Influence of a surface in the nonretarded interaction between two atoms

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In this work we obtain analytical expressions for the nonadditivity effects in the dispersive interaction between two atoms and a perfectly conducting surface of arbitrary shape in the nonretarded regime. We show that this three-body quantum-mechanical problem can be solved by mapping it onto a two-body electrostatic one. We apply the general formulas developed in this paper in several examples. First we rederive the London interaction as a particular case of our formalism. Then we investigate other interesting examples, such as the setup where two atoms lie inside a plane capacitor. Here we show that the nonadditivity is strikingly manifest since the planes lead to an exponential suppression of the interaction of the atoms. As a last example we deal with two atoms in the presence of a sphere, both grounded and isolated. We show that for realistic experimental parameters the nonadditivity may be relevant for the force in each atom.

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I. INTRODUCTION

One of the most interesting features of the van der Waals dispersive interactions is their nonadditivity [1–5], pointed out for the first time in [6] (for a pedagogical exposition see [7]). It means that the interaction between three bodies does not follow from the superposition principle or, equivalently, that the presence of a body influences the interaction of others. Although such effects have been known for many decades, their consequences are not yet fully uncovered. One of the fertile subjects that is being explored is the so-called Efimov quantum state [8], in which a resonant two-body force between identical bosons can produce bound states in a three-body system even if there is not any corresponding two-body bound state. The utilization of Feshbach resonances in ultracold-atom systems has allowed for an experimental probe of the Efimov states [9–11]. These are part of a larger program dedicated to the study of universal properties of few-body systems with a large scattering length [12–15]. It is also well known that the interaction between two Rydberg atoms can inhibit all but a single collective Rydberg excitation, a phenomenon called dipole blockade [16,17]. It has recently been shown that the inclusion of a third Rydberg atom can break the dipole blockade [18] due to nonadditivity effects.

Another very interesting consequence of nonadditivity is the (possibly strong) modification of the dispersion interactions between two or more atoms in the vicinities of macroscopic bodies. In [19] it was shown that the dispersive force between two atoms is greatly enhanced in the vicinity of a one-dimensional transmission line, while in [20–22] the influence of a magnetodielectric body in the interaction between atoms was studied. Moreover, there are numerous papers in the literature dedicated to the investigation of nonadditivity [23–26] in simple systems, with the three-atom problem actually being present in some textbooks [4,5]. Unfortunately, due to major calculational difficulties, the setups involving macroscopic bodies are rarely analyzed. A method developed by Eberlein and Zietal [27] enables us to evaluate the nonretarded dispersive interaction between one atom and a perfectly conducting surface of arbitrary shape, requiring only the knowledge of a classical Green's function that can be obtained from an electrostatic problem. This method has been applied in a variety of interesting problems [28–32] and in this paper we generalize it in order to obtain an analytic expression for the influence of a perfectly conducting surface on the nonretarded dispersive interaction between two atoms. As a particular case we show that in the absence of surfaces we recover the well-known interaction between two atoms, namely, London's formula.

This paper is organized as follows. In the next section we generalize Eberlein and Zietal's method to include a second atom in the system. We then identify the general expression for the nonadditivity term and show how it is related to an electrostatic problem of a *single* charge in the presence of the conducting body. In Sec. III we evaluate the influence of a surface on the interatomic interaction and in Sec. IV we analyze some examples. First we reobtain the interaction energy for two atoms in the presence of an infinite conducting plane. Then we investigate nonadditivity effects in the dispersive interaction between two atoms placed inside a parallel mirror cavity. This example is relevant experimentally [33] and we show that the nonadditivity effects are readily perceivable, leading to an exponential suppression of the van der Waals force between the atoms, provided they are kept apart by distances of the order of the separation between the plates or larger. A similar exponential attenuation was also obtained in [34] in the retarded regime for atoms inside a rectangular box. As a last example we present the calculation of two atoms in the presence of a conducting sphere, both grounded and isolated. We summarize in Sec. V.

II. INTERACTION ENERGY FOR TWO ATOMS AND A CONDUCTING SURFACE

In the nonretarded regime the electromagnetic field does not have to be quantized. Therefore, the interaction of an atom and a surface, which is usually dealt with within a quantum electrodynamics framework, can instead be approached by standard quantum-mechanics techniques, where the interaction Hamiltonian to be used in perturbation calculations is given by the instantaneous Coulomb interaction [35]. The convenience of the Eberlein-Zietal procedure consists in the mapping of a quantum-mechanical problem onto an electrostatic one, allowing us to solve the nonretarded interaction in the simpler electrostatic domain.

Let us begin by considering two atoms A and B at positions $\mathbf{r}_A$ and $\mathbf{r}_B$, respectively, in the presence of a grounded perfectly conducting surface $\mathcal{S}$. The electrostatic energy of the configuration is given by

$$U = \frac{1}{2} \int \rho(\mathbf{r})\Phi(\mathbf{r})d^3\mathbf{r}, \tag{1}$$

where $\rho(\mathbf{r})$ is the charge distribution and $\Phi(\mathbf{r})$ is the electrostatic potential, which satisfies the Poisson equation

$$\nabla^2 \Phi(\mathbf{r}) = -\frac{\rho(\mathbf{r})}{\varepsilon_0} \tag{2}$$

and vanishes at surface $\mathcal{S}$. The solution of Eq. (2) can be written in terms of the Green's function

