The Gauge Invariant Formulation for the Interaction of the Quantized Radiation Field with Matter

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It has been proved by the semi-classical gauge invariant formulation (GIF) that the correct interaction operator for coupling the field-free material states with the radiation field must be the *position form* regardless of the gauge chosen for expressing the electromagnetic potentials, in accordance with the well-established principle of gauge invariance. The semi-classical GIF is now extended to the quantized radiation field interacting with matter by defining the *energy operator* for the quantized radiation field in the presence of matter. It will be shown in this paper that the use of the energy operator guarantees the *position form* of the interaction operator even in the Coulomb gauge, contrary to the conventional approach in which the dark material Hamiltonian is used to get the interaction operator of the *momentum form*. The multipolar Hamiltonian is examined in the context of the quantum mechanical gauge transformation.

Introduction

The interaction of the radiation field with the matter provides important spectroscopic tools for probing the quantum mechanical structure of the matter. Theoretically, the interaction is described by an interaction operator for coupling the quantum mechanical material states with the radiation field. Within the electric-dipole approximation (EDA), many different forms of the interaction operator have been proposed for various reasons.[1] Among those operators, the *position form*, $\boldsymbol{E}(t)\cdot\boldsymbol{r}$, and the *momentum form*, $\boldsymbol{A}(t)\cdot\boldsymbol{p}$, are the best known forms of such interaction operators. The latter is often regarded as the more fundamental form since it appears to be resulted from more rigorous derivation employing fully quantized radiation field. The former is regarded as an approximation which is more practical in many calculations for various reasons.

In early 1950's, however, it was pointed out by Lamb that the different forms of the interaction operator are in fact related to the choice of gauge condition for the electromagnetic potentials to express the radiation field.[2] It was suggested that the position and momentum forms of the interaction operator correspond to the *Lamb gauge* and the *Coulomb gauge*, respectively.[3] Since the electromagnetic potentials in different gauges are related through a gauge transformation, these interaction operators must be equivalent according to *the principle of gauge invariance*.[1] Nevertheless, the controversy over the correct form of the interaction operator has been raised repeatedly. The apparent inequivalency among the different forms of the interaction operator has been pointed out in connection with the spectral line-shape and the transition probability involving nonlinear interactions, non-local potentials, spin-forbidden transitions, and many others.[3]

The controversy was finally resolved, at least within the semi-classical formulation, by the *gauge-invariant formulation* (GIF) proposed by Yang.[4] It was pointed out that the apparent difficulty arises from the use of the dark eigenstates, $\{|\phi_n^M\rangle\}$, of the field-free (dark) material Hamiltonian, $\boldsymbol{H}_M^0$, for expressing the material state, $|\Psi_M(t)\rangle$, in the presence of the radiation field. Since the phase of $|\Psi_M(t)\rangle$ changes with a gauge transformation, the expansion coefficients of $|\Psi_M(t)\rangle$ in terms of the (gauge independent) dark eigenstates, $\{|\phi_n^M\rangle\}$, inevitably become gauge dependent. The problem becomes more serious when the perturbation technique is used for finding the expression for the transition probability.

In order to overcome the difficulty, the *energy operator* for matter in the presence of the radiation, $\boldsymbol{H}_{MF}$, was introduced in the semi-classical gauge invariant formulation (GIF) proposed by Yang and others.[3,4] Within the EDA, $\boldsymbol{H}_{MF}$ is defined by a unitary transformation of $\boldsymbol{H}_M^0$ and it represents the instantaneous energy of the matter in the presence of the radiation field. Now, unlike the dark eigenstates, the phase of the eigenstates of $\boldsymbol{H}_{MF}$, $\{|\psi_n^M(t)\rangle\}$, also depends on the choice of the gauge by the same fashion as $|\Psi_M(t)\rangle$, thus making the expansion coefficients of $|\Psi_M(t)\rangle$ in terms of $\{|\psi_n^M(t)\rangle\}$ gauge invariant. It can also be shown that the rate of change of $\boldsymbol{H}_{MF}$ becomes the *power* in accordance with the classical Poynting's theorem.[5] The semi-classical GIF simply asserts the fact that the material energy eigenstates must acquire the phase factor, in the presence of the radiation field, which depends on the gauge chosen to express the radiation field.

According to the semi-classical GIF, *the interaction operator always takes on the position form at the EDA regardless of the gauge chosen for the radiation field.* The transition amplitudes defined in this fashion do not indeed depend on the gauge as they must be as a physically meaningful quantity according to the principle of gauge invariance. Although the semi-classical GIF results in the interaction operator which is incidentally identical to the form obtained from the conventional formulation with the Lamb gauge, it is important to emphasize that *the GIF does not prefer any particular gauge* for the radiation field.[6]

Although the semi-classical GIF has been successful in resolving the controversy over the form of the interaction operator, the radiation field, in addition to the material part, must also be quantized in order to develop the more rigorous description of the interaction between the radiation field and matter.[7] Such a fully quantized description becomes crucial for various nonlinear parametric processes including harmonic generations and wave mixings. Spontaneous emission step in these nonlinear parametric processes is conspicuously quantum mechanical in its nature.[8]

