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General framework for interatomic interaction energy of two ground-state atoms in a thermal bath

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We give a general derivation of the fourth-order DDC formalism (a formalism proposed by Dalibard, Dupont-Roc, and Cohen-Tannoudji) for calculating the interaction energy between two ground-state multilevel atoms which are coupled to electromagnetic fields in a thermal bath at temperature T. Both the contributions of the thermal field fluctuations and the radiation reaction of atoms are separately identified. As an application of the formalism, we revisit the interaction energy of two static ground-state two-level atoms in a free space and discover new behaviors such as the $\sim TL^{-2}$ behavior of the van der Waals interaction energy in the region where $\lambda^{3/4}T^{-1/4} \ll L \ll \lambda$, with λ the transition wavelength of the atoms and L the interatomic separation, and an oscillatory behavior of the Casimir-Polder interaction energy as L varies which is superimposed on the monotonic $\sim TL^{-6}$ dependence when $L \gg \lambda \gg T^{-1}$.

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I. INTRODUCTION

Spontaneous emission and energy-level shifts of an atom are remarkable phenomena arising from atom-field interaction, which can be attributed to vacuum fluctuations [1,2] or radiation reaction of the atom [3] or a combination of them [4,5]. However, different physical interpretations may be plagued by different problems, for example, the instability of ground-state atoms for the explanation of spontaneous emission as a result of only radiation reaction and the "spontaneous absorption" of ground-state atoms in vacuum for that of only vacuum fluctuations. The indeterminacy in physical interpretation originates from different choices of ordering of commuting operators of the atom and the field in a Heisenberg picture approach to the problem [6]. Noteworthily, this ambiguity was resolved by Dalibard, Dupont-Roc, and Cohen-Tannoudji (DDC) [7,8], who suggested that the symmetric operator ordering should be adopted such that the distinct contributions of vacuum fluctuations and radiation reaction of atoms to the variation rate of an arbitrary observable of the atom are separately Hermitian and thus each possesses an independent physical meaning. This approach which is later called the DDC formalism irons out the problem of instability for ground-state atoms when only radiation reaction is considered and the problem of spontaneous absorption of atoms when only vacuum fluctuations are taken into account.

The DDC formalism was then widely utilized to study the average variation rate of energy and radiative energy-level shifts of a single atom in various circumstances [9–25]. Recently, the DDC formalism was generalized to the case of two atoms in order to study the resonance interaction energy and the evolution of energy of a two-atom system in the symmetric or antisymmetric entangled state (the maximally entangled states) [26]. It is discovered that both the resonance interaction energy and the average variation rate of energy of a two-atom system in a maximally entangled state are wholly attributable to the radiation reaction of the atoms but irrelevant to the field fluctuations [26–29]. This is in sharp contrast to the radiative properties of a single atom which is a combined effect of both the field fluctuations and the radiation reaction of the atom.

Atoms in the maximally entangled state, although interesting in their own right, are not as commonplace as atoms in the ground state. Recently, the interaction between two ground-state atoms has attracted considerable interest [30–33]. The distinction between the interaction between two atoms in maximally entangled states and that in the ground state is that the former is a second-order effect while the latter is a fourth-order one from the point of view of perturbative calculations. Physically, the two differ as exchange of real photons is involved in the former while the latter is only a result of exchange of virtual photons during the atom-field interaction. Although there have already been investigations in the interatomic interaction between two ground-state atoms with different methods [34–40], attempts with the DDC formalism appear only very recently [30–33]. As a first step, a simplified model in which atoms are assumed to be in the monopole interaction with a scalar field in vacuum is considered, and by generalizing the DDC formalism, which is originally of the second order in perturbation, to the fourth order, it is

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shown that the interaction energy of two ground-state atoms is a joint effect of the vacuum fluctuations of the field and the radiation reaction of the atoms [32]. With this complete fourth-order DDC formalism [32], the interaction energy of two ground-state atoms in uniform acceleration is recently calculated and it is found that the interaction displays novel behaviors that reveal the long-range properties of vacuum altered by accelerating atoms through it [33]. Let us note here that this fourth-order DDC formalism has been further generalized from vacuum to a thermal bath [41].

However, one should bear in mind that the model in which atoms are in the monopole interaction with a scalar field is just a toy model that serves to qualitatively disclose the properties of the interatomic interaction in a simplified setting. To obtain quantitative behaviors of the interaction in the real world, one must study a realistic situation in which atoms are coupled to an electromagnetic field. The present paper aims to first establish, in Sec. II, the fourth-order DDC formalism that can deal with two multilevel ground-state atoms coupled to electromagnetic fields in a thermal bath.

Then, as an application of the generalized formalism, we separately calculate, in Sec. III, the contribution of thermal fluctuations and that of radiation reaction to the interaction energy of two static ground-state two-level atoms with the same transition frequency ω which are coupled with electromagnetic fields in a thermal bath at temperature T. Detailed analysis will be performed on the behaviors of the interaction energy in both the low- and high-temperature limits, i.e., when $T \ll \omega$ and $T \gg \omega$, which are equivalent to $\lambda \ll \beta$ and $\lambda \gg \beta$ with $\lambda = 2\pi\omega^{-1}$ and $\beta = T^{-1}$, the transition wavelength of the atoms and the characteristic wavelength of thermal photons, respectively.[1] Note that besides two characteristic lengths, λ and β, there is another characteristic length for the problem under consideration, i.e., the interatomic separation L. With this length scale, the low- and high-temperature limits can be further divided into three subregions, i.e., $L \ll \lambda \ll \beta$, $\lambda \ll L \ll \beta$, and $\lambda \ll \beta \ll L$ for the low-temperature limit, and $L \ll \beta \ll \lambda$, $\beta \ll L \ll \lambda$, and $\beta \ll \lambda \ll L$ for the high-temperature limit. We examine the behaviors of the interatomic interaction energy in all the six subregions, with the contributions of the thermal fluctuations and the radiation reaction separately identified. In comparison with the existing results in the literature [42–46], we discover behaviors of the interatomic interaction energy in the high-temperature limit, i.e., a unique behavior of $\sim TL^{-2}$ in the region where $\sqrt[4]{\lambda^3\beta} \ll L \ll \lambda$, and an oscillatory behavior which is superimposed on the monotonic $\sim TL^{-6}$ dependence on separation L when $\beta \ll \lambda \ll L$, which were both not reported before.

We then conclude in Sec. IV with a summary and some discussions. Throughout the paper, we exploit the units in which $\hbar = c = \varepsilon_0 = 1$ with $\hbar$ the reduced Plank constant, c the speed of light, and ε_0 the vacuum permittivity.

II. GENERIC FOURTH-ORDER DDC FORMALISM: THE THERMAL ELECTROMAGNETIC FIELD CASE

In this section, we give a detailed derivation of the fourth-order DDC formalism for calculating the interaction energy between two ground-state multilevel atoms which are coupled to electromagnetic fields in a thermal bath at temperature T. We label the two atoms by A and B and assume them to be in synchronous motion so that they share the same proper time τ. Then the Hamiltonian of the two atoms can be written as

$$H_S(\tau) = \sum_n \omega_n^A \sigma_{nn}^A(\tau) + \sum_n \omega_n^B \sigma_{nn}^B(\tau), \tag{1}$$

where $\sigma_{nn} = |n\rangle\langle n|$ and $|n\rangle$ denotes a series of stationary states of the atom with energy ω_n. In the Coulomb gauge, the electric-field operator reads [47]

$$\mathbf{E}(x) = \int d^3\mathbf{k}\, g_{\mathbf{k}} \sum_{\nu=1}^{2} i\omega_{\mathbf{k}} \boldsymbol{\epsilon}(\mathbf{k},\nu) \times [a_{\mathbf{k},\nu} e^{-i\omega_{\mathbf{k}} t} e^{i\mathbf{k}\cdot\mathbf{x}} - a^\dagger_{\mathbf{k},\nu} e^{i\omega_{\mathbf{k}} t} e^{-i\mathbf{k}\cdot\mathbf{x}}] \tag{2}$$

with $g_{\mathbf{k}} = [(2\pi)^3 2\omega_{\mathbf{k}}]^{-1/2}$, $\mathbf{k}$ and ν the wave vector and the polarization of the field modes respectively, $a_{\mathbf{k},\nu}$ and $a^\dagger_{\mathbf{k},\nu}$ the annihilation and creation operators, and $\boldsymbol{\epsilon}(\mathbf{k},\nu)$ the polarization vector satisfying

$$\sum_{\nu=1}^{2} \epsilon_p(\mathbf{k},\nu)\epsilon_q(\mathbf{k},\nu) + \frac{k_p k_q}{k^2} = \delta_{pq}. \tag{3}$$

Then the Hamiltonian of the field follows

$$H_F(t(\tau)) = \int d^3\mathbf{k}\, \omega_{\mathbf{k}} \sum_{\nu=1}^{2} a^\dagger_{\mathbf{k},\nu}(t(\tau)) a_{\mathbf{k},\nu}(t(\tau)) \frac{dt}{d\tau} \tag{4}$$

with $a_{\mathbf{k},\nu}(t) = a_{\mathbf{k},\nu} e^{-i\omega_{\mathbf{k}} t}$, and the Hamiltonian describing the atom-field interaction is given by

$$H_I(\tau) = -\boldsymbol{\mu}_A(\tau)\cdot \boldsymbol{E}(x_A(\tau)) - \boldsymbol{\mu}_B(\tau)\cdot \boldsymbol{E}(x_B(\tau)), \tag{5}$$

where $\boldsymbol{\mu}_\xi$ with $\xi = A, B$ is the dipole moment operator of atom ξ. Adding up the three Hamiltonians Eqs. (1), (4), and (5), we obtain the total Hamiltonian of the system: $H(\tau) = H_S(\tau) + H_F(\tau) + H_I(\tau)$.

As a result of the atom-field interaction, the electromagnetic field $\boldsymbol{E}(x)$ can be separated into two parts, i.e., a free part $\boldsymbol{E}^f(x)$ which exists even when there is no atom-field interaction, and a source part which arises from the atom-field interaction. Similarly, every operator of the atoms can also be separated into a free part and a source part. We next implement such separations using the Heisenberg equations of motion of the dynamical variables of the atoms and the field.

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[1] We have set the Boltzmann constant $k_B = 1$.

A. Evolution of dynamical variables of the atoms and the field

With the Hamiltonians of the atom-field system, we can derive the Heisenberg equations of motion for the atomic operator $\sigma^{\xi}_{mn}(\tau)$ and that of the field $a_{\mathbf{k},\nu}(t(\tau))$:

$$\frac{d}{d\tau}\sigma^{\xi}_{mn}(\tau)=i\big(\omega^{\xi}_{m}-\omega^{\xi}_{n}\big)\sigma^{\xi}_{mn}(\tau)-i\boldsymbol{E}(x_{\xi}(\tau))\cdot\big[\boldsymbol{\mu}_{\xi}(\tau),\sigma^{\xi}_{mn}(\tau)\big] \tag{6}$$

and

$$\frac{d}{d\tau}a_{\mathbf{k},\nu}(t(\tau))=-i\omega_{\mathbf{k}}a_{\mathbf{k},\nu}(t(\tau))\frac{dt}{d\tau}-i\sum_{\xi=A}^{B}\boldsymbol{\mu}_{\xi}(\tau)\cdot\{\boldsymbol{E}(x_{\xi}(\tau)),a_{\mathbf{k},\nu}(t(\tau))\}, \tag{7}$$

and then split each solution to the equations into a free part which is independent of the atom-field coupling and a source part which is induced by the coupling. Hereafter, $[\cdots,\cdots]$ represents the commutator of two sets of operators, and we denote the free parts and the source parts by superscripts "f" and "s" respectively. So, we can write

$$\sigma^{\xi}_{mn}(\tau)=\sigma^{\xi,f}_{mn}(\tau)+\sigma^{\xi,s}_{mn}(\tau), \tag{8}$$

$$a_{\mathbf{k},\nu}(t(\tau))=a^{f}_{\mathbf{k},\nu}(t(\tau))+a^{s}_{\mathbf{k},\nu}(t(\tau)) \tag{9}$$

with

$$\begin{cases}\sigma^{\xi,f}_{mn}(\tau)=\sigma^{\xi,f}_{mn}(\tau_0)e^{i(\omega^{\xi}_{m}-\omega^{\xi}_{n})(\tau-\tau_0)},\\ \sigma^{\xi,s}_{mn}(\tau)=-i\displaystyle\int_{\tau_0}^{\tau}d\tau_1\,\boldsymbol{E}(x_{\xi}(\tau_1))\cdot\big[\boldsymbol{\mu}_{\xi}(\tau_1),\sigma^{\xi}_{mn}(\tau_1)\big]e^{i(\omega^{\xi}_{m}-\omega^{\xi}_{n})(\tau-\tau_1)}\end{cases} \tag{10}$$

and

$$\begin{cases}a^{f}_{\mathbf{k},\nu}(t(\tau))=a^{f}_{\mathbf{k},\nu}(t(\tau_0))e^{-i\omega_{\mathbf{k}}[t(\tau)-t(\tau_0)]},\\ a^{s}_{\mathbf{k},\nu}(t(\tau))=-i\sum\limits_{\xi=A}^{B}\int_{\tau_0}^{\tau}d\tau_1\,\boldsymbol{\mu}_{\xi}(\tau_1)\cdot[\boldsymbol{E}(x_{\xi}(\tau_1)),a_{\mathbf{k},\nu}(t(\tau_1))]e^{-i\omega_{\mathbf{k}}[t(\tau)-t(\tau_1)]}.\end{cases} \tag{11}$$

