# van der Waals interactions in a magnetodielectric medium

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The van der Waals interaction between two ground-state atoms is calculated for two electrically or magnetically polarizable particles embedded in a dispersive magnetodielectric medium. Unlike previous calculations which infer the atom-atom interaction from the dilute-medium limit of the macroscopic, many-body van der Waals interaction, the interaction is calculated directly for the system of two atoms in a magnetodielectric medium. Two approaches are presented, the first based on the quantized electromagnetic field in a dispersive medium without absorption and the second on Green functions that allow for absorption. We show that the correct van der Waals interactions are obtained regardless of whether absorption in the host medium is explicitly taken into account.

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## I. INTRODUCTION

The van der Waals interaction between ground-state atoms in vacuum is often regarded as a consequence of the vacuum fluctuations of the electromagnetic field. It is well known that the interaction between two macroscopic, uncharged dielectric bodies cannot be obtained by the pairwise addition of this interatomic van der Waals interaction except in the dilute-medium limit. In this limit Lifshitz, for instance, obtained the retarded and unretarded pairwise van der Waals interaction between electrically polarizable atoms from a more general expression for the interaction energy of two dielectric half-spaces [1]. The van der Waals interaction obtained in this way is identical to that obtained more directly by Casimir and Polder [2] for the system of two atoms in vacuum.

The continuing interest in Casimir effects and the related effects of vacuum field fluctuations in the case of dielectric media has stimulated some interest in the van der Waals interactions of atoms embedded in magnetodielectric media. Recent work in this area [3–7], takes essentially the original approach of Abrikosov *et al.* [8] using Green functions for radiation in an *absorbing* medium and taking the dilute-medium limit to infer the atom-atom interaction. In this paper we obtain the atom-atom van der Waals interaction *directly*, rather than from a dilute-medium limit of an interaction between macroscopic bodies, and we show that this interaction can be correctly obtained *without explicit account of absorption in the host medium*.

We consider both electrically polarizable and magnetically polarizable atoms embedded in a magnetodielectric medium. A possible physical scenario to have a well-defined embedding of the atoms in the medium corresponds to two spherical cavities centered at the positions of the atoms, from which the medium has been removed [9]. The surrounding medium can be considered continuous as long as the radii of the cavities and the separation between them is much larger than the interatomic distances in the medium. In order to proceed to calculate the van der Waals interaction between the two atoms, one needs to quantize the electromagnetic field in this configuration, which is a complicated boundary problem [10]. In the following we will simplify the problem by assuming that the cavities have negligible radii, effectively embedding the atoms as pointlike particles in a medium approximated as a continuum. We will present two derivations of the van der Waals interaction between the two atoms embedded in the magnetodielectric medium. The first, presented in Sec. II, is based on the (electric or magnetic) dipole-dipole interaction induced in the two atoms by the "vacuum" field in the magnetodielectric medium. For this purpose we employ simple expressions for the quantized electromagnetic field in a dispersive magnetodielectric medium in which absorption is ignored [11]. The second, the subject of Sec. III, employs Green functions and takes explicit account of absorption [12]. The fact that the same results are obtained regardless of whether absorption is accounted for appears to us to be of some interest, and physical reasons for it are given in Sec. IV, which also includes some further discussion and a brief summary of our results.

## II. VAN DER WAALS INTERACTIONS FROM THE QUANTIZED FIELD WITHOUT ABSORPTION

We consider two identical atoms in a homogeneous and isotropic magnetodielectric medium which we regard as a continuum with real electric permittivity $\epsilon(\omega)$ and magnetic permeability $\mu(\omega)$, and therefore real refractive index $n(\omega)$. The calculation of the van der Waals interactions in this section will be based on the following expressions for the electric and magnetic fields in the nonabsorbing magnetodielectric medium [13]:

$$\mathbf{E}(\mathbf{r},t) = i\sum_{\mathbf{k}\lambda}\left(\frac{2\pi\hbar\omega_k\mu_k}{n_k\gamma_k V}\right)^{1/2}[a_{\mathbf{k}\lambda}(t)e^{i\mathbf{k}\cdot\mathbf{r}} - a^{\dagger}_{\mathbf{k}\lambda}(t)e^{-i\mathbf{k}\cdot\mathbf{r}}]\mathbf{e}_{\mathbf{k}\lambda}, \tag{1}$$

 

$$\mathbf{H}(\mathbf{r},t) = i\sum_{\mathbf{k}\lambda}\left(\frac{2\pi\hbar c^2}{\omega_k n_k \gamma_k \mu_k V}\right)^{1/2}[a_{\mathbf{k}\lambda}(t)e^{i\mathbf{k}\cdot\mathbf{r}} - a^{\dagger}_{\mathbf{k}\lambda}(t)e^{-i\mathbf{k}\cdot\mathbf{r}}]\mathbf{k} \times \mathbf{e}_{\mathbf{k}\lambda}. \quad (2)$$

We employ a standard notation in which $a_{\mathbf{k}\lambda}(t), a^{\dagger}_{\mathbf{k}\lambda}(t)$ are Heisenberg-picture photon annihilation and creation operators for the plane-wave mode with wave vector $\mathbf{k}$ [$|\mathbf{k}|=k = n_k\omega_k/c$, $n_k=n(\omega_k)$] and (linear) polarization unit vector $\mathbf{e}_{\mathbf{k}\lambda}$, [$\mathbf{k}\cdot\mathbf{e}_{\mathbf{k}\lambda}=0$, $\lambda=1,2$]. The refractive index is $n_k=(\epsilon_k\mu_k)^{1/2}$, where $\epsilon_k$ and $\mu_k$ are the electric permittivity and magnetic permeability, respectively, at frequency $\omega_k$. The group index is $\gamma_k \equiv n_k + \omega_k dn_k/d\omega_k$, and $V$ is the quantization volume for the box normalization of the plane-wave modes. The Hamiltonian is

$$H = H_A + H_B + H_F - \mathbf{d}_A(t)\cdot\mathbf{E}(\mathbf{r}_A,t) - \mathbf{d}_B(t)\cdot\mathbf{E}(\mathbf{r}_B,t) - \mathbf{m}_A(t)\cdot\mathbf{H}(\mathbf{r}_A,t) - \mathbf{m}_B(t)\cdot\mathbf{H}(\mathbf{r}_B,t), \quad (3)$$

where $H_A$, $H_B$, and $H_F$ are the Hamiltonian operators for atom $A$, atom $B$, and the electromagnetic field, respectively. The electric dipole moment operators are denoted by $\mathbf{d}(t)$ and the magnetic dipole moment operators by $\mathbf{m}(t)$.