In this paper, the semi-classical GIF is extended to the fully quantized description of the interaction of the radiation field with the matter. The full Hamiltonian for the matter and the radiation will be examined in detail. Especially, it will be emphasized that the radiation part of the Hamiltonian must be expressed in terms of the *displacement vector*, $\boldsymbol{D}(t)$, instead of the field strength, $\boldsymbol{E}(t)$, in order to take into account the effect of the polarization in the matter in the presence of the radiation field.[5] Then, it will be shown that *the energy operator for the radiation in the presence of the matter*, $\boldsymbol{H}_{FE}$, can also be defined through a unitary transformation of the free field Hamiltonian as in the semi-classical GIF. The use of $\mathbf{H}_{FE}$ together with $\mathbf{H}_{ME}$ for the matter, automatically guarantees the physically meaningful gauge invariant transition amplitudes for the combined system of the matter and the radiation field.

However, it has to be pointed out that in the fully quantized description arbitrary gauge transformation is not possible since the quantization rule for the radiation field is uniquely determined by the gauge condition chosen.[9] Therefore, the radiation field expressed in the Coulomb gauge will be extensively examined in this paper since it is the most convenient gauge for quantization. Contrary to the conventional result, it will be shown that the interaction operator at the EDA still takes on the *position form*, $\boldsymbol{E}(t)\cdot\boldsymbol{r}$, rather than the momentum form, $\boldsymbol{A}(t)\cdot\boldsymbol{p}$, even though the radiation is expressed in the Coulomb gauge.

Finally, the multipolar Hamiltonian formulation proposed by Power *et al.*[10] will be discussed briefly in the context of the gauge transformation. The multipolar Hamiltonian, $\boldsymbol{H}_{MP}$, was obtained through a unitary transformation of the free-field Hamiltonian quantized in the Coulomb gauge. $\boldsymbol{H}_{MP}$ contains the interaction terms directly expressed as a multipole series which is evidently gauge invariant. For this reason, it was suggested that the multipolar part of $\boldsymbol{H}_{MP}$ corresponds to the gauge invariant interaction operator to be used for defining the physically meaningful transition amplitudes. It will be shown, however, in this paper that the unitary transformation leading to $\boldsymbol{H}_{MP}$ does not correspond to a gauge transformation and that the so-called "multipolar" gauge is not acceptable as a gauge condition for representing the electromagnetic radiation.

The semi-classical GIF is briefly reviewed in Section II. The field energy operator, $\boldsymbol{H}_{FE}$, is defined and examined in Section III. In Section IV, the interaction operator, when the radiation field is expressed in the Coulomb gauge, is derived from the fully quantized GIF. Discussion on the multipolar Hamiltonian is given in Section V.

Semi-Classical Gauge Invariant Formulation

The semi-classical gauge invariant formulation proposed first by Yang[4] is briefly summarized in this Section. The more details can be found elsewhere.[3,8]

In the semi-classical theory, the time-evolution of the material system in the presence of the radiation field is described by the time-dependent Schrödinger equation,

$$i\hbar\frac{\partial\,\Psi_M(t)}{\partial t} = \boldsymbol{H}_M(\boldsymbol{r},t)\;\Psi_M(t) \tag{1}$$

where $\boldsymbol{H}_M(\boldsymbol{r},t)$ is the minimally coupled Hamiltonian for the matter in the presence of the radiation field,

$$\boldsymbol{H}_M(\boldsymbol{r},t) = \frac{1}{2m}\left[\boldsymbol{p}-\frac{q}{c}\boldsymbol{A}(\boldsymbol{r},t)\right]^2 - V(\boldsymbol{r}) + qA_o(\boldsymbol{r},t) \tag{2}$$

Here, $\boldsymbol{A}(\boldsymbol{r},t)$ and $A_o(\boldsymbol{r},t)$ are respectively the vector and scalar potentials for the radiation field, and $V(\boldsymbol{r})$ is the static Coulomb potential. In this equation, the matter is denoted as a system of a single charged particle with charge q and mass m for the sake of notational simplicity.

According to the principle of gauge invariance, the electromagnetic radiation must be invariant under the following gauge transformation on the electromagnetic potentials, $\boldsymbol{A}(\boldsymbol{r},t)$ and $A_o(\boldsymbol{r},t)$,

$$\boldsymbol{A}'(\boldsymbol{r},t) = \boldsymbol{A}(\boldsymbol{r},t) + \Delta\Lambda(\boldsymbol{r},t) \tag{3a}$$

$$A_o'(\boldsymbol{r},t) = A_o(\boldsymbol{r},t) - \frac{1}{c}\,\frac{\partial\Lambda(r,t)}{\partial t} \tag{3b}$$

where $\Lambda(\boldsymbol{r},t)$ is an arbitrary gauge transformation function from the (unprimed) original to the (primed) new potentials, $\{\boldsymbol{A}'(\boldsymbol{r},t), A_o'(\boldsymbol{r},t)\}$. In other words, the principle of gauge invariance requires that all physically meaningful properties must be equally described by either set of the electromagnetic potentials, $\{\boldsymbol{A}(\boldsymbol{r},t), A_o(\boldsymbol{r},t)\}$ or $\{\boldsymbol{A}'(\boldsymbol{r},t), A_o'(\boldsymbol{r},t)\}$.