As a result, $\boldsymbol{E}(x(\tau))$ is accordingly split into the free field $\boldsymbol{E}^{f}(x(\tau))$ and the source field $\boldsymbol{E}^{s}(x(\tau))$ with

$$\begin{cases}\boldsymbol{E}^{f}(x)=\int d^3\mathbf{k}\,g_{\mathbf{k}}\sum\limits_{\nu=1}^{2}i\omega_{\mathbf{k}}\boldsymbol{\epsilon}(\mathbf{k},\nu)\big[a^{f}_{\mathbf{k},\nu}(t)e^{i\mathbf{k}\cdot\mathbf{x}}-a^{\dagger f}_{\mathbf{k},\nu}(t)e^{-i\mathbf{k}\cdot\mathbf{x}}\big],\\ \boldsymbol{E}^{s}(x)=\int d^3\mathbf{k}\,g_{\mathbf{k}}\sum\limits_{\nu=1}^{2}i\omega_{\mathbf{k}}\boldsymbol{\epsilon}(\mathbf{k},\nu)\big[a^{s}_{\mathbf{k},\nu}(t)e^{i\mathbf{k}\cdot\mathbf{x}}-a^{\dagger s}_{\mathbf{k},\nu}(t)e^{-i\mathbf{k}\cdot\mathbf{x}}\big].\end{cases} \tag{12}$$

We now expand the full operators in the source operators $\sigma^{\xi,s}_{mn}(\tau)$ and $a^{s}_{\mathbf{k},\nu}(t(\tau))$ in the second line of Eqs. (10) and (11) in the coupling constant which is assumed to be small,

$$\sigma^{\xi}_{mn}(\tau)=\sigma^{\xi,f}_{mn}(\tau)+\sigma^{\xi,(1)}_{mn}(\tau)+\sigma^{\xi,(2)}_{mn}(\tau)+\sigma^{\xi,(3)}_{mn}(\tau)+\cdots, \tag{13}$$

$$a_{\mathbf{k},\nu}(t(\tau))=a^{f}_{\mathbf{k},\nu}(t(\tau))+a^{(1)}_{\mathbf{k},\nu}(t(\tau))+a^{(2)}_{\mathbf{k},\nu}(t(\tau))+a^{(3)}_{\mathbf{k},\nu}(t(\tau))+\cdots, \tag{14}$$

$$\boldsymbol{E}(x_{\xi}(\tau))=\boldsymbol{E}^{f}(x_{\xi}(\tau))+\boldsymbol{E}^{(1)}(x_{\xi}(\tau))+\boldsymbol{E}^{(2)}(x_{\xi}(\tau))+\boldsymbol{E}^{(3)}(x_{\xi}(\tau))+\cdots, \tag{15}$$

and put these expansions into the second line of Eqs. (10) and (11). Then we can iteratively express $\sigma^{\xi,(j)}_{mn}(\tau)$ and $a^{(j)}_{\mathbf{k},\nu}(t(\tau))$ in terms of free operators of the atoms and the field. For concrete expressions of the first- to third-order source operators, $\sigma^{\xi,(j)}_{mn}(\tau)$, $a^{(j)}_{\mathbf{k},\nu}(t(\tau))$ and $\boldsymbol{E}^{(j)}(x_{\xi}(\tau))$, please refer to Appendix A.

With these free and source operators, we next discuss the evolution of energy of the atoms by separately identifying contributions of the free field and the source field.

B. Variation rate of energy of atoms: Separation of contributions of field fluctuations and radiation reaction of atoms

To study the evolution of energy of the atoms, we first resort to the Heisenberg equations of motion of their Hamiltonians. Take that of atom A for an example, which is

$$\frac{d}{d\tau}H_A(\tau)=-i\left[\boldsymbol{\mu}_A(\tau)\cdot\boldsymbol{E}(x_A(\tau)),\sum_{n}\omega^{A}_{n}\sigma^{A}_{nn}(\tau)\right]. \tag{16}$$

Separating $\boldsymbol{E}(x_A(\tau))$ into the free part $\boldsymbol{E}^{f}(x_A(\tau))$ and the source part $\boldsymbol{E}^{s}(x_A(\tau))$, adopting the symmetric operator ordering [7,8] between the variables of the atom $\boldsymbol{\mu}_A(\tau)$ and those of field $\boldsymbol{E}^{f}(x_A(\tau))$ or $\boldsymbol{E}^{s}(x_A(\tau))$, and taking the average of the variation rate over the thermal state of the field, $|\beta\rangle$ with $\beta = T^{-1}$, we find

$$\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tot}^{\beta} = \left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta}+\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm rr}^{\beta} \tag{17}$$

with

$$\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta} = -\frac{i}{2}\sum_{n,p}\omega_n^A\langle\beta|\big\{E_p^f(x_A(\tau)),\,\big[\mu_{A,p}(\tau),\,\sigma_{nn}^A(\tau)\big]\big\}|\beta\rangle \tag{18}$$

and

$$\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm rr}^{\beta} = -\frac{i}{2}\sum_{n,p}\omega_n^A\langle\beta|\big\{E_p^s(x_A(\tau)),\,\big[\mu_{A,p}(\tau),\,\sigma_{nn}^A(\tau)\big]\big\}|\beta\rangle, \tag{19}$$

which we define as the contributions of thermal fluctuations [tf] and the radiation reaction of atoms [rr]. Hereafter, $\mu_{\xi,p}$ and $\{\cdots,\cdots\}$ denote the pth component of the dipole moment of atom ξ and the anticommutator of two sets of operators, respectively, and $\langle\cdots\rangle^{\beta}$ on the left-hand side stands for $\langle\beta|\cdots|\beta\rangle$.

Next, we expand operators on the right of Eq. (18) with the help of Eq. (13) and the relation

$$\boldsymbol{\mu}_\xi(\tau)=\sum_{m,n}^{m\neq n}(\boldsymbol{\mu}_\xi)_{mn}\sigma_{mn}^{\xi,f}(\tau)+\sum_{j=1}^{\infty}\sum_{m,n}^{m\neq n}(\boldsymbol{\mu}_\xi)_{mn}\sigma_{mn}^{\xi,(j)}(\tau), \tag{20}$$

and find that the first- to the fourth-order operators of $\langle\frac{dH_A(\tau)}{d\tau}\rangle_{\rm tf}^{\beta}$ are given by

$$\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta,(1)} = -\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,f}(\tau),\,\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle, \tag{21}$$

$$\begin{aligned}\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta,(2)} =& -\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,(1)}(\tau),\,\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle\\ &-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,f}(\tau),\,\sigma_{nn}^{A,(1)}(\tau)\big]\big\}|\beta\rangle,\end{aligned} \tag{22}$$

$$\begin{aligned}\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta,(3)} =& -\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,f}(\tau),\,\sigma_{nn}^{A,(2)}(\tau)\big]\big\}|\beta\rangle\\ &-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,(1)}(\tau),\,\sigma_{nn}^{A,(1)}(\tau)\big]\big\}|\beta\rangle\\ &-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,(2)}(\tau),\,\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle\end{aligned} \tag{23}$$

and

$$\begin{aligned}\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta,(4)} =& -\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,f}(\tau),\,\sigma_{nn}^{A,(3)}(\tau)\big]\big\}|\beta\rangle\\ &-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,(1)}(\tau),\,\sigma_{nn}^{A,(2)}(\tau)\big]\big\}|\beta\rangle\\ &-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,(2)}(\tau),\,\sigma_{nn}^{A,(1)}(\tau)\big]\big\}|\beta\rangle\\ &-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^f(x_A(\tau)),\,\big[\sigma_{pq}^{A,(3)}(\tau),\,\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle.\end{aligned} \tag{24}$$

Then with the first- to third-order atomic and field operators given in Appendix A, we can reexpress the above four equations in terms of the free operators of the atoms and the field. It is easy to see that $\langle\frac{dH_A(\tau)}{d\tau}\rangle_{\rm tf}^{\beta,(1)}$ and $\langle\frac{dH_A(\tau)}{d\tau}\rangle_{\rm tf}^{\beta,(3)}$ are zero, because the anticommutator $\{\cdots,\cdots\}$ on the right of Eq. (21) is obviously of the first power in $\boldsymbol{E}^f$ and that of Eq. (23) is of the third power in $\boldsymbol{E}^f$ according to Eqs. (A1) and (A6), and so they both vanish after taking the expectation value over $|\beta\rangle$. The second- and fourth-order average variation rates are generally nonzero and are respectively given by

$$\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta,(2)}=-\frac{1}{2}\sum_{n,p,q}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\,\langle\beta|\big\{E_p^f(x_A(\tau)),E_q^f(x_A(\tau_1))\big\}|\beta\rangle\big\{\mu_{A,p}^f(\tau),\big[\mu_{A,q}^f(\tau_1),\sigma_{nn}^{A,f}(\tau)\big]\big\}\tag{25}$$

and

$$\begin{aligned}\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm tf}^{\beta,(4)}=&\frac{1}{2}\sum_{n,p,q,r,s}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big\{E_p^f(x_A(\tau)),E_s^f(x_B(\tau_3))\big\}\\&\times\big\{E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big\}|\beta\rangle\big[\mu_{B,s}^f(\tau_3),\mu_{B,r}^f(\tau_2)\big]\big\{\mu_{A,q}^f(\tau_1),\big[\mu_{A,p}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}+F(\tau).\end{aligned}\tag{26}$$

Notice here that $F(\tau)$ is the abbreviation of a collection of terms of the zeroth and first power in $\boldsymbol{\mu}_B^f$ with $\boldsymbol{\mu}_\xi^f(\tau)\equiv\sum_{m,n}^{m\neq n}(\boldsymbol{\mu}_\xi)_{mn}\sigma_{mn}^{\xi,f}(\tau)$. Since the explicit expression of $F(\tau)$ is very tedious and more importantly it does not contribute to the interatomic interaction as we shall explain in the next subsection, we do not show it explicitly here.

Following similar procedures, we find $\langle\frac{dH_A(\tau)}{d\tau}\rangle_{\rm rr}^{\beta,(j)}$ which starts at the second order:

$$\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm rr}^{\beta,(2)}=-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(1)}(x_A(\tau)),\big[\sigma_{pq}^{A,f}(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle,\tag{27}$$

$$\begin{aligned}\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm rr}^{\beta,(3)}=&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(1)}(x_A(\tau)),\big[\sigma_{pq}^{A,(1)}(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle\\&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(1)}(x_A(\tau)),\big[\sigma_{pq}^{A,f}(\tau),\sigma_{nn}^{A,(1)}(\tau)\big]\big\}|\beta\rangle\\&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(2)}(x_A(\tau)),\big[\sigma_{pq}^{A,f}(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle,\end{aligned}\tag{28}$$

and

$$\begin{aligned}\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm rr}^{\beta,(4)}=&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(1)}(x_A(\tau)),\big[\sigma_{pq}^{A,(1)}(\tau),\sigma_{nn}^{A,(1)}(\tau)\big]\big\}|\beta\rangle\\&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(1)}(x_A(\tau)),\big[\sigma_{pq}^{A,f}(\tau),\sigma_{nn}^{A,(2)}(\tau)\big]\big\}|\beta\rangle\\&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(1)}(x_A(\tau)),\big[\sigma_{pq}^{A,(2)}(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle\\&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(2)}(x_A(\tau)),\big[\sigma_{pq}^{A,(1)}(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle\\&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(2)}(x_A(\tau)),\big[\sigma_{pq}^{A,f}(\tau),\sigma_{nn}^{A,(1)}(\tau)\big]\big\}|\beta\rangle\\&-\frac{i}{2}\sum_{n,p,q}\omega_n^A(\boldsymbol{\mu}_A)_{pq}\cdot\langle\beta|\big\{\boldsymbol{E}^{(3)}(x_A(\tau)),\big[\sigma_{pq}^{A,f}(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}|\beta\rangle.\end{aligned}\tag{29}$$

