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Macroscopic quantization in quantum optics and cavity quantum electrodynamics: Interatomic interactions

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We extend previous work on macroscopic canonical quantization leading to a multipolar Hamiltonian appropriate for application to quantum optics and cavity QED situations involving classical optical devices. In particular, we show that the electric displacement is the negative of the conjugate momentum field and that the Coulomb and polarization energies are equal to the sum of intra-atomic Coulomb and polarization energies and interatomic contact energies. The quantum Hamiltonian is now in a form in which the theory is manifestly gauge invariant. [S1050-2947(97)04307-2]

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I. INTRODUCTION

Experiments both in quantum optics generally and in cavity quantum electrodynamics QED in particular study the interaction of the quantum electromagnetic (EM) field with radiative atoms in the presence of classical optical devices such as resonator cavities, beam splitters, filters, and so on. In many situations the quantum nature of the dielectric atoms making up these classical optical devices is not important, as, for example, when nonlinearity, absorption, and dispersion can be ignored for EM field intensities and frequencies of interest.

To obtain the basic quantum Hamiltonian needed for theory, a canonical quantization for the full system of EM field, radiative atoms, and dielectric atoms can be performed [1] and then a formal elimination of the spatially inhomogeneous dielectric atoms constituting the optical system carried out later. However, it is often more convenient to base the canonical quantization on a classical Lagrangian in which the dielectric atoms of the optical device are treated macroscopically from the beginning. These atoms are replaced by the spatially dependent electric permittivity $\epsilon(\mathbf{R})$ [and magnetic permeability $\mu(\mathbf{R})$, if there is also magnetic material] that describes all the classical optical effects (apart from dispersion and absorption) for the device in question. This second approach will be referred to as macroscopic canonical quantization, to distinguish it from the first approach of microscopic canonical quantization, in which the dynamics of the dielectric atoms is initially included, albeit in terms of simple models. Such models for the dielectric material used in the microscopic approach include harmonic polarization fields [2,3] and polariton fields [4], which are then quantized along with the EM field and radiative atoms.

Previous work on macroscopic canonical quantization [5,6] used a Lagrangian that yielded the quantum Hamiltonian in the minimal coupling form. For quantum optical studies where often the electric dipole approximation is used, it is preferable to obtain the quantum Hamiltonian in the multipolar form, and this has recently been achieved by Dalton *et al.* [7]. The electric dipole approximation result was obtained also. This macroscopic canonical quantization work [5–7] is based on using a generalized radiation gauge condition in which the vector potential $\mathbf{A}(\mathbf{R})$ satisfies $\boldsymbol{\nabla}\cdot\epsilon\mathbf{A}=0$. In this gauge the scalar potential ϕ only depends on the positions of the charged particles in the radiative atoms.

In the case of free-space quantization there are actually several routes that one could use to derive the multipolar Hamiltonian [8]. One is to apply a unitary transformation to the minimal coupling Hamiltonian [9]. A second is to add a total time derivative to the original Lagrangian [1,10]. A third approach is via the use of gauge transformations [8] to the potentials $\mathbf{A},\phi$ in the original Lagrangian. All these methods give the same outcome; the method used here is the second one. The theory is of course gauge invariant, though this is not necessarily obvious if the quantum Hamiltonian involves the potentials $\mathbf{A},\phi$ rather than electric and magnetic fields.

In all of the previous work on macroscopic canonical quantization [5–7] for quantum optics and cavity QED situations, the basic picture of the quantum EM field that emerges is still the same photon model that applies in free space or microscopic canonical quantization. However, in the quantum optics of cavity situation or where any classical linear optics device is involved, the quantum EM field is equivalent to a set of independent quantum harmonic oscillators, one for each classical mode of the EM field, as modified by the classical linear optical device. The noncoupling of the quantum harmonic oscillators is associated with the use of the true modes (universe modes) for the optical system, which are obtained as solutions of a generalized Helmholtz equation. The vector potential is expanded in terms of such true mode functions and is required to satisfy the generalized radiation gauge condition.

In other work [11] idealizations of the modes for the optical device (or quasimodes) can be introduced. These are solutions of a generalized Helmholtz equation involving an artificially chosen permittivity function $\tilde{\epsilon}(\mathbf{R})$ [and permeability function $\tilde{\mu}(\mathbf{R})$] to produce the required quasimode functions. Naturally the vector potential still satisfies the generalized radiation gauge condition for the true $\epsilon(\mathbf{R})$. In this case macroscopic canonical quantization in terms of the quasimodes still yields a quantum multipolar Hamiltonian in

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which the photon model of quantum harmonic oscillators (one for each quasimode) still applies, but now the quantum harmonic oscillators are coupled so that energy exchanges between different quasimodes can now occur, as in the phenomenological model used in input-output theory [12].