In quantum mechanics, the time-dependent Schrödinger equation in Eq. (1) is *form invariant* under any gauge transformation, since the Hamiltonian, $\boldsymbol{H}_M(\boldsymbol{r},t)$, and the wave function, $|\Psi_M(t)\rangle$, are transformed respectively as following.[11]

$$\boldsymbol{H}_M'(\boldsymbol{r},t) = \boldsymbol{U}\boldsymbol{H}_M(\boldsymbol{r},t)\boldsymbol{U}^{\dagger} + i\hbar\frac{\partial\boldsymbol{U}}{\partial t}\boldsymbol{U}^{\dagger} \tag{4a}$$

$$|\Psi_M'(t)\rangle = \boldsymbol{U}|\Psi_M(t)\rangle \tag{4b}$$

where

$$\boldsymbol{U} = \exp\left[\frac{iq}{c\hbar}\Lambda(\boldsymbol{r},t)\right] \tag{4c}$$

Here, $\boldsymbol{H}_M'(\boldsymbol{r},t)$ and $|\Psi_M'(t)\rangle$ are the minimally coupled Hamiltonian and the wave function expressed in the new (primed) gauge, respectively. It is noted here that the minimally coupled Hamiltonian in the new (primed) gauge is not given as a simple unitary transformation of $\boldsymbol{H}_M(\boldsymbol{r},t)$ in the old (unprimed) gauge. Instead, there appears the additional term involving the time derivative of the transformation function, which guarantees the form invariance of the

Schrödinger equation. It is thus important to emphasize the fact that *the expectation value of the minimally coupled Hamiltonian which contains a time-dependent potential is not invariant under a gauge transformation*. The minimally coupled Hamiltonian controls the time-evolution of the material system interacting with the radiation through the form-invariant time-dependent Schrödinger equation given by Eq. (1).

The controversy over the form of the interaction operator arises from the phase factor of the wave function which is changed with a gauge transformation as shown in Eq. (4b). Unlike $|\Psi_M(t)\rangle$, the eigenfunctions, $\{|\phi_n^M\rangle\}$, of the field-free dark Hamiltonian, $\boldsymbol{H}_M^o(\boldsymbol{r}, t)$, are entirely independent of the choice of the gauge since there is no radiation field involved in $\boldsymbol{H}_M^o(\boldsymbol{r},t)$. Therefore, the expansion coefficients of the gauge dependent $|\Psi_M(t)\rangle$ in terms of the gauge independent $\{|\phi_n^M\rangle\}$ inevitably depend on the gauge chosen for $|\Psi_M(t)\rangle$. It is for this reason that the expansion coefficients defined in the conventional formulation cannot be accepted as physically meaningful *transition probability amplitudes* which must be invariant under any gauge transformation.

In the semi-classical gauge invariant formulation (GIF), the following basis defining operator, $\boldsymbol{H}_{ME}(\boldsymbol{r}, t)$, is employed

$$\boldsymbol{H}_{ME}(\boldsymbol{r},t)=\boldsymbol{H}_M(\boldsymbol{r},t)-qA_o(\boldsymbol{r},t)$$
$$=\frac{1}{2m}\left[\boldsymbol{p}-\frac{q}{c}\boldsymbol{A}(\boldsymbol{r},t)\right]^2+V(\boldsymbol{r})-qA_o(\boldsymbol{r},t) \quad (5)$$

The two operators, $\boldsymbol{H}_{ME}(\boldsymbol{r}, t)$ in the old (unprimed) gauge and $\boldsymbol{H}_{ME}'$ in the new (primed) gauge, are related by a simple unitary transformation,

$$\boldsymbol{H}_{ME}'=\boldsymbol{U}\,\boldsymbol{H}_{ME}\boldsymbol{U}^{+} \quad (6)$$

And, the eigenfunction, $|\psi_n^M(t)\rangle$, of $\boldsymbol{H}_{ME}(\boldsymbol{r}, t)$ satisfying the following equation,

$$\boldsymbol{H}_{ME}(\boldsymbol{r},t)\,|\psi_n^M(t)\rangle=E_n^M(t)|\psi_n^M(t)\rangle \quad (7)$$

is related to the eigenfunction $|\psi_n^{M'}(t)\rangle$ of $\boldsymbol{H}_{ME}'(\boldsymbol{r}, t)$ by the following relation,

$$|\psi_n^{M'}(t)\rangle=\boldsymbol{U}|\psi_n^M(t)\rangle \quad (8)$$

Here, it can be readily seen that upon a gauge transformation $|\psi_n^M(t)\rangle$ acquires the same additional phase factor as $|\Psi_M(t)\rangle$ as shown in Eq. (4b). Now, $|\Psi_M(t)\rangle$ can be expressed in terms of $\{|\psi_n^M(t)\rangle\}$, instead of the eigenstates of the field-free dark Hamiltonian, $\{|\psi_n^M(t)\rangle\}$, as following

$$|\Psi_M(t)\rangle=\sum_n a_n^M(t)\,|\psi_n^M(t)\rangle \quad (9)$$

The gauge invariance of the expansion coefficients, $\{a_n^M(t)\}$, is evident since both $|\Psi_M(t)\rangle$ and $|\psi_n^M(t)\rangle$ carry the same phase factor, as shown in Eqs. (4b) and (8), upon a gauge transformation. Furthermore, the following relation can be readily derived,

$$\frac{d}{dt}\langle\Psi_M(t)|\boldsymbol{H}_{ME}(\boldsymbol{r},t)|\Psi_M(t)\rangle=\langle\Psi_M(t)|\boldsymbol{P}(t)|\Psi_M(t)\rangle \quad (10)$$

where $\boldsymbol{P}(t)$ is the power operator given as

$$\boldsymbol{P}(t)=\frac{1}{2}[\boldsymbol{v}(t)\cdot q\boldsymbol{E}(\boldsymbol{r},t)+q\boldsymbol{E}(\boldsymbol{r},t)\cdot\boldsymbol{v}(t)] \quad (11)$$