$$\Phi(\mathbf{r}) = \frac{1}{\varepsilon_0} \int G(\mathbf{r},\mathbf{r}')\rho(\mathbf{r}')d^3\mathbf{r}', \tag{3}$$

where $G(\mathbf{r},\mathbf{r}')$ is the solution of the differential equation

$$\nabla^2 G(\mathbf{r},\mathbf{r}') = -\delta(\mathbf{r} - \mathbf{r}'), \tag{4}$$

subjected to the boundary condition

$$G(\mathbf{r},\mathbf{r}')|_{\mathbf{r}\in\mathcal{S}} = 0. \tag{5}$$

By substituting Eq. (3) into Eq. (1), we write the electrostatic energy as

$$U = \frac{1}{2\varepsilon_0} \int d^3\mathbf{r}\, d^3\mathbf{r}'\, \rho(\mathbf{r})G(\mathbf{r},\mathbf{r}')\rho(\mathbf{r}'). \tag{6}$$

An immediate particular solution of Eq. (4) is $1/4\pi|\mathbf{r} - \mathbf{r}'|$ which, however, does not obey the boundary condition (5). This suggests a decomposition of our Green's function in the form

$$G(\mathbf{r},\mathbf{r}') = \frac{1}{4\pi|\mathbf{r} - \mathbf{r}'|} + G_H(\mathbf{r},\mathbf{r}'), \tag{7}$$

where G_H satisfies the Laplace equation $\nabla^2 G_H = 0$ with the boundary condition

$$\left[\frac{1}{4\pi|\mathbf{r} - \mathbf{r}'|} + G_H(\mathbf{r},\mathbf{r}') \right]_{\mathbf{r}\in S} = 0. \tag{8}$$

The equations obeyed by G_H are analogous to those satisfied by the potential $\Phi_i(\mathbf{r})$ generated by the appropriate image charges in the electrostatic problem of a charge q at position $\mathbf{r}'$ in the presence of a perfectly conducting surface S. If we solve this electrostatic problem we will get G_H from the relation

$$G_H(\mathbf{r},\mathbf{r}') = \frac{\varepsilon_0 \Phi_i(\mathbf{r})}{q}. \tag{9}$$

The variable $\mathbf{r}'$ is implicitly present in $\Phi_i(\mathbf{r})$ since the image charges depend upon the position $\mathbf{r}'$ of the source charge. As it will become clear, G_H is the only function that must be calculated in order to evaluate the nonadditivity effects of our problem. Therefore, this method enables us to effectively replace a quantum-mechanical problem of two atoms in the presence of a conducting body by an electrostatic one of a single charge in the presence of the conducting body. To proceed further we must specify the charge distribution $\rho(\mathbf{r})$ appearing in Eq. (6). We model each atom, in a first approximation, as an electric point dipole, which we denote

by $\mathbf{d}$. Hence, the charge distribution is given by

$$\begin{aligned} \rho(\mathbf{r}) = & \lim_{\substack{\mathbf{h}_A \to 0 \\ q\mathbf{h}_A = \mathbf{d}_A}} q[\delta(\mathbf{r} - (\mathbf{r}_A + \mathbf{h}_A)) - \delta(\mathbf{r} - \mathbf{r}_A)] \\ & + \lim_{\substack{\mathbf{h}_B \to 0 \\ q\mathbf{h}_B = \mathbf{d}_B}} q[\delta(\mathbf{r} - (\mathbf{r}_B + \mathbf{h}_B)) - \delta(\mathbf{r} - \mathbf{r}_B)] \\ =: & \rho_A(\mathbf{r}) + \rho_B(\mathbf{r}), \end{aligned} \tag{10}$$

where the limit $\mathbf{h}_{A(B)} \to 0$ must be taken while keeping $q_{A(B)}\mathbf{h}_{A(B)}$ constant and equal to $\mathbf{d}_{A(B)}$. Note that the surface charges induced on the conductor do not contribute to the electrostatic energy (6) since G vanishes at the surface [36]. In the following we write the electrostatic energy of two point dipoles in the presence of a conducting surface. This allows us to write the quantum Hamiltonian interaction for two atoms and a conducting surface by promoting $\mathbf{d}$ to a quantum operator. Substituting the decomposition (7) and Eq. (10) into Eq. (6) we obtain

$$\begin{aligned} U = & \frac{1}{2\varepsilon_0} \int [\rho_A(\mathbf{r}) + \rho_B(\mathbf{r})]G(\mathbf{r},\mathbf{r}')[\rho_A(\mathbf{r}') + \rho_B(\mathbf{r}')]d^3\mathbf{r}'d^3\mathbf{r} \\ =: & U_A + U_B + U_{\text{crossed}}, \end{aligned} \tag{11}$$

where

$$U_i = \frac{1}{2\varepsilon_0} \int \rho_i(\mathbf{r})G(\mathbf{r},\mathbf{r}')\rho_i(\mathbf{r}')d^3\mathbf{r}'d^3\mathbf{r}, \tag{12}$$

with $i = A,B$, and

$$U_{\text{crossed}} = \frac{1}{\varepsilon_0} \int \rho_A(\mathbf{r})G(\mathbf{r},\mathbf{r}')\rho_B(\mathbf{r}')d^3\mathbf{r}'d^3\mathbf{r}. \tag{13}$$

