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van der Waals interaction of excited media

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The Casimir interaction between two media of ground-state atoms is well described with the help of the Lifshitz formula depending upon the permittivity of the media. We will show that this formula is in contradiction with experimental evidence for excited atoms. We calculate the Casimir force between two atoms if one or both of them are excited. We use methods of quantum electrodynamics specially derived for the problem. It enables us to take into account the excited-state radiation widths of atoms. Then we calculate the force between the excited atom and medium of ground-state atoms. The results are in agreement with the ones obtained by other authors who used perturbation theory or linear response theory. Generalization of our results to the case of the interaction between two media of excited atoms results in a formula, which is in not only in quantitative, but in qualitative contradiction with the Lifshitz formula. This contradiction disappears if the media of ground-state atoms are considered. Moreover, our result does not include the permittivity of the media. It includes a quantity which differs from the permittivity only for excited atoms. The main features of our results are as follows. The interaction is resonant, the force may be either attractive or repulsive depending on the resonant frequencies of the atoms of different media, and the value of the Casimir force may be several orders of magnitude lager than that predicted by the Lifshitz formula. The features mentioned here are in agreement with known experimental and theoretical evidence obtained by many authors for the interaction of a single excited atom with dielectric media.

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I. INTRODUCTION

The dispersion force between two atoms separated by a distance R large enough to neglect wave function overlap—the van der Waals or the Casimir force—has been studied in numerous works when the atoms are in ground states. If the distance R is smaller than the wavelength of the atom transitions, the force is described by the London formula [1]. If R is larger than the wavelength and the retardation effects are significant, the force is described by the Casimir formula [2], which was generalized later to arbitrary distances R by Casimir and Polder [3]. Numerous papers, where the Casimir interaction of ground-state atoms is considered, have been appearing for more than the last 50 years. For references see [4–6].

If one or both atoms are excited, the results for the Casimir force differ significantly from the ones mentioned above. If the atoms are in the ground state, the force is attractive. If one of the atoms is excited, the force is either attractive or repulsive depending on the transition frequencies of the atoms. Moreover, the force is resonant. To obtain these results the authors used either linear response theory [7] or perturbation methods of quantum electrodynamics [8]. But in both papers the excited energy-level widths of the atoms have not been taken into account. But if we deal with excited atoms and a resonant interaction, we should take into account the finite level widths of the atoms. Here we suggest a method which enables us to calculate the van der Waals potential taking into account such widths.

The interaction of an excited atom near a cold (nonexcited) dielectric surface is of great interest now. There are two theoretical approaches to the problem. The first one is based on linear response theory without explicit quantization of the electromagnetic field [9,10]. The second one is based on macroscopic quantum electrodynamics with the permittivity included in the Hamiltonian [11,12]. A review of recent works can be found in [11,12]. Both approaches result in a dependence of the Casimir force on the permittivity of the medium. Here the Casimir force is resonant and it can be either attractive or repulsive depending on the relation of excited atom and medium transition frequencies. For a dilute gas medium the results are in agreement with the ones obtained for a two-atom interaction [10]. The latest experiments [10,13,14] are in agreement with theoretical predictions.

The Casimir force between two dielectric media was found for the first time by Lifshitz [15] with the help of linear response theory. Another method of obtaining Lifshitz's result is based on Matsubara-temperature Green functions and is given in [16]. The results are identical and depend on the permittivities of the interacting media. The validity of the Lifshitz formula is discussed now for the case of the interaction between two real metals described by permittivities of the Drude model [17] and two magnetodielectric bodies embedded in another magnetodielectric body [18]. We examine the applicability of the Lifshitz formula to excited media. We will show that the result obtained with the help of the Lifshitz formula for excited media is in contradiction with the results of quantum electrodynamics and, moreover, they are in contradiction with the experimental evidence.

In Sec. II we consider electric-dipole interaction of two atoms, one of which is excited. We take into account the radiation width of energy levels. A specially developed method of quantum Green functions is implemented. We show that the results are in agreement with the ones obtained by different authors [1,7,8].

Section III is devoted to the interaction of an excited atom with a dielectric medium of dilute cold gas. We show that the Casimir force is expressed in terms of the coherent permittivity, but not the conventional one. But the results are in agreement with the ones expressed in terms of conventional permittivity [9–14]. If a ground-state atom interacts with an excited medium, the situation is different. We suppose that such a result cannot be obtained in terms of conventional permittivity.

In Sec. IV we calculate the Casimir force for a case of two media of diluted gases with excited atoms. The result obtained here is not expressed in terms of conventional permittivity (contrary to the Lifshitz formula), but in terms of coherent permittivity. We have shown that the results obtained with the help of quantum electrodynamics and the Lifshitz formula are not in agreement if the amount of excited atoms is significant. Moreover, the Lifshitz formula is in dramatic contradiction with the theoretical and experimental results obtained for the interaction of a single excited atom with a cold medium [9–14].

II. INTERACTION BETWEEN AN EXCITED ATOM AND A GROUND-STATE ATOM

We consider two nonidentical atoms A and B with infinite masses. We take atom A to be in the excited state and situated at a point with radius vector $\mathbf{R}_A$ and B in the ground state and situated at a point $\mathbf{R}_B$. We suppose the electromagnetic field to be in its vacuum state. The exchange interaction is negligible. Let us suppose for the sake of simplicity that the radiation width of excited level of atom A is negligible in comparison with the width of the excited level of atom B. The Hamiltonian of the system is as follows:

$$\hat{H} = \hat{H}_A + \hat{H}_B + \hat{H}_{ph} + \hat{H}_{int}, \tag{1}$$

where $\hat{H}_A = \Sigma_i \varepsilon_{Ai}\hat{b}_i^{\dagger}\hat{b}_i$, $\hat{H}_B = \Sigma_i \varepsilon_{Bi}\hat{\beta}_i^{\dagger}\hat{\beta}_i$ are the Hamiltonians of noninteracting atoms A and B, ε_i is the energy of the ith state of the corresponding atom, $\hat{b}_i(\hat{b}_i^{\dagger})$ and $\hat{\beta}_i(\hat{\beta}_i^{\dagger})$ are annihilation (creation) operators of the ith state of the corresponding atom, $\hat{H}_{ph} = \Sigma_{\mathbf{k}\lambda}\omega(\lambda)\left(\hat{\alpha}^{\dagger}_{\mathbf{k}\lambda}\hat{\alpha}_{\mathbf{k}\lambda} + \frac{1}{2}\right)$ is the Hamiltonian of the free electromagnetic field, $\mathbf{k}$ is the wave vector, $\lambda = 1,2,3$ is the index of polarization of the electromagnetic field, $\hat{\alpha}_{\mathbf{k}\lambda}$ ($\hat{\alpha}^{\dagger}_{\mathbf{k}\lambda}$) are annihilation (creation) operators of the electromagnetic field,

$$\hat{H}_{int} = -\int \hat{\psi}^{\dagger}(\mathbf{r} - \mathbf{R}_A)\hat{d}^{\nu}\hat{E}^{\nu}(\mathbf{r})\hat{\psi}(\mathbf{r} - \mathbf{R}_A)d\mathbf{r}$$

$$-\int \hat{\varphi}^{\dagger}(\mathbf{r} - \mathbf{R}_B)\hat{d}^{\nu}\hat{E}^{\nu}(\mathbf{r})\hat{\varphi}(\mathbf{r} - \mathbf{R}_B)d\mathbf{r} \tag{2}$$

is the interaction Hamiltonian, where

$$\hat{\psi} = \sum_i \psi_i(\mathbf{r} - \mathbf{R}_A)\hat{b}_i, \quad \hat{\varphi} = \sum_i \varphi_i(\mathbf{r} - \mathbf{R}_B)\hat{\beta}_i, \tag{3}$$

with $\psi_i(\mathbf{r} - \mathbf{R}_A)$ and $\varphi_i(\mathbf{r} - \mathbf{R}_B)$ being the wave functions of the ith state of the corresponding atoms. $\hat{d}^{\nu}$ is the operator of the dipole moment, $\hat{E}^{\nu}(\mathbf{r})$ is the operator of the free electromagnetic field,

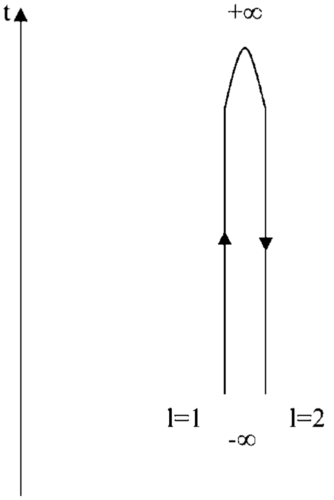

FIG. 1. Contour c.

$$\hat{E}^{\nu}(\mathbf{r}) = i\sum_{\mathbf{k}\lambda}\sqrt{\frac{2\pi\omega(\lambda)}{V}}e^{\nu}_{\mathbf{k}\lambda}(\hat{\alpha}_{\mathbf{k}\lambda}e^{i\mathbf{k}\cdot\mathbf{r}} - \hat{\alpha}^{\dagger}_{\mathbf{k}\lambda}e^{-i\mathbf{k}\cdot\mathbf{r}}), \tag{4}$$

where V is quantization volume, $e^{\nu}_{\mathbf{k}\lambda}$ is the polarization unit vector, $\omega(1,2) = k$, and $\omega(3) = 0$. Now our aim is to calculate the van der Waals potential for the system. It is evident that this potential is equal to the energy shift of a single atom resulting from the presence of the other atom. Consequently, we should calculate the energy shift of, say, an excited atom.