Here, $\boldsymbol{v}(t) = \boldsymbol{p} - (q/c)\,\boldsymbol{A}(\boldsymbol{r},t)$ is the velocity operator. Then, Eq. (10) corresponds to the *classical Poynting's theorem*, by which the rate of energy change is given by the power of the system, if $\boldsymbol{H}_{ME}(\boldsymbol{r},t)$ is regarded as the *energy operator* representing the *instantaneous energy of the material system in the presence of the radiation field*. Furthermore, $\boldsymbol{H}_{ME}(\boldsymbol{r},t)$ is reduced to the field-free dark Hamiltonian, $\boldsymbol{H}_M^o(\boldsymbol{r},t)$, in the absence of the field, $\boldsymbol{A}(\boldsymbol{r},t) = 0$. Now, the gauge invariant expansion coefficients, $\{a_n^M(t)\}$, defined in this fashion can be regarded as the *transition probability amplitudes between the energy eigenstates of the matter* regardless of a gauge chosen for the radiation field.

Furthermore, at the EDA, it can be shown that the energy operator in Eq. (5) is expressed as a unitary transformation of $\boldsymbol{H}_M^o(\boldsymbol{r}, t)$,

$$\boldsymbol{H}_{ME}(\boldsymbol{r},t)=\boldsymbol{U}_1\,\boldsymbol{H}_M^o\,\boldsymbol{U}_1^{+} \quad (12a)$$

and that an eigenstate of the energy operator is related to the corresponding dark eigenstate according to

$$E_n^M=\varepsilon_n^M \quad (12b)$$

$$|\psi_n^M(t)\rangle=\boldsymbol{U}_1(t)|\phi_n^M\rangle \quad (12c)$$

where ε_n^M is the eigenvalue of the dark Hamiltonian. Here,

$$U_1=\exp\left[\frac{iq}{c\hbar}\boldsymbol{r}\cdot\boldsymbol{A}(t)\right] \quad (12d)$$

Now, the equation of motion for $\{a_n^M(t)\}$ at the EDA can be written as

$$i\hbar\frac{\partial a_n^M(t)}{\partial t}=\varepsilon_n^M a_n^M(t)+\sum_m a_m^M(t)\langle\phi_n^M|-q\boldsymbol{r}\cdot\boldsymbol{E}(t)|\phi_m^M\rangle \quad (13)$$

The interaction operator is clearly in the position form. It is emphasized that Eq. (13) is valid no matter which gauge is chosen to expressed the radiation field.

According to the semi-classical GIF briefly summarized in this Section, the interaction operator for coupling the field-free dark eigenstates always takes on the position form regardless of the choice of the gauge for the radiation field. The position form obtained in this approach happens to coincide with the interaction operator obtained from the conventional formulation with the radiation field expressed in the Lamb gauge in which $\boldsymbol{A}(t) \approx 0$ and $A_o(t) \approx -q\boldsymbol{r}\cdot\boldsymbol{E}(t)$ at the EDA. In the semi-classical GIF, however, *no gauge condition is assumed at all* at the beginning and Eq. (13) is *valid for any gauge chosen*.

Energy Operator for the Radiation Field

In this Section, the Hamiltonian for the free radiation field in the absence of the matter is examined first and then the full Hamiltonian for the combined system of the matter and the radiation field is briefly discussed. The *energy operator* *for the field in the presence of the matter* is defined at the EDA.

Free Radiation Field. The Hamiltonian for the radiation field in the absence of the matter is given by[9]

$$\boldsymbol{H}_F^o(\boldsymbol{r},t) = \frac{1}{2}\{|\boldsymbol{E}(t)|^2 - |\boldsymbol{H}(t)|^2\} \tag{14}$$

When the Coulomb gauge ($\nabla \cdot \boldsymbol{A} = 0$ and $A_o = 0$) is chosen, the vector potential $\boldsymbol{A}(\boldsymbol{r}, t)$ can be written in the quantized form,

$$\boldsymbol{A}(r,t) = N\hat{e}\{\boldsymbol{a}e^{i(k\cdot r - \omega t)} + \boldsymbol{a}^{+}e^{-i(k\cdot r + \omega t)}\} \tag{15}$$

where $\boldsymbol{a}$ and $\boldsymbol{a}^{+}$ are the *creation* and *annihilation* operators of the radiation field, respectively, satisfying the commutation relation,

