

**Dynamical Casimir-Polder energy between an excited- and a ground-state atom**L. Rizzuto,<sup>1</sup> R. Passante,<sup>2,\*</sup> and F. Persico<sup>1</sup><sup>1</sup>*INFN and Dipartimento di Scienze Fisiche ed Astronomiche, Università degli Studi di Palermo, Via Archirafi 36, I-90123 Palermo, Italy*<sup>2</sup>*Istituto di Biofisica-Sezione di Palermo, Consiglio Nazionale delle Ricerche, Via Ugo La Malfa 153, I-90146 Palermo, Italy*  
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We consider the Casimir-Polder interaction between two atoms, one in the ground state and the other in its excited state. The interaction is time dependent for this system, because of the dynamical self-dressing and the spontaneous decay of the excited atom. We calculate the dynamical Casimir-Polder potential between the two atoms using an effective Hamiltonian approach. The results obtained and their physical meaning are discussed and compared with previous results based on a time-independent approach, which uses a nonnormalizable dressed state for the excited atom.

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**I. INTRODUCTION**

The existence of field fluctuations in the vacuum state is a remarkable prediction of quantum field theory. Vacuum fluctuations produce observable effects such as the Casimir force between two neutral mirrors or dielectrics in the vacuum [1] and the Casimir-Polder force between neutral atoms or molecules in their ground state [2]. The Casimir-Polder forces are long-range effects due to the interaction of the atoms with the common quantum radiation field. For intermolecular distances smaller than typical atomic transition wavelengths from the ground state, they reduce to van der Waals forces; for larger distances they decrease more rapidly than van der Waals forces due to retardation effects [2,3]. The physical origin of the Casimir-Polder force has been investigated in the past in terms of dressed vacuum fluctuations, radiation reaction field, or vacuum field correlations (for a review, see [4]). More recent studies have also considered the Casimir-Polder dispersion energy between two molecules, one in an excited state and the other in the ground state [5–7]. The van der Waals-like interaction between an excited atom and a dielectric surface has also been considered [8]. These calculations are based on fourth-order perturbation theory and they are time independent. In fact, the spontaneous decay of the excited atom, as well as its dynamical self-dressing, is not included in these calculations, the excited atom being treated as if it were in a stable state. The time-independent potential contains two terms: one resulting from virtual photon exchange and the other from the resonance due to the possibility of the emission of a resonant photon [6]. The term arising from the virtual photon exchange has the same structure as the Casimir-Polder potential for ground-state atoms. The resonant term is a polynomial in the inverse of the intermolecular separation  $R$ . Finally, the possibility of enhancement of van der Waals forces in nonequilibrium situations has recently been suggested [9]; this indicates that the matter is not entirely settled and explains our interest in Casimir-Polder forces in dynamical situations. The term “dynamic” in gen-

eral may refer to two situations, one time dependent and the other frequency dependent, which may also lead to dynamic potentials. This paper is concerned with the first case, i.e., explicitly time-dependent situations.

In this paper, we shall adopt a time-dependent approach for the calculation of the Casimir-Polder potential between a ground-state and an excited-state atom or molecule. This approach, which takes into account both the short time dynamical dressing and the spontaneous decay of the excited atomic state, will give a deeper understanding of the physical nature of the Casimir-Polder force.

As usual, the interaction energy between the excited- and the ground-state atom is assumed to stem from the response of the latter to the field emitted by the former. This idea has recently been used in the different context of the calculation of the Casimir-Polder force between partially dressed atoms [10]. We use perturbation theory, and this limits the validity of our results to times shorter than the lifetime of the excited atom. We find that this potential is zero before the “causality time”  $t=R/c$ , coherently with relativistic causality. For  $t>R/c$ , we find that the interaction energy contains three terms. Two of them were already obtained in previous time-independent calculations [6]. The third term is additional and it is time dependent; it describes the time dependence of the force when one atom is initially in its bare excited state. This term vanishes for times larger than the time scale of the dynamical dressing of the excited state, which coincides with the so-called Zeno time [11]; after the Zeno time (but at times shorter than the time scale of the spontaneous decay  $\gamma^{-1}$  of the excited atomic state), the interaction energy reduces to that obtained by time-independent calculations.

The paper is organized as follows. In Sec. II we describe our effective Hamiltonian approach, and in Sec. III we obtain the complete Casimir-Polder potential between the excited- and the ground-state atom, inclusive of the earlier (time-independent [6]) and of the additional (time-dependent) terms.