By the same line of reasoning as that for the thermal field contribution, we can also derive that only the second- and the fourth-order operators contribute. Then we have

$$\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm rr}^{\beta,(2)}=-\frac{1}{2}\sum_{n,p,q,\xi}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\,\langle\beta|\big[E_p^f(x_\xi(\tau_1)),E_q^f(x_A(\tau))\big]|\beta\rangle\big\{\mu_{\xi,p}^f(\tau_1),\big[\mu_{A,q}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}\tag{30}$$

and

$$\begin{aligned}\left\langle\frac{dH_A(\tau)}{d\tau}\right\rangle_{\rm rr}^{\beta,(4)}=&\frac{1}{2}\sum_{n,p,q,r,s}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_q^f(x_B(\tau_1)),E_p^f(x_A(\tau))\big]\\&\times\big[E_s^f(x_B(\tau_3)),E_r^f(x_A(\tau_2))\big]|\beta\rangle\big\{\mu_{B,q}^f(\tau_1),\mu_{B,s}^f(\tau_3)\big\}\big[\mu_{A,r}^f(\tau_2),\big[\mu_{A,p}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big]\\&+\frac{1}{2}\sum_{n,p,q,r,s}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau}d\tau_3\,\langle\beta|\big\{E_r^f(x_B(\tau_2)),E_s^f(x_A(\tau_3))\big\}\end{aligned}$$

$$
\begin{aligned}
&\times\big[E_q^f(x_B(\tau_1)),E_p^f(x_A(\tau))\big]|\beta\rangle\big[\mu_{B,r}^f(\tau_2),\mu_{B,q}^f(\tau_1)\big]\big[\mu_{A,s}^f(\tau_3),\big[\mu_{A,p}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big]\\
&+\frac{1}{2}\sum_{n,p,q,r,s}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_1}d\tau_3\,\langle\beta|\big[E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big]\\
&\times\big[E_s^f(x_B(\tau_3)),E_p^f(x_A(\tau))\big]|\beta\rangle\big[\mu_{B,r}^f(\tau_2),\mu_{B,s}^f(\tau_3)\big]\big\{\mu_{A,q}^f(\tau_1),\big[\mu_{A,p}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}\\
&+\frac{1}{2}\sum_{n,p,q,r,s}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big]\\
&\times\big[E_s^f(x_B(\tau_3)),E_p^f(x_A(\tau))\big]|\beta\rangle\big[\mu_{B,s}^f(\tau_3),\mu_{B,r}^f(\tau_2)\big]\big\{\mu_{A,q}^f(\tau_1),\big[\mu_{A,p}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}\\
&+\frac{1}{2}\sum_{n,p,q,r,s}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_q^f(x_A(\tau_1)),E_s^f(x_B(\tau_3))\big]\\
&\times\big[E_r^f(x_B(\tau_2)),E_p^f(x_A(\tau))\big]|\beta\rangle\big[\mu_{B,s}^f(\tau_3),\mu_{B,r}^f(\tau_2)\big\}\big\{\mu_{A,q}^f(\tau_1),\big[\mu_{A,p}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}\\
&+\frac{1}{2}\sum_{n,p,q,r,s}\omega_n^A\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_s^f(x_A(\tau_3)),E_r^f(x_B(\tau_2))\big]\\
&\times\big[E_q^f(x_B(\tau_1)),E_p^f(x_A(\tau))\big]|\beta\rangle\big[\mu_{B,r}^f(\tau_2),\mu_{B,q}^f(\tau_1)\big]\big\{\mu_{A,s}^f(\tau_3),\big[\mu_{A,p}^f(\tau),\sigma_{nn}^{A,f}(\tau)\big]\big\}+G(\tau),
\end{aligned}
\tag{31}
$$

where $G(\tau)$ is the abbreviation of a collection of terms of the zeroth, first, and third power in $\boldsymbol{\mu}_B^f$. We do not show it explicitly here as it does not contribute to the interatomic interaction potential either.

C. Effective Hamiltonians and the interatomic interaction energy

So far, we have derived the first- to fourth-order tf and rr contributions to the variation rate of the Hamiltonian of atom A, and those of atom B can be easily obtained by exchanging the indices A and B. Adding up the tf and rr contributions to the average variation rate of energy of both atoms, we then obtain $\langle\frac{d}{d\tau}H_S(\tau)\rangle_{\rm tf}^{\beta,(i)}$ and $\langle\frac{d}{d\tau}H_S(\tau)\rangle_{\rm rr}^{\beta,(i)}$ with $H_S^f(\tau)=\sum_{\xi,n}\omega_n^\xi\sigma_{nn}^{\xi,f}(\tau)$, which are respectively the contributions of the free field and the source field to the average variation rate of energy of the two-atom system. Following the tricky procedures similar to those in Ref. [32], we can further express them in the following form:

$$
\left\langle\frac{d}{d\tau}H_S(\tau)\right\rangle_{\rm tf,rr}^{\beta,(i)}=i\big[(H_S(\tau))_{\rm tf,rr}^{{\rm eff},(i)},H_S^f(\tau)\big]+\text{non-Hermitian terms}
\tag{32}
$$

where

$$
(H_S(\tau))_{\rm tf}^{{\rm eff},(2)}=\frac{i}{4}\sum_{p,q}\int_{\tau_0}^{\tau}d\tau_1\,\langle\beta|\big\{E_p^f(x_A(\tau)),E_q^f(x_A(\tau_1))\big\}|\beta\rangle\big[\mu_{A,q}^f(\tau_1),\mu_{A,p}^f(\tau)\big]+A\rightleftharpoons B\ \text{term}
\tag{33}
$$

and

$$
\begin{aligned}
(H_S(\tau))_{\rm tf}^{{\rm eff},(4)}=\frac{i}{8}\sum_{p,q,r,s}&\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big\{E_p^f(x_A(\tau)),E_s^f(x_B(\tau_3))\big\}\\
&\times\big[E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big]|\beta\rangle\big[\mu_{A,p}^f(\tau),\mu_{A,q}^f(\tau_1)\big]\big[\mu_{B,s}^f(\tau_3),\mu_{B,r}^f(\tau_2)\big]+\tilde{F}(\tau)+A\rightleftharpoons B\ \text{terms}
\end{aligned}
\tag{34}
$$

are the second- and fourth-order effective Hamiltonians of the two-atom system due to the thermal fluctuations, and

$$
\begin{aligned}
(H_S(\tau))_{\rm rr}^{{\rm eff},(2)}=&\frac{i}{4}\sum_{p,q}\int_{\tau_0}^{\tau}d\tau_1\,\langle\beta|\big[E_p^f(x_B(\tau_1)),E_q^f(x_A(\tau))\big]|\beta\rangle\big\{\mu_{B,p}^f(\tau_1),\mu_{A,q}^f(\tau)\big\}\\
&+\frac{i}{4}\sum_{p,q}\int_{\tau_0}^{\tau}d\tau_1\,\langle\beta|\big[E_p^f(x_A(\tau_1)),E_q^f(x_A(\tau))\big]|\beta\rangle\big\{\mu_{A,p}^f(\tau_1),\mu_{A,q}^f(\tau)\big\}+A\rightleftharpoons B\ \text{terms}
\end{aligned}
\tag{35}
$$

and

$$
\begin{aligned}
(H_S(\tau))_{\rm rr}^{{\rm eff},(4)}=\frac{i}{8}\sum_{p,q,r,s}&\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big]\\
&\times\big[E_s^f(x_B(\tau_3)),E_p^f(x_A(\tau))\big]|\beta\rangle\big\{\mu_{A,q}^f(\tau_1),\mu_{A,p}^f(\tau)\big\}\big[\mu_{B,r}^f(\tau_2),\mu_{B,s}^f(\tau_3)\big]
\end{aligned}
$$

$$
\begin{aligned}
&+\frac{i}{8}\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_q^f(x_A(\tau_1)),E_s^f(x_B(\tau_3))\big]\\
&\times\big[E_r^f(x_B(\tau_2)),E_p^f(x_A(\tau))\big]|\beta\rangle\big\{\mu^f_{A,q}(\tau_1),\mu^f_{A,p}(\tau)\big\}\big[\mu^f_{B,r}(\tau_2),\mu^f_{B,s}(\tau_3)\big]\\
&+\frac{i}{8}\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_s^f(x_A(\tau_3)),E_r^f(x_B(\tau_2))\big]\\
&\times\big[E_q^f(x_B(\tau_1)),E_p^f(x_A(\tau))\big]|\beta\rangle\big\{\mu^f_{A,s}(\tau_3),\mu^f_{A,p}(\tau)\big\}\big[\mu^f_{B,q}(\tau_1),\mu^f_{B,r}(\tau_2)\big]\\
&+\frac{i}{8}\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\langle\beta|\big[E_q^f(x_B(\tau_1)),E_p^f(x_A(\tau))\big]\\
&\times\big[E_s^f(x_B(\tau_3)),E_r^f(x_A(\tau_2))\big]|\beta\rangle\big[\mu^f_{A,p}(\tau),\mu^f_{A,r}(\tau_2)\big]\big\{\mu^f_{B,q}(\tau_1),\mu^f_{B,s}(\tau_3)\big\}\\
&+\frac{i}{8}\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau}d\tau_3\,\langle\beta|\big\{E_r^f(x_B(\tau_2)),E_s^f(x_A(\tau_3))\big\}\\
&\times\big[E_q^f(x_B(\tau_1)),E_p^f(x_A(\tau))\big]|\beta\rangle\big[\mu^f_{A,p}(\tau),\mu^f_{A,s}(\tau_3)\big]\big[\mu^f_{B,r}(\tau_2),\mu^f_{B,q}(\tau_1)\big]\\
&+\frac{i}{8}\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_1}d\tau_3\,\langle\beta|\big[E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big]\\
&\times\big[E_s^f(x_B(\tau_3)),E_p^f(x_A(\tau))\big]|\beta\rangle\big\{\mu^f_{A,q}(\tau_1),\mu^f_{A,p}(\tau)\big\}\big[\mu^f_{B,s}(\tau_3),\mu^f_{B,r}(\tau_2)\big]+\tilde{G}(\tau)\\
&+A\rightleftharpoons B\ \text{terms}
\end{aligned}
\tag{36}
$$

are the second- and fourth-order effective Hamiltonians of the two-atom system due to the radiation reaction. Then averaging these effective Hamiltonians in Eqs. (33)–(36) over $|g_Ag_B\rangle$ gives rise to the tf and rr contributions to the energy of the two atoms as a whole.

Now some comments are in order. First, for the second order, it is obvious that the terms in Eq. (33) are independent of the relative position of the two atoms (the interatomic separation), and thus $\langle g_Ag_B|(H_S(\tau))^{\text{eff},(2)}_{\text{tf}}|g_Ag_B\rangle$ actually equals to the second-order tf contribution to the energy shift of atom A plus that of atom B. Similarly, $\langle g_Ag_B|(H_S(\tau))^{\text{eff},(2)}_{\text{rr}}|g_Ag_B\rangle$ is the second-order rr contribution to the energy shift of atom A plus that of atom B, since the separation-dependent terms in Eq. (35) are proportional to $\mu^f_A\mu^f_B$ and their averages over $|g_Ag_B\rangle$ are vanishing. Second, for the fourth order, let us first note the fact that $\tilde{F}(\tau)$ in $(H_S(\tau))^{\text{eff},(4)}_{\text{tf}}$ [Eq. (34)] follows from $F(\tau)$ in $\langle\frac{d}{d\tau}H_A(\tau)\rangle^{\beta,(4)}_{\text{tf}}$ [Eq. (26)] and $\tilde{G}(\tau)$ in $(H_S(\tau))^{\text{eff},(4)}_{\text{rr}}$ [Eq. (36)] comes from $G(\tau)$ in $\langle\frac{d}{d\tau}H_A(\tau)\rangle^{\beta,(4)}_{\text{rr}}$ [Eq. (31)]. So $\tilde{F}(\tau)$ is also a collection of terms of the zeroth and first power in $\boldsymbol{\mu}^f_B$ and $\tilde{G}(\tau)$ is a collection of terms of the zeroth, first, and third power in $\boldsymbol{\mu}^f_B$. Therein, terms of the zeroth power in $\boldsymbol{\mu}^f_B$ only contribute to the high-order energy shift of atom A but are irrelevant to the interatomic interaction potential, and terms of the first and third power in $\boldsymbol{\mu}^f_B$ also do not contribute to the interatomic interaction potential because their averages over $|g_Ag_B\rangle$ are zero. We do not explicitly show such terms in Eqs. (34) and (36) but just abbreviate them by $\tilde{F}(\tau)$ and $\tilde{G}(\tau)$, partly because of the fact that they do not contribute to the interaction potential and partly because they are very lengthy.