$$[a, a^{+}] = 1 \tag{16}$$

In Eq. (15), $\hat{e}$, $\boldsymbol{k}$ and ω are the polarization unit vector, the wave vector, and the frequency of the radiation respectively, and $N = [\hbar c/2\omega V]^{1/2}$ is the normalization constant. Here, the radiation field is treated as completely monochromatic for the sake of notational simplicity. For the polychromatic radiation field, the vector potential in Eq. (15) must contain all the frequency and polarization components. With the vector potential given in Eq. (15), $\boldsymbol{E}(\boldsymbol{r},t)$ and $\boldsymbol{B}(\boldsymbol{r},t)$ in Eq. (14) are given as following,

$$\boldsymbol{E}(\boldsymbol{r},t) = \frac{i\omega N}{c}\hat{e}\{\boldsymbol{a}e^{i(k\cdot r - \omega t)} - \boldsymbol{a}^{+}e^{-i(k\cdot r - \omega t)}\} \tag{17a}$$

$$\boldsymbol{B}(\boldsymbol{r},t) = iN(\boldsymbol{k}\times\hat{\boldsymbol{e}})\{\boldsymbol{a}e^{i(k\cdot r - \omega t)} - \boldsymbol{a}^{\dagger}e^{-i(k\cdot r + \omega t)}\} \tag{17b}$$

Then, the free-field Hamiltonian in Eq. (14) can be expressed in the quantized form,

$$\boldsymbol{H}_F^o = \hbar\omega\left(\boldsymbol{a}\,\boldsymbol{a}^{+} + \frac{1}{2}\right) \tag{18}$$

Again, the right-hand side of the Eq. (18) must contain the summation over all the frequency and polarization components in the polychromatic radiation field. The Hamiltonian in the quantized form in Eq. (18) is unique for the Coulomb gauge. It is not possible to obtain the Hamiltonian quantized in any other gauge by direct transformation of the above Hamiltonian expressed in the Coulomb gauge since the quantization rule is completely different for every gauge condition chosen.

Full Hamiltonian for the Matter and the Radiation. The matter consisted of charged particles is polarized in the presence of the radiation field. Along with the external radiation field, the resulting polarization of the matter also becomes a source of the field acting on the charged particles in the matter. The overall field can be expressed in terms of the *displacement vector* (or *electric induction*), $\boldsymbol{D}(\boldsymbol{r},t)$, and the magnetic induction, $\boldsymbol{B}(\boldsymbol{r},t)$[5]

$$\boldsymbol{D}(\boldsymbol{r},t) = \boldsymbol{E}(\boldsymbol{r},t) - 4\pi\boldsymbol{P}(\boldsymbol{r},t) \tag{19a}$$

$$\boldsymbol{B}(\boldsymbol{r},t) = \boldsymbol{H}(\boldsymbol{r},t) + 4\pi\boldsymbol{M}(\boldsymbol{r},t) \tag{19b}$$

where $\boldsymbol{P}(\boldsymbol{r},t)$ and $\boldsymbol{M}(\boldsymbol{r},t)$ are respectively the electric polarization and the magnetization arising from the polarizable matter in the presence of the external radiation field.

Now the Hamiltonian for the field in the presence of the polarizable matter becomes[7]

$$\boldsymbol{H}_F(\boldsymbol{r},t) = \frac{1}{4\pi}\int\{\boldsymbol{E}(\boldsymbol{r},t)\cdot\boldsymbol{D}(\boldsymbol{r},t) - \boldsymbol{H}(\boldsymbol{r},t)\cdot\boldsymbol{B}(\boldsymbol{r},t)\}dV \tag{20}$$

and the full Hamiltonian for the matter and the radiation field is given as

$$\boldsymbol{H} = \boldsymbol{H}_M(\boldsymbol{r},t) + \boldsymbol{H}_F(\boldsymbol{r},t) \tag{21}$$

where $\boldsymbol{H}_M(\boldsymbol{r},t)$ is the minimally coupled Hamiltonian, given in Eq. (2), for the matter in the presence of the radiation field. The use of $\boldsymbol{D}(\boldsymbol{r},t)$ and $\boldsymbol{B}(\boldsymbol{r},t)$ for the energy of the radiation field in the presence of the polarizable matter is well established in classical theory on electromagnetism. In the quantum mechanical description of light-matter interaction, however, the polarization effect is often neglected in $\boldsymbol{H}_F(\boldsymbol{r},t)$ probably due to the fact that $\boldsymbol{D}(\boldsymbol{r},t)$ and $\boldsymbol{B}(\boldsymbol{r},t)$ are not easy to express in closed quantized forms. Instead, $\boldsymbol{H}_F(\boldsymbol{r},t)$ is often replaced by the Hamiltonian for the free field, $\boldsymbol{H}_F^o$, in Eq. (18). Then the interaction appears only in the material part of the Hamiltonian, $\boldsymbol{H}_M(\boldsymbol{r},t)$.

At the EDA, the electric polarization can be written as following,

$$\boldsymbol{P}(\boldsymbol{r},t) = q\boldsymbol{r}\cdot\boldsymbol{E}(t) \tag{22}$$

and the magnetization can often be ignored. Therefore, $\boldsymbol{H}_F(\boldsymbol{r},t)$ in Eq. (20) can be written as

$$\boldsymbol{H}_F(\boldsymbol{r},t) = \boldsymbol{H}_F^o + q\boldsymbol{r}\cdot\boldsymbol{E}(t) \tag{23}$$

at the EDA.