**II. THE EFFECTIVE HAMILTONIAN**

We consider two atoms  $A$  and  $B$  interacting with the electromagnetic radiation field in the Coulomb gauge;  $\mathbf{r}_A$  and  $\mathbf{r}_B$

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are their positions. Atom  $A$  is approximated as a two-level system. Its interaction with the radiation field, in the multipolar coupling scheme and within the dipole approximation, is described by the following Hamiltonian [4]:

$$H = \hbar\omega_0 S_z^A + \sum_{\mathbf{k}j} \hbar\omega_k a_{\mathbf{k}j}^\dagger a_{\mathbf{k}j} + \sum_{\mathbf{k}j} (\epsilon_{\mathbf{k}j} S_+^A - \epsilon_{\mathbf{k}j}^* S_-^A) (a_{\mathbf{k}j} e^{i\mathbf{k}\cdot\mathbf{r}_A} - a_{\mathbf{k}j}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}_A}), \quad (1)$$

where  $\omega_0 = ck_0$  is the transition frequency of the atom and  $S_z$ ,  $S_+$ , and  $S_-$  are the pseudospin atomic operators. The coupling constant  $\epsilon_{\mathbf{k}j}$  in the multipolar coupling scheme is given by

$$\epsilon_{\mathbf{k}j} = i \left( \frac{2\pi\hbar ck}{V} \right)^{1/2} \hat{\mathbf{e}}_{\mathbf{k}j} \cdot \boldsymbol{\mu}^A, \quad (2)$$

where  $\boldsymbol{\mu}^A$  is the transition dipole moment of atom  $A$  and  $\hat{\mathbf{e}}_{\mathbf{k}j}$  are the polarization unit vectors.

The use of the multipolar form of the interaction Hamiltonian is very convenient in our calculation. In fact, in this coupling scheme the momentum conjugate to the vector potential is the transverse displacement field which, outside the atoms, coincides with the total electric field [12] (transverse plus longitudinal). In this way, we directly obtain the total field generated by one atom, inclusive of the longitudinal components.

We assume that at  $t=0$  the atom  $A$  is in its bare excited state, while the atom  $B$  is in the ground state. The two atoms are in general different, and we consider a factorized state as the initial state. We are interested in the dynamical Casimir-Polder potential between these two atoms. Our calculation proceeds in two steps. First, we obtain the electromagnetic field emitted by the initially excited atom  $A$  and then we evaluate the interaction energy of the ground-state atom  $B$  with this field. We have already used a similar procedure to obtain the Casimir-Polder potential between ground-state atoms and shown its relation to the spatial correlations of vacuum fluctuations [13].

The interaction energy of the ground-state atom  $B$  with the field emitted by the excited atom  $A$  can be conveniently obtained by an effective interaction, which is quadratic in the field operators. The two atoms are in general different. This quadratic coupling can be obtained by a unitary transformation from the multipolar Hamiltonian, and it is given by [14,15]

$$H_{eff} = -\frac{1}{2} \sum_{\mathbf{k}j} \alpha^B(k) \langle \mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t) \cdot \mathbf{E}(\mathbf{r}_B, t) \rangle = -\frac{1}{2} \sum_{\mathbf{k}j} \sum_{\mathbf{k}'j'} \alpha^B(k) \times \langle \mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t) \cdot \mathbf{E}_{\mathbf{k}'j'}(\mathbf{r}_B, t) \rangle, \quad (3)$$

where the average in Eq. (3) has to be taken on the initial state of the system (atom  $A$  excited and the field in the vacuum state),  $\alpha^B(k)$  is the ground-state dynamic polarizability of the atom  $B$ , and

$$\mathbf{E}(\mathbf{r}_B, t) = \sum_{\mathbf{k}j} \mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t) = i \sum_{\mathbf{k}j} \sqrt{\frac{2\pi\hbar\omega_k}{V}} [\hat{\mathbf{e}}_{\mathbf{k}j} a_{\mathbf{k}j}(t) e^{i\mathbf{k}\cdot\mathbf{r}_B} - \hat{\mathbf{e}}_{\mathbf{k}j}^* a_{\mathbf{k}j}^\dagger(t) e^{-i\mathbf{k}\cdot\mathbf{r}_B}] \quad (4)$$

is the field operator evaluated at the position of atom  $B$ ,  $\mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t)$  being its  $(\mathbf{k}j)$  component, which includes a contribution coming from the presence of atom  $A$ . In this way, we obtain the Casimir-Polder potential between the atoms  $A$  and  $B$  from the response of atom  $B$  to the field emitted by atom  $A$ . We stress that the field operator  $\mathbf{E}$  in Eq. (4) is the transverse displacement field operator (that is, the momentum conjugate to the vector potential) which, outside the atoms, coincides with the total electric field operator [12]: longitudinal field contributions are already included in Eq. (4).