### A. van der Waals interaction between electrically polarizable particles

We first consider the van der Waals interaction between two electrically polarizable atoms separated by a distance $R$ in the magnetodielectric medium. The approach we will take follows closely that used by various authors for the derivation of the van der Waals interaction in the case in which the particles are in vacuum ($n=1$) [14]. The basic idea of this method is that quantum vacuum fluctuations of the electromagnetic field in the medium induce in the atoms fluctuating electric and magnetic dipoles that interact with each other. The electric dipole moment induced in an atom at $\mathbf{r}$ by an electric field is $\mathbf{d}(t)=\alpha_e(t)\mathbf{E}(\mathbf{r},t)$, where $\alpha_e$ is the (real) electric polarizability of the atom. (Since it is only the real part of the polarizability that determines shifts in energy levels, we can assume without loss of generality throughout this paper that the polarizabilities of the guest atoms are real.) The dipole interaction energy between the two induced, fluctuating electric dipoles is then

$$W_{ee}(R) = \sum_{\mathbf{k}\lambda}\alpha_e^A(\omega)\alpha_e^B(\omega)\langle \mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{E}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle V_{ij}^{ee}(\omega,\mathbf{R}), \quad (4)$$

where $\mathbf{R}=\mathbf{r}_B-\mathbf{r}_A$, $R=|\mathbf{R}|$. The two-point vacuum electric-field correlation function, summed over polarization states, follows easily from Eq. (1)

$$\sum_{\lambda}\langle \mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{E}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle = \frac{2\pi\hbar\omega_k\mu_k}{n_k\gamma_k V}(\delta_{ij} - \hat{\mathbf{k}}_i\hat{\mathbf{k}}_j)e^{-i\mathbf{k}\cdot\mathbf{R}}, \quad (5)$$

with $\hat{\mathbf{k}}_i=\mathbf{k}_i/k$. The interaction potential $V_{ij}^{ee}(\omega,\mathbf{R})$ between two oscillating electric dipoles embedded in the magnetodielectric medium is calculated as follows. From the Hamiltonian (3) and the Heisenberg equations of motion for the annihilation and creation operators, one obtains a formal expression for the electric field generated by the electric dipole $B$ at the position of the electric dipole $A$ as follows:

$$\mathbf{E}_e(\mathbf{r}_A,t) = \frac{i}{\pi c^3}\int_0^{\infty} d\omega n(\omega)\mu(\omega)\omega^3 \times\left[\mathbf{a}\frac{\sin kR}{kR} - \mathbf{b}\left(\frac{\sin kR}{k^3R^3} - \frac{\cos kR}{k^2R^2}\right)\right] \times\int_0^t dt' p_B(t')e^{i\omega(t'-t)} + \text{H.c.}, \quad (6)$$

where $\mathbf{a}=\mathbf{d}_B-(\mathbf{d}_B\cdot\hat{\mathbf{R}})\hat{\mathbf{R}}$, $\mathbf{b}=\mathbf{d}_B-3(\mathbf{d}_B\cdot\hat{\mathbf{R}})\hat{\mathbf{R}}$, and $\hat{\mathbf{R}}=\mathbf{R}/R$. We have used the notation $\mathbf{d}(t)=\mathbf{d}p(t)$, with $\mathbf{d}$ a unit vector specifying the direction of the electric dipole moment. We are interested in the case of two electric dipoles, both oscillating at frequency $\omega'$: $p_B(t)=C_Be^{-i\omega' t}+C_B^*e^{i\omega' t}$ and $p_A(t) = C_Ae^{-i\omega' t}+C_A^*e^{i\omega' t}$, where $C_A, C_B$ are arbitrary constants. We define the interaction $V^{ee}(\mathbf{R})=-\mathbf{d}_Ap_A(t)\cdot\mathbf{E}_e(\mathbf{r}_A,t)$. After performing the time integration for times $t\gg 1/\omega'$, and taking the time average of the resulting expression, we get

$$V^{ee}(\mathbf{R}) = -\frac{2}{\pi c^3}\,\text{Re}\int_0^{\infty} d\omega n(\omega)\mu(\omega)\omega^3\left[a\frac{\sin kR}{kR} - b\left(\frac{\sin kR}{k^3R^3} - \frac{\cos kR}{k^2R^2}\right)\right]\left[\frac{C_A^*C_B}{\omega-\omega'-i\eta} + \frac{C_AC_B^*}{\omega+\omega'-i\eta}\right], \quad (7)$$

where as usual $\eta\to 0^+$. Here $a=\mathbf{d}_A\cdot\mathbf{d}_B-(\mathbf{d}_A\cdot\hat{\mathbf{R}})(\mathbf{d}_B\cdot\hat{\mathbf{R}})$ and $b=\mathbf{d}_A\cdot\mathbf{d}_B-3(\mathbf{d}_A\cdot\hat{\mathbf{R}})(\mathbf{d}_B\cdot\hat{\mathbf{R}})$. We can also define the interaction as $V^{ee}(\mathbf{R})=-\mathbf{d}_Bp_B(t)\cdot\mathbf{E}_e(\mathbf{r}_B,t)$, which amounts to interchanging $A$ and $B$ above. This implies that we can take $C_A^*C_B=C_B^*C_A$. Note also that the time average $\overline{p_A(t)p_B(t)} = C_AC_B^*+C_A^*C_B$, so that we can write

$$V^{ee}(\mathbf{R}) = -\frac{1}{\pi c^3}\overline{p_Ap_B}\,\text{Re}\int_0^{\infty} d\omega n(\omega)\mu(\omega)\omega^3 \times\left[a\frac{\sin kR}{kR} - b\left(\frac{\sin kR}{k^3R^3} - \frac{\cos kR}{k^2R^2}\right)\right] \times\left(\frac{1}{\omega-\omega'-i\eta} + \frac{1}{\omega+\omega'-i\eta}\right). \quad (8)$$