For quantum optics and cavity QED situations involving a classical optical system the interaction between the radiative atoms and the quantum EM field described via the multipolar Hamiltonian [7,8] consists of an electric interaction term and a magnetic interaction term, both involving the quantum EM field linearly via the conjugate momentum field $\Pi(\mathbf{R})$ and the magnetic field $\nabla\times\mathbf{A}(\mathbf{R})$, respectively, together with reduced polarization densities $P_L'(\mathbf{R})$ and magnetization densities $M_L'(\mathbf{R})$, and also a diamagnetic interaction term, which involves the magnetic field quadratically. The electric and magnetic interaction energies are associated with one-photon processes, the diamagnetic energy with two-photon processes, and the dominant interaction is the electric dipole approximation to the electric interaction energy.

The atom EM field interaction terms involve the mode functions at the atomic positions. For the true mode quantization scheme each atom can interact with all true modes [7], whereas for the quasimode quantization scheme the mode functions can be chosen so that only cavitylike quasimodes have sufficient amplitude for the atom to interact with [11]. Thus in the quasimode approach only atom field coupling to a limited number of modes need be considered.

In the recent paper by Dalton *et al.* [7] the quantum multipolar Hamiltonian also contained a Coulomb energy term and a polarization energy term. In the free-space canonical quantization approach [1] these terms can be further developed to show that together they are equal to intra-atomic Coulomb energy and polarization energy terms along with interatomic contact energy terms. In this paper we show that this result also applies in canonical macroscopic quantization for quantum optics and cavity QED with classical optical devices. This development depends on finding the relationship between the scalar potential ϕ and the polarization and reduced polarization densities $\mathbf{P}_L(\mathbf{R})$, $\mathbf{P}_L'(\mathbf{R})$ and this relationship is found in the present paper, via the use of a generalization of the Helmholtz theorem.

Also, in previous work [7] the relationship between the electric displacement $\mathbf{D}(\mathbf{R})$ and the conjugate momentum field $\mathbf{\Pi}(\mathbf{R})$ was left in a form that did not exploit the relationship between ϕ, $\mathbf{P}_L$, and $\mathbf{P}_L'$ referred to above. In the free-space canonical quantization approach [1,7], the relationship $\mathbf{D}=-\mathbf{\Pi}$ applies for neutral radiative atoms. In the present paper we show that this relationship also applies in the case of macroscopic canonical quantization for cavity QED situations. The electric dipole approximation to the interaction now involves the usual $-\boldsymbol{\mu}\cdot\mathbf{D}/\epsilon$ form, familiar from free-space quantization.

Finally, the form for the equal-time commutator of the vector potential and the conjugate momentum field is expressed in terms of a generalized transverse Dirac delta function.

The plan of this paper is as follows. In Sec. II the generalized form of the Helmholtz theorem is developed, leading to the introduction of generalized transverse and longitudinal Dirac delta functions. In Sec. III the results are applied in macroscopic canonical quantization for cavity QED. In Sec. III A the reduced polarization density is expressed in terms of the generalized transverse component for $\mathbf{P}_L(\mathbf{R})/\epsilon(\mathbf{R})$, in Sec. III B the relationship between ϕ, $\mathbf{P}_L$, and $\mathbf{P}_L'$ is obtained, in Sec. III C the result for $\mathbf{D}$ is derived, in Sec. III D the equal-time commutator for $\mathbf{A}$, $\mathbf{\Pi}$ is obtained, in Sec. III E the Coulomb and polarization energy terms are combined, and in Sec. III F they are shown to be equal to the sum of intra-atomic Coulomb, polarization energies, and contact energy. In Sec. III G the quantum multipolar Hamiltonian is obtained, along with its electric dipolar approximation in Sec. III I. The results are summarized in Sec. IV.

II. GENERALIZED HELMHOLTZ THEOREM

The usual Helmholtz theorem writes a vector field $\mathbf{F}(\mathbf{R})$ as the sum of a transverse field $\mathbf{F}_\perp$ (with zero divergence) and a longitudinal field $\mathbf{F}_\parallel$ (with zero curl). For cavity QED macroscopic quantization via a spatially dependent electric permittivity function $\epsilon(\mathbf{R})$ requires the use of a generalized radiation gauge condition $\nabla\cdot\epsilon\mathbf{A}=0$ for the vector potential $\mathbf{A}(\mathbf{R})$. Thus the vector potential is no longer transverse.

It is therefore useful to consider a generalization of Helmholtz theorem in which the vector field is written as

$$\mathbf{F}(\mathbf{R})=\mathbf{F}_\perp^{(\epsilon)}(\mathbf{R})+\mathbf{F}_\parallel^{(\epsilon)}(\mathbf{R}),\tag{2.1}$$

where now with a specific function $\epsilon(\mathbf{R})$,

$$\nabla\cdot\epsilon\mathbf{F}_\perp^{(\epsilon)}=0,\tag{2.2}$$

$$\nabla\times\mathbf{F}_\parallel^{(\epsilon)}=0.\tag{2.3}$$

Thus as $\mathbf{F}_\perp^{(\epsilon)}$ is no longer transverse it will contain a longitudinal component and $\mathbf{F}_\parallel^{(\epsilon)}$, although longitudinal, is no longer the longitudinal component of $\mathbf{F}$.