To take into account the level widths of atoms we should use a nonperturbative approach. But methods based upon the linear response theory [9,10] or macroscopic quantum electrodynamics [11,12] are not suitable for us, since these methods involve classical polarizabilities of atoms. In a number of problems these methods yield the correct results [9–12], but as we are going to show, in the general case the van der Waals potential or the Casimir force cannot be expressed in terms of classical polarizabilities. To calculate the energy shift we will use the method of quantum Green functions similar to the one suggested by Keldysh for kinetics in a medium [16,19]. This method has no phenomenological elements, but on the other hand, it will be possible to take into account the energy-level widths of the atoms.

Let us consider the excited atom. Let

$$G^{A}_{ll'}(x,x') = -i\langle \hat{T}_c \hat{\psi}_l(x)\hat{\psi}^{\dagger}_{l'}(x')\hat{S}_c\rangle \tag{5}$$

be the Green function of atom A. Here $x = \{\mathbf{r}, t\}$, operators are in interaction representation [16],

$$\hat{S}_c = \hat{T}_c \exp\left\{\sum_{l=1,2}(-1)^l i\int_c \hat{H}_{int\ l}(t)dt\right\} \tag{6}$$

is the scattering operator, c is the contour of integration given in Fig. 1, $\hat{T}_c$ is the operator of time ordering for contour c [19], $\hat{H}_{intl}(t)$ is in interaction representation, and $\langle\cdots\rangle$ means averaging over the initial state of free atoms. Using Eqs. (2)–(4), we obtain

$$\hat{H}_{int\ l}(t) = -\int \hat{\psi}_l^\dagger(x)\hat{d}^\nu \hat{E}_l^\nu(x)\hat{\psi}_l(x)d\mathbf{r} - \int \hat{\varphi}_l^\dagger(x)\hat{d}^\nu \hat{E}_l^\nu(x)\hat{\varphi}_l(x)d\mathbf{r}, \tag{7}$$

where

$$\hat{\psi} = \sum_i \psi_i(\mathbf{r}-\mathbf{R}_A)e^{-i\varepsilon_{Ai}t}\hat{b}_i, \quad \hat{\varphi} = \sum_i \varphi_i(\mathbf{r}-\mathbf{R}_B)e^{-i\varepsilon_{Bi}t}\hat{\beta}_i, \tag{8}$$

$$\hat{E}^\nu(x) = i\sum_{\mathbf{k}\lambda} \sqrt{\frac{2\pi\omega(\lambda)}{V}} e_{\mathbf{k}\lambda}^\nu (\hat{\alpha}_{\mathbf{k}\lambda}e^{i\mathbf{k}\cdot\mathbf{r}}e^{-i\omega\lambda(\lambda)t} - \hat{\alpha}_{\mathbf{k}\lambda}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}e^{i\omega(\lambda)t}). \tag{9}$$

Using the Green function (5) it is easy to find the matrix of density of atom A:

$$\rho^A(x,x') = iG_{12}^A(x,x').$$

Representing the S matrix (6) as a perturbation expansion we come to the following system of equations (Appendix A):

$$\rho^{\mathrm{A}}(x,x') = \rho_c^A(x,x') + \rho_n^A(x,x'), \tag{10}$$

where $\rho_c^A(x,x')$ represents the coherent channel of interaction, with atom A returning to the initial state (e.g., elastic scattering). The matrix $\rho_n^A(x,x')$ represents the incoherent channel, where atom A does not return to the initial state after the interaction (e.g., spontaneous radiation, Raman scattering, etc.). Here we are not interested in the incoherent channel processes and we omit $\rho_n^A(x,x')$.

For the coherent channel we obtain equations similar to the ones derived in [20] for an electromagnetic field and in [21] for a system of atoms:

$$\begin{aligned}\rho_c^A(x,x') = {} & \rho_0^A(x,x') + \int dx_1dx_2 g_{11}^A(x,x_1)M_{11}(x_1,x_2)\rho_0^A(x_2,x') \\ & + \int dx_1dx_2\rho_0^A(x,x_1)M_{22}(x_1,x_2)g_{22}^A(x_2,x') \\ & + \int dx_1dx_2dx_3dx_4 g_{11}^A(x,x_1)M_{11}(x_1,x_2)\rho_0^A(x_2,x_3) \\ & \times M_{22}(x_3,x_4)g_{22}^A(x_4,x'),\end{aligned} \tag{11}$$

where $\rho_0^A(x,x')$ is the matrix of density of free atom A,

$$\rho_0^A(x,x') = \psi_0^*(\mathbf{r}')\psi_0(\mathbf{r})e^{-i\varepsilon_{A0}(t-t')}, \tag{12}$$

where 0 stands for the initial state of atom A, $g_r^A(x,x')$ $[g_a^A(x,x')]$ is the retarded [advanced] propagator of atom A, which obeys the equations [22]

$$\begin{aligned}g_{11}^A(x,x') = {} & g_{11}^{0A}(x,x') \\ & + \int dx_1dx_2 g_{11}^{0A}(x,x_1)M_{11}(x_1,x_2)g_{11}^A(x_2,x'),\end{aligned}$$

$$\begin{aligned}g_{22}^A(x,x') = {} & g_{22}^{0A}(x,x') \\ & + \int dx_1dx_2 g_{22}^{0A}(x,x_1)M_{22}(x_1,x_2)g_{22}^A(x_2,x').\end{aligned} \tag{13}$$

with

$$\begin{aligned}g_{11}^{0A}(x,x') & = -i\left\langle \hat{T}_c\hat{\psi}_1(x)\hat{\psi}_1^\dagger(x')\right\rangle_{\mathrm{vac}} \\ & = -i\theta(t-t')\sum_i \psi_i^*(\mathbf{r}')\psi_i(\mathbf{r})e^{-i\varepsilon_{Ai}(t-t')},\end{aligned}$$

$$\begin{aligned}g_{22}^{0A}(x,x') & = -i\left\langle \hat{T}_c\hat{\psi}_2(x)\hat{\psi}_2^\dagger(x')\right\rangle_{\mathrm{vac}} \\ & = -i\theta(t'-t)\sum_i \psi_i^*(\mathbf{r}')\psi_i(\mathbf{r})e^{-i\varepsilon_{Ai}(t-t')}\end{aligned} \tag{14}$$

being the retarded (advanced) propagator of free atom A. M_{11} and M_{22} are the mass operators,

$$M_{11}(x,x') = -ig_{11}^A(x,x')D_{11}^{\nu\nu'}(x',x)\hat{d}^\nu\hat{d}^{\nu'},$$

$$M_{22}(x,x') = -ig_{22}^A(x,x')D_{22}^{\nu\nu'}(x',x)\hat{d}^\nu\hat{d}^{\nu'}, \tag{15}$$

where $D_{11}^{\nu\nu'}(x',x)$ and $D_{22}^{\nu\nu'}(x',x)$ are photon propagators [22],

$$\begin{aligned}D_{11}^{\nu\nu'}(x',x) = {} & D_{11}^{0\nu\nu'}(x',x) + \int dx_1dx_2 \\ & \times \sum_{\nu_1\nu_2} D_{11}^{0\nu\nu_1}(x',x_1)\Pi_{11}^{\nu_1\nu_2}(x_1,x_2)D_{11}^{\nu\nu_1}(x_2,x),\end{aligned}$$

$$\begin{aligned}D_{22}^{\nu\nu'}(x',x) = {} & D_{22}^{0\nu\nu'}(x',x) + \int dx_1dx_2 \\ & \times \sum_{\nu_1\nu_2} D_{22}^{0\nu\nu_1}(x',x_1)\Pi_{22}^{\nu_1\nu_2}(x_1,x_2)D_{22}^{\nu\nu_1}(x_2,x),\end{aligned} \tag{16}$$

with

$$D_{11}^{0\nu\nu'}(x',x) = i\left\langle \hat{T}_c\hat{E}_1^\nu(x')\hat{E}_1^{\nu'}(x)\right\rangle_{\mathrm{vac}},$$

$$D_{22}^{0\nu\nu'}(x',x) = i\left\langle \hat{T}_c\hat{E}_2^\nu(x')\hat{E}_2^{\nu'}(x)\right\rangle_{\mathrm{vac}}. \tag{17}$$

In the frequency-coordinate domain these functions are equal [22]:

$$\begin{aligned}& D_{11}^{0\nu\nu'}(\omega,\mathbf{r}-\mathbf{r}') \\ & = \omega^2\left[\delta_{\nu\nu'}\left(1+\frac{i}{|\omega||\mathbf{r}-\mathbf{r}'|}-\frac{1}{\omega^2|\mathbf{r}-\mathbf{r}'|^2}\right)+\frac{(\mathbf{r}-\mathbf{r}')_\nu(\mathbf{r}-\mathbf{r}')_{\nu'}}{|\mathbf{r}-\mathbf{r}'|^2}\right. \\ & \left. \times\left(\frac{3}{\omega^2|\mathbf{r}-\mathbf{r}'|^2}-\frac{3i}{|\omega||\mathbf{r}-\mathbf{r}'|}-1\right)\frac{e^{i|\omega||\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}\right],\end{aligned}$$

$$D_{22}^{0\nu\nu'}(\omega,\mathbf{r}-\mathbf{r}') = [D_{11}^{0\nu\nu'}(\omega,\mathbf{r}-\mathbf{r}')]^*. \tag{18}$$

Now it is convenient to rewrite the integral equation (11)as a differential one (Appendix B):