### III. THE DYNAMICAL CASIMIR-POLDER POTENTIAL

The first step in obtaining the time-dependent Casimir-Polder potential, as outlined above, is to evaluate the average value of the operator  $\mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t) \cdot \mathbf{E}_{\mathbf{k}'j'}(\mathbf{r}_B, t)$  on the initial state, that is, the state with atom  $A$  excited and the field in the vacuum state. We obtain this quantity by solving at the second order in the coupling constant the Heisenberg equations of motion for the field operators and using the Hamiltonian (1), and then taking the average value on the state at  $t=0$ ; the calculation is sketched out in the Appendix. Our procedure closely follows that of Power and Thirunamachandran [15] for a multilevel atom, with the difference that we have specialized to a two-level case and that we deal explicitly with the case  $t > R/c$ . Substitution of Eq. (A8) into Eq. (3) yields the following expression for the average value of  $H_{eff}$ , which gives the Casimir-Polder potential between the two atoms:

$$\begin{aligned} \Delta E_{AB} = & -\frac{1}{2} \sum_{\mathbf{k}j\mathbf{k}'j'} \alpha^B(k) \langle \uparrow_A \{0_{\mathbf{k}j}\} | \mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t) \cdot \mathbf{E}_{\mathbf{k}'j'}(\mathbf{r}_B, t) | \uparrow_A \{0_{\mathbf{k}j}\} \rangle = \frac{1}{2} \left( \frac{2\pi c}{V} \right)^2 \sum_{\mathbf{k}j\mathbf{k}'j'} (\hat{\mathbf{e}}_{\mathbf{k}j} \cdot \hat{\mathbf{e}}_{\mathbf{k}'j'}) (\hat{\mathbf{e}}_{\mathbf{k}j} \cdot \boldsymbol{\mu}^A) (\hat{\mathbf{e}}_{\mathbf{k}'j'} \cdot \boldsymbol{\mu}^A) k k' \\ & \times \left[ \alpha^B(k) [F_t(\omega_0 + \omega_k) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_k t)} - F_t(\omega_0 - \omega_k) e^{-i(\mathbf{k}\cdot\mathbf{R} - \omega_k t)}] [F_t^*(\omega_0 - \omega_{k'}) e^{i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)} - F_t^*(\omega_0 + \omega_{k'}) e^{-i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)}] \right. \\ & + i\alpha^B(k) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_k t)} \left( \frac{1}{\omega_0 - \omega_k} \{ e^{i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)} [F_t(\omega_k + \omega_{k'}) - F_t(\omega_0 + \omega_{k'})] - e^{-i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)} [F_t(\omega_k - \omega_{k'}) - F_t(\omega_0 - \omega_{k'})] \} \right. \\ & \left. \left. + \frac{1}{\omega_0 + \omega_k} \{ e^{i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)} [F_t(\omega_k + \omega_{k'}) - F_t^*(\omega_0 - \omega_{k'})] - e^{-i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)} [F_t(\omega_k - \omega_{k'}) - F_t^*(\omega_0 + \omega_{k'})] \} \right) - i\alpha^B(k') e^{-i(\mathbf{k}\cdot\mathbf{R} - \omega_k t)} \right] \end{aligned}$$