The transverse and longitudinal components of a vector field satisfy the usual orthogonality condition with weight function unity. On the other hand, the generalized transverse component $\mathbf{F}_\perp^{(\epsilon)}$ and longitudinal components $\mathbf{F}_\parallel^{(\epsilon)}$ satisfy the generalized orthogonality condition applying for macroscopic quantization in cavity QED, with $\epsilon(\mathbf{R})$ the weight function:

$$\int d^3\mathbf{R}\ \epsilon(\mathbf{R})\mathbf{F}_\perp^{(\epsilon)}(\mathbf{R})\cdot\mathbf{F}_\parallel^{(\epsilon)}(\mathbf{R})=0.\tag{2.4}$$

The proof of this is simple enough—write $\epsilon\mathbf{F}_\perp^{(\epsilon)}$ as $\nabla\times\mathbf{W}$, $\mathbf{F}_\parallel^{(\epsilon)}$ as $\nabla\psi$ (where $\mathbf{W},\psi$ are suitable vector and scalar potentials), and then use $\nabla\cdot(\psi\nabla\times\mathbf{W})=\nabla\psi\cdot\nabla\times\mathbf{W}$, followed up with Gauss' theorem, after integrating each side over all space.

Expressions for the generalized transverse component $\mathbf{F}_\perp^{(\epsilon)}$ can be obtained in terms of normalized eigenfields $\mathbf{F}_\lambda(\mathbf{R})$ of any suitable self-adjoint operator L:

$$L\mathbf{F}_\lambda(\mathbf{R})=\lambda\mathbf{F}_\lambda(R),\tag{2.5}$$

where for arbitrary fields $\mathbf{U}$, $\mathbf{V}$:

$$\int d^3\mathbf{R}\ \epsilon(\mathbf{R})\mathbf{U}^*\cdot(L\mathbf{V})=\int d^3\mathbf{R}\ \epsilon(\mathbf{R})(LU)^*\cdot\mathbf{V}\tag{2.6}$$

and that also satisfy

$$\nabla\cdot\epsilon\mathbf{F}_\lambda(\mathbf{R})=0. \tag{2.7}$$

The eigenfields $\mathbf{F}_\lambda(\mathbf{R})$ can be shown to be orthogonal in the general sense as in Eq. (2.4) (see Ref. [7]) for different eigenvalues λ and can be finally chosen to be normalized also. The generalized orthogonality and normalization condition is

$$\int d^3\mathbf{R}\ \epsilon(\mathbf{R})\mathbf{F}^*_\lambda(\mathbf{R})\cdot\mathbf{F}_\mu(\mathbf{R})=\delta_{\lambda\mu}. \tag{2.8}$$

The eigenvalues λ are also real. As a specific example, any operator of the form

$$L=\frac{1}{\epsilon}\left(\nabla\times\frac{1}{\mu}\,(\nabla\times\cdots)\right), \tag{2.9}$$

with μ an arbitrary function, will lead to eigenfields satisfying the required conditions.

Expanding $\mathbf{F}^{(\epsilon)}_\perp$ in terms of the $\mathbf{F}_\lambda$ the expansion coefficients can easily be found using the generalized orthogonality and normalization expressions to give

$$\mathbf{F}^{(\epsilon)}_\perp(\mathbf{R})=\sum_\lambda\ C_\lambda\mathbf{F}_\lambda(\mathbf{R}), \tag{2.10}$$

with

$$C_\lambda=\int d^3\mathbf{R}'\ \epsilon(\mathbf{R}')\mathbf{F}^*_\lambda(\mathbf{R}')\cdot\mathbf{F}^{(\epsilon)}_\perp(\mathbf{R}'). \tag{2.11}$$

But analogous to the proof of Eq. (2.4),

$$\int d^3\mathbf{R}'\ \epsilon(\mathbf{R}')\mathbf{F}^*_\lambda(\mathbf{R}')\cdot\mathbf{F}^{(\epsilon)}_\parallel(\mathbf{R}')=0. \tag{2.12}$$

In view of this C_λ can be expressed in terms of the original field $\mathbf{F}(R)$ as

$$C_\lambda=\int d^3\mathbf{R}'\ \epsilon(\mathbf{R}')\mathbf{F}^*_\lambda(\mathbf{R}')\cdot\mathbf{F}(\mathbf{R}'). \tag{2.13}$$