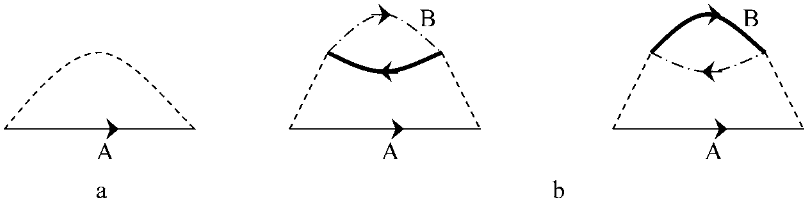

FIG. 2. Feynman's diagrams.

$$\rho_c^A(x,x') = \Psi(x)\Psi^*(x'),$$

$$\left(i\frac{\partial}{\partial t} - \hat{H}_A\right)\Psi(x) = \int M_{11}(x,x_1)\Psi(x_1)dx_1. \tag{19}$$

The coherent channel processes do not change the initial state of atom A, and consequently

$$\Psi(x) = \psi_0(\mathbf{r} - \mathbf{R}_A)f(t), \tag{20}$$

where the index 0 stands for the initial state of atom A. Substituting Eq. (20) into Eqs. (19) and neglecting nondiagonal elements of the mass operator we arrive at the equation

$$i\frac{\partial}{\partial t}f(t) - \varepsilon_{0\mathrm{A}}f(t) = \int_{t_0}^{\infty} M_{11}^{00}(t,t_1)f(t_1)dt_1,$$

$$M_{11}^{00}(t,t_1) = \int \psi_0^*(\mathbf{r} - \mathbf{R}_A)M_{11}(x,x_1)\psi_0(\mathbf{r}_1 - \mathbf{R}_A)d\mathbf{r}d\mathbf{r}_1; \tag{21}$$

here, we suppose that the interaction was switched on at t_0 ($t_0 \to -\infty$).

Using the pole approximation we find

$$\Psi(x) = \psi_0(\mathbf{r} - \mathbf{R}_A)e^{-i\varepsilon_{\mathrm{A}0}t}e^{-iM_{11}^{00}(\varepsilon_{\mathrm{A}0})(t-t_0)},$$

where $M_{11}^{00}(\varepsilon_{\mathrm{A}0}) = \int_{-\infty}^{\infty} M_{11}^{00}(t,t')e^{i\varepsilon_{\mathrm{A}0}(t-t')}d(t-t')$ is the Fourier transform of mass operator taken at point $E = \varepsilon_{A0}$.

Thus the density matrix of the coherent channel in the energy domain

$$\rho_c^A(E,E',\mathbf{r},\mathbf{r}') = \int_{t_0}^{\infty} \rho_c^A(x,x')e^{iEt - iE't'}dt\,dt', \quad t_0 \to -\infty,$$

is

$$\rho_c^A(E,E',\mathbf{r},\mathbf{r}') = \frac{\psi_0(\mathbf{r} - \mathbf{R}_A)\psi_0^*(\mathbf{r}' - \mathbf{R}_A)e^{i(E-E')t_0}}{[E - \varepsilon_{\mathrm{A}0} - M_{11}^{00}(\varepsilon_{\mathrm{A}0})][E' - \varepsilon_{\mathrm{A}0} - M_{22}^{00}(\varepsilon_{\mathrm{A}0})]}. \tag{22}$$

Such an equation for the case of the electromagnetic field was obtained [20].

Now we can easily calculate the energy shift of atom A and, consequently, the van der Waals potential

$$U(\mathbf{R}_A - \mathbf{R}_B) = \Delta E_A = \mathrm{Re}[M_{11}^{00}(\varepsilon_{\mathrm{A}0})] \tag{23}$$

and the energy-level width for atom A resulting from interaction with the vacuum and atom B:

$$\frac{\Gamma_{\mathrm{A}}}{2} = -Im[M_{11}^{00}(\varepsilon_{\mathrm{A}0})]. \tag{24}$$

We suppose that the Lamb shift due to the interaction with the vacuum is already taken into account in $\varepsilon_{\mathrm{A}0}$ and in expression (23) we take into account only interaction between atoms A and B. Using Eqs. (13), (15), and (16) we can draw Feynman's diagrams given in Fig. 2.

Here the solid line corresponds to g^A, the dashed line corresponds to $D_{ll'}^{0\nu\nu'}$, the dash-dotted line represents ρ^B, and the thick solid line represents g^B, which are the density matrix and propagator of atom B.

We suppose that the ground energy levels of atoms have no width; thus, we can replace ρ^B by ρ_0^B and g^A by g_0^A. The propagator g^B obeys Eq. (13). For the sake of simplicity, in Eq. (13) we take into account only the interaction of atom B with the vacuum, which is described by the mass operator given in Fig. 2(a), where the solid line represents g_0^B. The solution of Eq. (13) in the energy domain is

$$g_{11}^B(E,\mathbf{r},\mathbf{r}') = \sum_i \frac{\varphi_i(\mathbf{r} - \mathbf{R}_B)\varphi_i^*(\mathbf{r}' - \mathbf{R}_B)}{E - \varepsilon_{Bi} + i\dfrac{\gamma_{Bi}}{2}}, \tag{25}$$

with $\gamma_{iB}/2 = -\mathrm{Im}[M_{11}^{ii}(\varepsilon_{\mathrm{B}0})]$ being the radiation width of energy level i, while $M_{11}^{ii}(\varepsilon_{\mathrm{B}0})$ is described by the diagram shown in Fig. 2(a).

Thus, for the mass operator given in Fig. 2 with omitting terms whose contribution to the final result is zero, we find

$$M_{11}(x,x') = -i\hat{d}^{\nu}\hat{d}^{\nu'}g_r^{0A}(x,x')D_{11}^{0\nu\nu'}(x',x) + ig_r^{0A}(x,x')\int \hat{d}^{\nu}\hat{d}^{\nu'}\hat{d}^{\nu_1}\hat{d}^{\nu_2}[D_{11}^{0\nu\nu_1}(x,x_1)\rho_0^{\mathrm{B}}(x_1,x_2)g_r^B(x_2,x_1)D_{11}^{0\nu_2\nu'}(x_2,x') + D_{11}^{0\nu\nu_1}(x,x_1)\rho_0^{\mathrm{B}}(x_2,x_1)g_r^B(x_1,x_2)D_{11}^{0\nu_2\nu'}(x_2,x')]dx_1dx_2. \tag{26}$$

The first term corresponds to the interaction of atom A with the vacuum [Fig. 2(a)]; it results in a radiation-level width and Lamb shift. Consequently, we can omit this term. The second term [Fig. 2(b)] corresponds to the interaction between atoms A and B. In the energy domain we have

$$M_{11}(E,\mathbf{r},\mathbf{r}') = \frac{i}{(2\pi)^8} g_r^{0A}(E-\omega,\mathbf{r},\mathbf{r}') \int \hat{d}^{\nu}\hat{d}^{\nu'}\hat{d}^{\nu_1}\hat{d}^{\nu_2}(D_{11}^{0\nu\nu_1}(\omega,\mathbf{k}_1)\rho_0^{\mathrm{B}}(E'+\omega,\mathbf{r}_1,\mathbf{r}_2)g_r^{B}(E',\mathbf{r}_2,\mathbf{r}_1)D_{11}^{0\nu_2\nu'}(\omega,\mathbf{k}_2) + D_{11}^{0\nu\nu_1}(\omega,\mathbf{k}_1)\rho_0^{\mathrm{B}}(E'-\omega,\mathbf{r}_2,\mathbf{r}_1)g_r^{B}(E',\mathbf{r}_1,\mathbf{r}_2)D_{11}^{0\nu_2\nu'}(\omega,\mathbf{k}_2))e^{i\mathbf{k}_1(\mathbf{r}-\mathbf{r}_1)}e^{i\mathbf{k}_2(\mathbf{r}_2-\mathbf{r}')}dE'\,d\omega\,d\mathbf{r}_1\,d\mathbf{r}_2. \tag{27}$$

The Fourier transforms of g_r^{0A} and ρ_0^{B} could be easily found using and Eqs. (12) and (14):

$$\rho_0^{B}(E,\mathbf{r},\mathbf{r}') = 2\pi\varphi_0^{*}(\mathbf{r}')\varphi_0(\mathbf{r})\delta(E-\varepsilon_{B0}), \tag{28}$$

$$g_r^{0A}(E,\mathbf{r},\mathbf{r}') = \sum_i \frac{\psi_i^{*}(\mathbf{r}')\psi_i(\mathbf{r})}{E-\varepsilon_{Ai}+i0}. \tag{29}$$

Substituting Eqs. (25), (28), and (29) into Eq. (27), we find

$$M_{11}(E,\mathbf{r},\mathbf{r}') = \frac{i}{(2\pi)^7}\frac{\psi_g^{*}(\mathbf{r}'-\mathbf{R}_A)\psi_g(\mathbf{r}-\mathbf{R}_A)}{E-\omega-\varepsilon_{Ag}+i0}\int \hat{d}^{\nu}\hat{d}^{\nu'}\hat{d}^{\nu_1}\hat{d}^{\nu_2}\left(D_{11}^{0\nu\nu_1}(\omega,\mathbf{k}_1)\varphi_g^{*}(\mathbf{r}_2-\mathbf{R}_B)\varphi_g(\mathbf{r}_1-\mathbf{R}_B)\times\frac{\varphi_e(\mathbf{r}_2-\mathbf{R}_B)\varphi_e^{*}(\mathbf{r}_1-\mathbf{R}_B)}{\varepsilon_{\mathrm{B}g}-\varepsilon_{Be}-\omega+i\dfrac{\gamma_B}{2}}D_{11}^{0\nu_2\nu'}(\omega,\mathbf{k}_2) + D_{11}^{0\nu\nu_1}(\omega,\mathbf{k}_1)\varphi_g^{*}(\mathbf{r}_1-\mathbf{R}_B)\varphi_g(\mathbf{r}_2-\mathbf{R}_B)\times\frac{\varphi_e(\mathbf{r}_1-\mathbf{R}_B)\varphi_e^{*}(\mathbf{r}_2-\mathbf{R}_B)}{\varepsilon_{\mathrm{B}g}-\varepsilon_{Be}+\omega+i\dfrac{\gamma_B}{2}}D_{11}^{0\nu_2\nu'}(\omega,\mathbf{k}_2)\right)e^{i\mathbf{k}_1(\mathbf{r}-\mathbf{r}_1)}e^{i\mathbf{k}_2(\mathbf{r}_2-\mathbf{r}')}d\omega\,d\mathbf{r}_1\,d\mathbf{r}_2\,d\mathbf{k}_1\,d\mathbf{k}_2. \tag{30}$$

Here g and e stand for ground and excited states correspondingly.