Next we use the fact that $n$ and $\mu$ (or actually their real parts which are implicit here) are even functions of $\omega$ to rewrite this as

$$V^{ee}(\mathbf{R}) = -\frac{1}{\pi c^3}\overline{p_Ap_B}\int_{-\infty}^{\infty} d\omega n(\omega)\mu(\omega)\omega^3 \times\left[a\frac{\sin kR}{kR} - b\left(\frac{\sin kR}{k^3R^3} - \frac{\cos kR}{k^2R^2}\right)\right] \times\left(\frac{1}{\omega-\omega'-i\eta} + \frac{1}{\omega-\omega'+i\eta}\right).$$

Performing the trivial contour integrations, writing $\omega$ instead of $\omega'$ for the dipole frequencies, and using again $k$

$=n(\omega)\omega/c$, we obtain the electric dipole-dipole interaction tensor needed in Eq. (4)

$$V_{ij}^{ee}(\omega,\mathbf{R}) = \frac{1}{\epsilon_k}\frac{1}{R^3}[(\delta_{ij}-3\hat{\mathbf{R}}_i\hat{\mathbf{R}}_j)(\cos kR + kR\sin kR) - (\delta_{ij}-\hat{\mathbf{R}}_i\hat{\mathbf{R}}_j)k^2R^2\cos kR]. \quad (9)$$

Using Eqs. (5) and (9), and passing to the continuum limit $\Sigma_{\mathbf{k}}\to(V/8\pi^3)\int_0^\infty dk k^2\int d\Omega_{\mathbf{k}}=(V/8\pi^3c^3)\int_0^\infty d\omega\gamma_\omega n_\omega^2\omega^2\int d\Omega_{\mathbf{k}}$, we obtain the van der Waals interaction energy between electrically polarizable particles

$$W_{ee}(R) = -\frac{\hbar}{\pi c^3}\frac{1}{R^3}\int_0^\infty d\omega\alpha_e^A(\omega)\alpha_e^B(\omega)\frac{\omega^3\mu^2(\omega)}{n(\omega)}\left[kR\sin 2kR + 2\cos 2kR - 5\frac{\sin 2kR}{kR} - 6\frac{\cos 2kR}{k^2R^2} + 3\frac{\sin 2kR}{k^3R^3}\right]. \quad (10)$$

The integration path can be rotated using the fact that there are no poles in the upper half of the complex plane. We obtain finally

$$W_{ee}(R) = -\frac{\hbar}{16\pi R^6}\int_0^\infty du\alpha_e^A(iu)\alpha_e^B(iu)\frac{1}{\epsilon^2(iu)} \times F\left[\frac{2n(iu)uR}{c}\right]e^{-2n(iu)uR/c}, \quad (11)$$

where $F(x)=x^4+4x^3+20x^2+48x+48$. Recall that along the imaginary frequency axis the electric permittivity and refractive index are real and positive. The electric-electric van der Waals force that results from Eq. (11) is therefore always attractive, regardless of the frequency dependence of $\epsilon(\omega)$ and $n(\omega)$.

Our calculation based on the quantized fields (1) and (2) gives a van der Waals interaction (11) with the correct vacuum ($\epsilon=\mu=1$) limit [15], and the same is true for the other van der Waals interactions we calculate in this paper [16]. Moreover, for material systems ($\epsilon,\mu\neq 1$), they are in full agreement with those obtained recently by Tomaš [4], for instance. The main point of this section is to show that correct results for van der Waals interactions involving ground-state atoms in dispersive media can be obtained straightforwardly, without having to go to a dilute-medium limit of an interaction between macroscopic bodies, and without having to introduce complexities arising from absorption. We discuss this further in Sec. IV.

### B. van der Waals interaction between magnetically polarizable particles

We next use the same approach to calculate the van der Waals interaction between magnetic dipoles induced in the atoms by fluctuations of the zero-point magnetic field. For this the relation between an induced magnetic dipole moment at position $\mathbf{r}$ and the magnetic field is $\mathbf{m}(t)=\alpha_m(t)\mathbf{H}(\mathbf{r},t)$, where $\alpha_m$ is the (real) magnetic polarizability of the atom. The dipole interaction between the two induced, fluctuating magnetic dipoles is

$$W_{mm}(R) = \sum_{\mathbf{k}\lambda}\alpha_m^A(\omega)\alpha_m^B(\omega)\langle\mathbf{H}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{H}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle V_{ij}^{mm}(\omega,\mathbf{R}), \quad (12)$$

where the two-point vacuum magnetic field correlation function summed over polarizations is found from Eq. (2) to be

$$\sum_\lambda\langle\mathbf{H}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{H}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle = \frac{2\pi\hbar n_k\omega_k}{\mu_k\gamma_k V}(\delta_{ij}-\hat{\mathbf{k}}_i\hat{\mathbf{k}}_j)e^{-i\mathbf{k}\cdot\mathbf{R}}. \quad (13)$$

One can derive the magnetic dipole-dipole interaction tensor following steps similar to those above for the electric dipole-dipole interaction

$$V_{ij}^{mm}(\omega,\mathbf{R}) = \frac{1}{\mu_k}\frac{1}{R^3}[(\delta_{ij}-3\hat{\mathbf{R}}_i\hat{\mathbf{R}}_j)(\cos kR + kR\sin kR) - (\delta_{ij}-\hat{\mathbf{R}}_i\hat{\mathbf{R}}_j)k^2R^2\cos kR], \quad (14)$$

which differs from Eq. (9) simply by the replacement of $\epsilon_k$ by $\mu_k$. The details of the evaluation of Eq. (12) are essentially the same as for the electric van der Waals interaction and lead straightforwardly to the expression

$$W_{mm}(R) = -\frac{\hbar}{16\pi R^6}\int_0^\infty du\alpha_m^A(iu)\alpha_m^B(iu)\frac{1}{\mu^2(iu)} \times F\left[\frac{2n(iu)uR}{c}\right]e^{-2n(iu)uR/c}. \quad (15)$$

Recall that along the imaginary frequency axis the magnetic permeability is real and positive. The magnetic-magnetic van der Waals force that results from Eq. (15) is always attractive, regardless of the frequency dependence of $\mu(\omega)$ and $n(\omega)$.

