{11} \mathbf{P}^{\parallel} | \mathbf{X}_a \rangle \mathbf{d}_a | \varphi_{a_2} \otimes \varphi_{b_1} \rangle \\
&= \frac{1}{2} \langle \varphi_{a_1} | \mathbf{d}_a | \varphi_{a_2} \rangle \cdot (-\omega_0 + i\delta) \langle \mathbf{X}_a | \mathbf{P}^{\parallel} ([-\omega_0 + i\delta - H_e^{1/2}]^{-1} \mathbf{Q}_e)_{11} \mathbf{P}^{\parallel} | \mathbf{X}_b \rangle \langle \varphi_{b_2} | \mathbf{d}_b | \varphi_{b_1} \rangle \\
&\quad + \frac{1}{2} \langle \varphi_{b_2} | \mathbf{d}_b | \varphi_{b_1} \rangle \cdot (\omega_0 + i\delta) \langle \mathbf{X}_b | \mathbf{P}^{\parallel} ([\omega_0 + i\delta - H_e^{1/2}]^{-1} \mathbf{Q}_e)_{11} \mathbf{P}^{\parallel} | \mathbf{X}_a \rangle \langle \varphi_{a_1} | \mathbf{d}_a | \varphi_{a_2} \rangle. \quad (C2)
\end{aligned}$$

Using

$$\langle \mathbf{X}_b | \mathbf{P}^{\parallel} ([\zeta - H_e^{1/2}]^{-1} \mathbf{Q}_e)_{11} \mathbf{P}^{\parallel} | \mathbf{X}_a \rangle = \langle \mathbf{X}_a | \mathbf{P}^{\parallel} ([\zeta - H_e^{1/2} \mathbf{Q}_e]^{-1})_{11} \mathbf{P}^{\parallel} | \mathbf{X}_b \rangle, \quad (C3)$$

this simplifies into

$$\begin{aligned}
&\langle \varphi_{a_1} \otimes \varphi_{b_2} | X_1 | \varphi_{a_2} \otimes \varphi_{b_1} \rangle \\
&= \frac{1}{2} \langle \varphi_{a_1} | \mathbf{d}_a | \varphi_{a_2} \rangle \langle \varphi_{b_2} | \mathbf{d}_b | \varphi_{b_1} \rangle : \langle \mathbf{X}_a | \mathbf{P}^{\parallel} \{ (-\omega_0 + i\delta) [-\omega_0 + i\delta - H_e^{1/2}]^{-1} \mathbf{Q}_e + (\omega_0 + i\delta) [\omega_0 + i\delta - H_e^{1/2}]^{-1} \mathbf{Q}_e \}_{11} \mathbf{P}^{\parallel} | \mathbf{X}_b \rangle. \quad (C4)
\end{aligned}$$

But, observing that $\omega_0 > 0$ and $H_e^{1/2} > 0$ and denoting a principal value by \mathcal{P} ,

$$\begin{aligned}
&(-\omega_0 + i\delta) [-\omega_0 + i\delta - H_m^{(0)} - H_e^{1/2}]^{-1} \mathbf{Q}_e + (\omega_0 + i\delta) [\omega_0 + i\delta - H_e^{1/2}]^{-1} \mathbf{Q}_e \\
&= \omega_0 \{ [\omega_0 - i\delta + H_e^{1/2}]^{-1} \mathbf{Q}_e + [\omega_0 + i\delta - H_e^{1/2}]^{-1} \mathbf{Q}_e \} \\
&= \omega_0 \left\{ \mathcal{P} \frac{1}{\omega_0 + H_e^{1/2}} + i\pi \delta(\omega_0 + H_e^{1/2}) + \mathcal{P} \frac{1}{\omega_0 - H_e^{1/2}} - i\pi \delta(\omega_0 - H_e^{1/2}) \right\} \mathbf{Q}_e \\
&= \left\{ 2\omega_0^2 \mathcal{P} \frac{1}{\omega_0^2 - H_e} - i\pi \omega_0 \delta(\omega_0 - H_e^{1/2}) \right\} \mathbf{Q}_e \\
&= \left\{ 2\omega_0^2 \mathcal{P} \frac{1}{\omega_0^2 - H_e} - i\pi \omega_0 (\omega_0 + H_e^{1/2}) \delta(\omega_0^2 - H_e) \right\} \mathbf{Q}_e \\
&= 2\omega_0^2 \left\{ \mathcal{P} \frac{1}{\omega_0^2 - H_e} - i\pi \delta(\omega_0^2 - H_e) \right\} \mathbf{Q}_e \\
&= 2\omega_0^2 [\omega_0^2 + i\delta - H_e]^{-1} \mathbf{Q}_e = -2\mathbf{P}_e + 2\omega_0^2 [\omega_0^2 + i\delta - H_e]^{-1}. \quad (C5)
\end{aligned}$$