To obtain this last equation we used that $G(\mathbf{r},\mathbf{r}') = G(\mathbf{r}',\mathbf{r})$, whose validity follows from Green's identity [37]. Since the Green's function is the same in the case of one atom or two atoms, U_i represents the interaction energy between point dipole i and the surface $\mathcal{S}$ in the absence of the other dipole [38]. To unveil the physical meaning of U_{AB} let us employ the decomposition (7)

$$\begin{aligned} U_{\text{crossed}} = & \frac{1}{\varepsilon_0} \int \underbrace{\frac{\rho_A(\mathbf{r})\rho_B(\mathbf{r}')}{4\pi|\mathbf{r} - \mathbf{r}'|}d^3\mathbf{r}'d^3\mathbf{r}}_{U_{AB}} \\ & + \underbrace{\frac{1}{\varepsilon_0} \int \rho_A(\mathbf{r})G_H(\mathbf{r},\mathbf{r}')\rho_B(\mathbf{r}')d^3\mathbf{r}'d^3\mathbf{r}}_{U_{ABS}}. \end{aligned} \tag{14}$$

The first term on the right-hand side of Eq. (14), U_{AB}, does not depend on the surface. It describes the interaction between two point dipoles in vacuum. The last term U_{ABS} depends conjointly on both dipoles and the surface. Therefore, the complete interaction energy can be written as

$$U = U_A + U_B + U_{AB} + U_{ABS}. \tag{15}$$

We see at once one positive aspect of this formalism: It enables us to study separately the so-called nonadditivity of dispersive forces, which is totally contained in the last term. Substituting Eq. (10) into Eq. (12), performing a Taylor expansion, and discarding divergent self-interaction terms, we obtain

$$\begin{aligned} U_A &= \frac{1}{2\varepsilon_0}(\mathbf{d}_A \cdot \nabla')(\mathbf{d}_A \cdot \nabla)G_H(\mathbf{r},\mathbf{r}')|_{\mathbf{r}=\mathbf{r}'=\mathbf{r}_A}, \\ U_B &= \frac{1}{2\varepsilon_0}(\mathbf{d}_B \cdot \nabla')(\mathbf{d}_B \cdot \nabla)G_H(\mathbf{r},\mathbf{r}')|_{\mathbf{r}=\mathbf{r}'=\mathbf{r}_B}, \end{aligned} \tag{16}$$

which in turn allows us to retrieve the Eberlein-Zietal formula for the interaction of a single atom with a conducting surface [27] as a particular case. Similarly, U_{AB} and U_{ABS} are given by

$$U_{AB} = \frac{1}{\varepsilon_0}(\mathbf{d}_B \cdot \nabla')(\mathbf{d}_A \cdot \nabla)\frac{1}{4\pi |\mathbf{r} - \mathbf{r}'|}\bigg|_{\mathbf{r}=\mathbf{r}_A, \mathbf{r}'=\mathbf{r}_B},$$

$$U_{ABS} = \frac{1}{\varepsilon_0}(\mathbf{d}_B \cdot \nabla')(\mathbf{d}_A \cdot \nabla)G_H(\mathbf{r},\mathbf{r}')\bigg|_{\mathbf{r}=\mathbf{r}_A, \mathbf{r}'=\mathbf{r}_B}. \tag{17}$$

Until now, we have been working within classical electrostatics. The passage to the corresponding quantum-mechanical problem is made by promoting $\mathbf{d}$ to an operator in Eqs. (12) and (17) in order to obtain the quantum interaction Hamiltonian for two atoms and a conducting surface. Therefore, the previous decomposition (15) can be recast into the form

$$\hat{H}_{\text{int}} = \hat{H}_A + \hat{H}_B + \hat{H}_{AB} + \hat{H}_{ABS}, \tag{18}$$

where the operators on the right-hand side are obtained from U_A, U_B, U_{AB}, and U_{ABS} by changing $\mathbf{d}$ to the quantum-mechanical operator $\hat{\mathbf{d}}$. Note that, as mentioned before, the interaction Hamiltonian does not involve field operators since we are in the nonretarded regime. To obtain the interaction energy for the dispersive interaction between the atoms (assumed to be in the ground state) and the surface we proceed perturbatively. In first order of perturbation theory we have $E_{nr}^{(1)} := \langle \hat{H}_{\text{int}} \rangle$, where $\langle \cdots \rangle$ denotes the expectation value of the operator inside the brackets in the ground state $|0_A,0_B\rangle$ of the atoms. In this order only the first two terms in Eq. (18) contribute, since for atoms with no permanent dipole moment we have

$$\langle 0_A,0_B|\hat{d}_i^A \hat{d}_j^B|0_A,0_B\rangle = \langle 0_A|\hat{d}_i^A|0_A\rangle\langle 0_B|\hat{d}_j^B|0_B\rangle = 0. \tag{19}$$