If we introduce a generalization of the transverse Dirac delta function via

$$\delta^{(\epsilon)}_\perp(\mathbf{R},\mathbf{R}')_{ij}=\sum_\lambda\ \mathbf{F}_{\lambda i}(\mathbf{R})F^*_{\lambda j}(\mathbf{R}')\epsilon(\mathbf{R}'). \tag{2.14}$$

then we can write the generalized transverse component

$$\mathbf{F}^{(\epsilon)}_\perp(\mathbf{R})_i=\int d^3\mathbf{R}'\ \delta^{(\epsilon)}_\perp(\mathbf{R},\mathbf{R}')_{ij}F_j(\mathbf{R}'). \tag{2.15}$$

The generalized longitudinal Dirac delta function is defined by

$$\delta^{(\epsilon)}_\parallel(\mathbf{R},\mathbf{R}')_{ij}=\delta_{ij}\delta(\mathbf{R}-\mathbf{R}')-\delta^{(\epsilon)}_\perp(\mathbf{R},\mathbf{R}')_{ij} \tag{2.16}$$

so that the generalized longitudinal component is

$$\mathbf{F}^{(\epsilon)}_\parallel(\mathbf{R})_i=\int d^3\mathbf{R}'\ \delta^{(\epsilon)}_\parallel(\mathbf{R},\mathbf{R}')_{ij}F_{\mathbf{j}}(\mathbf{R}'). \tag{2.17}$$

The uniqueness of $\mathbf{F}^{(\epsilon)}_\perp$ (and hence of $\mathbf{F}^{(\epsilon)}_\parallel$) is guaranteed by the unique result Eq. (2.13) for the coefficients C_λ.

III. APPLICATION IN MACROSCOPIC CANONICAL QUANTIZATION FOR CAVITY QED

In this application $\epsilon(\mathbf{R})$ is the permittivity function, $\mu(\mathbf{R})$ the permeability function, the eigenfields are the vector mode functions $\mathbf{A}_k$ and Eq. (2.5) is the generalized Helmholtz equation with λ_k equal to ω_k^2, the square of the angular frequency. The vector potential $\hat{\mathbf{A}}(\mathbf{R})$ is expanded in terms of the $\mathbf{A}_k$ as

$$\hat{\mathbf{A}}(\mathbf{R})=\sum_k\ \hat{q}_k\mathbf{A}_k(\mathbf{R}), \tag{3.1}$$

where the $\hat{q}_k$ act as the generalized coordinates. The operator nature of the various quantities will be shown explicitly.

The generalized transverse Dirac delta function will be

$$\delta^{(\epsilon)}_\perp(\mathbf{R},\mathbf{R}')_{ij}=\sum_k\ A_{ki}(\mathbf{R})A^*_{kj}(\mathbf{R}')\epsilon(\mathbf{R}'). \tag{3.2}$$

A similar function to this is used by Vogel and Welsch [13].

A. Reduced polarization density

The reduced polarization density $\mathbf{P}'_L(\mathbf{R})$ is defined as [see Eq. (53) of Ref. [7]]

$$\mathbf{P}'_L(\mathbf{R})=\sum_k\ \epsilon(\mathbf{R})\mathbf{A}_k(\mathbf{R})\int d^3\mathbf{R}'\ \mathbf{A}^*_k(\mathbf{R}')\cdot\hat{\mathbf{P}}_L(\mathbf{R}'), \tag{3.3}$$

where $\hat{\mathbf{P}}_L(\mathbf{R})$ is the polarization density associated with the radiative atoms [see Eqs. (3) and (7) of Ref. [7]].

If we consider the field $\hat{\mathbf{P}}_L(\mathbf{R})/\epsilon(\mathbf{R})$ then it is easy to see that

$$\hat{\mathbf{P}}'_L(\mathbf{R})=\epsilon(\mathbf{R})\left(\frac{\hat{\mathbf{P}}_L(\mathbf{R})}{\epsilon(\mathbf{R})}\right)^{(\epsilon)}_\perp \tag{3.4}$$

so that the reduced polarization density is related to the generalized transverse component of the field $\hat{\mathbf{P}}_L/\epsilon$.

B. The scalar potential ϕ

Consider the field given by $\hat{\mathbf{P}}_L/\epsilon-\hat{\mathbf{P}}'_L/\epsilon$. We can show that this field has zero curl so that it is a generalized longitudinal field. For

$$\nabla\times\left(\frac{\hat{\mathbf{P}}_L}{\epsilon}\right)=\nabla\times\left(\frac{\hat{\mathbf{P}}_L}{\epsilon}\right)^{(\epsilon)}_\perp \tag{3.5}$$

since $(\hat{\mathbf{P}}_L/\epsilon)^{(\epsilon)}_\parallel$ has a zero curl. But from Eq. (3.4) above

$$\nabla\times\left(\frac{\hat{\mathbf{P}}'_L}{\epsilon}\right)=\nabla\times\left(\frac{\hat{\mathbf{P}}_L}{\epsilon}\right)^{(\epsilon)}_\perp \tag{3.6}$$

so taking the difference between each side of (3.5) and (3.6) we find that

$$\nabla\times\left(\frac{\hat{\mathbf{P}}_L}{\epsilon}-\frac{\hat{\mathbf{P}}'_L}{\epsilon}\right)=0. \tag{3.7}$$