Now it is clear that, up to the fourth order, the tf and rr contributions to the interatomic interaction potential respectively come from the averages of those separation-dependent terms explicitly listed in $(H_S(\tau))^{\text{eff},(4)}_{\text{tf}}$ [Eq. (34)] and $(H_S(\tau))^{\text{eff},(4)}_{\text{rr}}$ [Eq. (36)] over $|g_Ag_B\rangle$, and they can be further simplified to

$$
(\delta E)_{\text{tf}}=2i\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,C^F_{ps}(x_A(\tau),x_B(\tau_3))\chi^F_{qr}(x_A(\tau_1),x_B(\tau_2))\chi^A_{pq}(\tau,\tau_1)\chi^B_{rs}(\tau_2,\tau_3)+A\rightleftharpoons B\ \text{term}
\tag{37}
$$

and

$$
\begin{aligned}
(\delta E)_{\text{rr}}=\;&2i\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\chi^F_{ps}(x_A(\tau),x_B(\tau_3))\chi^F_{qr}(x_A(\tau_1),x_B(\tau_2))C^A_{pq}(\tau,\tau_1)\chi^B_{rs}(\tau_2,\tau_3)\\
&+2i\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\chi^F_{qs}(x_A(\tau_1),x_B(\tau_3))\chi^F_{pr}(x_A(\tau),x_B(\tau_2))C^A_{pq}(\tau,\tau_1)\chi^B_{sr}(\tau_3,\tau_2)\\
&+2i\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\chi^F_{rs}(x_B(\tau_2),x_A(\tau_3))\chi^F_{pq}(x_A(\tau),x_B(\tau_1))C^A_{ps}(\tau,\tau_3)\chi^B_{qr}(\tau_1,\tau_2)\\
&+2i\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\chi^F_{rs}(x_A(\tau_2),x_B(\tau_3))\chi^F_{pq}(x_A(\tau),x_B(\tau_1))\chi^A_{pr}(\tau,\tau_2)C^B_{qs}(\tau_1,\tau_3)
\end{aligned}
$$

$$+2i\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau}d\tau_3\,C^F_{rs}(x_B(\tau_2),x_A(\tau_3))\chi^F_{pq}(x_A(\tau),x_B(\tau_1))\chi^A_{ps}(\tau,\tau_3)\chi^B_{qr}(\tau_1,\tau_2)$$

$$+2i\sum_{p,q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_1}d\tau_3\,\chi^F_{ps}(x_A(\tau),x_B(\tau_3))\chi^F_{qr}(x_A(\tau_1),x_B(\tau_2))C^A_{pq}(\tau,\tau_1)\chi^B_{sr}(\tau_3,\tau_2)+A\rightleftharpoons B\text{ terms.} \tag{38}$$

Note that the contribution of zero-point fluctuations is included in the tf contribution [Eq. (37)]. In these two equations, we have defined the symmetric correlation function and the linear susceptibility of the field along the trajectories of the atoms in the thermal bath by

$$C^F_{pq}(x_A(\tau),x_B(\tau'))\equiv\tfrac{1}{2}\langle\beta|\big\{E^f_p(x_A(\tau)),E^f_q(x_B(\tau'))\big\}|\beta\rangle, \tag{39}$$

$$\chi^F_{pq}(x_A(\tau),x_B(\tau'))\equiv\tfrac{1}{2}\langle\beta|\big[E^f_p(x_A(\tau)),E^f_q(x_B(\tau'))\big]|\beta\rangle\theta(\tau-\tau')\ , \tag{40}$$

with $\theta(x)$ being the Heaviside function defined as $\theta(x)=1$ if $x>0$ and 0 if $x\leqslant 0$, and the symmetric and antisymmetric statistical functions of the two atoms in their ground states $C^\xi_{pq}(\tau,\tau')$ and $\chi^\xi_{pq}(\tau,\tau')$ by

$$C^\xi_{pq}(\tau,\tau')\equiv\tfrac{1}{2}\langle g_\xi|\big\{\mu^f_{\xi,p}(\tau),\mu^f_{\xi,q}(\tau')\big\}|g_\xi\rangle, \tag{41}$$

$$\chi^\xi_{pq}(\tau,\tau')\equiv\tfrac{1}{2}\langle g_\xi|\big[\mu^f_{\xi,p}(\tau),\mu^f_{\xi,q}(\tau')\big]|g_\xi\rangle, \tag{42}$$

which can be further simplified to

$$C^\xi_{pq}(\tau,\tau')=\frac{1}{2}\sum_{e_\xi}[\langle g_\xi|\mu_{\xi,p}(0)|e_\xi\rangle\langle e_\xi|\mu_{\xi,q}(0)|g_\xi\rangle e^{i\omega^\xi_{ge}(\tau-\tau')}+\langle g_\xi|\mu_{\xi,q}(0)|e_\xi\rangle\langle e_\xi|\mu_{\xi,p}(0)|g_\xi\rangle e^{-i\omega^\xi_{ge}(\tau-\tau')}], \tag{43}$$

$$\chi^\xi_{pq}(\tau,\tau')=\frac{1}{2}\sum_{e_\xi}[\langle g_\xi|\mu_{\xi,p}(0)|e_\xi\rangle\langle e_\xi|\mu_{\xi,q}(0)|g_\xi\rangle e^{i\omega^\xi_{ge}(\tau-\tau')}-\langle g_\xi|\mu_{\xi,q}(0)|e_\xi\rangle\langle e_\xi|\mu_{\xi,p}(0)|g_\xi\rangle e^{-i\omega^\xi_{ge}(\tau-\tau')}] \tag{44}$$

with $\omega^\xi_{ge}=\omega^\xi_g-\omega^\xi_e$ and the sum being over the complete set of atomic states. Note that in obtaining Eqs. (37) and (38), we have also used the following relation:

$$\langle\beta|E^f_p(x_1)E^f_q(x_2)E^f_r(x_3)E^f_s(x_4)|\beta\rangle=\langle\beta|E^f_p(x_1)E^f_q(x_2)|\beta\rangle\langle\beta|E^f_r(x_3)E^f_s(x_4)|\beta\rangle+\langle\beta|E^f_p(x_1)E^f_r(x_3)|\beta\rangle\langle\beta|E^f_q(x_2)E^f_s(x_4)|\beta\rangle$$
$$+\langle\beta|E^f_p(x_1)E^f_s(x_4)|\beta\rangle\langle\beta|E^f_q(x_2)E^f_r(x_3)|\beta\rangle, \tag{45}$$

which simplifies every four-point correlation function of the field in the thermal state $|\beta\rangle$ into a product of two two-point correlation functions.

It is worth pointing out now that since the effective Hamiltonians Eqs. (34) and (36) are derived by separating contributions from the couplings of the free field and the source field with the induced dipole moments to the average variation rate of the Hamiltonian of the two-atom system, the term "thermal fluctuation (radiation reaction) contribution" refers to the contribution resulting from the interaction between the free (source) field and the atoms with induced dipole moments, regardless of whether they are induced by the free field or by radiative fields of the atoms [see Eqs. (18) and (19)]. Noteworthily, the radiation reaction contribution at the fourth order is not exactly the same as that at the second order since it now also contains the contribution of the source radiation (radiative fields) from one atom on the other rather than just that of radiative fields from the dipole moments induced only by the free fields at the second order (see Refs. [7,8] for instances). In this sense, one may think that "source-radiation contribution" may be an appropriate label too. We choose to hold on to the original "radiation reaction contribution" for the following two reasons. First, in our derivation of the fourth-order DDC formalism, the two atoms are treated as a whole, and as a result, the action of the radiative field emitted from one atom on the other can be understood as the radiation reaction of the whole two-atom system. So, it is still legitimate to call it "radiation reaction contribution" in this sense. Second, it may be conducive to avoiding misunderstanding about the origin of the source field, since it conveys clearly that the back-reaction of the radiative fields is generated by induced dipole moments of the atoms rather than by possible intrinsic permanent ones.

Finally, a sum of the tf and rr contributions, Eqs. (37) and (38), leads to the total interaction energy: $(\delta E)_{\text{tot}}=(\delta E)_{\text{tf}}+(\delta E)_{\text{rr}}$. So, the interatomic interaction energy of two ground-state atoms, which is a fourth-order effect, is generally ascribed to joint contributions of the field fluctuations and the radiation reaction of atoms, and this is in sharp contrast to the resonance interaction energy of two atoms in the symmetric or antisymmetric entangled state which is a second-order effect and totally caused by the radiation reaction of atoms but irrelevant to the field fluctuations [26–29].

III. INTERATOMIC INTERACTION ENERGY OF TWO STATIC GROUND-STATE ATOMS IN A THERMAL BATH IN A FREE SPACE

As an application of the generic fourth-order DDC formalism derived in the preceding section, we now concretely calculate the interaction energy of two ground-state two-level atoms in a thermal bath. The trajectories of the two atoms are given by

$$t_A = t_B = \tau, \quad x_A = x_B = 0, \quad y_A = y_B = 0, \quad z_A = 0,\ z_B = L \tag{46}$$

with $L > 0$ being the interatomic separation.

To compute the tf and rr contributions, we have to first calculate the symmetric correlation function and the linear susceptibility of the field. For this purpose, we first evaluate the two-point correlation function of the electric field $\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle$. Applying the expansion of $\boldsymbol{E}^f(x)$ in Eq. (12) to $\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle$, we obtain, after some simplifications (see Appendix B),

$$\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle = -\frac{1}{4\pi^2}(\partial_0\partial_0'\delta_{pq} - \partial_p\partial_q')\sum_{m=-\infty}^{\infty}\frac{1}{(\Delta t - im\beta - i\varepsilon)^2 - |\Delta\mathbf{x}|^2} \tag{47}$$

with $\Delta t = t - t'$, $|\Delta\mathbf{x}| = \sqrt{(x-x')^2 + (y-y')^2 + (z-z')^2}$, and ε a positive infinitesimal. Combining this two-point function and the definitions of the symmetric correlation function and the linear susceptibility of the field [Eqs. (39) and (40)] and performing the Fourier transform, we arrive at

$$C_{pq}^F(x_A(\tau), x_B(\tau')) = \frac{1}{4\pi^2L^3}\int_0^\infty d\omega'\,\frac{e^{\beta\omega'}+1}{e^{\beta\omega'}-1}\mathcal{G}_{pq}(\omega'L)\cos[\omega'(\tau-\tau')], \tag{48}$$

$$\chi_{pq}^F(x_A(\tau), x_B(\tau')) = -\frac{i}{4\pi^2L^3}\int_0^\infty d\omega'\,\mathcal{G}_{pq}(\omega'L)\sin[\omega'(\tau-\tau')]\theta(\tau-\tau') \tag{49}$$

with

$$\begin{cases}\mathcal{G}_{xx}(x) = \mathcal{G}_{yy}(x) = x\cos x + (x^2-1)\sin x,\\ \mathcal{G}_{zz}(x) = -2x\cos x + 2\sin x,\end{cases} \tag{50}$$

and all other components not listed vanishing.

For simplicity, we now assume that the dipole transition matrix elements of the atoms are real, i.e., $\langle g_\xi|\mu_{\xi,p}(0)|e_\xi\rangle = \langle e_\xi|\mu_{\xi,p}(0)|g_\xi\rangle$, and denote them by $(\mu_p^\xi)_{ge}$. Then the statistical functions of the atoms, Eqs. (43) and (44), are further simplified to

$$C_{pq}^\xi(\tau,\tau') = \big(\mu_p^\xi\big)_{ge}\big(\mu_q^\xi\big)_{ge}\cos[\omega_\xi(\tau-\tau')], \tag{51}$$

$$\chi_{pq}^\xi(\tau,\tau') = -i\big(\mu_p^\xi\big)_{ge}\big(\mu_q^\xi\big)_{ge}\sin[\omega_\xi(\tau-\tau')]. \tag{52}$$

Before evaluating the tf and rr contributions, let us note that we are considering the interatomic interaction during the time interval $\Delta\tau = \tau - \tau_0$ which is assumed to be much larger than the correlation time τ_c of the fluctuating fields adapted to the two-atom system and much less than the relaxation time of the atoms for the DDC approach to be applicable [8,48]. Once these conditions are fulfilled, a large enough $\Delta\tau$ as compared with τ_c can be treated as infinity for computational convenience. We note that such a treatment of integrals over $\Delta\tau$ has been adopted in the existing extensive studies of the radiative properties of atoms in various circumstances with the DDC formalism [11–25,49–52]. Now we substitute Eqs. (48), (49), (51), and (52) into Eqs. (37) and (38), and perform the triple integrals with respect to τ_1, τ_2, and τ_3 for an infinitely long time interval $\Delta\tau$ to find

$$(\delta E)_{\mathrm{tf}} = -\frac{1}{4\pi^4L^6}\sum_{p,q,r,s}\big(\mu_p^A\big)_{ge}\big(\mu_q^B\big)_{ge}\big(\mu_r^A\big)_{ge}\big(\mu_s^B\big)_{ge}\int_0^\infty d\omega_1\int_0^\infty d\omega_2\coth(\beta\omega_1/2)\frac{\omega_A\omega_B\omega_2\,\mathcal{G}_{pq}(\omega_1L)\mathcal{G}_{rs}(\omega_2L)}{\big(\omega_1^2-\omega_A^2\big)\big(\omega_1^2-\omega_B^2\big)\big(\omega_2^2-\omega_1^2\big)} \tag{53}$$

and

$$\begin{aligned}(\delta E)_{\mathrm{rr}} = {}& -\frac{1}{4\pi^4L^6}\sum_{p,q,r,s}\big(\mu_p^A\big)_{ge}\big(\mu_q^B\big)_{ge}\big(\mu_r^A\big)_{ge}\big(\mu_s^B\big)_{ge}\int_0^\infty d\omega_1\int_0^\infty d\omega_2\,\mathcal{G}_{pq}(\omega_1L)\mathcal{G}_{rs}(\omega_2L)\\ &\times\left[\frac{2\omega_1\omega_2\big(\omega_1^2-\omega_A^2-\omega_A\omega_B-\omega_B^2\big)}{(\omega_A+\omega_B)\big(\omega_1^2-\omega_A^2\big)\big(\omega_1^2-\omega_B^2\big)\big(\omega_2^2-\omega_1^2\big)} + \frac{\omega_A\omega_B\omega_2\coth(\beta\omega_1/2)}{\big(\omega_1^2-\omega_A^2\big)\big(\omega_1^2-\omega_B^2\big)\big(\omega_2^2-\omega_1^2\big)}\right].\end{aligned} \tag{54}$$