From now on we omit the carets to denote quantum operators in order to not overburden the notation. Evaluating $\langle H_A \rangle$ and $\langle H_B \rangle$ we obtain

$$E_{nr}^{(1)}(\mathbf{r}_A,\mathbf{r}_B) = \frac{1}{2\varepsilon_0}\sum_m \langle (d_m^A)^2 \rangle \nabla_m \nabla_m' G_H(\mathbf{r},\mathbf{r}')\bigg|_{\mathbf{r}=\mathbf{r}'=\mathbf{r}_A}$$
$$+ \frac{1}{2\varepsilon_0}\sum_m \langle (d_m^B)^2 \rangle \nabla_m \nabla_m' G_H(\mathbf{r},\mathbf{r}')\bigg|_{\mathbf{r}=\mathbf{r}'=\mathbf{r}_B}, \tag{20}$$

where we employed $\langle d_m d_n \rangle = \delta_{mn}\langle d_m^2 \rangle$, valid for the orthonormal basis, which are used throughout this paper. In other words, in this approximation the atoms do not perceive each other and the interaction of the system is the direct superposition of the interaction between each atom and the surface S. The nonadditivity effects we are looking for appear only at second order

$$E_{nr}^{(2)}(\mathbf{r}_A,\mathbf{r}_B) = -\sum_I {}' \frac{\langle 0_A,0_B|H_{\text{int}}|I\rangle\langle I|H_{\text{int}}|0_A,0_B\rangle}{E_I - (E_0^A + E_0^B)}, \tag{21}$$

where the prime indicates that we must sum over all possible states $|I\rangle \neq |0_A,0_B\rangle$. Here E_0^A and E_0^B are the ground-state energies of the atoms A and B, respectively. Denoting the possible states of atom A by $|r\rangle$ and the possible states of atom

B by $|s\rangle$, we may write the previous formula as

$$E_{nr}^{(2)}(\mathbf{r}_A,\mathbf{r}_B) = -\sum_{r,s} {}' \frac{\langle 0_A,0_B|H_{\text{int}}|r,s\rangle\langle r,s|H_{\text{int}}|0_A,0_B\rangle}{E_{0r}^A + E_{0s}^B}, \tag{22}$$

where $E_{0r}^A = E_r^A - E_0^A$ and $E_{0s}^B = E_s^B - E_0^B$. Equation (18) shows that we would have, in principle, 16 terms to deal with in Eq. (22). Fortunately, as we shall see, most of them either vanish or are irrelevant for our purposes. For the sake of clarity, let us analyze the terms separately. We begin by the term quadratic in H_A, to wit

$$E_A^{(2)}(\mathbf{r}_A) = -\sum_{r,s} {}' \frac{\langle 0_A,0_B|H_A|r,s\rangle\langle r,s|H_A|0_A,0_B\rangle}{E_{0r}^A + E_{0s}^B}$$
$$= -\sum_r {}' \frac{\langle 0_A|H_A|r\rangle\langle n|H_A|0_A\rangle}{E_{0r}^A}, \tag{23}$$

where we used the fact that only intermediate states with $|s\rangle = |0_B\rangle$ survive in the summation. This term depends only on atom A and stands for the second-order contribution to the interaction between atom A and the surface. It is clear that such a term does not contribute to the interatomic force and is then irrelevant for nonadditivity effects. For this reason we shall neglect $E_A^{(2)}(\mathbf{r}_A)$ henceforth, along with the analogous term for atom B.

All the other ten terms involving H_i vanish. Indeed, the crossed term involving H_A and H_B

$$-\sum_{r,s} {}' \frac{\langle 0,0|H_A|r,s\rangle\langle r,s|H_B|0,0\rangle}{E_{0r}^A + E_{0s}^B} = 0 \tag{24}$$

because the term $\langle 0,0|H_A|r,s\rangle$ is not zero only for intermediate states with $s = 0$, while $\langle r,s|H_B|0,0\rangle$ vanishes in such cases. From Eq. (17) we see that the same argument applies to the crossed term

$$-\sum_{r,s} {}' \frac{\langle 0,0|H_A|r,s\rangle\langle r,s|H_{ABS}|0,0\rangle}{E_{0r}^A + E_{0s}^B}$$
$$= -\frac{1}{\varepsilon_0}\sum_{r,s} {}' \frac{\langle 0,0|H_A|r,s\rangle\langle r,s|d_i^A d_j^B|0,0\rangle}{E_{0r}^A + E_{0s}^B}$$
$$\times \nabla_i \nabla_j' G_H(\mathbf{r},\mathbf{r}')\big|_{\mathbf{r}=\mathbf{r}_A, \mathbf{r}'=\mathbf{r}_B}, \tag{25}$$

so the summation in Eq. (22) becomes

$$E_{nr}^{(2)}(\mathbf{r}_A,\mathbf{r}_B) = E_A^{(2)}(\mathbf{r}_A) + E_B^{(2)}(\mathbf{r}_B)$$
$$+ E_{\text{Lon}}(\mathbf{r}_A,\mathbf{r}_B) + E_{na}(\mathbf{r}_A,\mathbf{r}_B), \tag{26}$$

where

$$E_{\text{Lon}} = -\sum_{r,s} {}' \frac{\langle 0,0|H_{AB}|r,s\rangle\langle r,s|H_{AB}|0,0\rangle}{E_{0r}^A + E_{0s}^B} \tag{27}$$

and

$$E_{na} = -\sum_{r,s} {}' \frac{\langle 0,0|H_{AB}|r,s\rangle\langle r,s|H_{ABS}|0,0\rangle}{E_{0r}^A + E_{0s}^B}$$
$$- \sum_{r,s} {}' \frac{\langle 0,0|H_{ABS}|r,s\rangle\langle r,s|H_{AB}|0,0\rangle}{E_{0r}^A + E_{0s}^B}$$
$$- \sum_{r,s} {}' \frac{\langle 0,0|H_{ABS}|r,s\rangle\langle r,s|H_{ABS}|0,0\rangle}{E_{0r}^A + E_{0s}^B}. \tag{28}$$