Now we should substitute Eq. (30) into Eq. (21) and take into account the integral in the dipole approximation

$$\int e^{i\mathbf{k}\cdot\mathbf{r}}\psi_i^{*}(\mathbf{r}-\mathbf{R})\hat{d}^{\nu}\psi_j(\mathbf{r}-\mathbf{R})d\mathbf{r} = d_{ij}^{\nu}e^{i\mathbf{k}\cdot\mathbf{R}},$$

where d_{ij}^{ν} is the matrix element of dipole moment:

$$M_{11}^{00}(\varepsilon_{\mathrm{A}e}) = \frac{i}{(2\pi)}\frac{1}{\varepsilon_{\mathrm{A}e}-\omega-\varepsilon_{Ag}+i0}\times\int\left(D_{11}^{0\nu\nu_1}(\omega,\mathbf{R}_B-\mathbf{R}_A)\frac{d_{eg}^{\nu A}d_{ge}^{\nu'\mathrm{A}}d_{eg}^{\nu_1 B}d_{ge}^{\nu_2 B}}{\varepsilon_{\mathrm{B}g}-\varepsilon_{Be}-\omega+i\dfrac{\gamma_B}{2}}\times D_{11}^{0\nu_2\nu'}(\omega,\mathbf{R}_A-\mathbf{R}_B) + D_{11}^{0\nu\nu_1}(\omega,\mathbf{R}_B-\mathbf{R}_A)\times\frac{d_{eg}^{\nu A}d_{ge}^{\nu'\mathrm{A}}d_{ge}^{\nu_1 B}d_{eg}^{\nu_2 B}}{\varepsilon_{\mathrm{B}g}-\varepsilon_{Be}+\omega+i\dfrac{\gamma_B}{2}}D_{11}^{0\nu_2\nu'}(\omega,\mathbf{R}_A-\mathbf{R}_B)\right)d\omega. \tag{31}$$

Using the symmetry property of $D_{11}^{0\nu\nu'}$ function $D_{11}^{0\nu\nu'}(\omega)$ $=D_{11}^{0\nu\nu'}(-\omega)$, which is evident from Eq. (18), we can rewrite Eq. (31) in terms of the coherent polarizabilities introduces in [22] and widely discussed in [23],

$$M_{11}^{00}(\varepsilon_{\mathrm{A}e}) = \frac{i}{4\pi}\int_{-\infty}^{\infty} D_{11}^{0\nu\nu_1}(\omega,\mathbf{R}_B-\mathbf{R}_A)D_{11}^{0\nu_2\nu'}(\omega,\mathbf{R}_A-\mathbf{R}_B)\times\alpha_A^{(c)\nu\nu'}(\omega)\alpha_B^{(c)\nu_1\nu_2}(\omega)d\omega, \tag{32}$$

or using Eqs. (23) and (24), we find

$$U(\mathbf{R}_A-\mathbf{R}_B) = \mathrm{Re}\left[\frac{i}{4\pi}\int_{-\infty}^{\infty} D_{11}^{0\nu\nu_1}(\omega,\mathbf{R}_B-\mathbf{R}_A)D_{11}^{0\nu_2\nu'}(\omega,\mathbf{R}_A-\mathbf{R}_B)\times\alpha_A^{(c)\nu\nu'}(\omega)\alpha_B^{(c)\nu_1\nu_2}(\omega)d\omega\right] \tag{33}$$

and

$$\frac{\Gamma_{\mathrm{A}}}{2} = -\mathrm{Im}\left[\frac{i}{4\pi}\int_{-\infty}^{\infty} D_{11}^{0\nu\nu_1}(\omega,\mathbf{R}_B-\mathbf{R}_A)D_{11}^{0\nu_2\nu'}(\omega,\mathbf{R}_A-\mathbf{R}_B)\times\alpha_A^{(c)\nu\nu'}(\omega)\alpha_B^{(c)\nu_1\nu_2}(\omega)d\omega\right], \tag{34}$$

with the coherent polarizability for the ground-state atom,

$$\alpha_g^{(c)\nu\nu'}(\omega) = \frac{d_{ge}^{\nu}d_{eg}^{\nu'}}{\omega_{eg}-\omega-i\dfrac{\gamma}{2}} + \frac{d_{eg}^{\nu}d_{ge}^{\nu'}}{\omega_{eg}+\omega-i\dfrac{\gamma}{2}}, \tag{35}$$

and for the excited atom,

$$\alpha_e^{(c)\nu\nu'}(\omega) = \frac{d_{eg}^{\nu} d_{ge}^{\nu}}{-\omega_{eg} - \omega - i\dfrac{\gamma}{2}} + \frac{d_{ge}^{\nu} d_{eg}^{\nu'}}{-\omega_{eg} + \omega - i\dfrac{\gamma}{2}}. \qquad (36)$$

Here we introduce $\omega_A = \varepsilon_{eA} - \varepsilon_{gA}$ and $\omega_B = \varepsilon_{eB} - \varepsilon_{gB}$.

The conventional polarizabilities of atoms are well known [22]:

$$\alpha_g^{\nu\nu'}(\omega) = \frac{d_{ge}^{\nu} d_{eg}^{\nu'}}{\omega_{eg} - \omega - i\dfrac{\gamma}{2}} + \frac{d_{eg}^{\nu} d_{ge}^{\nu'}}{\omega_{eg} + \omega + i\dfrac{\gamma}{2}}, \qquad (37)$$

$$\alpha_e^{\nu\nu'}(\omega) = \frac{d_{eg}^{\nu} d_{ge}^{\nu'}}{-\omega_{eg} - \omega - i\dfrac{\gamma}{2}} + \frac{d_{ge}^{\nu} d_{eg}^{\nu'}}{-\omega_{eg} + \omega + i\dfrac{\gamma}{2}}. \qquad (38)$$

Equation (33) coincides with the result well known from the classical textbook [22] for the case of neglecting the radiation width ($\gamma_B/2 \to 0$) and supposing that atom A is not excited ($\omega_A \to -\omega_A$). Equations (35) and (37) as well as Eqs. (36) and (38) are different due to the sign of the imaginary part of the denominators of the second terms. The signs of the imaginary parts of the denominators of the conventional polarizabilities (37) and (38) are connected with their analytical properties. They should be analytical in the upper part of the complex plane, while the signs of the imaginary parts of coherent polarizabilities (35) and (36) are the result of the causality principle. If we change the corresponding signs in our calculations, we will come to violation of the causality principle in quantum electrodynamics [22]. As was shown in [23–25] the signs in the denominators of coherent polarizabilities could be changed only due to the presence of an incoherent channel, which describes the processes of spontaneous and induced radiation (at any rate the initial state of the atoms should be changed). But in our case of the van der Waals interaction, the incoherent channel does not contribute to the result.

After averaging over all possible orientations of dipole moments of atoms we can write [22]

$$d_{eg}^{\nu_1} d_{ge}^{\nu_2} \to \frac{|d_{eg}|^2}{3} \delta_{\nu_1 \nu_2}. \qquad (39)$$

Let us consider a case of small distance between the atoms, $R \ll \lambda$, where R is the distance between the atoms and λ is the wavelength of radiation of atoms. Substituting Eq. (18) into Eq. (33) and taking into account Eq. (39) as well as $R \ll \lambda$:

$$U(R) = \mathrm{Re}\left[\frac{3i}{2\pi R^6} \int_{-\infty}^{\infty} \alpha_A^{(c)}(\omega) \alpha_B^{(c)}(\omega) d\omega\right]. \qquad (40)$$

After substituting Eqs. (35) and (36) into Eq. (40), we find

$$U_{eg}(R) = \frac{2}{3R^6} \frac{(\omega_A - \omega_B)|d_{eg}^A|^2 |d_{eg}^B|^2}{(\omega_A - \omega_B)^2 + \left(\dfrac{\gamma_B}{2}\right)^2}. \qquad (41)$$

For the case of ($\gamma_B/2 \to 0$) we come to the formula obtained in [7,8]. Here we should mention that the van der Waals interaction between the excited and ground-state atoms could be either attractive or repulsive depending on the sign of $\omega_A - \omega_B$.