### C. van der Waals interaction between an electrically polarizable particle and a magnetically polarizable particle

In calculating $W_{ee}(R)$ and $W_{mm}(R)$ it has not been necessary to account for the fact that the field operators in Eqs. (4) and (12) do not commute. Because of this noncommutativity, it is more appropriate to write $W_{ee}(R)$, for instance, in the symmetrized form

$$\begin{aligned} W_{ee}(R) &= \frac{1}{2}\sum_{\mathbf{k}\lambda}\alpha_e^A(\omega)\alpha_e^B(\omega)[\langle\mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{E}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle + \langle\mathbf{E}_j(\mathbf{r}_B,\mathbf{k}\lambda)\mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\rangle]V_{ij}^{ee}(\omega,\mathbf{R}) \\ &= \mathrm{Re}\sum_{\mathbf{k}\lambda}\alpha_e^A(\omega)\alpha_e^B(\omega)\langle\mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{E}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle V_{ij}^{ee}(\omega,\mathbf{R}). \end{aligned} \quad (16)$$

The forms (5) and (9), however, show that symmetrization is actually not required because the summation over $\mathbf{k}$ does not require us to distinguish between $\langle\mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{E}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle$ and $\langle\mathbf{E}_j(\mathbf{r}_B,\mathbf{k}\lambda)\mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\rangle$.

The situation in the case of the van der Waals interaction between an electrically polarizable particle and a magnetically polarizable particle, however, is different because the electric-magnetic correlation function summed over polarization states,

$$\sum_{\lambda} \langle \mathbf{E}_i(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{H}_j(\mathbf{r}_B,\mathbf{k}\lambda)\rangle = \frac{2\pi\hbar\omega_k}{\gamma_k V}\epsilon_{ijl}\hat{\mathbf{k}}_l e^{-i\mathbf{k}\cdot\mathbf{R}}, \quad (17)$$

is not purely real when summed over $\mathbf{k}$. ($\epsilon_{ijl}$ is the Levi-Civita tensor.) Moreover, the interaction tensor in this case, which we calculate to be

$$V_{ij}^{em}(\omega,\mathbf{R}) = \frac{\omega^3}{c^3}n^2(\omega)\epsilon_{ijp}\hat{\mathbf{R}}_p\left[\frac{\sin kR}{k^2R^2} - \frac{\cos kR}{kR}\right] \quad (18)$$

in a manner directly analogous to the electric-electric and magnetic-magnetic tensors, is antisymmetric.

Let $\mathbf{E}_m(\mathbf{r}_A,t)$ be the electric field operator at $\mathbf{r}_A$ due to a magnetic dipole at $\mathbf{r}_B$. We write the interaction between the fluctuating electric and magnetic dipole moments in the symmetrized form

$$W_{em}(R) = -\text{Re}\sum_{\mathbf{k}\lambda}\alpha_e^A(\omega)\langle \mathbf{E}_i^{(+)}(\mathbf{r}_A,\mathbf{k}\lambda)\mathbf{E}_{mi}^{(-)}(\mathbf{r}_A,\mathbf{k}\lambda)\rangle, \quad (19)$$

where $\mathbf{E}^{(+)}(\mathbf{r}_A,t)$ is the positive-frequency (photon annihilation) part of the source-free ("vacuum") electric field operator at $\mathbf{r}_A$, and $\mathbf{E}_m^{(-)}(\mathbf{r}_A,t)$ is the negative-frequency (photon creation) part of the electric field produced by the magnetic dipole moment at $\mathbf{r}_B$. This electric field is induced by the source-free magnetic field $\mathbf{H}(\mathbf{r}_B,t)$, so that the evaluation of Eq. (19) involves the electric-magnetic correlation function (17). The calculation is essentially just the same as that presented by Farina *et al.* [17] for the case where the two particles are in free space, except of course that in our case the refractive index $n(\omega)$ appears

$$W_{em}(R) = \frac{\hbar}{4\pi c^2R^4}\int_0^\infty du u^2\alpha_e^A(iu)\alpha_m^B(iu) \times G\left[\frac{2n(iu)uR}{c}\right]e^{-2n(iu)uR/c}, \quad (20)$$

where $G(x)=(x+2)^2$. $n(iu)$ is real and positive, so that $W_{em}(R)$ is always repulsive, regardless of the frequency dependence of the refractive index.

## III. VAN DER WAALS INTERACTIONS FROM THE QUANTIZED FIELD WITH ABSORPTION

In this section we will calculate the van der Waals interactions considered in the previous section for two atoms embedded in a magnetodielectric medium, *but now taking absorption in the host medium into account*. We use the quantization procedure for the EM field in a dispersive and absorbing medium based on the Green-function formulation [12]. The dyadic Green function $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ satisfies [12]

$$\left[\boldsymbol{\nabla}\times\kappa(\mathbf{r},\omega)\boldsymbol{\nabla}\times - \frac{\omega^2}{c^2}\epsilon(\mathbf{r},\omega)\right]\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = \delta(\mathbf{r},\mathbf{r}',\omega), \quad (21)$$

as well as the appropriate boundary conditions. Here $\kappa(\mathbf{r},\omega)=\mu^{-1}(\mathbf{r},\omega)$. In an infinite, homogeneous material,