Finally, we are left with just four terms to analyze. The one contained in E_{Lon} is independent of the surface, so it accounts for the interaction between the atoms in the vacuum. The other three terms, expressed in E_{na}, are the key elements in this paper and contain the nonadditivity effects. Before delving deeper in this contribution, though, let us turn to E_{Lon} in order to rewrite it in a more familiar way. From Eq. (17) we see that

$$
\begin{aligned}
H_{AB} &= \frac{1}{4\pi\varepsilon_0}\big(d_i^A d_j^B\big)\nabla_j'\nabla_i \frac{1}{|\mathbf{r}-\mathbf{r}'|}\bigg|_{\mathbf{r}=\mathbf{r}_A,\mathbf{r}'=\mathbf{r}_B} \\
&= \frac{d_i^A d_j^B}{4\pi\varepsilon_0 R_{AB}^3}[\delta_{ij}-3(\hat{R}_{AB})_i(\hat{R}_{AB})_j],
\end{aligned}
\tag{29}
$$

where $\mathbf{R}_{AB}:=\mathbf{r}_A-\mathbf{r}_B$. Substituting (29) into (27) we get

$$
\begin{aligned}
E_{\text{Lon}} = -\sum_{r,s}{}' \frac{\langle 0,0|d_i^A d_j^B|r,s\rangle\langle r,s|d_m^A d_n^A|0,0\rangle}{(4\pi\varepsilon_0)^2 R_{AB}^6\big(E_{0r}^A+E_{0s}^B\big)} \\
\times[\delta_{ij}-3(\hat{R}_{AB})_i(\hat{R}_{AB})_j][\delta_{mn}-3(\hat{R}_{AB})_m(\hat{R}_{AB})_n].
\end{aligned}
\tag{30}
$$

As usual, we adopt the notation $\langle 0|d_i^A|r\rangle=d_i^{0r}$ and, for the atom B, $\langle 0|d_j^B|s\rangle=d_j^{0s}$. Therefore, we write

$$
\langle 0,0|d_i^A d_j^B|r,s\rangle\langle r,s|d_m^A d_n^A|0,0\rangle = d_i^{0r}d_m^{r0}d_j^{0s}d_n^{s0}.
\tag{31}
$$

As we are dealing with freely rotating atoms, the above transition elements must be averaged over all directions. Furthermore, assuming isotropy of the atoms, we have

$$
\overline{d_i^{0r}d_m^{r0}} = \delta_{im}\frac{|\mathbf{d}_A^{0r}|^2}{3},
\tag{32}
$$

where the overline denotes the average over all directions. An analogous equation holds for atom B. So, combining the Eqs. (31) and (32) and substituting them into (30), we obtain London's result in its most common form [39]

$$
E_{\text{Lon}} = -\frac{1}{24\pi^2\varepsilon_0^2 R_{AB}^6}\sum_{r,s}{}' \frac{|\mathbf{d}_A^{0r}|^2|\mathbf{d}_B^{0s}|^2}{\big(E_{0r}^A+E_{0s}^B\big)}.
\tag{33}
$$

Having reobtained this important expression as a particular case of our general expressions, we turn to the nonadditive terms in next section.

III. THE NONADDITIVITY TERM

Let us now focus on the nonadditive effects. Equation (28) is the only one that depends simultaneously on both atoms and the surface; it reflects and contains the nonadditivity effects that are inherent to the van der Waals dispersive interaction. The mathematical treatment of this term is completely analogous to that given to E_{Lon} in the preceding section. In such a way, the first two terms on the right-hand side of Eq. (28) are equal and are given by

$$
\begin{aligned}
&-\sum_{r,s}{}' \frac{\langle 0,0|H_{AB}|r,s\rangle\langle r,s|H_{ABS}|0,0\rangle}{E_{0r}^A+E_{0s}^B} \\
&= -\sum_{r,s}{}' \frac{\langle 0,0|H_{ABS}|r,s\rangle\langle r,s|H_{AB}|0,0\rangle}{E_{0r}^A+E_{0s}^B}
\end{aligned}
$$

$$
\begin{aligned}
&= -\frac{1}{36\pi\varepsilon_0^2 R_{AB}^3}\sum_{r,s}{}' \frac{|\mathbf{d}_A^{0r}|^2|\mathbf{d}_B^{0s}|^2}{\big(E_{0r}^A+E_{0s}^B\big)}\big\{\mathcal{G}_{ii}^H(\mathbf{r}_A,\mathbf{r}_B) \\
&\quad -3(\hat{R}_{AB})_i(\hat{R}_{AB})_j\mathcal{G}_{ij}^H(\mathbf{r}_A,\mathbf{r}_B)\big\},
\end{aligned}
\tag{34}
$$

while the last term on the right-hand side of Eq. (28) is

$$
-\sum_{r,s}{}' \frac{\langle 0,0|H_{ABS}|r,s\rangle\langle r,s|H_{ABS}|0,0\rangle}{E_{0r}^A+E_{0s}^B}
$$

$$
= -\frac{1}{9\varepsilon_0^2}\sum_{r,s,i,j}{}' \frac{|\mathbf{d}_A^{0r}|^2|\mathbf{d}_B^{0s}|^2}{\big(E_{0r}^A+E_{0s}^B\big)}\big[\mathcal{G}_{ij}^H(\mathbf{r}_A,\mathbf{r}_B)\big]^2,
\tag{35}
$$

where we defined

$$
\mathcal{G}_{ij}^H(\mathbf{r}_1,\mathbf{r}_2) = \nabla_i\nabla_j'G_H(\mathbf{r},\mathbf{r}')|_{\mathbf{r}=\mathbf{r}_1,\mathbf{r}'=\mathbf{r}_2}.
\tag{36}
$$