$$\begin{aligned} & \times \left( \frac{1}{\omega_0 - \omega_k} \left\{ e^{-i(\mathbf{k}' \cdot \mathbf{R} - \omega_{k'} t)} [F_t^*(\omega_k + \omega_{k'}) - F_t^*(\omega_0 + \omega_{k'})] - e^{i(\mathbf{k}' \cdot \mathbf{R} - \omega_{k'} t)} [F_t^*(\omega_k - \omega_{k'}) - F_t^*(\omega_0 - \omega_{k'})] \right\} \right. \\ & \left. + \frac{1}{\omega_0 + \omega_k} \left\{ e^{-i(\mathbf{k}' \cdot \mathbf{R} - \omega_{k'} t)} [F_t^*(\omega_k + \omega_{k'}) - F_t(\omega_0 - \omega_{k'})] - e^{i(\mathbf{k}' \cdot \mathbf{R} - \omega_{k'} t)} [F_t^*(\omega_k - \omega_{k'}) - F_t(\omega_0 + \omega_{k'})] \right\} \right), \end{aligned} \quad (5)$$

where the complex function  $F_t(x)$  is defined in Eq. (A7).

We first perform integrations and summations over  $\mathbf{k}' j'$  in the continuum limit, obtaining

$$\begin{aligned} \Delta E(A, B) = & \frac{\pi}{V} \mu_m^A \mu_n^A \sum_{\mathbf{k}j} (\hat{\mathbf{e}}_{\mathbf{k}j})_\ell (\hat{\mathbf{e}}_{\mathbf{k}j})_m \times \left\{ \alpha^B(k) i c [F_t(\omega_0 - \omega_k) e^{-i(\mathbf{k} \cdot \mathbf{R} - ckt)} - F_t(\omega_0 + \omega_k) e^{i(\mathbf{k} \cdot \mathbf{R} - ckt)}] e^{-ik_0 R} F_{\ell n}^R \frac{e^{ik_0 R}}{R} + \alpha^B(k) e^{i\mathbf{k} \cdot \mathbf{R}} \right. \\ & \times \left[ \frac{1}{k_0 - k} \left( F_{\ell n}^R \frac{e^{-ikR}}{R} - e^{i(k_0 - k)ct} F_{\ell n}^R \frac{e^{-ik_0 R}}{R} \right) + \frac{1}{k_0 + k} \left( F_{\ell n}^R \frac{e^{-ikR}}{R} - e^{-i(k_0 + k)ct} F_{\ell n}^R \frac{e^{ik_0 R}}{R} \right) \right] + e^{-i\mathbf{k} \cdot \mathbf{R}} \left[ \frac{1}{k_0 - k} \left( \alpha^B(k) F_{\ell n}^R \frac{e^{ikR}}{R} \right. \right. \\ & \left. \left. - \alpha^B(k_0) e^{-i(k_0 - k)ct} F_{\ell n}^R \frac{e^{ik_0 R}}{R} \right) + \frac{1}{k_0 + k} \left( \alpha^B(k) F_{\ell n}^R \frac{e^{ikR}}{R} - \alpha^B(k_0) e^{i(k_0 + k)ct} F_{\ell n}^R \frac{e^{-ik_0 R}}{R} \right) \right] \right\} \Theta(ct - R), \end{aligned} \quad (6)$$

where we have defined the differential operator acting on the variable  $\mathbf{R}$  as

$$F_{\ell n}^R = (-\delta_{n\ell} \nabla^2 + \nabla_\ell \nabla_n). \quad (7)$$

The presence of the  $\Theta$  function in Eq. (6) ensures relativistic causality in the propagation of the field generated by atom  $A$  and consequently in the interaction between the two atoms. The  $\Theta$  function results from integrals over  $k$  of the following kind:

$$P \int_{-\infty}^{\infty} dk \frac{e^{ikx}}{k + k_0} \alpha(k) = i\pi [2\Theta(x) - 1] e^{-ik_0 x} \alpha(k_0). \quad (8)$$

After lengthy calculations which include integration over  $\mathbf{k}j$  of some of the terms containing  $1/(k_0 - k)$ , Eq. (6) can be expressed in the more compact form

$$\begin{aligned} \Delta E(A, B) = & \frac{2\pi}{V} \mu_m^A \mu_n^A \sum_{\mathbf{k}j} (\hat{\mathbf{e}}_{\mathbf{k}j})_\ell (\hat{\mathbf{e}}_{\mathbf{k}j})_m \frac{k}{k_0 - k} \\ & \times \text{Re} \left[ e^{i\mathbf{k} \cdot \mathbf{R}} \left( 2\alpha^B(k) F_{\ell n}^R \frac{e^{-ikR}}{R} - [\alpha^B(k) \right. \right. \\ & \left. \left. + \alpha^B(k_0)] e^{i(k_0 - k)ct} F_{\ell n}^R \frac{e^{-ik_0 R}}{R} \right) \right] \Theta(ct - R). \end{aligned} \quad (9)$$