$$G_{ij}(\mathbf{r},\mathbf{r}',\omega) = \frac{\mu(\omega)}{4\pi k^2(\omega)}\left[k^2(\omega)(\delta_{ij}-\hat{\mathbf{R}}_i\hat{\mathbf{R}}_j) - (\delta_{ij}-3\hat{\mathbf{R}}_i\hat{\mathbf{R}}_j) \times\left(\frac{1}{R^2}-\frac{ik(\omega)}{R}\right)\right]\frac{e^{ik(\omega)R}}{R}, \quad (22)$$

where $\mathbf{R}=\mathbf{r}-\mathbf{r}'$, $R=|\mathbf{R}|$, $\hat{\mathbf{R}}=\mathbf{R}/R$, and $k(\omega)=n(\omega)\omega/c$. The refractive index of the medium is given by $n^2(\omega)=\epsilon(\omega)\mu(\omega)$, with $\epsilon(\omega)=\epsilon'(\omega)+i\epsilon''(\omega)$ the complex electric permittivity and $\mu(\omega)=\mu'(\omega)+i\mu''(\omega)$ the complex magnetic permeability.

The quantized electric field in dispersive, absorbing media may be written in the form [12]

$$\mathbf{E}(\mathbf{r},\omega) = \sum_{\lambda=e,m}\int d^3\mathbf{r}'\mathbf{G}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{f}_\lambda(\mathbf{r}',\omega) + \text{H.c.}, \quad (23)$$

where the operators $\mathbf{f}_\lambda$ are bosonic operators satisfying the usual commutation relations

$$[f_{\lambda,i}(\mathbf{r},\omega),f_{\lambda',j}^\dagger(\mathbf{r}',\omega')] = \delta_{\lambda,\lambda'}\delta_{ij}\delta(\omega-\omega')\delta(\mathbf{r}-\mathbf{r}'), \quad (24)$$

$$[f_{\lambda,i}(\mathbf{r},\omega),f_{\lambda',j}(\mathbf{r}',\omega')] = 0.$$

These operators may be regarded as being variables of the system composed of the EM field *and* the medium including the dissipative system. The electric and magnetic dyadic Green functions are defined in terms of the full Green function as

$$\mathbf{G}_e(\mathbf{r},\mathbf{r}',\omega) = i\frac{\omega^2}{c^2}\sqrt{\frac{\hbar}{\pi}\text{Im}\,\epsilon(\mathbf{r}',\omega)}\mathbf{G}(\mathbf{r},\mathbf{r}',\omega), \quad (25)$$

$$\mathbf{G}_m(\mathbf{r},\mathbf{r}',\omega) = -i\frac{\omega}{c}\sqrt{-\frac{\hbar}{\pi}\text{Im}\,\kappa(\mathbf{r}',\omega)}[\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\times\nabla_{\mathbf{r}'}]. \quad (26)$$

Note that for an absorbing medium $\text{Im}\,\epsilon(\mathbf{r},\omega)>0$, $\text{Im}\,\mu(\mathbf{r},\omega)>0$, and $\text{Im}\,\kappa(\mathbf{r},\omega)<0$. The quantized magnetic field, similarly, may be written as

$$\mathbf{H}(\mathbf{r},\omega) = \sum_{\lambda=e,m}\frac{c}{i\omega\mu(\omega)}\int d^3\mathbf{r}'\nabla_{\mathbf{r}}\times\mathbf{G}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{f}_\lambda(\mathbf{r}',\omega) + \text{H.c.}, \quad (27)$$

and the total Hamiltonian for the free field is

$$H = \sum_{\lambda=e,m} \int d^3\mathbf{r} \int_0^\infty d\omega \hbar \omega \mathbf{f}_\lambda^\dagger(\mathbf{r},\omega) \cdot \mathbf{f}_\lambda(\mathbf{r},\omega). \tag{28}$$

We will require the following two-point vacuum field correlation functions obtained from these expressions:

$$\langle \mathbf{E}_i(\mathbf{r},\omega)\mathbf{E}_j^\dagger(\mathbf{r}',\omega')\rangle = \frac{\hbar}{\pi}\frac{\omega^2}{c^2}\delta(\omega-\omega')\mathrm{Im}[G(\mathbf{r},\mathbf{r}',\omega)]_{ij}, \tag{29}$$

$$\langle \mathbf{H}_i(\mathbf{r},\omega)\mathbf{H}_j^\dagger(\mathbf{r}',\omega')\rangle = \frac{\hbar}{\pi}\frac{1}{|\mu(\omega)|^2}\delta(\omega-\omega') \times \mathrm{Im}[\nabla_{\mathbf{r}} \times \nabla_{\mathbf{r}'} \times G(\mathbf{r},\mathbf{r}',\omega)]_{ij}, \tag{30}$$

$$\langle \mathbf{E}_i(\mathbf{r},\omega)\mathbf{H}_j^\dagger(\mathbf{r}',\omega')\rangle = -\frac{1}{i\omega\mu^*(\omega)}\frac{\hbar}{\pi}\frac{\omega^2}{c}\delta(\omega-\omega') \times \mathrm{Im}[\nabla_{\mathbf{r}'} \times G(\mathbf{r},\mathbf{r}',\omega)]_{ij}. \tag{31}$$