Since the field $\hat{\mathbf{P}}_L/\epsilon-\hat{\mathbf{P}}'_L/\epsilon$ is longitudinal it can be expressed as the gradient of a scalar potential. Suppose we choose the scalar potential ϕ to satisfy

$$\epsilon\hat{\nabla}\phi=\hat{\mathbf{P}}_L-\hat{\mathbf{P}}'_L \tag{3.8}$$

or $\hat{\nabla}\phi=\hat{\mathbf{P}}_L/\epsilon-\hat{\mathbf{P}}'_L/\epsilon$, then since $\hat{\mathbf{P}}'_L$ is transverse [and using Eqs. (6) and (27) of Ref. [7]] we have

$$\nabla\cdot(\epsilon\hat{\nabla}\phi)=\nabla\cdot\hat{\mathbf{P}}_L-\nabla\cdot\hat{\mathbf{P}}'_L=-\hat{\rho}_L, \tag{3.9}$$

showing that ϕ satisfies the generalized Poisson equation [Eq. (2) of Ref. [7]]. Hence it is the scalar potential ϕ for the EM field. The uniqueness of the result for $\epsilon\hat{\nabla}\phi$ is demonstrated in Appendix I of Ref. [7].

Then with the vector potential **A** Eq. (3.1) satisfying the generalized inhomogeneous wave equation [Eq. (21) of Ref. [7]] it is easy to show that the electric field **E** and magnetic field **B** given by

$$\hat{\mathbf{E}}=-\nabla\hat{\phi}-\dot{\hat{\mathbf{A}}} \tag{3.10}$$

$$\hat{\mathbf{B}}=\nabla\times\hat{\mathbf{A}} \tag{3.11}$$

will satisfy Maxwell's equations [Eq. (13) of Ref. [7]] for electrically neutral, nonconducting dielectric, and magnetic media.

C. The electric displacement $\hat{\mathbf{D}}$

In general [see Eq. (79) of Ref. [7]] the electric displacement $\hat{\mathbf{D}}$ is related to the conjugate momentum field $\hat{\mathbf{\Pi}}$ for the case of electrically neutral dielectric and radiative atoms via

$$\hat{\mathbf{D}}=\hat{\mathbf{D}}_\perp=-\hat{\Pi}-(\epsilon\hat{\nabla}\phi)_\perp+(\hat{\mathbf{P}}_L)_\perp-(\hat{\mathbf{P}}'_L), \tag{3.12}$$

where in terms of the vector mode functions

$$\hat{\Pi}(\mathbf{R})=\sum_k \hat{p}_k\epsilon(\mathbf{R})A_k(\mathbf{R}) \tag{3.13}$$

with $\hat{p}_k$ being the generalized momentum coordinates.

In view of Eq. (3.8), however, the last three terms in (3.12) cancel out, giving the relationship between the electric displacement and the conjugate momentum field

$$\hat{\mathbf{D}}(\mathbf{R})=-\hat{\Pi}(\mathbf{R}), \tag{3.14}$$

which is the usual result obtained in free-space quantization (see, for example, Ref. [1]).

D. Equal-time commutation rule

The equal-time commutator between the components of the vector potential $\hat{\mathbf{A}}$ and the conjugate momentum field $\hat{\mathbf{\Pi}}$ can be written in terms of the transverse generalized Dirac delta function. From Eq. (74) of Ref. [7]

$$[\hat{A}_i(\mathbf{R}),\hat{\Pi}_j(\mathbf{R}')]=i\hbar\sum_k A_{ki}(\mathbf{R})A^*_{kj}(\mathbf{R}')\epsilon(\mathbf{R}')$$
$$=i\hbar\,\delta_\perp^{(\epsilon)}(\mathbf{R},\mathbf{R}')_{ij}. \tag{3.15}$$

This now more clearly expresses the commutator as the generalization of the transverse Dirac delta function.

E. Coulomb energy and polarization energy

From Eqs. (23) and (62) of Ref. [7] the Coulomb energy $\hat{V}_{\text{Coul}}$ and the polarization energy $\hat{V}_{\text{pol}}$ are given by

$$\hat{V}_{\text{Coul}}=\int d^3\mathbf{R}\,\frac{\epsilon\hat{\nabla}\phi\cdot\epsilon\hat{\nabla}\phi}{2\epsilon}, \tag{3.16}$$

$$\hat{V}_{\text{pol}}=\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}'_L\cdot\hat{\mathbf{P}}'_L}{2\epsilon}. \tag{3.17}$$