After summation over  $(\mathbf{k}j)$  in the continuum limit and some algebraic manipulations where the analytical properties of the dynamical polarizability  $\alpha^B(k)$  are used, we finally get

$$\begin{aligned} \Delta E(A, B) = & \left[ -\mu_m^A \mu_n^A \alpha^B(k_0) F_{\ell n}^R \frac{1}{R} F_{\ell m}^{\bar{R}} \frac{1}{\bar{R}} \cos k_0(R - \bar{R}) \right. \\ & + \frac{\hbar c}{2\pi} F_{\ell n}^R \frac{1}{R} F_{\ell m}^{\bar{R}} \frac{1}{\bar{R}} \int_0^\infty du e^{-u(R + \bar{R})} \alpha_{mn}^A(iu) \alpha^B(iu) \\ & \left. + \frac{1}{\pi} \mu_m^A \mu_n^A F_{\ell n}^R \frac{1}{R} F_{\ell m}^{\bar{R}} \frac{1}{\bar{R}} \right] \end{aligned}$$

$$\begin{aligned} & \times \left( \cos k_0(ct - R) \int_0^\infty du [\alpha^B(iu) + \alpha^B(iu_0)] e^{-uct} \right. \\ & \times \frac{2k_0 \sinh u\bar{R}}{k_0^2 + u^2} + \sin k_0(ct - R) \int_0^\infty du [\alpha^B(iu) \\ & \left. + \alpha^B(iu_0)] e^{-uct} \frac{2u \sinh u\bar{R}}{k_0^2 + u^2} \right) \Bigg|_{R=\bar{R}} \Theta(ct - R), \end{aligned} \quad (10)$$

where the variable  $\bar{R}$ , which is put equal to  $R$  after the action of the differential operator  $F_{\ell m}^{\bar{R}}$ , has been conveniently introduced in order to distinguish the variables on which the operators  $F_{\ell n}^R$  and  $F_{\ell m}^{\bar{R}}$  operate.  $\alpha_A(iu)$  is the dynamical polarizability of the excited state of the atom  $A$ , extended to imaginary frequencies  $iu$ ,

$$\alpha_{mn}^A(iu) = \frac{2k_0 \mu_m^A \mu_n^A}{\hbar c (k_0^2 + u^2)}. \quad (11)$$

For a two-level system, the dynamical polarizability of the excited state coincides with that of the ground state except for a change of its sign. The experimental observability of time dependences of the form implied by expression (10) has been discussed in [16].

We notice from Eq. (10) that the first two terms inside the square brackets are time independent, whereas the third term depends on time. This time-dependent term contains, inside the  $u$  integrals, an exponential factor decreasing with time. For a given  $R$ , this term rapidly vanishes to zero with a time scale of the order of  $k_0^{-1}/c = \omega_0^{-1}$ . This means that, for this given  $R < ct$  after a transient in which there is a time-dependent Casimir-Polder interaction, then the interatomic interaction stabilizes to

$$\Delta E(A,B) = \left( -\mu_m^A \mu_n^A \alpha_B(k_0) F_{\ell n}^R \frac{1}{R} F_{\ell m}^R \frac{1}{\bar{R}} \cos k_0(R - \bar{R}) + \frac{\hbar c}{2\pi} F_{\ell n}^R \frac{1}{R} F_{\ell m}^{\bar{R}} \frac{1}{\bar{R}} \int_0^\infty du e^{-u(R+\bar{R})} \alpha_{mn}^A(iu) \alpha^B(iu) \right)_{R=\bar{R}}, \quad (12)$$