The electric and magnetic fields at the position of the atom $A$ are given by the sum of the "vacuum" contributions, $\mathbf{E}_0(\mathbf{r}_A,\omega)$ and $\mathbf{H}_0(\mathbf{r}_A,\omega)$, plus the fields generated by the atom $B$ which contain both electric dipole and magnetic dipole components. An electric dipole located at position $\mathbf{r}_B$ generates fields at position $\mathbf{r}_A$ given by

$$\mathbf{E}_e(\mathbf{r}_A,\omega) = \omega^2\alpha_e^B(\omega)\mathbf{G}(\mathbf{r}_A,\mathbf{r}_B,\omega)\cdot\mathbf{E}_0(\mathbf{r}_B,\omega) + \mathrm{H.c.}, \tag{32}$$

$$\mathbf{H}_e(\mathbf{r}_A,\omega) = -i\omega c\kappa(\omega)\alpha_e^B(\omega) \times [\nabla_{\mathbf{r}_A} \times \mathbf{G}(\mathbf{r}_A,\mathbf{r}_B,\omega)]\cdot\mathbf{E}_0(\mathbf{r}_B,\omega) + \mathrm{H.c.}, \tag{33}$$

while the fields generated by a magnetic dipole at position $\mathbf{r}_B$ are

$$\mathbf{E}_m(\mathbf{r}_A,\omega) = -i\omega\kappa(\omega)\alpha_m^B(\omega) \times [\nabla_{\mathbf{r}_A} \times \mathbf{G}(\mathbf{r}_A,\mathbf{r}_B,\omega)]\cdot\mathbf{H}_0(\mathbf{r}_B,\omega) + \mathrm{H.c.}, \tag{34}$$

$$\mathbf{H}_m(\mathbf{r}_A,\omega) = c\kappa(\omega)k^2(\omega)\alpha_m^B(\omega)\mathbf{G}(\mathbf{r}_A,\mathbf{r}_B,\omega)\cdot\mathbf{H}_0(\mathbf{r}_B,\omega) + \mathrm{H.c.} \tag{35}$$

The vacuum expectation value of the van der Waals energy may be written as the sum of three contributions, one purely electric, one purely magnetic, and one mixed. Using the above expressions for the two-point correlation functions of the EM field, one can easily find each of these terms. The purely electric part stems from the $\mathbf{p}\cdot\mathbf{E}$ interaction, and is found to be

$$\begin{aligned} W_{ee}(R) &= -\frac{1}{2}\int_0^\infty\int_0^\infty d\omega d\omega'\,\alpha_e^A(\omega)e^{i(\omega-\omega')t}\langle\hat{\mathbf{E}}_0(\mathbf{r}_A,\omega)\cdot\hat{\mathbf{E}}_e^\dagger(\mathbf{r}_A,\omega')\rangle + \mathrm{H.c.} \\ &= -\frac{\hbar}{\pi}\int_0^\infty d\omega\alpha_e^A(\omega)\alpha_e^B(\omega)\omega^4\,\mathrm{Re}[G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij}\,\mathrm{Im}[G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij}. \end{aligned} \tag{36}$$

Comparing this expression with Eq. (4) we see that $\omega^2\,\mathrm{Im}[G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij}$ is related to the electric dipole-dipole interaction tensor $V_{ij}^{ee}(\omega,\mathbf{R})$, and that $\omega^2\,\mathrm{Re}[G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij}$ is related to the solid-angle integration of the two-point vacuum electric-field correlation function summed over polarization states, given in Eq. (5). After rotation in the complex plane ($\omega\to iu$), we can rewrite this expression as

$$W_{ee}(R) = -\frac{\hbar}{2\pi}\int_0^\infty du\alpha_e^A(iu)\alpha_e^B(iu)u^4\,\mathrm{Tr}[G(\mathbf{r}_A,\mathbf{r}_B,iu)\cdot G(\mathbf{r}_A,\mathbf{r}_B,iu)]. \tag{37}$$

The purely magnetic part, similarly, comes from the $\mathbf{m}\cdot\mathbf{H}$ interaction:

$$\begin{aligned} W_{mm}(R) &= -\frac{1}{2}\int_0^\infty\int_0^\infty d\omega d\omega'\,\alpha_m^A(\omega)e^{i(\omega-\omega')t}\langle\hat{\mathbf{H}}_0(\mathbf{r}_A,\omega)\cdot\hat{\mathbf{H}}_m^\dagger(\mathbf{r}_A,\omega')\rangle + \mathrm{H.c.} \\ &= -\frac{\hbar}{\pi}\int_0^\infty d\omega\alpha_m^A(\omega)\alpha_m^B(\omega)\frac{c}{|\mu(\omega)|^2}\,\mathrm{Re}[\kappa(\omega)k^2(\omega)G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij}\,\mathrm{Im}[\nabla_{\mathbf{r}_A}\times\nabla_{\mathbf{r}_B}\times G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij} \\ &= -\frac{\hbar}{2\pi}\int_0^\infty du\alpha_m^A(iu)\alpha_m^B(iu)u^4\frac{\epsilon^2(iu)}{\mu^2(iu)}\mathrm{Tr}[G(\mathbf{r}_A,\mathbf{r}_B,iu)\cdot G(\mathbf{r}_A,\mathbf{r}_B,iu)], \end{aligned} \tag{38}$$

where Eq. (21) and a rotation in the complex plane were used in obtaining the last equality. Finally, there are two electric-magnetic terms, one arising from the $\mathbf{p}\cdot\mathbf{E}$ interaction, and one from the $\mathbf{m}\cdot\mathbf{H}$ interaction. They result in the mixed interaction

$$W_{em}(R) = -\frac{1}{2}\int_0^\infty\int_0^\infty d\omega d\omega' e^{i(\omega-\omega')t}[\alpha_e^A(\omega)\langle\hat{\mathbf{E}}_0(\mathbf{r}_A,\omega)\cdot\hat{\mathbf{E}}_m^\dagger(\mathbf{r}_A,\omega')\rangle + \alpha_m^A(\omega)\langle\hat{\mathbf{H}}_0(\mathbf{r}_A,\omega)\cdot\hat{\mathbf{H}}_e^\dagger(\mathbf{r}_A,\omega')\rangle + \text{H.c.}]$$
$$= \frac{\hbar}{\pi}\int_0^\infty d\omega[\alpha_e^A(\omega)\alpha_m^B(\omega) + \alpha_m^A(\omega)\alpha_e^B(\omega)]\frac{\omega^2}{c}\,\mathrm{Re}[\kappa^2(\omega)\nabla_{\mathbf{r}_A}\times G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij}\,\mathrm{Im}[\nabla_{\mathbf{r}_B}\times G(\mathbf{r}_A,\mathbf{r}_B,\omega)]_{ij}$$
$$= \frac{\hbar}{2\pi}\int_0^\infty du[\alpha_e^A(iu)\alpha_m^B(iu) + \alpha_m^A(iu)\alpha_e^B(iu)]u^2\mathrm{Tr}[\nabla_{\mathbf{r}_A}\times G(\mathbf{r}_A,\mathbf{r}_B,iu)\cdot\nabla_{\mathbf{r}_B}\times G(\mathbf{r}_A,\mathbf{r}_B,iu)]. \quad (39)$$