Assuming that the atoms are isotropically polarizable, then we have [38]

$$\big(\mu_p^\xi\big)_{ge}\big(\mu_q^\xi\big)_{ge} = \delta_{pq}\big|\mu_p^\xi\big|_{ge}^2 = \tfrac{1}{3}\delta_{pq}|\boldsymbol{\mu}^\xi|_{ge}^2. \tag{55}$$

Using this relation together with Eq. (50) in Eqs. (53) and (54), and performing the ω_2 integration with the technique of contour integration and the residue theorem, the tf and rr contributions are finally simplified to

$$\begin{aligned}(\delta E)_{\mathrm{tf}} = {}& \frac{\mu_A^2\mu_B^2}{144\pi^2\left(\omega_A^2-\omega_B^2\right)L^6}\{\omega_B[\mathcal{P}(\omega_A L)\sin(2\omega_A L)-\mathcal{Q}(\omega_A L)\cos(2\omega_A L)]-\omega_A[\mathcal{P}(\omega_B L)\sin(2\omega_B L)-\mathcal{Q}(\omega_B L)\cos(2\omega_B L)]\}\\ &-\frac{\mu_A^2\mu_B^2}{36\pi^3L^6}\int_0^\infty du\,\frac{\omega_A\omega_B[\mathcal{P}(uL)\cos(2uL)+\mathcal{Q}(uL)\sin(2uL)]}{\left(\omega_A^2-u^2\right)\left(\omega_B^2-u^2\right)(e^{\beta u}-1)}-\frac{\mu_A^2\mu_B^2}{72\pi^3L^6}\int_0^\infty du\,\frac{\omega_A\omega_B[i\mathcal{P}(iuL)+\mathcal{Q}(iuL)]}{\left(\omega_A^2+u^2\right)\left(\omega_B^2+u^2\right)}e^{-2uL}\end{aligned} \tag{56}$$

and

$$\begin{aligned}(\delta E)_{\mathrm{rr}} = {}& -\frac{\mu_A^2\mu_B^2}{144\pi^2\left(\omega_A^2-\omega_B^2\right)L^6}\{\omega_B[\mathcal{P}(\omega_A L)\sin(2\omega_A L)-\mathcal{Q}(\omega_A L)\cos(2\omega_A L)]-\omega_A[\mathcal{P}(\omega_B L)\sin(2\omega_B L)-\mathcal{Q}(\omega_B L)\cos(2\omega_B L)]\}\\ &-\frac{\mu_A^2\mu_B^2}{36\pi^3L^6}\int_0^\infty du\,\frac{\omega_A\omega_B[\mathcal{P}(uL)\cos(2uL)+\mathcal{Q}(uL)\sin(2uL)]}{\left(\omega_A^2-u^2\right)\left(\omega_B^2-u^2\right)(e^{\beta u}-1)}-\frac{\mu_A^2\mu_B^2}{72\pi^3L^6}\int_0^\infty du\,\frac{\omega_A\omega_B[i\mathcal{P}(iuL)+\mathcal{Q}(iuL)]}{\left(\omega_A^2+u^2\right)\left(\omega_B^2+u^2\right)}e^{-2uL}\end{aligned} \tag{57}$$

with $\mathcal{P}(x) = -6x + 2x^3$ and $\mathcal{Q}(x) = 3 - 5x^2 + x^4$. Hereafter, we abbreviate $|\boldsymbol{\mu}^\xi|_{ge}^2$ by μ_ξ^2.

Adding Eqs. (56) and (57) up, we obtain the total interatomic interaction energy of the two ground-state atoms in a thermal bath at temperature T (β^{-1}):

$$(\delta E)_{\mathrm{tot}} = -\frac{\mu_A^2\mu_B^2}{18\pi^3L^6}\int_0^\infty du\,\frac{\omega_A\omega_B[\mathcal{P}(uL)\cos(2uL)+\mathcal{Q}(uL)\sin(2uL)]}{\left(\omega_A^2-u^2\right)\left(\omega_B^2-u^2\right)(e^{\beta u}-1)}-\frac{\mu_A^2\mu_B^2}{36\pi^3L^6}\int_0^\infty du\,\frac{\omega_A\omega_B[i\mathcal{P}(iuL)+\mathcal{Q}(iuL)]}{\left(\omega_A^2+u^2\right)\left(\omega_B^2+u^2\right)}e^{-2uL}. \tag{58}$$

Obviously, the total interaction energy contains two terms, with the temperature-independent term being the interaction energy of two static ground-state two-level atoms in vacuum and the temperature-dependent term representing the thermal corrections.

As analytical results of the above three equations are difficult to obtain, we discuss in the following the total interatomic interaction energy as well as the tf and rr contributions to it in two limiting cases, i.e., the low-temperature case in which the temperature of the thermal bath T is much smaller than the transition frequency of the atoms ω_ξ and the high-temperature case in which $T \gg \omega_\xi$. For simplicity, we assume that the two atoms are identical with the same energy gap ω.

A. Low-temperature limit $T \ll \omega$

The low-temperature limit, $T \ll \omega$ or equivalently $\beta \gg \lambda$ with $\beta = T^{-1}$, can be further divided into three typical subregions: $L \ll \lambda \ll \beta$, which is a van der Waals subregion, and $\lambda \ll L \ll \beta$ and $\lambda \ll \beta \ll L$, which are two Casimir-Polder subregions.[2]

1. The van der Waals subregion $L \ll \lambda \ll \beta$

The tf contribution Eq. (56) in the $L \ll \lambda \ll \beta$ subregion can be approximated by

$$\begin{aligned}(\delta E)_{\mathrm{tf}} \simeq {}& -\frac{7\omega^4}{192\pi^3L^3}+\frac{11\omega^6\ln(2\omega L)}{120\pi^3L}\\ &-\frac{(607\text{–}660\gamma)\omega^6}{7200\pi^3L}-\frac{11\pi^3T^6}{945L}\end{aligned} \tag{59}$$

with γ the Euler constant. Here and after, we express the tf and rr contributions as well as the total interatomic interaction energy in units of $\alpha(A)\alpha(B)$ with $\alpha(\xi) \equiv 2\mu_\xi^2/3\omega$ the polarizability of atom ξ. So, the tf contribution leads to a separation dependence of $\sim L^{-3}$ in the leading order for the van der Waals interaction energy. The rr contribution Eq. (57) is, however, approximated by

$$\begin{aligned}(\delta E)_{\mathrm{rr}} \simeq {}& -\frac{3\omega}{64\pi^2L^6}+\frac{\omega^3}{64\pi^2L^4}-\frac{7\omega^4\alpha^2}{192\pi^3L^3}\\ &+\frac{3\omega^5}{64\pi^2L^2}+\frac{11\omega^6\ln(2\omega L)}{120\pi^3L}\\ &-\frac{(607\text{–}660\gamma)\omega^6}{7200\pi^3L}-\frac{11\pi^3T^6}{945L},\end{aligned} \tag{60}$$

which results in a separation-dependence of $\sim L^{-6}$ for the van der Waals interaction energy.

From the above two equations we can see that the rr contribution dominates over the tf contribution and thermal effects on both the tf and rr contributions (referring to those temperature-dependent terms) are very small since they appear at the fourth order at most for the tf contribution and at even higher orders for the rr contribution. As a result, the interatomic van der Waals interaction energy mainly comes from the rr contribution which, in the leading order, gives the total interaction energy of the two atoms in vacuum, i.e.,

$$(\delta E)_{\mathrm{tot}} \simeq (\delta E)_{\mathrm{rr}} \simeq -\frac{3\omega}{64\pi^2L^6}. \tag{61}$$

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[2] By convention, we refer to regions with $L \ll \lambda$ as van der Waals regions and those with $L \gg \lambda$ as Casimir-Polder regions.

2. The Casimir-Polder subregion $\lambda \ll L \ll \beta$

In the $\lambda \ll L \ll \beta$ region, the tf and rr contributions to the Casimir-Polder interaction energy can respectively be simplified to

$$(\delta E)_{\rm tf} \simeq \frac{\omega^6 g_1(\omega L)}{64\pi^2 L}\sin(2\omega L) + \frac{\omega^5 h_1(\omega L)}{128\pi^2 L^2}\cos(2\omega L) - \frac{23}{128\pi^3 L^7} - \frac{11\pi^3 T^6}{945 L}, \tag{62}$$

$$(\delta E)_{\rm rr} \simeq -\frac{\omega^6 g_1(\omega L)}{64\pi^2 L}\sin(2\omega L) - \frac{\omega^5 h_1(\omega L)}{128\pi^2 L^2}\cos(2\omega L) - \frac{23}{128\pi^3 L^7} - \frac{11\pi^3 T^6}{945 L} \tag{63}$$

with $g_1(x) = 1 - 3x^{-2} + 3x^{-4}$ and $h_1(x) = 1 - 7x^{-2} + 3x^{-4}$. Note that each of the above two contributions includes both oscillatory and nonoscillatory terms with the amplitudes of oscillatory terms much greater than the nonoscillatory terms, and thus an attractive or repulsive and even vanishing interaction force could result from the tf or rr contribution. Jointly, they however yield a monotonic behavior for the total interaction energy:

$$(\delta E)_{\rm tot} \simeq -\frac{23}{64\pi^3 L^7} - \frac{22\pi^3 T^6}{945 L}. \tag{64}$$

The first term on the right, which is identical to that of the two atoms in vacuum, exhibits a separation dependence of L^{-7} which is one order higher than that of the van der Waals interaction energy in vacuum and that in the $L \ll \lambda \ll \beta$ subregion in the thermal bath. This is the effect of retardation. The second term is much smaller than the first term and this indicates that thermal effects in this case lead to a slight enhancement of the interaction energy.

3. The Casimir-Polder subregion $\lambda \ll \beta \ll L$

When the interatomic separation is very large, i.e., $\lambda \ll \beta \ll L$, the tf and rr contributions are respectively approximated by

$$(\delta E)_{\rm tf} \simeq \frac{\omega^6 g_1(\omega L)}{64\pi^2 L}\sin(2\omega L) + \frac{\omega^5 h_1(\omega L)}{128\pi^2 L^2}\cos(2\omega L) - \frac{3T}{32\pi^2 L^6} \tag{65}$$

and

$$(\delta E)_{\rm rr} \simeq -\frac{\omega^6 g_1(\omega L)}{64\pi^2 L}\sin(2\omega L) - \frac{\omega^5 h_1(\omega L)}{128\pi^2 L^2}\cos(2\omega L) - \frac{3T}{32\pi^2 L^6}. \tag{66}$$

Similar to their counterparts in the $\lambda \ll L \ll \beta$ subregion, here both the tf and rr contributions contain oscillatory and nonoscillatory terms. However, there is also a significant distinction, i.e., the leading-order nonoscillatory terms in the above two equations are temperature dependent while those in Eqs. (62) and (63) are temperature independent.

As the oscillatory terms in the tf and rr contributions cancel out perfectly, the total interaction energy turns out to be

$$(\delta E)_{\rm tot} \simeq -\frac{3T}{16\pi^2 L^6}, \tag{67}$$

which is temperature dependent in the leading order. This result is in sharp contrast to that in subregions where $L \ll \lambda \ll \beta$ and $\lambda \ll L \ll \beta$ since there the interaction energy in the leading order is temperature independent, meaning that thermal effects are negligible.