Substituting Eqs. (34) and (35) into Eq. (28) we obtain

$$
E_{na} = E_{na}^{(1)}+E_{na}^{(2)},
\tag{37}
$$

with

$$
\begin{aligned}
E_{na}^{(1)} = -\frac{\Lambda_{AB}}{18\pi\varepsilon_0^2 R^3}\big[\operatorname{Tr}\mathcal{G}^H(\mathbf{r}_A,\mathbf{r}_B) \\
-3(\hat{R}_{AB})_i(\hat{R}_{AB})_j\mathcal{G}_{ij}^H(\mathbf{r}_A,\mathbf{r}_B)\big],
\end{aligned}
\tag{38}
$$

$$
E_{na}^{(2)} = -\frac{\Lambda_{AB}}{9\varepsilon_0^2}\sum_{i,j}\big[\mathcal{G}_{ij}^H(\mathbf{r}_A,\mathbf{r}_B)\big]^2,
\tag{39}
$$

where

$$
\Lambda_{AB} = \sum_{r,s}{}' \frac{|\mathbf{d}_A^{0r}|^2|\mathbf{d}_B^{0s}|^2}{\big(E_{0r}^A+E_{0s}^B\big)}.
\tag{40}
$$

Equations (38) and (39) constitute the main result of this paper. We now proceed to evaluate the influence exerted by the surface on the interatomic interaction by analyzing the ratio

$$
\begin{aligned}
\frac{E_{na}}{E_{\text{Lon}}} &= \frac{4\pi R_{AB}^3}{3}\big[\operatorname{Tr}\mathcal{G}^H(\mathbf{r}_A,\mathbf{r}_B)-3(\hat{R}_{AB})_i(\hat{R}_{AB})_j\mathcal{G}_{ij}^H(\mathbf{r}_A,\mathbf{r}_B)\big] \\
&\quad +\frac{8\pi^2 R_{AB}^6}{3}\sum_{i,j}\big[\mathcal{G}_{ij}^H(\mathbf{r}_A,\mathbf{r}_B)\big]^2.
\end{aligned}
\tag{41}
$$

The influence of the surface on the atomic interaction, in this order of perturbation theory, is a purely geometrical effect since it does not depend on the internal structure of the atoms. The formulas developed in this section show that in order to calculate the nonadditivity effects all we need is G_H, which we can get by mapping our problem onto an electrostatic one, according to Eq. (9). In the next section we will illustrate this method by treating some examples.

IV. APPLICATIONS

A. Two atoms and a conducting infinite plane

As a first example let us consider two atoms in front of an infinite conducting plane as illustrated in Fig. 1. We choose our coordinate axes so as to have the conducting plane at $z=0$ and both atoms in the $\mathcal{X}\mathcal{Z}$ plane. From Eq. (9) we get

$$
G_H(\mathbf{r},\mathbf{r}') = -\frac{1}{4\pi\sqrt{(x-x')^2+(y-y')^2+(z+z')^2}}.
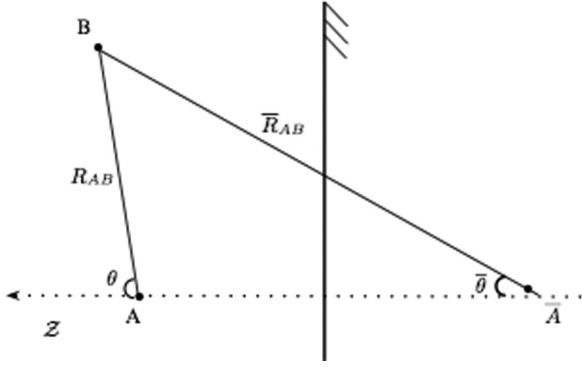\tag{42}
$$

FIG. 1. Two atoms A and B in the presence of an infinite conducting plane. Here $\bar{A}$ is the image of A and the image of B is not represented; R_{AB} is the distance between the atoms and $\bar{R}_{AB}$ is the distance between B and the image of A, denoted by $\bar{A}$.

Employing Eq. (36), we see that the only nonvanishing terms are

$$\mathcal{G}_{xx} = \frac{3\sin^2\bar{\theta} - 1}{4\pi\,\bar{R}_{AB}^3}, \tag{43}$$

$$\mathcal{G}_{yy} = -\frac{1}{4\pi\,\bar{R}_{AB}^3}, \tag{44}$$

$$\mathcal{G}_{zz} = \frac{1 - 3\cos^2\bar{\theta}}{4\pi\,\bar{R}_{AB}^3}, \tag{45}$$

$$\mathcal{G}_{xz} = \frac{3\sin\bar{\theta}\cos\bar{\theta}}{4\pi\,\bar{R}_{AB}^3} = -\mathcal{G}_{zx}, \tag{46}$$

where we used $R_{AB}\sin\theta = \bar{R}_{AB}\sin\bar{\theta}$. Substituting these expressions into Eq. (39) we arrive at

$$E_{na}^{(2)} = -\frac{\Lambda_{AB}}{24\pi^2\varepsilon_0^2\,\bar{R}_{AB}^6} = E_{\text{Lon}}(\bar{R}), \tag{47}$$

where we used Eq. (33). This term stands for the London interactions between either of the two atoms and the image of the other. To evaluate the next term, note that $\hat{R}_x = \sin\theta$ and $\hat{R}_z = \cos\theta$, while the y component vanishes. Hence Eq. (38) yields

$$E_{na}^{(1)} = -\frac{\Lambda_{AB}}{72\pi^2\varepsilon_0^2 R_{AB}^3\bar{R}_{AB}^3}(2 - 3\sin^2\theta - 3\sin^2\bar{\theta}). \tag{48}$$