Recalling that

$$([\omega_0^2 + i\delta - H_e]^{-1})_{11} = [\omega_0^2 \varepsilon(\mathbf{x}, \omega_0) - \mathbf{h}_0]^{-1} = \mathbf{R}_e(\omega_0), \quad (C6)$$

$$\begin{aligned}
&= \omega_0^2 \langle \varphi_{a_1} | \mathbf{d}_a | \varphi_{a_2} \rangle \langle \varphi_{b_2} | \mathbf{d}_b | \varphi_{b_1} \rangle : \langle \mathbf{X}_a | \mathbf{P}^{\parallel} \mathbf{R}_e(\omega_0) \mathbf{P}^{\parallel} | \mathbf{X}_b \rangle \\
&\quad - \langle \varphi_{a_1} \otimes \varphi_{b_2} | V_{stat} | \varphi_{a_2} \otimes \varphi_{b_1} \rangle. \quad (C7)
\end{aligned}$$

we obtain

$$\begin{aligned}
&\langle \varphi_{a_1} \otimes \varphi_{b_2} | X_1 | \varphi_{a_2} \otimes \varphi_{b_1} \rangle \\
&= \omega_0^2 \langle \varphi_{a_1} | \mathbf{d}_a | \varphi_{a_2} \rangle \langle \varphi_{b_2} | \mathbf{d}_b | \varphi_{b_1} \rangle : \langle \mathbf{X}_a | \mathbf{P}^{\parallel} \mathbf{R}_e(\omega_0) \mathbf{P}^{\parallel} | \mathbf{X}_b \rangle \\
&\quad - \langle \varphi_{a_1} | \mathbf{d}_a | \varphi_{a_2} \rangle \langle \varphi_{b_2} | \mathbf{d}_b | \varphi_{b_1} \rangle : \langle \mathbf{X}_a | (\mathbf{P}_e)_{11} | \mathbf{X}_b \rangle \\
&\quad \langle \varphi_{out} | T(z) | \varphi_{in} \rangle = \omega_0^2 \langle \varphi_{a_1} | \mathbf{d}_a | \varphi_{a_2} \rangle \\
&\quad \times \langle \varphi_{b_2} | \mathbf{d}_b | \varphi_{b_1} \rangle : \langle \mathbf{X}_a | \mathbf{R}_e(\omega_0) | \mathbf{X}_b \rangle. \quad (C8)
\end{aligned}$$

APPENDIX D: THE AHARONOV-AU INVARIANCE PRINCIPLE

Consider a quantity

$$F = \langle \varphi_{out} | X | \varphi_{in} \rangle, \quad (D1)$$

on a Hilbert space \mathcal{H} , where φ_{in} and φ_{out} are eigenstates of a self-adjoint operator H_0 at the same eigenvalue E , $H_0\varphi_{in} = E\varphi_{in}$, $H_0\varphi_{out} = E\varphi_{out}$. Then, if $U = \exp[i\Phi]$ is a unitary operator,

$$F = \langle \hat{\varphi}_{out} | \hat{X} | \hat{\varphi}_{in} \rangle, \quad \hat{X} = \hat{X}(U) = UXU^{-1}, \quad \hat{\varphi}_j = U\varphi_j. \quad (D2)$$

But we can also write

$$\begin{aligned} F &= \langle \varphi_{out} | \exp[-iH_0t] \hat{X}(U) \exp[iH_0t] | \varphi_{in} \rangle \\ &= \langle \varphi_{out} | \hat{X}[U(t)] | \varphi_{in} \rangle, \\ U(t) &= \exp[-iH_0t] U \exp[iH_0t]. \end{aligned} \quad (D3)$$

Now, suppose that

$$\hat{X}[U(t)] \xrightarrow{t \rightarrow \infty} \hat{X}(1) = X. \quad (D4)$$

This is the case if, for $f \in \mathcal{H}$, $U(t)f \rightarrow_{t \rightarrow \infty} f$, or $\Phi(t)f = \exp[-iH_0t]\Phi\exp[iH_0t]f \rightarrow_{t \rightarrow \infty} 0$. Then

$$F = \langle \varphi_{out} | \hat{X} | \varphi_{in} \rangle, \quad (D5)$$

i.e., the original states φ_{in} and φ_{out} give the same result in both representations. Note that we cannot directly consider $U(t)\varphi_j$, since in general the φ_j 's are not in \mathcal{H} (for example, plane-wave states in scattering processes). A typical case is a transition amplitude $F = \langle \varphi_{out} | T(E+i0) | \varphi_{in} \rangle$. Now consider the switch from the primed system to the hatted one. Then $U = \Omega_F^*$ and

$$\begin{aligned} &\exp[-iH_f^{(0)}t] \Omega_F^* \exp[iH_f^{(0)}t] \\ &= \Omega_F^* \exp[-iH_f t] \exp[iH_f^{(0)}t] \xrightarrow{t \rightarrow \infty} \Omega_F^* \Omega_F = 1. \end{aligned} \quad (D6)$$

Thus, if φ_{in} and φ_{out} are products of matter states and field states with the field states being eigenstates of $H_f^{(0)}$ at the same eigenvalue, then

$$\langle \varphi_{out} | \hat{X} | \varphi_{in} \rangle = \langle \varphi_{out} | X' | \varphi_{in} \rangle, \quad (D7)$$

hence extending Eq. (5.13).

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