Substituting for $\epsilon\hat{\nabla}\phi$ from Eq. (3.8) we find that

$$\hat{V}_{\text{Coul}}+\hat{V}_{\text{pol}}=\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}_L\cdot\hat{\mathbf{P}}_L}{2\epsilon}-\int\left(\frac{\hat{\mathbf{P}}_L}{\epsilon}-\frac{\hat{\mathbf{P}}'_L}{\epsilon}\right)\cdot\hat{\mathbf{P}}'_L\,d^3\mathbf{R}. \tag{3.18}$$

But $\hat{\mathbf{P}}_L/\epsilon-\hat{\mathbf{P}}'_L/\epsilon$ is purely longitudinal, whereas $\hat{\mathbf{P}}'_L$ is transverse. Hence the integral of their scalar products is zero and

$$\hat{V}_{\text{Coul}}+\hat{V}_{\text{pol}}=\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}_L(\mathbf{R})\cdot\hat{\mathbf{P}}_L(\mathbf{R})}{2\epsilon(\mathbf{R})}, \tag{3.19}$$

which now only involves the (nonreduced) polarization density $\hat{\mathbf{P}}_L(\mathbf{R})$ for the radiative atoms. A similar result can be derived for free-space quantization.

F. Contact energy and intra-atomic Coulomb and polarization energies

As in the case of free-space quantization the polarization density $\hat{\mathbf{P}}_L$ can be expressed as the sum of contributions from the separate radiative atoms ξ, whose charge density is $\rho_{L\xi}$:

$$\hat{\mathbf{P}}_L(\mathbf{R})=\sum_\xi \hat{\mathbf{P}}_{L\xi}(\mathbf{R}). \tag{3.20}$$

Substituting into Eq. (3.19) we see that the sum of the Coulomb and polarization energies can be written as

$$\hat{V}_{\text{Coul}}+\hat{V}_{\text{pol}}=\sum_\xi\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}_{L\xi}\cdot\hat{\mathbf{P}}_{L\xi}}{2\epsilon}+\sum_{\xi\neq\eta}\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}_{L\xi}\cdot\hat{\mathbf{P}}_{L\eta}}{2\epsilon}. \tag{3.21}$$

The last term is the contact energy $\hat{V}_{\text{cont}}$ given by

$$\hat{V}_{\text{cont}}=\sum_{\xi\neq\eta}\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}_{L\xi}\cdot\hat{\mathbf{P}}_{L\eta}}{2\epsilon} \tag{3.22}$$

and is normally ignored unless the two different radiative atoms are overlapping, which is not usually the case.

The first term can be written using the arguments of Sec. III F in reverse, as the sum of the intra-atomic Coulomb $\hat{V}^{\mathrm{IA}}_{\mathrm{Coul}}$ and polarization energies $\hat{V}^{\mathrm{IA}}_{\mathrm{pol}}$:

$$\hat{V}^{\mathrm{IA}}_{\mathrm{Coul}}=\sum_{\xi}\int d^3\mathbf{R}\,\frac{(\epsilon\hat{\boldsymbol{\nabla}}\phi)_{\xi}\cdot(\epsilon\hat{\boldsymbol{\nabla}}\phi)_{\xi}}{2\epsilon},\tag{3.23}$$

$$\hat{V}^{\mathrm{IA}}_{\mathrm{pol}}=\sum_{\xi}\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}'_{L\xi}\cdot\hat{\mathbf{P}}'_{L\xi}}{2\epsilon},\tag{3.24}$$

where

$$\hat{\mathbf{P}}'_{L\xi}(\mathbf{R})=\sum_{k}\epsilon(\mathbf{R})\mathbf{A}_k(\mathbf{R})\int d^3\mathbf{R}'\;\mathbf{A}^*_{\mathbf{k}}(\mathbf{R}')\cdot\hat{\mathbf{P}}_{L\xi}(\mathbf{R}'),\tag{3.25}$$

$$(\epsilon\hat{\boldsymbol{\nabla}}\phi)_{\xi}=\hat{\mathbf{P}}_{L\xi}-\hat{\mathbf{P}}'_{L\xi},\tag{3.26}$$

and thus

$$\boldsymbol{\nabla}\cdot(\epsilon\hat{\boldsymbol{\nabla}}\phi)_{\xi}=-\rho_{L\xi}.\tag{3.27}$$

Hence we have the result

$$\hat{V}_{\mathrm{Coul}}+\hat{V}_{\mathrm{pol}}=\hat{V}^{\mathrm{IA}}_{\mathrm{coul}}+\hat{V}^{\mathrm{IA}}_{\mathrm{pol}}+\hat{V}_{\mathrm{cont}}.\tag{3.28}$$