The traces appearing in the integrands of Eqs. (37)–(39) can be explicitly computed given the form of the dyadic Green function evaluated at the imaginary frequency $\omega = iu$. The final result for the complete van der Waals interaction energy between two ground-state atoms embedded in an absorbing and dispersive medium is then

$$W(R) = -\frac{\hbar}{16\pi R^6}\int_0^\infty du e^{-2n(iu)uR/c}F\left[\frac{2n(iu)R}{c}\right] \times\left[\frac{\alpha_e^A(iu)\alpha_e^B(iu)}{\epsilon^2(iu)} + \frac{\alpha_m^A(iu)\alpha_m^B(iu)}{\mu^2(iu)}\right] + \frac{\hbar}{4\pi c^2R^4}\int_0^\infty du u^2 e^{-2n(iu)uR/c}G\left[\frac{2n(iu)R}{c}\right] \times[\alpha_e^A(iu)\alpha_m^B(iu) + \alpha_m^A(iu)\alpha_e^B(iu)], \quad (40)$$

where again $F(z) = z^4 + 4z^3 + 20z^2 + 48z + 48$ and $G(z) = (z+2)^2$. This is identical to the complete van der Waals interaction obtained in Sec. II.

## IV. DISCUSSION

Since the van der Waals interaction between electrically polarizable particles is the most important, a rough model for the modification of the vacuum interaction by the medium might be of interest. Let us consider a two-level model in which the polarizabilities are

$$\alpha_e^A(\omega) = \alpha_e^B(\omega) \equiv \alpha(\omega) = \frac{2\omega_0 d^2/3\hbar}{\omega_0^2 - \omega^2}, \quad (41)$$

where $d$ and $\omega_0$ are, respectively, the (real) transition electric dipole moment and the transition angular frequency, and similarly

$$n(\omega) = [1 + 4\pi N\alpha(\omega)]^{1/2}, \quad (42)$$

where $N$ is the atomic density of the host medium and we take $\mu = 1$. It is useful to normalize Eq. (11) to the familiar, nonretarded London form of the interaction:

$$W_L(R) = -\frac{3\hbar\omega_0\alpha^2(0)}{4R^6} = -\frac{3\hbar\omega_0}{4R^6}\left(\frac{2d^2}{3\hbar\omega_0}\right)^2. \quad (43)$$

We define

$$D(R) \equiv \frac{W_{ee}(R)}{W_L(R)} = \frac{4}{3\pi}\int_0^\infty dy\left(\frac{1}{y^2+1}\right)^2\frac{1}{\epsilon^2}[n^4r^4y^4 + 2n^3r^3y^3 + 5n^2r^2y^2 + 6nry + 3]e^{-2nry}, \quad (44)$$

where $r \equiv \omega_0 R/c$ and $\epsilon$ and $n$ are evaluated at $i\omega_0 y$

$$\epsilon(i\omega_0 y) = n^2(i\omega_0 y) = 1 + \frac{C}{y^2+1}, \quad (45)$$

where $C = 8\pi N d^2/3\hbar\omega_0$. In the limit $C=0$ and $r\to 0$, $D\to 1$; for $r\to\infty$, $D\to 23/3\pi r$, or $W_{ee} = -23\hbar c\alpha^2(0)/4\pi R^7$, the famous Casimir-Polder result. Figure 1 plots $D(R)$ for $C=0$ (vacuum) and $C=3$.

This simple model is not in any sense meant to be a realistic, quantitative description of the van der Waals interaction between two atoms embedded in a dielectric. It does, however, suggest that the predominant effect of the host medium on the van der Waals interaction is to weaken it, without substantially changing the distance dependence in either the nonretarded or retarded regimes. More realistic models of the van der Waals interaction in a liquid, for example, must take into account local field corrections, as has been discussed, for instance, by Abrikosov *et al.* [8] and McLachlan [18]. See also [19] for local field corrections in magnetodielectric media. It might be noted that no local field correction ap-

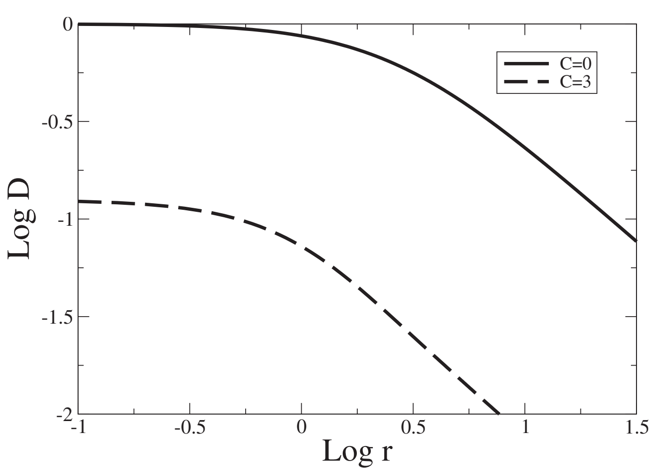


FIG. 1. The van der Waals interaction (11) divided by the London interaction for a two-level model. The upper curve is for two atoms in vacuum ($C=0$), and the lower curve is for two atoms in a dielectric defined by Eq. (45) with $C=3$.

pears in the Lifshitz theory, which is based on a continuum model in which there are no spaces between atoms and therefore no local field corrections. This is also clear from the fact that the (electric-electric) van der Waals interaction obtained from the dilute-medium limit of the Lifshitz theory involves no local field correction.

