Time evolution of Heisenberg operators of nuclei and electrons of QED system based on field theory

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Abstract. The present status of the development of our code, QEDynamics, is reported. In this code, the time evolution simulation is carried out for Heisenberg operators based on quantum electrodynamics in a manner of quantum field theory. For this simulation, the treatment of the photon is important, particularly infrared and ultra-violet photons. As a result, renormalization is one of essential ingredients, since quantum field theory is adopted.

1. Introduction
For the description of the nature, QED (quantum electrodynamics) is the most celebrated theory. For example, the prediction of the anomalous magnetic dipole moment of electron by perturbative calculations shows extreme accordance with experimental observations [1, 2, 3]. Although the standard model of particle physics is now established completely by the discovery of a Higgs-like particle, the precision of this model is not so improved compared with QED due to the uncertainties of parameters. Hence, QED is suitable enough for the precise description of the interaction between photon and matter.

QED is extremely successful in perturbative calculations for free particles. The application to bound states is intensely studied, while the sophisticated calculation method has not been established due to its nonperturbativity. QED is based on quantum field theory, where all phenomena are essentially described by field operators. In computational simulations, operators are difficult to handle. The exact simulation of field theory is performed by lattice calculation, such as lattice QCD. However, QED is not suitable for the lattice computation, since the way to take the continuum limit has not been established. Hence, the computation of QED bound states has been developed as approximation in most studies. Some treatments, Bethe-Salpeter formalism or NRQED [4], are frequently discussed. In quantum chemistry, the QED correction to Hamiltonian for ab initio calculation is often used [5]. In most studies, the photon is treated as potential, and however this is not suitable for quantum system studies, for example systems with single photon.

In this work, we propose a new computation method of the time evolution of quantum systems based on QED and report the present status of the development of our program code, QEDynamics [6]. In our method, field operators are treated in Heisenberg picture. The evolution of the operators is calculated as the transition between modes prepared by the method of quantum chemistry. Since the aim of our work is the study of bound states, such as molecules,
nuclei as well as electrons are treated as quantum fields. The QED including nuclei is formulated with the gauge invariance as Rigged QED [7, 8, 9].

2. Theory and computational details
In this paper, electron fields are treated as two-component Schrödinger fields. This treatment is called as primary Rigged QED [10]. Since the four-component relativistic treatment requires much larger computation resources, this approximation supports longer time evolution calculations and larger system calculations. It is noted that even a two-component treatment requires much larger computation resources, this approximation supports longer time evolution is called as primary Rigged QED [10]. Since the four-component relativistic treatment is called as primary Rigged QED [10]. Since the four-component relativistic treatment requires much larger computation resources, this approximation supports longer time evolution calculations and larger system calculations. It is noted that even a two-component treatment requires much larger computation resources, this approximation supports longer time evolution is already included partially in (3)-(7). Further effects of the time evolution of operators

The electron and nucleus fields are expanded as

\[ \hat{\Psi}(t, \vec{x}) = \sum_m \hat{e}_m(t) \psi_m(\vec{x}), \quad (1) \]

\[ \hat{\chi}_a(t, \vec{x}) = \sum_m \hat{f}_am(t) \chi_am(\vec{x}), \quad (2) \]

where \( \hat{e}_m(\hat{f}_am) \) is the annihilation operator of the electron (nucleus) and \( \psi_m(\chi_am) \) is its expansion function. The photon field is given by [7, 8, 9]

\[ A^\mu(t, \vec{x}) = \hat{A}^\mu_{rd}(t, \vec{x}) + \hat{A}^\mu_A(t, \vec{x}) + \hat{A}^\mu_M(t, \vec{x}). \quad (3) \]

Here radiation photon, \( \hat{A}^\mu_{rd} \), is derived by the quantization for a free field, while \( \hat{A}^\mu_A \) and \( \hat{A}^\mu_M \) are photons produced by dynamical effects of the system and environment, respectively. These terms are particularly important for the description of infrared photons.

The definition of \( \hat{A}^\mu_{rd} \) is given by

\[ \hat{A}^\mu_{rd}(t, \vec{x}) = \sqrt{\frac{e}{\pi \sqrt{2\hbar}}} \sum_{k, \sigma} \int \frac{d^3k}{\sqrt{2k_0}} \hat{A}^\mu(t, \vec{x}, \vec{k}, \sigma), \quad (4) \]

\[ \hat{A}^\mu_A(t, \vec{x}, \vec{k}, \sigma) = \hat{a}(\vec{k}, \sigma) e^{\mu}(\vec{k}, \sigma) \exp[-ik_x x^x/\hbar] + \hat{a}^\dagger(\vec{k}, \sigma) e^{\mu}(\vec{k}, \sigma) \exp[ik_x x^x/\hbar], \quad (5) \]

where \( \hat{a}^\dagger \) and \( \hat{a} \) are the creation and annihilation operators of the photon, and \( \epsilon \) is its polarization. The integration in this definition is reduced to a summation for the implementation of this computation.