which is time independent. We note that the time scale  $\omega_0^{-1}$  of the dynamical Casimir-Polder potential is the same as at the nonexponential early stage of the spontaneous decay of the excited atom (Zeno time). Details of the time-dependent term in Eq. (10) may depend on the choice of the initial state at  $t=0$ , in our case a bare excited state. Other possible choices, for example a partially dressed state, might yield a different expression for this term, but we expect that the general properties of the time-dependent energy should not change. The time-dependent energy in Eq. (10) yields a time-dependent force between the two atoms, which in principle is observable. During this stage of the decay, the self-dressing of the atom occurs [17]. This indicates that the time-dependent part of the potential is related to the interaction of atom  $B$  with the dynamical photon cloud of atom  $A$ , which is generated during its self-dressing. Our result (10) is valid only up to times of the order of  $\gamma^{-1}$  or smaller, where  $\gamma$  is the decay rate of the excited state, because of the limitation of the perturbation theory we have used. However, for atomic systems the time interval between  $k_0^{-1}/c$  and  $\gamma^{-1}$  is typically quite a long interval. Equation (12) coincides with the result obtained by Power and Thirunamachandran using a time-independent approach based on a nonnormalizable dressed excited state for atom  $A$  [6,7]. This part of the potential has two components: one has the same form as the potential for ground-state atoms, and the other is spatially oscillating and is related to the fact that the excited atom can emit a resonant photon.

#### IV. CONCLUSIONS

We have considered the Casimir-Polder intermolecular interaction between two atoms, one in its ground state and the other excited. The latter is assumed to be at  $t=0$  in its bare excited state. We have used an effective Hamiltonian approach, and the interaction energy between the two atoms stems from the interaction of the ground-state atom (through its dynamical polarizability) with the field generated by the excited atom. The interaction energy yielding the Casimir-Polder potential is time dependent because of the dynamical

self-dressing processes of the excited atom; there is also a contribution to the potential from the resonance related to the possibility of emission of a resonant photon by the excited atom. We find that for times  $t \gg \omega_0^{-1}$ , that is, for times larger than the inverse of the transition frequency of the excited atom, and for  $t > R/c$  the Casimir-Polder interaction becomes time independent. In this limit its expression coincides with that already obtained by Power and Thirunamachandran using a time-independent approach and based on a nonnormalizable dressed state for the excited atom. We argue that the time-dependent part of the potential that we obtain is due to virtual photons that are emitted by the excited atom in the very early stages of its decay.

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#### APPENDIX: ITERATIVE SOLUTION OF THE HEISENBERG EQUATIONS

In this appendix we outline the iterative solution of the Heisenberg equations describing the interaction of atom  $A$  with the radiation field, using the Hamiltonian (1) for the part pertaining to atom  $A$ . The Heisenberg equations for the field and atomic operators are

$$\dot{a}_{\mathbf{k}j}(t) = -i\omega_{\mathbf{k}} a_{\mathbf{k}j}(t) + \frac{i}{\hbar} [\epsilon_{\mathbf{k}j} S_+^A(t) - \epsilon_{\mathbf{k}j}^* S_-^A(t)] e^{-i\mathbf{k}\cdot\mathbf{r}_A}, \quad (A1)$$

$$\dot{S}_+^A(t) = i\omega_0 S_+^A(t) + \frac{2i}{\hbar} S_z^A(t) \sum_{\mathbf{k}j} \epsilon_{\mathbf{k}j}^* [a_{\mathbf{k}j}(t) e^{i\mathbf{k}\cdot\mathbf{r}_A} - a_{\mathbf{k}j}^\dagger(t) e^{-i\mathbf{k}\cdot\mathbf{r}_A}]. \quad (A2)$$

The iterative solution of these equations and their Hermitian conjugates yields the perturbative expansion of the field operators

$$a_{\mathbf{k}j}(t) = a_{\mathbf{k}j}^{(0)}(t) + a_{\mathbf{k}j}^{(1)}(t) + a_{\mathbf{k}j}^{(2)}(t) + \dots, \quad (A3)$$

where

$$a_{\mathbf{k}j}^{(0)}(t) = a_{\mathbf{k}j}(0) e^{-i\omega_{\mathbf{k}} t}, \quad (A4)$$

$$a_{\mathbf{k}j}^{(1)}(t) = \frac{i}{\hbar} e^{-i\omega_{\mathbf{k}} t} [\epsilon_{\mathbf{k}j} S_+^A(0) F_t^*(\omega_0 + \omega_{\mathbf{k}}) - \epsilon_{\mathbf{k}j}^* S_-^A(0) F_t^*(\omega_0 - \omega_{\mathbf{k}})] e^{-i\mathbf{k}\cdot\mathbf{r}_A}, \quad (A5)$$