We have now the complete expression for the nonadditivity terms up to second order in perturbation theory for the two-atom–conducting-plane case. The sum $E_{na}^{(1)} + E_{na}^{(2)}$ coincides precisely with the result obtained by Power and Thirunamachandran [40].

B. Two atoms inside a plane capacitor

This constitutes the main example of this paper. As we presently show, the nonadditivity effects are noticeable even at large interatomic separations as they strongly suppress the interatomic interaction.

Let us consider two atoms A and B between two infinite perfectly conducting planes parallel to each other. It is convenient to choose our coordinates in such a way as to
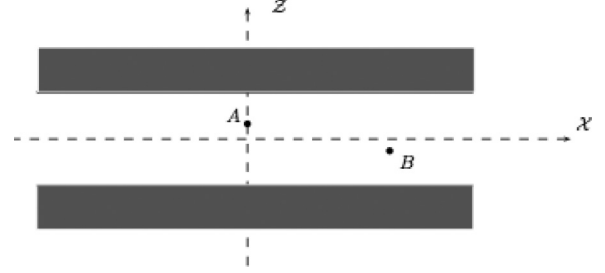

FIG. 2. Two atoms A and B inside a perfectly conducting plate conductor. We choose the plane $z = 0$ midway between the plates. Without any loss of generality, atom A is put at $(0,0,z_A)$ and B at $(x_B,0,z_B)$.

have the planes at $z = -D/2$ and $z = D/2$ and both atoms in the $\mathcal{X}\mathcal{Z}$ plane as illustrated in Fig. 2. The G_H is obtained through the solution of the electrostatic problem of one charge in the presence of a plane capacitor, which can be done again by employing the image method. In this case, however, we must deal with an infinite series of images and the potential generated by this series is very slowly convergent. It is therefore more convenient to write G_H in another form. We follow Ref. [41] to find that for this geometry the Green's function may be written as

$$G(\mathbf{r},\mathbf{r}') = \frac{1}{\pi D}\sum_{n=1}^{\infty}\cos\frac{n\pi z}{D}\cos\frac{n\pi z'}{D}K_0\left(\frac{n\pi|\boldsymbol{\rho}-\boldsymbol{\rho}'|}{D}\right), \tag{49}$$

where $\boldsymbol{\rho} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}}$, $\boldsymbol{\rho}' = x'\hat{\mathbf{x}} + y'\hat{\mathbf{y}}$, and K_0 is a modified Bessel function of the second kind [42]. Note that in Eq. (49) we have the complete Green's function [see Eq. (7)] instead of G_H. It turns out, however, that in this example it pays off to work directly with G and in the end we isolate G_H in order to get the nonadditivity contribution.

The asymptotic expansion of Eq. (49), valid in the region $|\boldsymbol{\rho}-\boldsymbol{\rho}'| \gtrsim D$, is given by

$$G(\mathbf{r},\mathbf{r}') = \frac{1}{4\pi}\sqrt{\frac{8}{|\boldsymbol{\rho}-\boldsymbol{\rho}'|D}}\cos\frac{\pi z}{D}\cos\frac{\pi z'}{D}e^{-\pi|\boldsymbol{\rho}-\boldsymbol{\rho}'|/D}. \tag{50}$$

Working directly with G, the four terms depicted in Eqs. (30), (34), and (35) can be naturally assembled in just one term

$$E_{\text{Lon}} + E_{na} = -\frac{\Lambda_{AB}}{9\varepsilon_0^2}\sum_{i,j}[\mathcal{G}_{ij}(\mathbf{r}_A,\mathbf{r}_B)]^2, \tag{51}$$

where

$$\mathcal{G}_{ij}(\mathbf{r}_A,\mathbf{r}_B) = \frac{1}{4\pi}\nabla_i\nabla_j'\left\{\cos\frac{\pi z}{D}\cos\frac{\pi z'}{D}e^{-\pi|\boldsymbol{\rho}-\boldsymbol{\rho}'|/D}\right.$$
$$\left.\times\sqrt{\frac{8}{|\boldsymbol{\rho}-\boldsymbol{\rho}'|D}}\right\}\Bigg|_{\mathbf{r}=\mathbf{r}_A,\mathbf{r}'=\mathbf{r}_B}. \tag{52}$$

Hence, when the atoms are separated by a distance of the order of D or larger, the nonadditivity effects shield the atoms' interaction exponentially. This is a remarkable result and it is particularly interesting when both atoms are equidistant
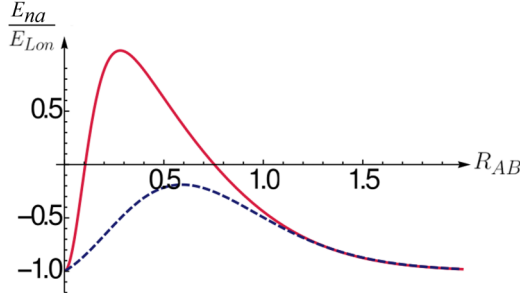
FIG. 3. (Color online) Nonadditive part of the interaction energy (normalized by the London interaction energy) as a function of the distance between atoms A and B. The atoms are both equidistant to the plates. Here R_{AB} is measured in units of D. The blue dashed curve shows the asymptotic behavior obtained via Eq. (50) while the red solid curve presents the exact result obtained through (49).