On the other hand, \( \hat{A}^\mu_{A,M} \) is given with creation-annihilation operators of matter fields. These photons are divided into two parts, scalar one, \( \hat{A}^\mu_{A,M} \), and vector one, \( \hat{A}^\mu_{A,M} \). The scalar part is given by

\[ \hat{A}^\mu_{A,M}(t, \vec{x}) = Z_e e \int d^3s \frac{\hat{\Psi}^\dagger(t, \vec{s}) \hat{\Psi}(t, \vec{s})}{|\vec{x} - \vec{s}|} + \sum_a Z_a e \int d^3s \frac{\hat{\chi}^a(t, \vec{s}) \hat{\chi}_a(t, \vec{s})}{|\vec{x} - \vec{s}|}. \quad (6) \]

The vector part is induced by the transverse component of currents,

\[ \hat{A}^\mu_{A,M}(t, \vec{x}) = \frac{1}{c} \int d^3s \frac{\hat{\tilde{\gamma}}_{ET}(u, \vec{s})}{|\vec{x} - \vec{s}|} + \frac{1}{c} \int d^3s \frac{\hat{\tilde{\gamma}}_{NT}(u, \vec{s})}{|\vec{x} - \vec{s}|}, \quad (7) \]

where \( u = t - |\vec{r} - \vec{s}|/c \) is introduced for the retardation effect.

Next, the description of the evolution of the fields is introduced. For the photon field, the evolution is already included partially in (3)-(7). Further effects of the time evolution of operators
Beyond this approximation is not considered in this article. These effects are discussed in our next study. The equations of motion of the electron and nucleus fields are given by

$$
i\hbar \frac{d}{dt} \hat{\Psi} = \left[ \frac{1}{2m_e} \left( -i\hbar \partial_t - \frac{Ze\hbar c}{c} \hat{A}^i \right)^2 + Ze\hbar e\hat{A}_0 \right] \hat{\Psi}, \quad (8)$$

$$
i\hbar \frac{d}{dt} \hat{\chi}_a = \left[ \frac{1}{2m_a} \left( -i\hbar \partial_t - \frac{Z_a\hbar e}{c} \hat{A}^i \right)^2 + Z_a\hbar e\hat{A}_0 \right] \hat{\chi}_a. \quad (9)$$

By these equations, the equations of motion for annihilation operators are derived as

$$
i\hbar \frac{d}{dt} \hat{c}_m(t) = \sum_n \int d^3\vec{x} \hat{\psi}_m^\dagger(\vec{x}) \left[ \frac{1}{2m_e} \left( -i\hbar \partial_t - \frac{Ze\hbar e}{c} \hat{A}^i \right)^2 + Ze\hbar e\hat{A}_0 \right] \hat{\psi}_n(\vec{x})\hat{c}_n(t), \quad (10)$$

$$
i\hbar \frac{d}{dt} \hat{f}_{am}(t) = \sum_n \int d^3\vec{x} \hat{\chi}_m^\dagger(\vec{x}) \left[ \frac{1}{2m_a} \left( -i\hbar \partial_t - \frac{Z_a\hbar e}{c} \hat{A}^i \right)^2 + Z_a\hbar e\hat{A}_0 \right] \hat{\chi}_n(\vec{x})\hat{f}_{an}(t). \quad (11)$$

The creation operators at $t$ can be derived as Hermitian conjugate operators. These equations are computed in a numerical manner. In the manner, the continuum time is discretized by finite time step, $\delta t$. The operators at $t$ are calculated as an expansion by the operators at $t = 0$. This expansion should not be limited only at the first order of creation-annihilation operators. For example, the operator, $\hat{A}_0$, contains electron annihilation and creation operators, and $\hat{A}_{rad}^i$ includes the creation-annihilation operators of the photon. Hence, the operators, $\hat{c}_m$ and $\hat{f}_{am}$, should be expanded as the polynomials of creation-annihilation operators of the electron, nucleus and photon. Accordingly, the coefficients of these polynomials are computed by our numerical code.