G. Quantum Hamiltonian

From Eq. (80) of Ref. [7] and using Eq. (3.28) the full quantum multipolar Hamiltonian can be written in the form

$$\hat{H}'=\hat{H}_A+\hat{H}_F+\hat{H}_{AF}+\hat{H}_{AA}.\tag{3.29}$$

The terms are as follows: in the expressions for the quantum Hamiltonian the α charged particle in the ξ atom has position $\hat{\mathbf{r}}_{\xi\alpha}$ momentum $\hat{\mathbf{p}}_{\xi\alpha}$, mass $M_{\xi\alpha}$, and charge $q_{\xi\alpha}$. The atomic Hamiltonian $\hat{H}_A$ can be written as the sum of terms $\hat{H}_{A\xi}$ for each separate atom:

$$\hat{H}_A=\sum_{\xi}\hat{H}_{A\xi},\tag{3.30}$$

with

$$\hat{H}_{A\xi}=\sum_{\alpha}\frac{\hat{\mathbf{p}}^2_{\xi\alpha}}{2M_{\xi\alpha}}+\int d^3\mathbf{R}\,\frac{(\epsilon\hat{\boldsymbol{\nabla}}\phi)_{\xi}\cdot(\epsilon\hat{\boldsymbol{\nabla}}\phi)_{\xi}}{2\epsilon}+\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}'_{L\xi}\cdot\hat{\mathbf{P}}'_{L\xi}}{2\epsilon},\tag{3.31}$$

giving the atomic Hamiltonian for the ξ atom as the sum of the kinetic energy, the intra-atomic Coulomb energy and the intra-atomic polarization energy for this atom. The field Hamiltonian can be written as the sum of quantum harmonic oscillator terms, one for each mode of the field:

$$\hat{H}_F=\sum_{k}\left(\hat{a}^{\dagger}_k\hat{a}_k+\frac{1}{2}\right)\hbar\omega_k.\tag{3.32}$$

The atom field interaction $\hat{H}_{AF}$ can be written as the sum of terms for each separate atom, and each atom ξ interacts with the field through an electric interaction $\hat{H}_{E\xi}$, a magnetic interaction $\hat{H}_{M\xi}$, and a diamagnetic interaction $\hat{H}_{D\xi}$. Thus

$$\hat{H}_{AF}=\sum_{\xi}(\hat{H}_{E\xi}+\hat{H}_{M\xi}+\hat{H}_{D\xi}),\tag{3.33}$$

with

$$\hat{H}_{E\xi}=\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}'_{L\xi}\cdot\hat{\Pi}}{\epsilon},\tag{3.34}$$

$$\hat{H}_{M\xi}=\int d^3\mathbf{R}\;\hat{\mathbf{M}}'_{L\xi}\cdot\boldsymbol{\nabla}\times\hat{\mathbf{A}},\tag{3.35}$$

$$\hat{H}_{D\xi}=\sum_{\alpha}\frac{q^2_{\xi\alpha}}{2M_{\xi\alpha}}\int_0^1 u\,du\{\hat{B}[\mathbf{R}_{\xi}+u(\mathbf{r}_{\xi\alpha}-\mathbf{R}_{\xi})]\times(\mathbf{r}_{\xi\alpha}-\mathbf{R}_{\xi})\}^2.\tag{3.36}$$

The reduced magnetization density $\hat{\mathbf{M}}'_L$ [see Eq. (37) of Ref. [7]] is written as the sum of contributions from the separate radiative atoms

$$\hat{\mathbf{M}}'_L(\mathbf{R})=\sum_{\xi}\hat{\mathbf{M}}'_{L\xi}(\mathbf{R}).\tag{3.37}$$

The atom-atom interaction $\hat{H}_{AA}$ is given by the contact energy term and is the sum over pairs of different atoms of contact terms:

$$\hat{H}_{AA}=\sum_{\xi\neq\eta}\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}_{L\xi}\cdot\hat{\mathbf{P}}_{L\eta}}{2\epsilon}.\tag{3.38}$$

In view of Eqs. (3.11) and (3.14) the electric and magnetic interaction terms may also be written as

$$\hat{H}_{E\xi}=-\int d^3\mathbf{R}\,\frac{\hat{\mathbf{P}}'_{L\xi}\cdot\hat{\mathbf{D}}}{\epsilon},\tag{3.39}$$

$$\hat{H}_{M\xi}=-\int d^3\mathbf{R}\;\hat{\mathbf{M}}'_{L\xi}\cdot\hat{\mathbf{B}}.\tag{3.40}$$

These are the forms of the electric and magnetic interaction energies that are familiar from free-space quantization, except of course that the reduced polarization density and the electric displacement and magnetic field intensities would have expansions involving the vector mode functions $\mathbf{A}_k(\mathbf{R})$. In this form the theory is manifestly gauge invariant.