Now some comments are in order. Let us note that there has already been an analysis on the interaction energy in the low-temperature limit $T \ll \omega$ [42], and our results above are fully consistent with those of Ref. [42]. Besides, the TL^{-6} behavior of the interaction energy in the $\lambda \ll \beta \ll L$ Casimir-Polder subregion has also been reported in other works [43–46]. However, with the fourth-order DDC formalism, we are able to identify how the field fluctuations and the radiation reaction of the atoms separately contribute to the total interaction energy.

B. High-temperature limit $T \gg \omega$

Let us now turn our attention to the high-temperature limit, i.e., $T \gg \omega$ or equivalently $\beta \ll \lambda$. Here we also have three interatomic separation subregions: two van der Waals subregions, i.e., $L \ll \beta \ll \lambda$ and $\beta \ll L \ll \lambda$, and one Casimir-Polder subregion, i.e., $\beta \ll \lambda \ll L$. We next analyze the interatomic interaction energy in these three subregions.

1. The van der Waals subregion $L \ll \beta \ll \lambda$

In the $L \ll \beta \ll \lambda$ subregion, the tf and rr contributions are respectively given by

$$(\delta E)_{\rm tf} \simeq -\frac{7\omega^4}{192\pi^3 L^3} - \frac{11\omega^4 T^2}{120\pi^3 L}, \tag{68}$$

$$(\delta E)_{\rm rr} \simeq -\frac{3\omega}{64\pi^2 L^6} + \frac{\omega^3}{64\pi^2 L^4} - \frac{7\omega^4}{192\pi^3 L^3} - \frac{11\omega^4 T^2}{120\pi^3 L}, \tag{69}$$

and the total van der Waals interaction energy is given by

$$(\delta E)_{\rm tot} \simeq -\frac{3\omega}{64\pi^2 L^6} + \frac{\omega^3}{64\pi^2 L^4} - \frac{7\omega^4}{96\pi^3 L^3} - \frac{11\omega^4 T^2}{60\pi^3 L}. \tag{70}$$

Obviously, the total interaction energy mainly comes from the rr contribution as $(\delta E)_{\rm tf} \ll (\delta E)_{\rm rr}$. The thermal corrections to the tf and rr contributions appear at high orders and are thus negligible. Although thermal corrections here are still small, they however appear at orders lower than those in the van der Waals subregion in the low-temperature limit, i.e., $L \ll \lambda \ll \beta$ [please see Eqs. (59) and (60)], indicating that thermal effects in the $L \ll \beta \ll \lambda$ van der Waals subregion in the high-temperature limit are more significant than those in the $L \ll \lambda \ll \beta$ van der Waals subregion in the low-temperature limit.

2. The van der Waals subregion $\beta \ll L \ll \lambda$

In the second van der Waals subregion in the high-temperature limit, $\beta \ll L \ll \lambda$, the tf contribution is approximated by

$$(\delta E)_{\rm tf} \simeq -\frac{\omega^4 T}{32\pi^2 L^2} + \frac{7\omega^4}{192\pi^3 L^3}. \tag{71}$$

In sharp contrast to its counterpart in the first subregion in the high-temperature limit, i.e., Eq. (68), here the leading term is temperature dependent. Moreover, the second temperature-independent term changes sign as compared with its counterpart in the tf contribution in the $L \ll \lambda \ll \beta$ subregion in the low-temperature limit and that in the $L \ll \beta \ll \lambda$ subregion in the high-temperature limit. This sign change can also be regarded as a result of strong thermal effects.

Approximately, the rr contribution in this subregion is given by

$$(\delta E)_{\rm rr} \simeq -\frac{\omega^4 T}{32\pi^2 L^2} - \frac{3\omega}{64\pi^2 L^6} + \frac{\omega^3}{64\pi^2 L^4} + \frac{7\omega^4}{192\pi^3 L^3}, \tag{72}$$

and the leading term can be either the first or the second term depending on a more delicate relation among the three characteristic lengths L, λ, and β. To be specific, when $\beta \ll L \ll \sqrt[4]{\lambda^3\beta}$,

$$(\delta E)_{\rm rr} \simeq -\frac{3\omega}{64\pi^2 L^6}. \tag{73}$$

This is much larger than the tf contribution [Eq. (71)] and thus constitutes the dominant contribution to the total interaction energy, leading to $(\delta E)_{\rm tot} \sim (\delta E)_{\rm rr}$. This behavior is the same as that of the leading term of the total interaction energy in the $L \ll \lambda \ll \beta$ van der Waals region in the low-temperature limit and that in the $L \ll \beta \ll \lambda$ van der Waals subregion in the high-temperature limit [refer to Eqs. (61) and (70)].

When $\sqrt[4]{\lambda^3\beta} \ll L \ll \lambda$, the rr contribution becomes equally important as the tf contribution, so

$$(\delta E)_{\rm rr} \simeq (\delta E)_{\rm tf} \simeq -\frac{\omega^4 T}{32\pi^2 L^2}, \tag{74}$$

and as a result the total interaction energy is

$$(\delta E)_{\rm tot} \simeq -\frac{\omega^4 T}{16\pi^2 L^2}, \tag{75}$$

which is twice that of the tf or rr contribution alone and scales as $\sim TL^{-2}$. This behavior of the van der Waals interaction energy of two ground-state atoms in a thermal bath is a result that has not been reported before.

3. *The Casimir-Polder subregion $\beta \ll \lambda \ll L$*

When $\beta \ll \lambda \ll L$, which is a typical Casimir-Polder region, we obtain the following tf and rr contributions:

$$\begin{aligned}(\delta E)_{\rm tf} \simeq\ & \frac{T\omega^5 g_2(\omega L, TL)}{32\pi^2 L}\sin(2\omega L)\\ &+ \frac{T\omega^4 h_2(\omega L, TL)}{32\pi^2 L^2}\cos(2\omega L) - \frac{3T}{32\pi^2 L^6},\end{aligned} \tag{76}$$

$$\begin{aligned}(\delta E)_{\rm rr} \simeq\ & \frac{T\omega^5 g_3(\omega L, TL)}{32\pi^2 L}\sin(2\omega L)\\ &+ \frac{T\omega^4 h_3(\omega L, TL)}{32\pi^2 L^2}\cos(2\omega L) - \frac{3T}{32\pi^2 L^6},\end{aligned} \tag{77}$$

and the total interaction energy

$$\begin{aligned}(\delta E)_{\rm tot} \simeq\ & \frac{T\omega^5 g_4(\omega L, TL)}{16\pi^2 L}\sin(2\omega L)\\ &+ \frac{T\omega^4 h_4(\omega L, TL)}{16\pi^2 L^2}\cos(2\omega L) - \frac{3T}{16\pi^2 L^6},\end{aligned} \tag{78}$$

in which we have defined

$$\begin{cases} g_2(x,y) = 1 - 4x^{-2} + 6x^{-4} - \frac{1}{2}xy^{-1}(1 - 3x^{-2} + 3x^{-4}),\\ g_3(x,y) = 1 - 4x^{-2} + 6x^{-4} - \frac{3}{2}xy^{-1}(1 - 3x^{-2} + 3x^{-4}),\\ g_4(x,y) = 1 - 4x^{-2} + 6x^{-4} - xy^{-1}(1 - 3x^{-2} + 3x^{-4}),\end{cases} \tag{79}$$

and

$$\begin{cases} h_2(x,y) = 1 - 6x^{-2} + 3x^{-4} - \frac{1}{4}xy^{-1}(1 - 7x^{-2}),\\ h_3(x,y) = 1 - 6x^{-2} + 3x^{-4} - \frac{3}{4}xy^{-1}(1 - 7x^{-2}),\\ h_4(x,y) = 1 - 6x^{-2} + 3x^{-4} - \frac{1}{2}xy^{-1}(1 - 7x^{-2}).\end{cases} \tag{80}$$

Equations (76)–(78) show that not only the tf and rr contributions but also the total Casimir-Polder interaction energy of the two atoms in the $\beta \ll \lambda \ll L$ subregion contain both oscillatory and nonoscillatory terms. As the amplitudes of the oscillatory terms are much greater than the nonoscillatory terms, both the tf and rr contributions as well as the total interaction energy can be positive or negative and even vanishing, depending on the interatomic separation L and the temperature T. Noteworthily, this oscillatory behavior of the Casimir-Polder interaction energy in the high-temperature limit, which is superimposed on the monotonic TL^{-6} behavior, has not been reported before either.

Finally, it is worth pointing out that the $\sim TL^{-6}$ behavior of the interaction energy was obtained for the two atoms in a thermal bath when $L \gg \beta$ [43,45,46]. However, our results indicate that although the interaction energy indeed scales as TL^{-6} in the Casimir-Polder subregion $\lambda \ll \beta \ll L$ in the low-temperature limit [see our Eq. (67)], it is not a universal behavior for $L \gg \beta$. In fact, the actual behavior in the high-temperature limit depends on some delicate relations among L, β, and λ, and the interaction energy behaves differently in different subregions of $L \gg \beta$. To be specific, in the leading order, it displays a temperature-independent behavior of L^{-6} in $\beta \ll L \ll \sqrt[4]{\lambda^3\beta}$, and a temperature-dependent one of TL^{-2} in $\sqrt[4]{\lambda^3\beta} \ll L \ll \lambda$, and oscillates with the interatomic separation in $\beta \ll \lambda \ll L$. And only in $\beta \ll \lambda \ll L$ can the interaction energy be approximated by TL^{-6} for some special values of L and β such that the sum of the oscillatory terms in Eq. (78) vanishes or is comparatively very small.

IV. SUMMARY

In this paper, we have given a detailed derivation for the fourth-order DDC formalism for calculating the interaction energy between two ground-state multilevel atoms which are coupled with electromagnetic fields in a thermal bath. As an application of our generalized fourth-order DDC formalism, we concretely calculated the interaction energy of two static ground-state two-level atoms with the same transition frequency ω in a thermal bath at an arbitrary temperature T in an unbounded space, and analyzed in detail the behaviors of the total interaction energy as well as those of the contributions of the thermal field fluctuations and the radiation reaction of the atoms to it in both the low- and high-temperature limits. The main results are summarized as follows.

In the low-temperature limit in which $T \ll \omega$, there are three subregions, i.e., the $L \ll \lambda \ll \beta$ van der Waals subregion and two Casimir-Polder subregions, i.e., the regions where $\lambda \ll L \ll \beta$ and $\lambda \ll \beta \ll L$. We showed that the interaction energy behaves as $\sim L^{-6}$ in the $L \ll \lambda \ll \beta$ van der Waals subregion, as $\sim L^{-7}$ due to the effect of retardation in the first Casimir-Polder subregion, i.e., when $\lambda \ll L \ll \beta$, and as $\sim TL^{-6}$ in the second Casimir-Polder subregion, i.e., when $\lambda \ll \beta \ll L$. Although these behaviors of the total interaction energy in the low-temperature limit we obtained with the DDC formalism are perfectly consistent with what was derived with other approaches [42], our analysis shows that the interaction energy is mainly attributable to the radiation reaction of the atoms in the $L \ll \lambda \ll \beta$ van der Waals subregion while it is equally contributed by both the thermal field fluctuations and the radiation reaction of the atoms in two Casimir-Polder subregions.

In the high-temperature limit in which $T \gg \omega$, there are also three subregions, i.e., two van der Waals subregions, $L \ll \beta \ll \lambda$ and $\beta \ll L \ll \lambda$, and a Casimir-Polder subregion, $\beta \ll \lambda \ll L$. We find that the total interaction energy scales as $\sim L^{-6}$ in the first van der Waals subregion $L \ll \beta \ll \lambda$ and is mainly ascribed to the rr contribution. The second van der Waals subregion $\beta \ll L \ll \lambda$ can be further divided into two sub-subregions $\beta \ll L \ll \sqrt[4]{\lambda^3\beta}$ and $\sqrt[4]{\lambda^3\beta} \ll L \ll \lambda$. In the former, the interaction energy still scales as $\sim L^{-6}$ and is also mainly ascribed to the rr contribution, while in the latter it displays a unique behavior of $\sim TL^{-2}$ as a result of equal tf and rr contributions. In the Casimir-Polder subregion, i.e., $\beta \ll \lambda \ll L$, both the tf and rr contributions as well as the total interaction energy oscillate with interatomic separation, resulting in an either positive or negative and even vanishing total interaction energy depending on the interatomic separation and the temperature. The TL^{-2} scaling when $\sqrt[4]{\lambda^3\beta} \ll L \ll \lambda$ and the oscillatory behavior when $\beta \ll \lambda \ll L$ are two new findings of the present paper.

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APPENDIX A: THE FIRST- TO THIRD-ORDER SOURCE OPERATORS OF THE ATOMS AND THE FIELD

In this Appendix, we show how to derive the source operators of the atoms and the field by iteration in a perturbative approach. For the source operators of the atoms, we only take those of atom A as an example, since the source operators of atom B can be easily obtained by exchanging the indices A and B.