In the time evolution computation in this expansion procedure, the unitary operator, $\hat{U} = \exp(-i\hat{H}\delta t)$, is impossible to use, since the Hamiltonian is not a number but an operator with creation-annihilation operators. As a result, we solve the equation of motion by linear approximation,

$$
\hat{c}_m(t_{i+1}) \simeq \hat{c}_m(t_i) + \delta t \frac{d\hat{c}_m}{dt}(t_i), \quad (12)
$$

$$
\hat{f}_{am}(t_{i+1}) \simeq \hat{f}_{am}(t_i) + \delta t \frac{d\hat{f}_{am}}{dt}(t_i), \quad (13)
$$

where $t_{i+1} = t_i + \delta t$. The value of $\delta t$ should be small enough to satisfy the condition that this linear expansion is reliable. The order of the polynomials of the operators at $t = 0$ increases exponentially in an instant. In addition, $\hat{A}_{A,M}$ is defined with the operators at past steps in order to represent retardation effects. To calculate these terms exactly, all expansion coefficients of the all past operators are required to store, and storing this data is not realistic due to their huge size. Hence, the order of operator polynomials is restricted to some order. In the present work, we adopt the minimal order expansion as a trial computation. For the electron field, the creation-annihilation operator of the electron at $t = 0$ is limited to third order, that of the photon is up to the second order, and those of nucleus fields are restricted to the zeroth order. This nucleus contribution to the electron field is given by using the expectation value of nucleus fields. In other words, this nucleus contribution is given by not an operator but a number. The third order electron operator terms are required to represent the exchange interaction, and hence this is considered to be minimal. For nucleus fields, the limitation of the operator order is chosen as the similar criterion. The same nucleus field is limited to the third order, and other species and the electron field are replaced by the expectation values.
The calculation of \( \hat{A}_{A,M} \) is tremendous task, since this term is difficult to compute in an analytic form. Hence, we use the expectation value of this term by a numerical integration and a difference method. (One approach to compute this term in a smart form is mentioned in [10].) The effects of \( \hat{A}_{A,M} \) are quantitatively small in spite of their qualitative importance. Hence we calculate these terms once fifties time steps and until \( \hat{A}_{A,M} \) is updated the same value of \( \hat{A}_{A,M} \) is used. The transverse component of current in equation (7) is computed as [8]

\[
\hat{j}^{eT}(t, \vec{x}) = \hat{j}^{e}(t, \vec{x}) - \hat{j}^{eL}(t, \vec{x}), \tag{14}
\]

\[
\hat{j}^{e} = \frac{1}{2m_e} \left[ i\hbar\hat{\Psi}^\dagger \hat{D} \hat{\Psi} - i\hbar \left( \hat{D} \hat{\Psi} \right)^\dagger \hat{\Psi} \right], \tag{15}
\]

\[
\hat{j}^{eL} = \frac{1}{4\pi} \text{grad} \frac{\partial}{\partial t} \hat{A}_0, \tag{16}
\]

where the nuclear current has the same form. The difficulty of the analytic calculation of \( \hat{A}_{A,M} \) resides in \( \text{grad} \frac{\partial}{\partial t} \hat{A}_0 \). The values of \( \langle \hat{j}^{eT} \rangle \) are calculated on grid points and stored. The numerical integration in \( \hat{A}_{A,M} \) is performed based on this stored data. The study about \( \hat{A}_{A,M} \) will be reported in our future work in detail.

After the evolution of operators is calculated, we can calculate physical quantities, such as charge density and energy density. Of course, these quantities depends on states, i.e. ket vector. Ket vector is defined with creation operators and the vacuum state \( |0\rangle \). For the photon, coherent states are chosen in the present version of our code to suppress data size. For matter fields, the choice of \( \psi_m \) and \( \chi^{am} \) is important for efficient and accurate computations. In this work, we simply use Hartree-Fock orbitals as \( \psi_m \) for electrons. The extension to post Hartree-Fock calculation including correlation effects is not so difficult, and we implement this in the near future. For nuclei, we use wave packets as \( \chi^{am} \) so that the orthogonality, \( \int \chi^{\dagger}_{am} \chi^{an} d^3x = \delta_{mn} \), is satisfied.