Energy Operator at the Electric Dipole Approximation. In the semiclassical GIF, the energy operator, $\boldsymbol{H}_{ME}$, for the matter in the presence of the radiation field is given as the unitary transformation of the dark Hamiltonian, $\boldsymbol{H}_o$, as shown in Eq. (12a). It will be now shown that the energy operator, $\boldsymbol{H}_{FE}$, for the field in the presence of the matter can be defined through the same unitary transformation of the free-field Hamiltonian, $\boldsymbol{H}_F^o$, in Eq. (18).

When two quantum mechanical operators $\boldsymbol{Q}$ and $\boldsymbol{S}$ do not commute with each other, it can be readily shown that the unitary transformation of $\boldsymbol{Q}$ with $\boldsymbol{U}_1 = \exp[i\boldsymbol{S}]$ becomes

$$\boldsymbol{U}_1\boldsymbol{Q}\boldsymbol{U}_1^{+} = \boldsymbol{Q} + [i\boldsymbol{S}, \boldsymbol{Q}] + \frac{1}{2!}[i\boldsymbol{S}, [i\boldsymbol{S}, \boldsymbol{Q}]] + \frac{1}{3!}[i\boldsymbol{S}, [i\boldsymbol{S}, [i\boldsymbol{S}, \boldsymbol{Q}]]] - \cdots \tag{24}$$

When $\boldsymbol{U}_1 = \exp[(iq/c\hbar)\boldsymbol{r}\cdot\boldsymbol{A}(t)]$ (*i.e.* $\boldsymbol{S} = (iq/c\hbar)\boldsymbol{r}\cdot\boldsymbol{A}(t)$), which corresponds to the transformation function in Eq. (12d) at the EDA, the commutation relation in Eq. (16) can be used to derive the following relations.

$$[i\boldsymbol{S}, \boldsymbol{p}] = \frac{iq}{c\hbar}A(t) \tag{25a}$$

$$[i\boldsymbol{S}, \boldsymbol{a}] = -\frac{iq}{c\hbar}(\boldsymbol{r}\cdot\hat{e})e^{i\omega t} \tag{25b}$$

$$[i\boldsymbol{S},\ \boldsymbol{a}^{\dagger}]=\frac{iq}{c\hbar}\ (\boldsymbol{r}\cdot\hat{e})e^{-i\omega t} \qquad (25c)$$

Since $[i\boldsymbol{S}, [\cdots [i\boldsymbol{S}, \boldsymbol{a}]]] = [i\boldsymbol{S}, [\cdots [i\boldsymbol{S}, \boldsymbol{a}^{\dagger}]]] = 0$ and $[i\boldsymbol{S}, [\cdots [i\boldsymbol{S}, \boldsymbol{p}]]] = 0$.

Thus, it can be readily confirmed that the relation in Eq. (12a) is still valid when the vector potential in U_1 is expressed in the quantized form in Eq. (15). This means that the energy operator for the matter in the presence of the radiation field as defined by Eq. (5) is related to the field-free dark material Hamiltonian by the unitary transformation in Eq. (12a) even if the radiation field is quantized in the Coulomb gauge. Thus, the eigenstates of the energy operator retains the same phase factor as in the semi-classical approach.

Now, it is interesting to find the unitary transformation of $\boldsymbol{H}_F^o$ with U_1, which may be regarded as the *energy operator*, $\boldsymbol{H}_{FE}$, for the *radiation field in the presence of the matter* in analogy with the energy operator, H_{ME}, for the matter in the presence of the radiation field given by Eq. (5). By writing $U_1 a^{\dagger} a\ U_1^{\dagger} = (U_1 a^{\dagger}\ U_1^{\dagger})\ (U_1 a U_1^{\dagger})$ along with Eqs. (25a) and (25b), one can easily obtain the following result.

$$\boldsymbol{H}_{FE}=U_1\boldsymbol{H}_F^o U_1^{\dagger}=\boldsymbol{H}_F^o+q\boldsymbol{r}\cdot\boldsymbol{E}(t)-\left(\frac{q^2\omega}{c^2\hbar}\right)(\boldsymbol{r}\cdot\hat{e})^2 \qquad (26)$$

Here, Eq. (17a) has been used for $\boldsymbol{E}(t)$ at the electric dipole approximation.

Now, the following relations can be readily verified.

$$E_n^F=\varepsilon_n^F \qquad (27a)$$

$$|\psi_n^F(t)\rangle=\boldsymbol{U}_1(t)|\phi_n^F\rangle \qquad (27b)$$

where $\{E_n^F\}$ and $\{|\psi_n^F(t)\rangle\}$ are the eigenvalues and eigenfunctions of $\boldsymbol{H}_{FE}$ while $\{\varepsilon_n^F\}$ and $\{|\phi_n^F\rangle\}$ are the eigenvalues and eigenfucntions of $\boldsymbol{H}_o^F$, the Hamiltonian for the free field in the absence of the matter. The above relations are reminiscent of the relations for the material energy operator in the presence of the radiation field as given in Eqs. (12b) and (12c). That is, the eigenvalues of $\boldsymbol{H}_{FE}$ and $\boldsymbol{H}_o^F$ are the same, but the eigenfunctions differ by a phase factor. Thus, in view of the successful role of $\boldsymbol{H}_{ME}$ as the basis defining Hamiltonian for achieving the gauge invariant transition probability amplitude in the semi-classical approach, one can expect the similar role of $\boldsymbol{H}_{FE}$ in the fully quantized description of the radiation field interacting with the matter.