First, for the first order, it is straightforward to deduce from Eqs. (10) and (11) that the first-order operator in $\sigma_{mn}^{A,s}(\tau)$ and that in $a_{\mathbf{k},\nu}^{s}(t(\tau))$ are

$$\sigma_{mn}^{A,(1)}(\tau) = -\frac{i}{2}\sum_{p}\int_{\tau_0}^{\tau} d\tau_1 \big\{E_p^f(x_A(\tau_1)), \big[\mu_{A,p}^f(\tau_1), \sigma_{mn}^{A,f}(\tau)\big]\big\} \tag{A1}$$

and

$$a_{\mathbf{k},\nu}^{(1)}(t(\tau)) = -\frac{i}{2}\sum_{\xi,q}\int_{\tau_0}^{\tau} d\tau_1 \big\{\mu_{\xi,q}^f(\tau_1), \big[E_q^f(x_\xi(\tau_1), a_{\mathbf{k},\nu}^f(t(\tau))\big]\big\} \tag{A2}$$

where $\boldsymbol{\mu}_{\xi}^{f}(\tau) \equiv \sum_{m,n}^{m\neq n} (\boldsymbol{\mu}_\xi)_{mn}\sigma_{mn}^{\xi,f}(\tau)$. Then an application of the above $a_{\mathbf{k},\nu}^{(1)}(t(\tau))$ to the second formula in Eq. (12) gives rise to the following first-order source field operator:

$$\mathbf{E}^{(1)}(x(\tau)) = -\frac{i}{2}\sum_{\xi,q}\int_{\tau_0}^{\tau} d\tau_1 \big\{\mu_{\xi,q}^f(\tau_1), \big[E_q^f(x_\xi(\tau_1), \mathbf{E}^f(x(\tau))\big]\big\}. \tag{A3}$$

Second, for the second order, according to the formulas in the second line in Eqs. (10) and (11), we have

$$\begin{aligned}\sigma_{mn}^{A,(2)}(\tau) = & -\frac{i}{2}\sum_{p}\int_{\tau_0}^{\tau} d\tau_1 \big\{E_p^{(1)}(x_A(\tau_1)), \big[\mu_{A,p}^f(\tau_1), \sigma_{mn}^{A,f}(\tau)\big]\big\} - \frac{i}{2}\sum_{p}\int_{\tau_0}^{\tau} d\tau_1 \big\{E_p^f(x_A(\tau_1)), \big[\mu_{A,p}^{(1)}(\tau_1), \sigma_{mn}^{A,f}(\tau)\big]\big\} \\ & -\frac{i}{2}\sum_{p}\int_{\tau_0}^{\tau} d\tau_1 \big\{E_p^f(x_A(\tau_1)), \big[\mu_{A,p}^f(\tau_1), \sigma_{mn}^{A,(1)}(\tau)\big]\big\}\end{aligned} \tag{A4}$$

and

$$
\begin{aligned}
a^{(2)}_{\mathbf{k},\nu}(t(\tau)) = {} & -\frac{i}{2}\sum_{\xi,q}\int_{\tau_0}^{\tau} d\tau_1\big\{\mu^{(1)}_{\xi,q}(\tau_1), \big[E^f_q(x_\xi(\tau_1)), a^f_{\mathbf{k},\nu}(t(\tau))\big]\big\} - \frac{i}{2}\sum_{\xi,q}\int_{\tau_0}^{\tau} d\tau_1\big\{\mu^f_{\xi,q}(\tau_1), \big[E^{(1)}_q(x_\xi(\tau_1), a^f_{\mathbf{k},\nu}(t(\tau))\big]\big\} \\
& -\frac{i}{2}\sum_{\xi,q}\int_{\tau_0}^{\tau} d\tau_1\big\{\mu^f_{\xi,q}(\tau_1), \big[E^f_q(x_\xi(\tau_1), a^{(1)}_{\mathbf{k},\nu}(t(\tau))\big]\big\}.
\end{aligned} \tag{A5}
$$

Then the use of the relation Eq. (20) and the first-order source operators Eqs. (A1)–(A3) in the above two equations leads to

$$
\begin{aligned}
\sigma^{A,(2)}_{mn}(\tau) = {} & -\frac{1}{2}\sum_{p,q}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2 \big[E^f_q(x_A(\tau_2)), E^f_p(x_A(\tau_1))\big]\big\{\mu^f_{A,q}(\tau_2), \big[\mu^f_{A,p}(\tau_1), \sigma^{A,f}_{mn}(\tau)\big]\big\} \\
& -\frac{1}{2}\sum_{p,q}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2 \big\{E^f_q(x_A(\tau_2)), E^f_p(x_A(\tau_1))\big\}\big[\mu^f_{A,q}(\tau_2), [\mu^f_{A,p}(\tau_1), \sigma^{A,f}_{mn}(\tau)]\big] \\
& -\sum_{p,q}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2 \big[E^f_q(x_B(\tau_2)), E^f_p(x_A(\tau_1))\big]\mu^f_{B,q}(\tau_2)\big[\mu^f_{A,p}(\tau_1), \sigma^{A,f}_{mn}(\tau)\big]
\end{aligned} \tag{A6}
$$

and

$$
a^{(2)}_{\mathbf{k},\nu}(t(\tau)) = -\sum_{\xi,q,r}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2\, E^f_r(x_\xi(\tau_2))[E^f_q(x_\xi(\tau_1)), a^f_{\mathbf{k},\nu}(t(\tau))][\mu^f_{\xi,r}(\tau_2), \mu^f_{\xi,q}(\tau_1)]. \tag{A7}
$$

Applying the above $a^{(2)}_{\mathbf{k},\nu}(t(\tau))$ in the second formula in Eq. (12) yields the following second-order source field operator:

$$
\mathbf{E}^{(2)}(x(\tau)) = -\sum_{\xi,q,r}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2\, E^f_r(x_\xi(\tau_2))\big[E^f_q(x_\xi(\tau_1)), \mathbf{E}^f(x(\tau))\big]\big[\mu^f_{\xi,r}(\tau_2), \mu^f_{\xi,q}(\tau_1)\big]. \tag{A8}
$$

Third, for the third order, we first expand $\sigma^{A,(3)}_{mn}(\tau)$ and $a^{(3)}_{\mathbf{k},\nu}(t(\tau))$, according to formulas in the second line in Eqs. (10) and (11), as

$$
\begin{aligned}
\sigma^{A,(3)}_{mn}(\tau) = {} & -\frac{i}{2}\sum_{p}\int_{\tau_0}^{\tau} d\tau_1\big\{\big\{E^{(2)}_p(x_A(\tau_1)), \big[\mu^f_{A,p}(\tau_1), \sigma^{A,f}_{mn}(\tau)\big]\big\} + \big\{E^f_p(x_A(\tau_1)), \big[\mu^{(2)}_{A,p}(\tau_1), \sigma^{A,f}_{mn}(\tau)\big]\big\} \\
& + \big\{E^f_p(x_A(\tau_1)), \big[\mu^f_{A,p}(\tau_1), \sigma^{A,(2)}_{mn}(\tau)\big]\big\} + \big\{E^{(1)}_p(x_A(\tau_1)), \big[\mu^{(1)}_{A,p}(\tau_1), \sigma^{A,f}_{mn}(\tau)\big]\big\} \\
& + \big\{E^{(1)}_p(x_A(\tau_1)), \big[\mu^f_{A,p}(\tau_1), \sigma^{A,(1)}_{mn}(\tau)\big]\big\} + \big\{E^f_p(x_A(\tau_1)), \big[\mu^{(1)}_{A,p}(\tau_1), \sigma^{A,(1)}_{mn}(\tau)\big]\big\}\big\}
\end{aligned} \tag{A9}
$$

and

$$
\begin{aligned}
a^{(3)}_{\mathbf{k},\nu}(t(\tau)) = {} & -\frac{i}{2}\sum_{\xi,q}\int_{\tau_0}^{\tau} d\tau_1\big\{\big\{\mu^{(2)}_{\xi,q}(\tau_1), \big[E^f_q(x_\xi(\tau_1)), a^f_{\mathbf{k},\nu}(t(\tau))\big]\big\} + \big\{\mu^f_{\xi,q}(\tau_1), \big[E^{(2)}_q(x_\xi(\tau_1)), a^f_{\mathbf{k},\nu}(t(\tau))\big]\big\} \\
& + \big\{\mu^f_{\xi,q}(\tau_1), \big[E^f_q(x_\xi(\tau_1)), a^{(2)}_{\mathbf{k},\nu}(t(\tau))\big]\big\} + \big\{\mu^{(1)}_{\xi,q}(\tau_1), \big[E^{(1)}_q(x_\xi(\tau_1)), a^f_{\mathbf{k},\nu}(t(\tau))\big]\big\} \\
& + \big\{\mu^{(1)}_{\xi,q}(\tau_1), \big[E^f_q(x_\xi(\tau_1)), a^{(1)}_{\mathbf{k},\nu}(t(\tau))\big]\big\} + \big\{\mu^f_{\xi,q}(\tau_1), \big[E^{(1)}_q(x_\xi(\tau_1)), a^{(1)}_{\mathbf{k},\nu}(t(\tau))\big]\big\}\big\}.
\end{aligned} \tag{A10}
$$

Then using the zeroth- to second-order expressions of all operators we have derived so far in the above two equations, we obtain

$$
\begin{aligned}
\sigma^{A,(3)}_{mn}(\tau) = {} & i\sum_{p,q,r}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2 \int_{\tau_0}^{\tau_2} d\tau_3\, E^f_r(x_B(\tau_3))\big[E^f_q(x_B(\tau_2)), E^f_p(x_A(\tau_1))\big]\big[\mu^f_{B,r}(\tau_3), \mu^f_{B,q}(\tau_2)\big] \\
& \times\big[\mu^f_{A,p}(\tau_1), \sigma^{A,f}_{mn}(\tau)\big] + \text{ terms of the zeroth and first power of } \boldsymbol{\mu}^f_B
\end{aligned} \tag{A11}
$$

and

$$
\begin{aligned}
a^{(3)}_{\mathbf{k},\nu}(t(\tau)) = {} & i\sum_{q,r,s}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2 \int_{\tau_0}^{\tau_1} d\tau_3 \,\big[E^f_r(x_B(\tau_2)), E^f_q(x_A(\tau_1))\big]\big[E^f_s(x_B(\tau_3)), a^f_{\mathbf{k},\nu}(t(\tau))\big]\mu^f_{A,q}(\tau_1)\big[\mu^f_{B,r}(\tau_2), \mu^f_{B,s}(\tau_3)\big] \\
& + i\sum_{q,r,s}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2 \int_{\tau_0}^{\tau_2} d\tau_3 \,\big[E^f_r(x_B(\tau_2)), E^f_q(x_A(\tau_1))\big]\big[E^f_s(x_B(\tau_3)), a^f_{\mathbf{k},\nu}(t(\tau))\big]\mu^f_{A,q}(\tau_1)\big[\mu^f_{B,s}(\tau_3), \mu^f_{B,r}(\tau_2)\big] \\
& + i\sum_{q,r,s}\int_{\tau_0}^{\tau} d\tau_1 \int_{\tau_0}^{\tau_1} d\tau_2 \int_{\tau_0}^{\tau_2} d\tau_3 \,\big[E^f_q(x_A(\tau_1)), E^f_s(x_B(\tau_3))\big]\big[E^f_r(x_B(\tau_2)), a^f_{\mathbf{k},\nu}(t(\tau))\big]\mu^f_{A,q}(\tau_1)\big[\mu^f_{B,s}(\tau_3), \mu^f_{B,r}(\tau_2)\big]
\end{aligned}
$$

$$+i\sum_{q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\big[E_s^f(x_A(\tau_3)),E_r^f(x_B(\tau_2))\big]\big[E_q^f(x_B(\tau_1)),a_{\mathbf{k},\nu}^f(t(\tau))\big]\mu_{A,s}^f(\tau_3)\big[\mu_{B,r}^f(\tau_2),\mu_{B,q}^f(\tau_1)\big]$$
$$+\text{terms of the zeroth, first, and third power of }\boldsymbol{\mu}_B^f. \tag{A12}$$