Before concluding this section, we mention the renormalization of fields. In the exact form of our method, all quantum effects of the system are included, and hence fields must face divergence. This divergence problem should be solved by the renormalization as ordinary quantum field theory. However, the ordinary renormalization scheme assumes the existence of asymptotic fields. We do not know the asymptotic state for a bound state in quantum field theory. We must adopt a novel prescription of renormalization for our method. For this purpose, we use conservation laws, such as energy and charge. For each time step, we calculate quantities, which must conserve, and if the values are deviated from the true values the operators are renormalized by the common normalization factor. We have coded the renormalization routine using particle number conservation. However other conservation quantities, total energy, momentum, and angular momentum, have not been coded yet. Hence for the calculation shown in this article we adopt the prescription used in NRQED for bound states [12]. In this prescription, cutoff, for the ultra-violet divergence, is used, since the nonrelativistic Lagrangian is incorrect for very high energy, for example higher energy than the electron mass. For higher energy than the cutoff scale, the renormalization for a free particle is adopted, since binding energy is negligible for such a high energy. Therefore effects by virtual ultra-violet photons from the second order operator expansion are appropriately renormalized.

3. Results
In this article, we show our results for hydrogen atom as a demonstration. The purpose of the usage of this model is to show the validity of the interaction by radiation photons. We can of course calculate larger models, and however the behavior of operators is slightly worse due to
the insufficient renormalization of the present version of our code, since interactions among the system are much stronger. The results of molecules will be reported in the near future.

In our computation, the initial state of the hydrogen atom is prepared by our own self-consistent field simulation code which can calculate both electronic and nuclear wave functions. The initial state is prepared in a Hartree-Fock calculation. The basis set of the electron is cc-pVDZ [13]. For the nuclear one, we use our (3s1p) basis, where one exponent of Gaussian (1s1p) is chosen as the same in [14] and other two exponents are chosen to have larger and smaller values. Incident photons are assumed to move toward the positive $y$ direction with kinetic energy, 10 hartree. The ket vector of the photon is the coherent state whose eigenvalue is 0.01. The time step width is chosen as $\delta t = 10^{-5}$ a.u. (1 a.u.$= 2.4 \times 10^{-17}$ s) and the cutoff scale is chosen as 20 a.u. for virtual photons. This cutoff scale is much lower than $m_e c^2$ for the sake of reliable computation.

In figure 1, the electron density distribution at $t = 2.0$ a.u. is shown on $y - z$ plane ($x = 0$). The density evolution is shown as the difference from the density at $t = 0$. The response of the electron is correctly transverse to the photon momentum, and its distribution pattern is plausible. In figure 2, the evolution of the electron density is shown as a function of time. The electron density at $(x, y, z) = (0, 0, 0.8)$ bohr is depicted. Two time scale can be seen in this figure. The shorter time scale, whose period is about 0.6 a.u., corresponds to the photon oscillation of $\exp(\pm i E_{\text{photon}} t)$. The longer time scale, whose period is about 5 a.u., is attributed to the energy difference between the ground and excited states. This oscillation behavior is seen to be disturbed slightly. We consider that this may be due to the effects of the nuclear motion and $\vec{A}_{A}$.

Finally, we should mention the thermalization of Hamiltonian. The initial state we prepared is not the QED initial state but the state which is derived by variational calculations in quantum mechanics. As the real QED initial state, we should calculate it in our QED program. For the calculation without external incident photon, we can derive the QED state, where the space is filled by real and virtual photons, after long enough time steps are computed. For this computation, an appropriate definition is required for the condition of sufficient thermalization. In addition, infinite real and virtual photon modes, which are described by both $\hat{A}_{\text{rad}}$ and $\hat{A}_{A}$,
Figure 2. The evolution of the electron density as a function of time. The electron density at $(x, y, z) = (0, 0, 0.8)$ bohr is depicted. The units are atomic units.

are required to be treated. For these issues, we are now trying some ideas. Hence, the results shown in this article are not for the QED initial state.

4. Summary
We have proposed our computation method of the time evolution of quantum systems based on Rigged QED and reported the present status of the development of our program code, QEDynamics [6]. In our method, the evolution of Heisenberg operators of nuclei and electrons is calculated as coefficients of polynomials of creation-annihilation operators of electrons, nuclei, and photons. For the simulation based on QED, the treatment of the photon is one of the most important points, particularly infrared and ultra-violet photons. For the treatment of the ultra-violet photon, we have used the cutoff. In this article, the results for the hydrogen atom is shown as a demonstration. For larger atoms and molecules, where internal interactions are much stronger, the renormalization is extremely important in viewpoints of quantum field theory. The renormalization is only partially implemented in this version of our code. We improve the treatment of the renormalization method in the near future.

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