Now let us consider an opposite case. Let atom A be a ground-state one and atom B be an excited one. In this case we should use Eq. (26), but we should substitute Eqs. (22) and (14) into Eq. (26) but not Eqs. (28) and (25). If $t \ll \tau$, where t is the time of interaction and τ is the lifetime of excited state of atom B, we come to the evident result

$$U_{ge}(R) = \frac{2}{3R^6} \frac{(\omega_B - \omega_A)|d_{eg}^A|^2 |d_{eg}^B|^2}{(\omega_A - \omega_B)^2 + \left(\dfrac{\gamma_B}{2}\right)^2}. \qquad (42)$$

The results for the case of both the ground-state atoms could be obtained analogously:

$$U_{gg}(R) = -\frac{2}{3R^6} \frac{(\omega_A + \omega_B)|d_{eg}^A|^2 |d_{eg}^B|^2}{(\omega_A + \omega_B)^2 + \left(\dfrac{\gamma_B}{2}\right)^2}. \qquad (43)$$

Evidently the result (43) corresponds to attraction of atoms. This result coincides with the London formula [1] if ($\gamma_B/2 \to 0$).

III. INTERACTION BETWEEN AN ATOM AND A DIELECTRIC SURFACE

For the sake of simplicity we will consider a dielectric semi-infinite body of a dilute gas of atoms. Our aim is to compare the results for the van der Waals force obtained with the help of the Lifshitz formula and the one obtained with the help of quantum electrodynamics, taking into account pair interactions between atoms.

(i) Let us consider an excited atom A near a surface of a gas of ground-state atoms B. Taking into account only pair interactions, we can obtain a formula for the interaction potential by integrating Eq. (41) with respect to the volume of the medium, with γ_B being a collision width of excited energy level of the atoms of the gas:

$$U_1(z_0) = \int dV \frac{2}{3R^6} \frac{(\omega_A - \omega_B)|d_{eg}^A|^2 |d_{eg}^B|^2}{(\omega_A - \omega_B)^2 + \left(\dfrac{\gamma_B}{2}\right)^2} n,$$

where n is the number of density of atoms of the medium.

If the atom is separated by a distance of z_0 from the interface, the result of integrating is

$$U_1(z_0) = \frac{\pi}{9{z_0}^3} \frac{(\omega_A - \omega_B)|d_{eg}^A|^2 |d_{eg}^B|^2}{(\omega_A - \omega_B)^2 + \left(\frac{\gamma_B}{2}\right)^2} n. \tag{44}$$

(ii) Let us consider the interaction of ground-state atom A and a ground gaseous medium of atoms B. Using expression (43), we find

$$U_2(z_0) = -\frac{\pi}{9{z_0}^3} |d_{eg}^A|^2 |d_{eg}^B|^2 \frac{(\omega_A + \omega_B)n}{(\omega_A + \omega_B)^2 + \left(\frac{\gamma_B}{2}\right)^2}. \tag{45}$$

The results (44) and (45) are in agreement with the well-known experimental and theoretical results [9–14].

(iii) Let us consider the interaction of ground-state atom A and an excited gaseous medium of atoms B. Using expressions (42) and (43), we find

$$U_3(z_0) = \frac{\pi}{9{z_0}^3} |d_{eg}^A|^2 |d_{eg}^B|^2 \times \left(\frac{(\omega_B - \omega_A)n_e}{(\omega_A - \omega_B)^2 + \left(\frac{\gamma_B}{2}\right)^2} - \frac{(\omega_A + \omega_B)n_g}{(\omega_A + \omega_B)^2 + \left(\frac{\gamma_B}{2}\right)^2} \right), \tag{46}$$

where n_e and n_g are the density numbers of excited and ground-state atoms. The result of such a kind, as far as we know, is obtained for the first time.

Using Eq. (40), we can rewrite expressions (44)–(46) in the following way:

$$U(z_0) = \mathrm{Re}\left[\frac{i}{16\pi z_0^3} \int_{-\infty}^{\infty} \alpha_A^{(c)}(\omega)[\varepsilon^{(c)}(\omega) - 1] d\omega \right]. \tag{47}$$

Here we introduce the coherent permittivity

$$\varepsilon^{(c)}(\omega) = 1 + 4\pi[n_e \alpha_e^{(c)}(\omega) + n_g \alpha_g^{(c)}(\omega)], \tag{48}$$

where $\alpha_e^{(c)}(\omega)$ and $\alpha_g^{(c)}(\omega)$ are the coherent polarizabilities given by Eqs. (35) and (36).

The conventional permittivity could be constructed of the conventional polarizabilities (37) and (38) and as follows:

$$\varepsilon(\omega) = 1 + 4\pi[n_e \alpha_e(\omega) + n_g \alpha_g(\omega)]. \tag{49}$$

We see that the van der Waals potential of an atom interacting with a dielectric interface is expressed in terms of the coherent permittivity, but not in terms of the conventional one. But for the case of a single excited atom interacting with a nonexcited medium, the result obtained by the linear response theory is in agreement with the result obtained by quantum electrodynamics, Eq. (44). The situation differs dramatically if a ground-state atom interacts with a medium of excited atoms. The result (46) obtained with the help of quantum electrodynamics cannot be obtained in the framework of linear response theory or other phenomenological approaches requiring conventional permittivities to describe media [9–12]. It results from the dependence of the conventional permittivity of the media, which must be included in the result obtained with the help of the phenomenological approach, on a difference of numbers of the density of ground-state and excited atoms:

$$\varepsilon(\omega) - 1 = 4\pi[n_e \alpha_e(\omega) + n_g \alpha_g(\omega)] \propto n_g - n_e.$$

But the result obtained here without phenomenology (46) does not depend on such a difference.

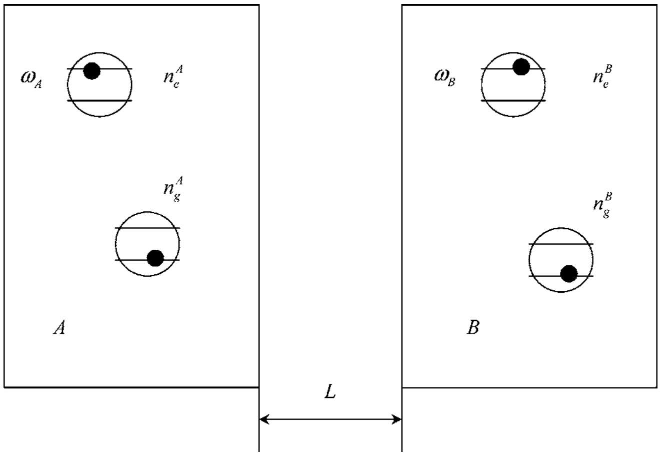

FIG. 3. Interacting media.

We must stress that a similar situation, where the result is expressed in terms of the coherent permittivity, but not the conventional one, appears in other phenomena. It has been shown [20,24] that the reflection coefficient of resonant radiation reflected from a gas medium containing excited atoms is expressed in terms of the coherent permittivity. The correlation function $\langle E^{\nu}(\mathbf{r}', t) E^{\nu'}(\mathbf{r}, t) \rangle$ of an electromagnetic field in a hot medium depends on the coherent permittivity as well [23].

IV. INTERACTION BETWEEN TWO MEDIA OF EXCITED ATOMS

Let us consider the simplest case of two media of dilute gases separated by a distance of L (Fig. 3). Let both the media contain excited atoms.

To find the van der Waals potential per unit area we should integrate Eq. (47) with respect to dz_0, taking into account pair interactions of atoms of both the media. The result is evident:

$$u(L) = \mathrm{Re}\left[\frac{i}{128\pi^2 L^2} \int_{-\infty}^{\infty} [\varepsilon_A^{(c)}(\omega) - 1][\varepsilon_B^{(c)}(\omega) - 1] d\omega \right], \tag{50}$$

where $\varepsilon_A^{(c)}(\omega)$ and $\varepsilon_B^{(c)}(\omega)$ are the coherent permittivities of media A and B, which are expressed through the coherent permittivities (48). Differentiating Eq. (50) by L, we can find the van der Waals force per unit area:

$$F(L) = \frac{\partial}{\partial L} u(L),$$

$$F(L)=\mathrm{Re}\left[-\frac{i}{64\pi^2L^3}\int_{-\infty}^{\infty}[\varepsilon_A^{(c)}(\omega)-1][\varepsilon_B^{(c)}(\omega)-1]d\omega\right]. \tag{51}$$

Substituting Eqs. (48), (35), and (36) into Eq. (51) and calculating the integral with respect to $d\omega$, we find

$$F(L)=\frac{\pi}{9L^3}|d_{eg}^A|^2|d_{eg}^B|^2\left(n_g^An_g^B\frac{(\omega_A+\omega_B)}{(\omega_A+\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}-(n_e^An_g^B-n_g^An_e^B)\frac{(\omega_A-\omega_B)}{(\omega_A-\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}-n_e^An_e^B\frac{(\omega_A+\omega_B)}{(\omega_A+\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}\right). \tag{52}$$

We consider a case of thermal equilibrium, with atoms obeying the Boltzman distribution

$$n_e^A=n_g^Ae^{-\omega_A/T},\quad n_e^B=n_g^Be^{-\omega_B/T},$$

with $n^A=n_g^A+n_e^A$, $n^B=n_g^B+n_e^B$ being the total numbers of density, which is supposed to be constant:

$$F(L,T)=\frac{\pi}{9L^3}|d_{eg}^A|^2|d_{eg}^B|^2\frac{n^An^B}{\left[1+\exp\left(-\frac{\omega_A}{T}\right)\right]\left[1+\exp\left(-\frac{\omega_B}{T}\right)\right]}\times\left(\frac{(\omega_A+\omega_B)\left[1-\exp\left(-\frac{\omega_A}{T}\right)\exp\left(-\frac{\omega_B}{T}\right)\right]}{(\omega_A+\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}-\frac{\left[\exp\left(-\frac{\omega_A}{T}\right)-\exp\left(-\frac{\omega_B}{T}\right)\right](\omega_A-\omega_B)}{(\omega_A-\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}\right). \tag{53}$$