Accordingly, the third-order source field operator follows:

$$\begin{aligned}\mathbf{E}^{(3)}(x(\tau))={}&i\sum_{q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_1}d\tau_3\,\big[E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big]\big[E_s^f(x_B(\tau_3)),\mathbf{E}^f(x(\tau))\big]\mu_{A,q}^f(\tau_1)\big[\mu_{B,r}^f(\tau_2),\mu_{B,s}^f(\tau_3)\big]\\&+i\sum_{q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\big[E_r^f(x_B(\tau_2)),E_q^f(x_A(\tau_1))\big]\big[E_s^f(x_B(\tau_3)),\mathbf{E}^f(x(\tau))\big]\mu_{A,q}^f(\tau_1)\big[\mu_{B,s}^f(\tau_3),\mu_{B,r}^f(\tau_2)\big]\\&+i\sum_{q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\big[E_q^f(x_A(\tau_1)),E_s^f(x_B(\tau_3))\big]\big[E_r^f(x_B(\tau_2)),\mathbf{E}^f(x(\tau))\big]\mu_{A,q}^f(\tau_1)\big[\mu_{B,s}^f(\tau_3),\mu_{B,r}^f(\tau_2)\big]\\&+i\sum_{q,r,s}\int_{\tau_0}^{\tau}d\tau_1\int_{\tau_0}^{\tau_1}d\tau_2\int_{\tau_0}^{\tau_2}d\tau_3\,\big[E_s^f(x_A(\tau_3)),E_r^f(x_B(\tau_2))\big]\big[E_q^f(x_B(\tau_1)),\mathbf{E}^f(x(\tau))\big]\mu_{A,s}^f(\tau_3)\big[\mu_{B,r}^f(\tau_2),\mu_{B,q}^f(\tau_1)\big]\\&+\text{terms of the zeroth, first, and third power of }\boldsymbol{\mu}_B^f.\end{aligned} \tag{A13}$$

Here we have only listed in $\sigma_{mn}^{A,(3)}(\tau)$, $a_{\mathbf{k},\nu}^{(3)}(t(\tau))$, and $\mathbf{E}^{(3)}(x(\tau))$ the terms in the second power of $\boldsymbol{\mu}_B^f$ and abbreviated others by "terms of the zeroth, first, and third power of $\boldsymbol{\mu}_B^f$" since they do not contribute to the interatomic interaction potential for reasons we now explain. As we have shown in Sec. II B, $\sigma_{mn}^{A,(3)}(\tau)$ and $\mathbf{E}^{(3)}(x(\tau))$ are respectively needed for derivation of the fourth-order effective Hamiltonians [see Eqs. (24) and (29)], and "terms of the zeroth and first power in $\boldsymbol{\mu}_B^f$" collected in $F(\tau)$ in Eq. (26) and "terms of the zeroth, first, and third power in $\boldsymbol{\mu}_B^f$" collected in $G(\tau)$ in Eq. (31) follow from "terms of the zeroth and first power of $\boldsymbol{\mu}_B^f$" of $\sigma_{mn}^{A,(3)}(\tau)$ and "terms of the zeroth, first, and third power of $\boldsymbol{\mu}_B^f$" of $\mathbf{E}^{(3)}(x(\tau))$, respectively. As we have shown in Sec. II C, these characters of $F(\tau)$ and $G(\tau)$ further carry onto $\tilde{F}(\tau)$ and $\tilde{G}(\tau)$ in the fourth-order effective Hamiltonians [Eqs. (34) and (36)], which after averaging over the atomic state $|g_Ag_B\rangle$ give rise to zero or interatomic-separation-independent contributions to the interaction potential. Because of this fact and that they are very lengthy, we abbreviate them by "terms of the zeroth and first power in $\boldsymbol{\mu}_B^f$" and "terms of the zeroth, first, and third power in $\boldsymbol{\mu}_B^f$" respectively.

APPENDIX B: DERIVATION OF THE TWO-POINT CORRELATION FUNCTION OF THE ELECTRIC FIELD $\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle$

In this Appendix, we show how to calculate the two-point correlation function of the electric field $\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle$, i.e., Eq. (47).

Using the expansion of $\boldsymbol{E}^f(x)$ [the first line of Eq. (12)] and the relation Eq. (3) in the following formula,

$$\begin{aligned}\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle&\equiv Z^{-1}\mathrm{Tr}\big[e^{-\beta H_F^f}E_p^f(x)E_q^f(x')\big]\\&=-Z^{-1}\sum_{\nu_1,\nu_2=0}^{2}\int d^3\mathbf{k}_1\int d^3\mathbf{k}_2\;g_{\mathbf{k}_1}g_{\mathbf{k}_2}\omega_{\mathbf{k}_1}\omega_{\mathbf{k}_2}\epsilon_p(\mathbf{k}_1,\nu_1)\epsilon_q(\mathbf{k}_2,\nu_2)\mathrm{Tr}\big\{e^{-\beta H_F^f}\\&\quad\times\big[a_{\mathbf{k}_1,\nu_1}^f e^{-i\omega_{\mathbf{k}_1}t}e^{i\mathbf{k}_1\cdot\mathbf{x}}-\text{H.c.}\big]\big[a_{\mathbf{k}_2,\nu_2}^f e^{-i\omega_{\mathbf{k}_2}t'}e^{i\mathbf{k}_2\cdot\mathbf{x}'}-\text{H.c.}\big]\big\},\end{aligned} \tag{B1}$$

where $H_F^f=\sum_\nu\int d^3\mathbf{k}\omega_\mathbf{k}a_{\mathbf{k},\nu}^{\dagger,f}a_{\mathbf{k},\nu}^f$ is the free Hamiltonian of the electric field, $Z\equiv\mathrm{Tr}(e^{-\beta H_F^f})=\prod_\mathbf{k}(1-e^{-\beta\omega_\mathbf{k}})^{-1}$, and "H.c." denotes the Hermitian conjugate, we obtain, after some algebraic simplifications,

$$\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle=\frac{1}{16\pi^3}\int d^3\mathbf{k}\,\frac{1}{k}(k^2\delta_{pq}-k_pk_q)\left[\sum_{m=0}^{\infty}e^{-\beta m\omega_\mathbf{k}}e^{-i\omega_\mathbf{k}(t-t')}e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')}+\sum_{m=1}^{\infty}e^{-\beta m\omega_\mathbf{k}}e^{i\omega_\mathbf{k}(t-t')}e^{-i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')}\right]. \tag{B2}$$

This equation can be further transformed into

$$\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle=\frac{1}{16\pi^3}(\partial_0\partial_0'\delta_{pq}-\partial_p\partial_q')\int d^3\mathbf{k}\,\frac{1}{k}\left[\sum_{m=0}^{\infty}e^{-\beta m\omega_\mathbf{k}}e^{-i\omega_\mathbf{k}(t-t')}e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')}+\sum_{m=1}^{\infty}e^{-\beta m\omega_\mathbf{k}}e^{i\omega_\mathbf{k}(t-t')}e^{-i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')}\right]. \tag{B3}$$

Then perform the integration and we obtain

$$\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle = -\frac{1}{4\pi^2}(\partial_0\partial_0'\delta_{pq} - \partial_p\partial_q')\left[\sum_{m=0}^{\infty}\frac{1}{(\Delta t - im\beta - i\varepsilon)^2 - |\Delta\mathbf{x}|^2} + \sum_{m=1}^{\infty}\frac{1}{(\Delta t + im\beta)^2 - |\Delta\mathbf{x}|^2}\right] \tag{B4}$$

with $\Delta t = t - t'$, $|\Delta\mathbf{x}| = \sqrt{(x-x')^2 + (y-y')^2 + (z-z')^2}$, and ε a positive infinitesimal. Combining the two summations, we finally arrive at

$$\langle\beta|E_p^f(x)E_q^f(x')|\beta\rangle = -\frac{1}{4\pi^2}(\partial_0\partial_0'\delta_{pq} - \partial_p\partial_q')\sum_{m=-\infty}^{\infty}\frac{1}{(\Delta t - im\beta - i\varepsilon)^2 - |\Delta\mathbf{x}|^2}, \tag{B5}$$

which is Eq. (47).

[1] T. A. Welton, Phys. Rev. **74**, 1157 (1948).
[2] G. Compagno, R. Passante, and F. Persico, Phys. Lett. A **98**, 253 (1983).
[3] J. R. Ackerhalt, P. L. Knight, and J. H. Eberly, Phys. Rev. Lett. **30**, 456 (1973).
[4] P. W. Milonni and W. A. Smith, Phys. Rev. A **11**, 814 (1975).
[5] P. W. Milonni, Phys. Scr. **T21**, 102 (1988).
[6] P. W. Milonni, J. R. Ackerhalt, and W. A. Smith, Phys. Rev. Lett. **31**, 958 (1973).
[7] J. Dalibard, J. Dupont-Roc, and C. Cohen-Tannoudji, J. Phys. France **43**, 1617 (1982).
[8] J. Dalibard, J. Dupont-Roc, and C. Cohen-Tannoudji, J. Phys. France **45**, 637 (1984).
[9] D. Meschede, W. Jhe, and E. A. Hinds, Phys. Rev. A **41**, 1587 (1990).
[10] W. Jhe, Phys. Rev. A **43**, 5795 (1991).
[11] J. Audretsch and R. Müller, Phys. Rev. A **50**, 1755 (1994).
[12] J. Audretsch and R. Müller, Phys. Rev. A **52**, 629 (1995).
[13] J. Audretsch, R. Müller, and M. Holzmann, Class. Quantum Grav. **12**, 2927 (1995).
[14] R. Passante, Phys. Rev. A **57**, 1590 (1998).
[15] J. L. Tomazelli and L. C. Costa, Int. J. Mod. Phys. A **18**, 1079 (2003).
[16] H. Yu and S. Lu, Phys. Rev. D **72**, 064022 (2005); **73**, 109901(E) (2006).
[17] Z. Zhu, H. Yu, and S. Lu, Phys. Rev. D **73**, 107501 (2006).
[18] Z. Zhu and H. Yu, Phys. Lett. B **645**, 459 (2007).
[19] L. Rizzuto, Phys. Rev. A **76**, 062114 (2007).
[20] L. Rizzuto and S. Spagnolo, Phys. Rev. A **79**, 062110 (2009).
[21] L. Rizzuto and S. Spagnolo, J. Phys.: Conf. Ser. **161**, 012031 (2009).
[22] Z. Zhu and H. Yu, Phys. Rev. A **82**, 042108 (2010).
[23] L. Rizzuto and S. Spagnolo, Phys. Scr. **T143**, 014021 (2011).
[24] W. Zhou and H. Yu, Phys. Rev. A **86**, 033841 (2012).
[25] Q. Li, H. Yu, and W. Zhou, Ann. Phys. (NY) **348**, 144 (2014).
[26] L. Rizzuto, M. Lattuca, J. Marino, A. Noto, S. Spagnolo, W. Zhou, and R. Passante, Phys. Rev. A **94**, 012121 (2016).
[27] W. Zhou, R. Passante, and L. Rizzuto, Phys. Rev. D **94**, 105025 (2016).
[28] W. Zhou and H. Yu, Phys. Rev. D **96**, 045018 (2017).
[29] W. Zhou, L. Rizzuto, and R. Passante, Phys. Rev. A **97**, 042503 (2018).
[30] J. Marino, A. Noto, and R. Passante, Phys. Rev. Lett. **113**, 020403 (2014).
[31] G. Menezes, C. Kiefer, and J. Marino, Phys. Rev. D **95**, 085014 (2017).
[32] W. Zhou, S. Cheng, and H. Yu, Phys. Rev. A **103**, 012227 (2021).
[33] S. Cheng, W. Zhou, and H. Yu, Phys. Lett. B **834**, 137440 (2022).
[34] F. London, Z. Phys. **63**, 245 (1930); Z. Phys. Chem. Abt. B **11**, 222 (1930).
[35] H. B. G. Casimir and D. Polder, Phys. Rev. **73**, 360 (1948).
[36] R. R. McLone and E. A. Power, Proc. R. Soc. A **286**, 573 (1965).
[37] E. A. Power and T. Thirunamachandran, Phys. Rev. A **48**, 4761 (1993).
[38] D. P. Craig and T. Thirunamachandran, *Molecular Quantum Electrodynamics* (Dover, New York, 1998).
[39] B. R. Holstein, Am. J. Phys. **69**, 441 (2001).
[40] H. Safari, S. Y. Buhmann, D.-G. Welsch, and H. T. Dung, Phys. Rev. A **74**, 042101 (2006).
[41] S. Cheng, W. Zhou, and H. Yu, Commun. Theor. Phys. **74**, 125103 (2022).
[42] A. D. McLachlan, Proc. R. Soc. A **274**, 80 (1963).
[43] T. H. Boyer, Phys. Rev. A **11**, 1650 (1975).
[44] B. W. Ninham and J. Daicic, Phys. Rev. A **57**, 1870 (1998).
[45] H. Wennerström, J. Daicic, and B. W. Ninham, Phys. Rev. A **60**, 2581 (1999).
[46] G. H. Goedecke and R. C. Wood, Phys. Rev. A **60**, 2577 (1999).
[47] W. Greiner and J. Reinhardt, *Field Quantization* (Springer-Verlag, Berlin, 1996), p. 177.
[48] C. Cohen-Tannoudji, Phys. Scr. **T12**, 19 (1986).
[49] G. Menezes and N. F. Svaiter, Phys. Rev. A **92**, 062131 (2015).
[50] G. Menezes and N. F. Svaiter, Phys. Rev. A **93**, 052117 (2016).
[51] G. Menezes, Phys. Rev. D **94**, 105008 (2016).
[52] W. Zhou and H. Yu, Phys. Rev. D **101**, 025009 (2020).