In summary, the atomic Hamiltonian $\hat{H}_A$ describes the free evolution of noninteracting radiative atoms, each with its own Hamiltonian $\hat{H}_{A\xi}$ taking intra-atomic Coulomb and polarization energies into account; the field Hamiltonian $\hat{H}_F$ describes the free evolution of noninteracting true modes of the EM field, each with its own quantum harmonic oscillator Hamiltonian $(\hat{a}_k^\dagger \hat{a}_k + \frac{1}{2})\hbar\omega_k$, as in the photon model; the atom-field Hamiltonian $\hat{H}_{AF}$ describes the coupling of each atom separately with the quantum EM field, with a linear coupling for each atom with the electric field via $\hat{H}_{E\xi}$ and with the magnetic field via $\hat{H}_{M\xi}$—these are associated with one-photon processes, and with a quadratic diamagnetic coupling with the quantum EM field via $\hat{H}_{D\xi}$—this is associated with two-photon processes; the atom-atom interaction $\hat{H}_{AA}$ describes the direct coupling between pairs of atoms via the contact terms.

The interaction between different atoms in this cavity QED situation is predominantly via the quantum EM field and here mainly via one-photon processes associated with the leading term (dipole interaction) of the electric interaction $\hat{H}_{E\xi}$. Generally speaking, with well-separated atoms the contact terms can be ignored and in the usual situations the magnetic and diamagnetic interactions $\hat{H}_{M\xi}, \hat{H}_{D\xi}$ have smaller effects than $\hat{H}_{E\xi}$. This situation is the same as for free-field quantization in terms of the multipolar Hamiltonian [1].

H. Electric dipole approximation

To obtain the electric dipole approximation result we now ignore the magnetic interaction energy, the diamagnetic interaction energy, and the contact energy. The intra-atomic polarization energy can be retained. In addition as in Ref. [7], the polarization density $\hat{\mathbf{P}}_L(\mathbf{R})$ is given in its dipolar approximation,

$$\hat{\mathbf{P}}_L(\mathbf{R}) = \sum_{\xi} \hat{\boldsymbol{\mu}}_{\xi} \delta(\mathbf{R} - \mathbf{R}_{\xi}) \tag{3.41}$$

and as in Ref. [7] the reduced polarization density becomes

$$\hat{\mathbf{P}}'_L(\mathbf{R}) = \sum_{k\xi} \hat{\boldsymbol{\mu}}_{\xi} \cdot \mathbf{A}_k^*(\mathbf{R}_{\xi}) \epsilon(\mathbf{R}) \mathbf{A}_k(\mathbf{R}). \tag{3.42}$$

The quantum Hamiltonian $\hat{H}'_{E1}$ in the electric dipole approximation is then

$$\hat{H}'_{E1} = \hat{H}_A + \hat{H}_F + \hat{V}_{E1} \tag{3.43}$$

with [as in Ref. [7], Eq. (85)]

$$\hat{V}_{E1} = \sum_{k\xi} \left(\frac{\hbar\omega_k}{2}\right)^{1/2} \frac{1}{i} [\hat{a}_k \hat{\boldsymbol{\mu}}_{\xi} \cdot \mathbf{A}_k(\mathbf{R}_{\xi}) - \hat{a}_k^\dagger \hat{\boldsymbol{\mu}}_{\xi} \cdot \mathbf{A}_k^*(\mathbf{R}_{\xi})], \tag{3.44}$$

$$= \sum_{\xi} \frac{\hat{\boldsymbol{\mu}}_{\xi} \cdot \hat{\Pi}(\mathbf{R}_{\xi})}{\epsilon(\mathbf{R}_{\xi})} \tag{3.45}$$

$$= -\sum_{\xi} \frac{\hat{\boldsymbol{\mu}}_{\xi} \cdot \hat{\mathbf{D}}(\mathbf{R}_{\xi})}{\epsilon(\mathbf{R}_{\xi})}. \tag{3.46}$$

In view of Eq. (3.14) the electric dipole interaction can now also be expressed in terms of the electric displacement $\hat{\mathbf{D}}(\mathbf{R}_{\xi})$ at the radiative atom, as in the free-space quantization situation [1].

IV. SUMMARY

The key results may be summarized as follows. Previous work on macroscopic canonical quantization for quantum optics and cavity QED situations involving classical optical devices has been extended to show that (a) The Coulomb and polarization energies are equal to the sum of intra-atomic Coulomb energies, intra-atomic polarization energies, and interatomic contact energies. (b) The electric displacement is the negative of the conjugate momentum field. (c) The quantum Hamiltonian is now in a form in which the theory is manifestly gauge invariant. (d) The equal-time commutator of the vector potential and the conjugate momentum field are given in terms of a generalized transverse Dirac delta function.

This development depends on a generalization of Helmholtz theorem in which generalized transverse and longitudinal Dirac delta functions are introduced. This enables the relationship between the scalar potential ϕ and the polarization and reduced polarization densities $\mathbf{P}_L, \mathbf{P}'_L$ to be obtained.

The work described here and in Ref. [7] is restricted to the case where the permittivity function $\epsilon(\mathbf{R})$ is frequency independent (and real). Extension of the theory to deal with macroscopic canonical quantization in the multipolar form for quantum optics and cavity QED cases where dispersion (and absorption) are incorporated via a (complex) frequency-dependent permittivity function would be very worthwhile.

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