It is interesting to compare our result (52) with the one derived from the Lifshitz formula [15,16]. For dilute gases the Lifshitz formula is

$$F_L(L)=\frac{1}{32\pi^2L^3}\int_0^{\infty}[\varepsilon_A(iu)-1][\varepsilon_B(iu)-1]du, \tag{54}$$

with $\varepsilon_A(iu)$ and $\varepsilon_B(iu)$ being the conventional permittivities (49), which are expressed in terms of the conventional polarizabilities (35) and (38). After integrating with respect to du supposing that $\omega_A,\omega_B\gg\gamma_B$ we find

$$F_L(L)=\frac{\pi}{9L^3}|d_{eg}^A|^2|d_{eg}^B|^2(n_g^A-n_e^A)(n_g^B-n_e^B)\frac{(\omega_A+\omega_B)}{(\omega_A+\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}. \tag{55}$$

The temperature dependence of the Casimir force resulting from the Lifshitz formula is

$$F_L(L,T)=\frac{\pi}{9L^3}|d_{eg}^A|^2|d_{eg}^B|^2\times\frac{n^An^B\left[1-\exp\left(-\frac{\omega_A}{T}\right)\right]\left[1-\exp\left(-\frac{\omega_B}{T}\right)\right]}{\left[1+\exp\left(-\frac{\omega_A}{T}\right)\right]\left[1+\exp\left(-\frac{\omega_B}{T}\right)\right]}\times\frac{(\omega_A+\omega_B)}{(\omega_A+\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}. \tag{56}$$

The difference between expression resulting from quantum electrodynamics, Eq. (52), and the one obtained with the help of the Lifshitz formula is dramatic. The results coincide only for the case of cold media where the density numbers of excited atoms are negligible (Fig. 4). If the temperatures are high enough for the media to contain excited atoms, the dependences (52) and (55) differ qualitatively (Fig. 5). The temperature dependences of the Casimir forces are shown in Fig. 6. Now let us return to the case of the interaction of a single excited atom and the dielectric nonexcited media discussed in Sec. III. Using expression (55) of the Casimir force obtained by Lifshitz, one can easily find a corresponding

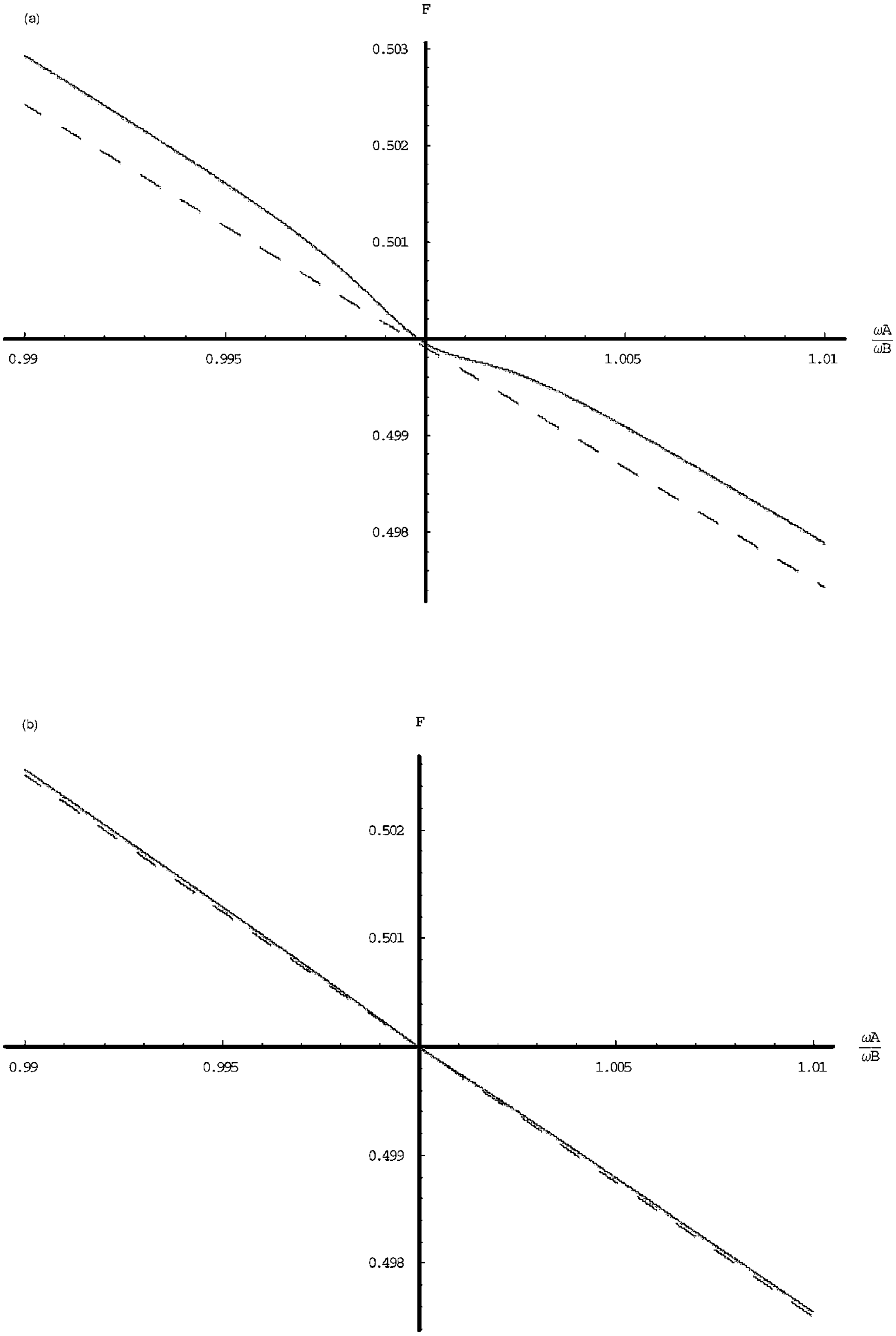

FIG. 4. Normalized Casimir force calculated by means of quantum electrodynamics (53) (solid line) and the Lifshitz formula (56) (dashed line). (a) $T/\omega_B=0.1$, $\gamma_B/\omega_B=0.002$ and (b) $T/\omega_B=0.08$, $\gamma_B/\omega_B=0.002$.

expression for the potential of a single excited atom interacting with a cold medium:

$$U_L(z_0)=\frac{\pi}{9z_0^3}|d_{eg}^A|^2|d_{eg}^B|^2\frac{(\omega_A+\omega_B)n}{(\omega_A+\omega_B)^2+\left(\frac{\gamma_B}{2}\right)^2}. \quad (57)$$

Disagreement of the results of quantum electrodynamics, Eq. (44), and the consequence of the Lifshitz formula (57) for a case of excited atom near a cold medium is dramatic (Fig. 7). We see that the van der Waals potential obtained by means of quantum electrodynamics corresponds to resonant attraction (repulsion) for red (blue) detuned atomic transition frequencies $\omega_A<\omega_B$ ($\omega_A>\omega_B$). This is a well-known result. The van der Waals potential resulting from the Lifshitz formula corresponds to repulsion for all atom frequencies. The difference between the results is impressive. At some points it is about three orders of magnitude. But our result (44) coincides with the well-known theoretical results [9–12] and experimental ones [10,13,14]. Thus failure of the Lifshitz formula for a case of excited atoms is clear. If we consider the case of a ground-state atom near an interface of cold dielec-

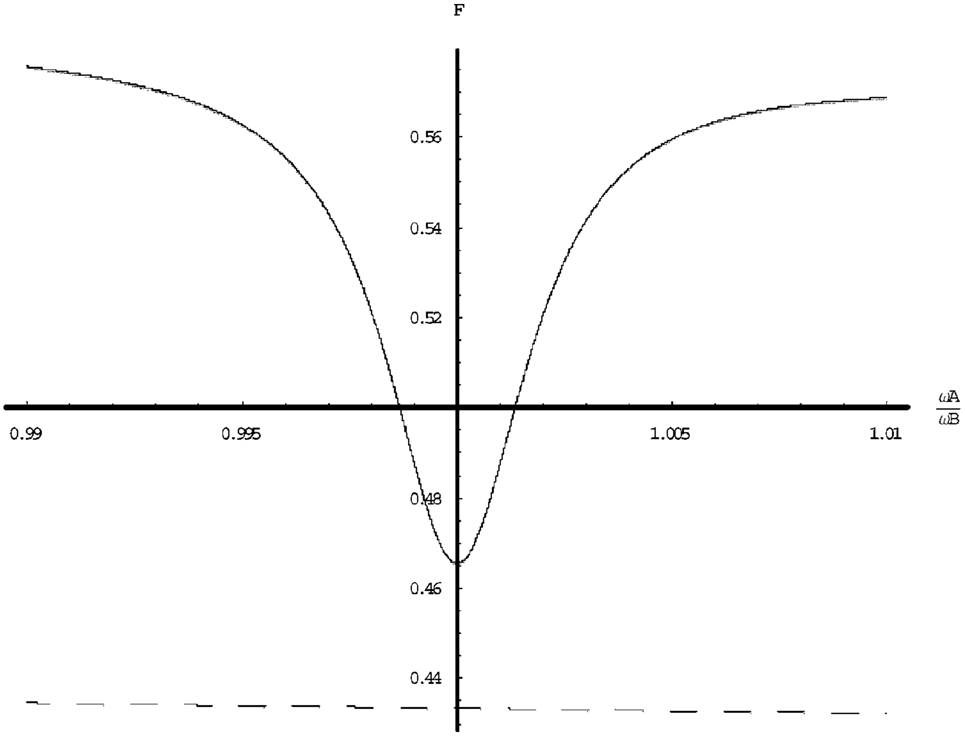

FIG. 5. Normalized Casimir force calculated by means of quantum electrodynamics (53) (solid line) and the Lifshitz formula (56) (dashed line). $T/\omega_B = 0.3$, $\gamma_B/\omega_B = 0.002$.

tric, the results obtained by using the Lifshitz formula (55) and quantum electrodynamics (45) are obviously the same.