$$\begin{aligned}
a_{\mathbf{k}j}^{(2)}(t) = & -\frac{2i}{\hbar^2} S_z^A(0) e^{-i\omega_k t} e^{-i\mathbf{k}\cdot\mathbf{r}_A} \sum_{\mathbf{k}'j'} \left[ a_{\mathbf{k}'j'}(0) e^{i\mathbf{k}'\cdot\mathbf{r}_A} \left( \boldsymbol{\epsilon}_{\mathbf{k}j}^* \boldsymbol{\epsilon}_{\mathbf{k}'j'} \frac{F_t(\omega_k - \omega_{k'}) - F_t(\omega_0 + \omega_k)}{\omega_0 + \omega_{k'}} + \boldsymbol{\epsilon}_{\mathbf{k}j}^* \boldsymbol{\epsilon}_{\mathbf{k}'j'} \frac{F_t(\omega_k - \omega_{k'}) - F_t^*(\omega_0 - \omega_k)}{\omega_0 - \omega_{k'}} \right) \right. \\
& \left. - a_{\mathbf{k}'j'}^\dagger(0) e^{-i\mathbf{k}'\cdot\mathbf{r}_A} \left( \boldsymbol{\epsilon}_{\mathbf{k}j} \boldsymbol{\epsilon}_{\mathbf{k}'j'}^* \frac{F_t(\omega_k + \omega_{k'}) - F_t(\omega_0 + \omega_k)}{\omega_0 - \omega_{k'}} + \boldsymbol{\epsilon}_{\mathbf{k}j} \boldsymbol{\epsilon}_{\mathbf{k}'j'}^* \frac{F_t(\omega_k + \omega_{k'}) - F_t^*(\omega_0 - \omega_k)}{\omega_0 + \omega_{k'}} \right) \right], \quad (\text{A6})
\end{aligned}$$

where we have defined the function

$$F_t(x) = \int_0^t dt' e^{ixt'}. \quad (\text{A7})$$

Using Eqs. (A4)–(A6), we obtain the following expression for the average value of the field operators present in Eq. (3) on the initial state of the system (atom A+ field)  $|\uparrow_A\{0_{\mathbf{k}j}\}\rangle$ :

$$\begin{aligned}
\langle \mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t) \cdot \mathbf{E}_{\mathbf{k}j}(\mathbf{r}_B, t) \rangle = & -\left(\frac{2\pi c}{V}\right)^2 (\hat{\mathbf{e}}_{\mathbf{k}j} \cdot \hat{\mathbf{e}}_{\mathbf{k}'j'}) (\hat{\mathbf{e}}_{\mathbf{k}j} \cdot \boldsymbol{\mu}^A) (\hat{\mathbf{e}}_{\mathbf{k}'j'} \cdot \boldsymbol{\mu}^A) k k' \left\{ [F_t(\omega_0 + \omega_k) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_k t)} - F_t(\omega_0 - \omega_k) e^{-i(\mathbf{k}\cdot\mathbf{R} - \omega_k t)}] \right. \\
& \times [F_t^*(\omega_0 - \omega_{k'}) e^{i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)} - F_t^*(\omega_0 + \omega_{k'}) e^{-i(\mathbf{k}'\cdot\mathbf{R} - \omega_{k'} t)}] + \left[ -\left(\frac{F_t(\omega_k + \omega_{k'}) - F_t(\omega_0 + \omega_{k'})}{i(\omega_0 - \omega_k)} \right. \right. \\
& + \left. \left. \frac{F_t(\omega_k + \omega_{k'}) - F_t^*(\omega_0 - \omega_{k'})}{i(\omega_0 + \omega_k)} \right) e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{R} - i(\omega_k + \omega_{k'})t} + \left( \frac{F_t(\omega_k - \omega_{k'}) - F_t(\omega_0 - \omega_{k'})}{i(\omega_0 - \omega_k)} \right. \right. \\
& \left. \left. + \frac{F_t(\omega_k - \omega_{k'}) - F_t^*(\omega_0 + \omega_{k'})}{i(\omega_0 + \omega_k)} \right) e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R} - i(\omega_k - \omega_{k'})t} + \text{c.c.}(\mathbf{k} \leftrightarrow \mathbf{k}') \right] \left. \right\} \quad (\text{A8})
\end{aligned}$$

(the last term indicates the complex conjugate of the terms inside the square bracket after exchange between  $\mathbf{k}$  and  $\mathbf{k}'$ ).  $\mathbf{R} = \mathbf{r}_B - \mathbf{r}_A$  is the interatomic separation.

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