Interaction Operator

It is now possible to derive the interaction operator for coupling the quantized radiation field with the quantum mechanical matter states by using $\boldsymbol{H}_{FE}$ in Eq. (26) as the basis defining Hamiltonian.

The total Hamiltonian for the combined system of the matter and the radiation field expressed in the Coulomb gauge at the EDA can be written as following.

$$\boldsymbol{H}=\boldsymbol{H}_M+\boldsymbol{H}_F=\boldsymbol{H}_{ME}+\boldsymbol{H}_{FE}-\left(\frac{q^2\omega}{c^2\hbar}\right)(\boldsymbol{r}\cdot\hat{e})^2 \qquad (28)$$

$\boldsymbol{H}_M$, $\boldsymbol{H}_F$, $\boldsymbol{H}_{ME}$ and $\boldsymbol{H}_{FE}$ are given by Eqs. (2), (24), (5), (26), respectively. The last term in the above equation is added in order to cancel out the extra term appeared in the unitary transformation of $\boldsymbol{H}_o^F$ in Eq. (26).

The time evolution of the combined system is described by the time-dependent Schrödinger equation.

$$i\hbar\frac{\partial|\Psi(t)\rangle}{\partial t}=H|\Psi(t)\rangle \qquad (29)$$

where $|\Psi(t)\rangle$ represents the quantum mechanical state of the combined system. It is now possible to express $|\Psi(t)\rangle$ in terms of the direct product of the eigenstates of $\boldsymbol{H}_{ME}$ and $\boldsymbol{H}_{FE}$. That is,

$$|\Psi(t)\rangle=\sum_{n,m}a_{n,m}(t)\ |\psi_n^M(t)\rangle\ |\psi_m^F(t)\rangle \qquad (30)$$

where the indices n and m represents the eigenstates of the matter and the radiation field, respectively. Then, the equation of motion for $\{a_{n,m}(t)\}$ is given by

$$i\hbar\frac{\partial a_{n,m}(t)}{\partial t}=(\varepsilon_n^M+\varepsilon_m^F)a_{n,m}(t)$$
$$+\sum_n\sum_m a_{n,m}(t)$$
$$\times\langle\psi_n^M\psi_m^F|\left[-i\hbar\frac{\partial}{\partial t}-\left(\frac{q^2\omega}{c^2\hbar}\right)(\boldsymbol{r}\cdot\hat{e})^2\right]|\psi_m^F\psi_n^M\rangle \qquad (31)$$

As explained above, the eigenstates of $\boldsymbol{H}_{ME}$ and $\boldsymbol{H}_{FE}$ are related to the eigenstates of $\boldsymbol{H}_o^M$ and $\boldsymbol{H}_o^F$ by the same unitary transformation with U_1 given by Eq. (12d). Thus, the above equation can be rewritten as following.

$$i\hbar\frac{\partial a_{n,m}(t)}{\partial t}=(\varepsilon_n^M+\varepsilon_m^F)a_{n,m}(t)$$
$$+\sum_n\sum_m a_{n,m}(t)$$
$$\times\langle\psi_n^M\psi_m^F|\ U_1\left[-i\hbar\frac{\partial}{\partial t}-\left(\frac{q^2\omega}{c^2\hbar}\right)(\boldsymbol{r}\cdot\hat{e})^2\right]U_1^{\dagger}\ |\psi_m^F\psi_n^M\rangle \qquad (32)$$

Since

$$U_1\frac{\partial}{\partial t}U_1^{\dagger}=-iT-\frac{1}{2!}[iS,\ iT]-\frac{1}{3!}[iS,\ [iS,\ iT]]-\cdots \qquad (33)$$

where $T=\frac{\partial S}{\partial t}$ and $S=(iq/c\hbar)\boldsymbol{r}\cdot\boldsymbol{A}(t)$, it can be proven that

$$i\hbar U_1\frac{\partial}{\partial t}U_1^{\dagger}=q\boldsymbol{r}\cdot\boldsymbol{E}(t)-\left(\frac{q^2\omega}{c^2\hbar}\right)(\boldsymbol{r}\cdot\hat{e})^2 \qquad (34)$$

So. Eq. (32) becomes

$$i\hbar\frac{\partial a_{n,m}(t)}{\partial t}=(\varepsilon_n^M\cdot\varepsilon_m^F)a_{n,m}(t)$$
$$+\sum_n\sum_m a_{n,m'}(t)\cdot\phi_n^M\phi_m^F|-q\boldsymbol{r}\cdot\boldsymbol{E}(t)|\phi_m^F\phi_n^M \qquad (35)$$

in which the interaction operator is clearly the *position* form, not the momentum form, *even though the Coulomb gauge has been employed* from the very beginning. Although the position form of the interaction operator is not at all conspicuous at the Hamiltonian level given in Eq. (28), the position form of the interaction operator is nevertheless recovered at last mainly from the time derivatives of the time-dependent phase factors of the energy eigenstates.