Our results for the electric-electric, magnetic-magnetic, and electric-magnetic van der Waals interactions between two atoms embedded in a dispersive magnetodielectric medium are in agreement with those obtained previously [3–7]. Unlike previous derivations, however, we obtained the interactions directly rather than inferring them from the dilute-medium limit of van der Waals interactions between macroscopic bodies, where the interactions can be obtained by pairwise summations of interatomic interactions [1–8].

What is perhaps more interesting, however, concerns the role of absorption in the host medium. In the preceding section we accounted for dissipation (absorption) in the host magnetodielectric medium, as in previous work. In Sec. II, however, we ignored any possibility that the medium could be absorbing: we worked with expressions for the electric and magnetic fields that derive directly from the assumption that the medium is nonabsorbing [13].

The same situation holds, for instance, in the case of the Lifshitz formula for the van der Waals interaction between infinite, plane-parallel dielectric media. Lifshitz's original derivation, and various derivations that followed, include the imaginary (dissipative) part of the dielectric function $\epsilon(\omega)$. Derivations of the Lifshitz formula based on changes in zero-point field energy arising from the dielectric media, however, make no reference to dissipation [20–22]. In other words, the Lifshitz formula can be derived without explicit accounting for absorption.

Ginzburg [23] has also noted that (macroscopic) van der Waals interactions can be correctly derived based on changes in zero-point field energy, without accounting for absorption. He remarks, in connection with such derivations [21], that "oddly enough there is no mention that they consider directly only transparent media," and then gives reasons why the van der Waals (electric-electric) interaction for real media can be obtained by presuming nonabsorbing media: "Firstly, the permittivities… are functions. Secondly, the function $\epsilon(\omega)$ is always real on the imaginary axis." The results (11), (15), and (20), for instance, all involve permittivities and permeabilities on the imaginary axis.

A simple and more physical explanation can be given for why (ground-state) van der Waals interactions calculated for nonabsorbing media apply directly to real (absorbing) media, as we have found for the electric-electric, magnetic-magnetic, and electric-magnetic van der Waals interactions between two atoms embedded in a magnetodielectric medium. At zero temperature, for instance, any atom of the host medium is in its ground state and can absorb radiation that is resonant with one of its transitions to an excited state. It cannot, of course, absorb from the vacuum field: in this case the fluctuations in the field that might induce absorption are exactly cancelled by fluctuations in the atom itself [22]. The same is true when the atom is part of a magnetodielectric in which is embedded, as in examples considered in this paper, two guest atoms. Any atom of the host medium still finds itself in a vacuum field state, regardless of the nature or the number of guest atoms. The host and guest atoms modify the modes of the field from the simple plane waves of a pure vacuum, but the field remains in a vacuum state $|\text{vac}\rangle$ ($f_{\lambda,i}|\text{vac}\rangle=0$ for any mode). Just as in free space, therefore, there is no absorption unless it is possible to populate one or more field modes; this would be the case only if there were an applied external field or if one or more atoms is excited, leading to the possibility that a different atom could absorb its emitted (*real*) photon. In other words, absorption by the host medium would play a role if we were to consider a van der Waals interaction involving excited atoms. Otherwise, one can expect to obtain correct van der Waals interactions without having to account for the absorption that is always present in a real medium. This expectation applies, of course, regardless of how many atoms are involved and regardless of the shape of any macroscopic bodies for which the van der Waals forces are to be calculated.

Of course the polarizabilities, permittivities, and permeabilities in our expressions for $W_{ee}(R)$, $W_{mm}(R)$, and $W_{em}(R)$ will be absorption-dependent. For example, a more general form for the real part of the polarizability $\alpha_e^A(\omega)$ than that used in Eq. (41) is

$$\alpha_e^A(\omega) = \frac{2\omega_0 d^2}{\hbar}\frac{\omega_0^2-\omega^2}{(\omega_0^2-\omega^2)^2+4\beta^2\omega^2}, \tag{46}$$

where $\beta$ is the linewidth characterizing the absorption associated with the imaginary part of the polarizability. Similarly, the permittivity and permeability of the host medium will have both real and imaginary parts as a consequence of absorption, which of course cannot be zero at all frequencies. But our analyses show that the only effect of host-medium absorption on the van der Waals interactions is through such functional dependence of the permittivity and the permeability on the parameters that determine the absorption coefficient for *real* photons.

In light of recent interest in negative-index media [24,25], let us reconsider specifically the most important of the van der Waals interactions we have calculated in this paper, namely, that between two electrically polarizable atoms. First we note that the expressions for the quantized fields in Sec. II are directly applicable to negative-index media [13]. In a negative-index medium $n$, $\epsilon$, and $\mu$ are all negative at some frequency or range of frequencies. This would at first glance suggest that the dipole-dipole interaction $V_{ij}^{ee}(\omega,\mathbf{R})$ [Eq. (9)] changes sign at frequencies for which the refractive index is negative. To see that this is not the case, note that Eq. (8) is unchanged whenever $n(\omega)$, $\epsilon(\omega)$, and $\mu(\omega)$ all change sign within any frequency range. This means that the dipole-dipole interaction in a negative-index medium does not change sign, and, in particular, that Eq. (9) is directly applicable, in general, provided we just replace $n(\omega)$, $\epsilon(\omega)$, and $\mu(\omega)$ by their absolute values. It follows similarly that the van der Waals interaction does not change sign or undergo any other significant change in a negative-index medium. The same conclusion applies to the magnetic-magnetic and mixed van der Waals interactions, and is in agreement with the conclusions of Buhmann *et al.* [5].

The pairwise electric-electric and magnetic-magnetic van der Waals interactions are always attractive, whereas the pairwise electric-magnetic interaction is always repulsive. These results apply also in the case of negative-index media, at least to the extent that such media can be modeled as continua. It is well known, however, that nonpairwise van der Waals interactions can be repulsive [26]. Evidently, repulsive Casimir effects such as those recently suggested by Henkel and Joulain [27] and by Leonhardt and Philbin [28] in the case of negative-index media must in some way involve either nonpairwise interactions or electric-magnetic van der Waals interactions.


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