V. SUMMARY

Using a specially developed method of Green functions, which enabled us to take into account the energy-level widths of atoms, we calculated the van der Waals potential for a two-atom dipole-dipole interaction if the atoms are in the following initial states: one atom is excited and the other is in the ground state; both atoms are in the ground state. We generalized well-known results obtained in the framework of perturbation theory and linear response theory [7,8,22] to the case of a finite energy-level width of atoms. The results are not expressed in terms of the conventional polarizabilities of atoms, Eqs. (37) and (38), but they contain the so-called coherent polarizabilities (35) and (36) with different analytical properties in the upper complex semiplane.

The analysis of the interaction between two atoms enabled us to calculate the van der Waals potential for the interaction of a single atom with a semi-infinite medium. We considered the case of a dilute gas medium and took into account only pair interactions of atoms. The result obtained for the case of excited atom and medium of ground-state atoms is in complete agreement with theoretical works, which used a linear response approach or macroscopic quan-

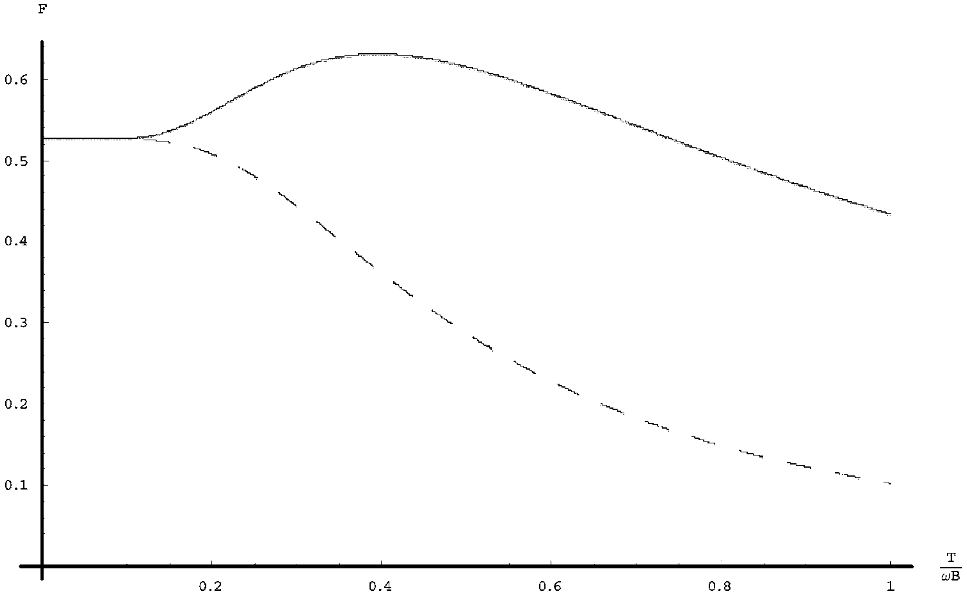

FIG. 6. Normalized Casimir force calculated by means of quantum electrodynamics (53) (solid line) and the Lifshitz formula (56) (dashed line). $\omega_A/\omega_B = 0.9$, $\gamma_B/\omega_B = 0.002$.

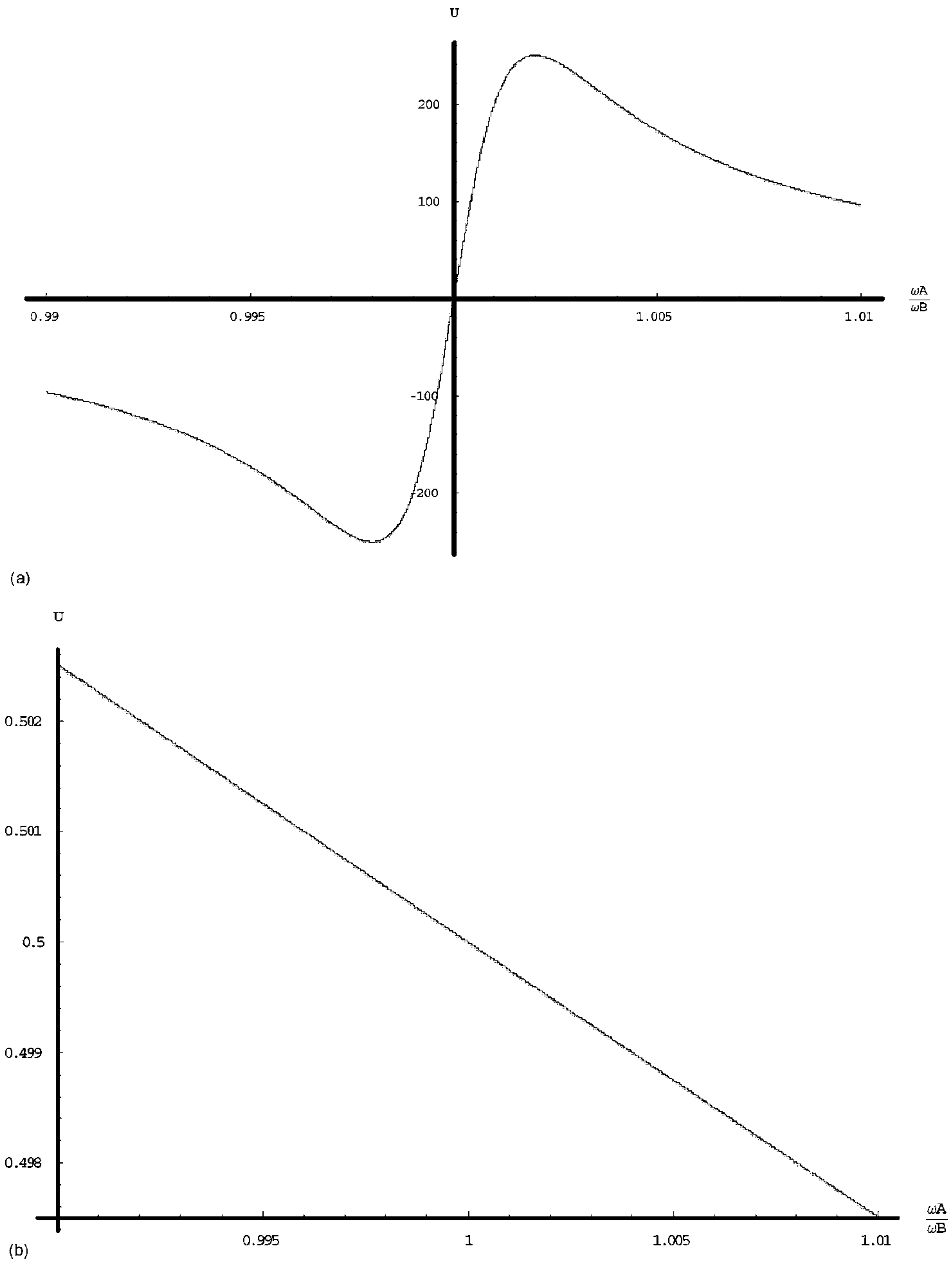

FIG. 7. Normalized Casimir potential for excited atom and cold media interaction calculated by means of (a) quantum electrodynamics (44) and (b) the Lifshitz formula (57). $\gamma_B/\omega_B=0.002$.

tum electrodynamics (i.e., conventional polarizability) [9–12], and experimental works [10,13,14], while in our paper it is not expressed in terms of conventional polarizabilities. What is the reason for such an agreement? The authors of the above-mentioned papers used the linear response theory or macroscopic QED to describe a medium; as a result, it was described in terms of conventional permittivity (49), while the excited atom was described with the help of the Heisenberg equation of motion. Thus the function corresponding to the excited atom possesses analytical properties of the coherent polarizability (36). But for ground-state atoms, the first term of the conventional polarizability(37) is resonant and it coincides with the first term of the coherent polarizability. As a result, the formulas obtained in this paper and [9–12] are in complete agreement.

The situation is different if a ground-state atom is placed in the vicinity of a medium of excited atoms. The result obtained in this paper cannot, as far as we know, be obtained with the help of the linear response theory and cannot be expressed in terms of conventional permittivity.

In the last section we compared the results obtained with the help of the Lifshitz formula and quantum electrodynamics for the van der Waals interaction of two media of excited atoms. Here we considered dilute gas media and took into account only pair interactions. It was shown that the results coincide only if the media do not contain excited atoms. If the concentrations of excited atoms are significant, the difference of the results is dramatic. The result obtained in this paper is expressed in terms of coherent permittivity, while the Lifshitz formula depends on conventional permittivity. We compared the results of the Lifshitz formula calculated for a case of a single excited atom near ground-state medium and showed that it is not in agreement with theoretical and experimental results [9–14]. The difference is dramatic.