Multipolar Hamiltonian

In late 1970's, Power *et al. proposed the so-called the multipolar Hamiltonian*, $\boldsymbol{H}_{MP}$, in which the interaction operators of the form of the classical multipolar series are clearly exposed at the Hamiltonian level.[10-12] They suggested that the multipolar Hamiltonian corresponds to the minimally coupled Hamiltonian expressed in the "multipolar gauge" as they called it. They obtained the multipolar gauge by the following unitary transformation of the minimally coupled Hamiltonian given in Eq. (2).

$$\boldsymbol{H}_{MP}(\boldsymbol{r},t)\quad \boldsymbol{U}\ \boldsymbol{H}_M(\boldsymbol{r},t)\boldsymbol{U}^{+}$$
$$=\boldsymbol{H}_o-\mu\cdot\boldsymbol{E}(t)-\boldsymbol{M}(\boldsymbol{r})\cdot\boldsymbol{B}(\boldsymbol{r}) \qquad (36)$$

with

$$\boldsymbol{U}=\exp\left[\frac{iq}{c\hbar}\boldsymbol{r}\cdot\boldsymbol{A}(\boldsymbol{r},t)\right] \qquad (37)$$

Here, the quantization of the radiation field is expressed in the Coulomb gauge signified by the commutation relation given in Eq. (16). They claimed that the unitary transformation in Eq. (36) corresponds to the gauge transformation from the Coulomb gauge to the so-called multipolar gauge.

There are two serious difficulties in accepting the multipolar Hamiltonian as a physically meaningful operator. First of all, as clearly indicated in Eq. (37), the transformation function used to get $\boldsymbol{H}_{MP}$ from $\boldsymbol{H}_M$ is time-dependent. Because of the time-dependent nature of the transformation, the time-evolution of the wavefunction in the transformed representation now have to be given by the following equation.

$$i\hbar\frac{\partial|\Psi_{MP}(t)}{\partial t}\quad \boldsymbol{H}_{MP}(\boldsymbol{r},t)\ \Psi_{MP}(t)\quad - i\hbar\frac{\partial\boldsymbol{U}^{+}}{\partial t}\ \Psi_{MP}(t) \qquad (38)$$

The last term does not automatically vanish even at the EDA because the time derivative of the vector potential in the Coulomb gauge must be directly related to the electric field strength which cannot vanish. Thus, the wavefunction in the transformed representation does not satisfy the usual time-dependent Schrödinger equation. In other words, it is no longer possible to use the usual time-dependent Schrödinger equation once $\boldsymbol{H}_M$ is tranformed into $\boldsymbol{H}_{MP}$ by the time-dependent transformation function.

The other difficulty with the multipolar gauge is that the vector and scalar potentials implied from $\boldsymbol{H}_{MP}$ does not really corresponds to the acceptable legitimate gauge condition. According to Power *et al.*,[10] the multipolar gauge is identified by $\boldsymbol{A}(\boldsymbol{r},t)$ = 0 and qA_o being expressed by the multipole series. The vanishing vector potential implies $\boldsymbol{B}(\boldsymbol{r},t)$ = $\nabla\times\boldsymbol{A}(\boldsymbol{r},t)$ = 0 for the radiation field, which is clearly not acceptable as realistic description of the radiation field beyond the EDA. It is a quite different matter for the Lamb gauge mentioned above in which $\boldsymbol{A}(\boldsymbol{r},t)$ is also null, since the Lamb gauge is defined in a limited sense of the EDA.

Concluding Remarks

The quantum mechanical description of the interaction of radiation field with matter is examined from the point of view of the gauge invariance. The full Hamiltonian for the combined system of the radiation field with the matter has been closely examined with the quantized radiation field. It is pointed out that in the presence of the matter consisted of charged particles the displacement vector which includes the polarization effect due to the charged particles must be used. The resulting Hamiltonian expressed in the Coulomb gauge at the electric dipole approximation (EDA) is presented. Then, the basis defining energy operator for the radiation field in the presence of the matter is defined by the unitary transformation of the free field Hamiltonian, analogous to the energy operator for matter in the presence of radiation field. The use of such gauge-dependent basis defining function guarantees the gauge invariance of the time-evolution of the combined system of the radiation field in the presence of the matter as in the semi-classical description. It has been proved that the interaction operator for coupling the dark eigenstates of the free material eigenstates and of the free field eigenstates is given by the position form at the EDA even if the Coulomb gauge is employed for the radiation field. In the conventional formulation using the free material and free field eigenstates, the Coulomb gauge is believed to result in the momentum form of the interaction operator.

It has also been shown that the multipolar Hamiltonian proposed by Power *et al.* has serious theoretical difficulties. Since the transformation of the minimally coupled Hamiltonian in the Coulomb gauge to the multipolar Hamiltonian is inherently time-dependent due to the vector potential involved, an additional term must be added to the time-dependent Schrödinger equation in order to maintain the validity of the description of the time-evolution of the system. Furthermore, it is also pointed out that the so-called multipolar gauge which is believed to be the gauge condition corresponding to the multipolar Hamiltonian cannot be accepted as a legitimate gauge condition for the radiation field.

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