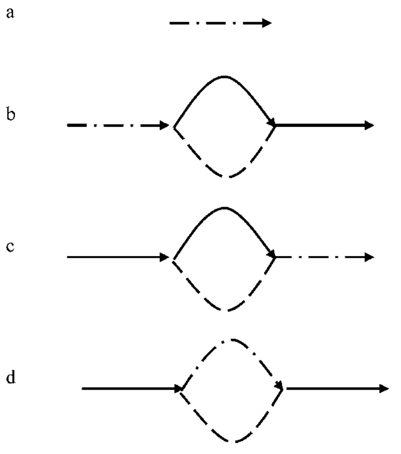

FIG. 8. Feynman's diagram corresponding to Eq. (A1). (a) First term, (b) second term, (c) third term, and (d) fourth term. The solid line corresponds to g^{0A}, the dashed line corresponds to $D_{ll'}^{0\nu\nu'}$, and the dash-dotted line represents ρ_0^A.

Quantum electrodynamics results in a *resonant* interaction (attractive or repulsive), while the Lifshitz formula gives us *nonresonant* repulsion only. The graphs are given in Fig. 7. The difference may be up to three orders of magnitude.

Thus we state that the Lifshitz formula is applicable only for ground-state (cold) media. If the media are hot enough to possess excited atoms in a significant amount, the Casimir interaction cannot be described by the Lifshitz formula, at any rate, for distances smaller than the wavelength of the atom transition. It cannot be described even in terms of the conventional permittivities of media. To describe the Casimir interaction of excited media one should use coherent permittivities.

ACKNOWLEDGMENTS

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APPENDIX A: METHOD OF GREEN FUNCTIONS

Here we use a specially elaborated method of quantum Green functions, which enables us to take into account the energy-level width of atoms. This method resembles a very-well-known method of kinetic Green's functions suggested by Keldysh [19].

Now we will outline some basic principles of the diagram technique.

A Green function of atom A is given by Eq. (5), with Eq. (6) being the scattering operator taken on the contour given in Fig. 1. The density matrix of atom A is

$$\rho^A(x,x') = iG_{12}^A(x,x').$$

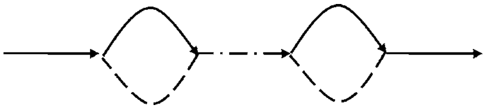

FIG. 9. Feynman's diagram of the fourth order of the coherent channel.

To derive Eqs. (10) and (11) we should expand the scattering operator (6) and substitute the result into Eq. (5). One should mention that all odd orders of the expansion are equal to zero since we have only one operator of the electromagnetic field in each term of the interaction Hamiltonian (7). The first two orders of perturbation theory read

$$\rho^A(x,x') = \rho_0^A(x,x') - \frac{1}{2}\left\langle \hat{T}_c \int dx_1 dx_2 \hat{\psi}_1(x)\hat{\psi}_2^+(x')(-1)^{l_1+l_2} \times \hat{\psi}_{l_1}^+(x_1)\hat{E}_{l_1}^{\nu_1}(x_1)\hat{d}^{\nu_1}\hat{\psi}(x_1)\hat{\psi}_{l_2}^+(x_2)\hat{E}_{l_2}^{\nu_2}(x_2)\hat{d}^{\nu_2}\hat{\psi}(x_2)\right\rangle.$$

Using Wick's theorem, we find

$$\begin{aligned}\rho^A(x,x') = \rho_0^A(x,x') &- i\int dx_1 dx_2 \rho_0^A(x,x_1)\hat{d}^\nu\hat{d}^{\nu'} g_{22}^{0A}(x_1,x_2) \\ &\times D_{22}^{0\nu\nu'}(x_2,x_1) g_{22}^{0A}(x_2,x') \\ &- i\int dx_1 dx_2 g_{11}^{0A}(x,x_1)\hat{d}^\nu\hat{d}^{\nu'} g_{11}^{0A}(x_1,x_2) \\ &\times D_{11}^{0\nu\nu'}(x_2,x_1)\rho_0^A(x_2,x') \\ &- i\int dx_1 dx_2 g_{11}^{0A}(x,x_1)\hat{d}^\nu\hat{d}^{\nu'}\rho_0^A(x_1,x_2) \\ &\times D_{21}^{0\nu\nu'}(x_2,x_1) g_{22}^{0A}(x_2,x'),\end{aligned} \tag{A1}$$

with the Green functions given by Eqs. (14) and (17).

We take into account that for a single atom all the normal products of orders higher than 2 are equal to zero,

$$\langle \hat{N}\hat{\psi}_{l_1}(x)\hat{\psi}_{l_2}^+(x')\hat{\psi}_l^+(x_1)\hat{\psi}_{l'}(x_2)\cdots\rangle = 0,$$

while the second order of the normal product represents the density matrix of the initial state of the atom:

$$\rho^0(x,x') = \langle \hat{N}\hat{\psi}_{l_1}(x)\hat{\psi}_{l_2}^+(x')\rangle.$$

We can take into account that $g_{12}^0 = -i\langle\hat{\psi}^\dagger\hat{\psi}\rangle_{\text{vacuum}} = 0$.

We can draw Feynman's diagram corresponding to Eq. (A1) [Fig. 8]. All the disconnected diagrams are canceling.

The first term of Eq. (A1) is represented by diagram (a). It corresponds to the matrix of density of the initial state of the atom. The second and third terms are represented as diagrams (b) and (c). They correspond to the processes of coherent channel, with the resultant state of atom being the same as the initial one; it means that the resultant state is described by the same wave function. As an example of such processes, we can consider elastic scattering of a photon or interaction of an atom with the electromagnetic vacuum. The last term given by diagram (d) represents the process of the incoherent channel, with the initial state changing as a result of such a process. As an example, we may consider the processes of the inelastic scattering of a photon or spontaneous and induced radiation of an atom. It is significant that these channels (coherent and incoherent) are separated and the complete matrix of density is equal to the sum of contributions of these channels. The same separation of channels appears in the Γ-operator technique [20].

Now we take into account the higher orders of the perturbation technique and use the well-known Dyson equation for photon and electron propagators and (13) and (16). It is easy to show that we should substitute complete electron and photon propagators satisfying Dyson equations (13) and (16) into Eq. (A1) instead of free-field propagators and add a term appearing in the fourth order of perturbation technique and shown in Fig. 9. Neglecting the incoherent channel, which has nothing to do with the van der Waals interaction, we come to Eq. (11), with the mass operators given by formulas (15).

APPENDIX B: DERIVATION OF Eq. (19)

Here we will derive the differential equation (19) using the integral one (11).

It is easy to show [22] that the free-electron propagators and the density matrix satisfy the equations

$$g_{11}^{0(-1)}g_{11}^{0}(x,x')=\delta(x-x'),\quad g_{22}^{0}(x,x')g_{22}^{0(-1)}=\delta(x-x'),\tag{B1}$$

$$g_{11}^{0(-1)}\rho^{0}(x,x')=0,\quad \rho^{0}(x,x')g_{22}^{0(-1)}=0,\tag{B2}$$

$$g_{11}^{0A,B(-1)}=\left(i\frac{\partial}{\partial t}-\hat{H}_{A,B}\right),\quad g_{22}^{0A,B(-1)}=\left(i\frac{\partial}{\partial t'}-\hat{H}_{A,B}\right).\tag{B3}$$

Using Eqs. (B1)–(B3) and (13), we find

$$g_{11}^{0A(-1)}g_{11}^{A}(x,x')=\delta(x-x')+\int dx_2M_{11}(x,x_2)g_{11}^{A}(x_2,x'),$$

$$g_{22}^{A}(x,x')g_{22}^{0A(-1)}=\delta(x-x')+\int dx_1g_{22}^{A}(x,x_1)M_{22}(x_1,x').\tag{B4}$$

Now Eq. (11) can be rewritten as

$$\begin{aligned}g_{11}^{0A(-1)}\rho_c^{A}(x,x')g_{22}^{0A(-1)}&=\int dx_2dx_3M_{11}(x,x_2)\rho_0^{A}(x_2,x_3)M_{22}(x_3,x')\\&+\int dx_1dx_2dx_3dx_4M_{11}(x,x_2)\rho_0^{A}(x_2,x_3)M_{22}(x_3,x_4)g_{22}^{A}(x_4,x_1)M_{22}(x_1,x')\\&+\int dx_1dx_2dx_3dx_4M_{11}(x,x_4)g_{11}^{A}(x_4,x_1)M_{11}(x_1,x_2)\rho_0^{A}(x_2,x_3)M_{22}(x_3,x')\\&+\int dx_1dx_2dx_3dx_4dx_5dx_6M_{11}(x,x_5)g_{11}^{A}(x_5,x_1)M_{11}(x_1,x_2)\rho_0^{A}(x_2,x_3)M_{22}(x_3,x_4)g_{22}^{A}(x_4,x_6)M_{22}(x_6,x').\end{aligned}$$

Using Dyson equations (13) and formula (11), we come to the equation

$$g_{11}^{0A(-1)}\rho_c^{A}(x,x')g_{22}^{0A(-1)}=\int dx_1dx_2M_{11}(x,x_1)\rho_c^{A}(x_1,x_2)M_{22}(x_2,x').\tag{B5}$$

Equation (B5) can be easily solved if one represents the density matrix as $\rho_c^{A}(x,x')=\Psi(x)\Psi^{*}(x')$, with $\Psi(x)$ being the wave function of atom A in the Shrödinger picture. Such a representation is evident since the coherent channel describes the processes which return the atoms to the initial states; consequently, the final state of atoms can be described in terms of wave functions (pure state) if the initial state is pure. Taking into account formulas (B3) we come to Eq. (19).

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