from the plates. In this case, we see immediately by symmetry that the force exerted by the surfaces in each atom separately vanishes. In spite of that, the plates leave their mark on the interatomic force by suppressing exponentially an interaction that would fall with $1/R_{AB}^6$ in their absence (with no plates we would have only the London interaction). The results displayed here remain valid when several atoms are present, because up to second order there are no terms in the interaction energy that can couple more than two atoms. This can be seen by employing a reasoning similar to the one used to show that the terms in Eqs. (24) and (25) vanish. Hence, if a gas is rarefied enough so that its atoms are separated on average by a distance D or more, then up to second order of perturbation the capacitor strongly shields the interatomic interaction, effectively producing an ideal gas behavior. Even when this condition is not strictly satisfied our results show that the atoms interact only with the atoms that are closer than D, leading us to the expectation that a gas put between parallel conducting plates behaves more ideally than otherwise. Since the interatomic interaction is the main feature responsible for the gas-liquid phase transition, we conjecture that putting a gas between conducting plates could lower the liquefaction temperature.

Finally, we isolate the nonadditivity effects through Eqs. (38) and (39). In order to obtain G_H we must subtract $1/|\mathbf{r} - \mathbf{r}'|$ from Eq. (49). In Fig. 3 we plot the nonadditivity effects normalized by the London energy for both the complete expression and the asymptotic expansion [obtained using Eq. (50) for G_H] in the situation where both atoms are at $z = 0$ (equidistant from the conducting planes). As expected from our previous discussion, for large distances the ratio goes to -1, showing that the nonadditivity cancels out the London interaction between the atoms. Finally, for illustration purposes, in Fig. 4 we show the plot for the ρ component of the force on atom A.

C. Two atoms and a conducting grounded sphere

In this section we analyze the nonretarded interaction between two atoms and a grounded perfectly conducting sphere. Since the application of the image method to the sphere is well known, we shall use it to obtain G_H. Placing the origin
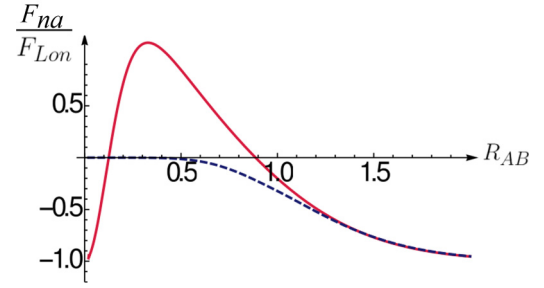

FIG. 4. (Color online) Nonadditive part of the force (normalized by the force between the atoms in vacuum) as a function of the distance between atom A and atom B. The atoms are both equidistant to the plates. Here R_{AB} is measured in units of D. The blue dashed curve shows the asymptotic behavior obtained via Eq. (50) while the red solid curve presents the exact result obtained through (49).

of our coordinates at the center of the sphere of radius a and setting $\mathbf{r}'$ as the position of the physical charge, we have to put an image charge $q_i = -\frac{a}{r'}q$ at position $\mathbf{r}'_i = \frac{a^2}{r'^2}\mathbf{r}'$ [43]. Therefore, from Eq. (9) we have

$$G_H(\mathbf{r},\mathbf{r}') = -\frac{a}{4\pi r'|\mathbf{r} - \mathbf{r}'_i|} = -\frac{a}{4\pi\sqrt{r^2 r'^2 - 2\mathbf{r}\cdot\mathbf{r}'a^2 + a^4}}. \quad (53)$$

Following the same scheme outlined in the preceding sections, we now use this function to evaluate the quantum dispersive interaction between the atoms and the sphere. Calculating $\mathcal{G}_{ij}$ from Eq. (36), we obtain after some algebra

$$\mathcal{G}_{ij}^H = -\frac{3a\left(x_i^A r_B^2 - x_i^B a^2\right)\left(x_j^B r_A^2 - x_j^A a^2\right)}{4\pi\left[r_A^2 r_B^2 - 2\mathbf{r}_A\cdot\mathbf{r}_B a^2 + a^4\right]^{5/2}}$$
$$+ \frac{a\left(2x_i^A x_j^B - \delta_{ij}a^2\right)}{4\pi\left[r_A^2 r_B^2 - 2\mathbf{r}_A\cdot\mathbf{r}_B a^2 + a^4\right]^{3/2}}, \quad (54)$$

where x_i^A stands for the ith Cartesian coordinate of $\mathbf{r}_A$, $r_A = |\mathbf{r}_A|$, with analogous notation for the coordinates of B. It is convenient to orient the axis in order to have atom A at $(0,0,r_A)$ and atom B at $(0,r_B \sin\theta, r_B\cos\theta)$. Employing Eq. (39) we get the complete expression for the nonadditivity terms for any configuration of the atoms, but for the sake of clarity we write explicitly only two particular cases. When the two atoms are aligned with the center of the sphere, we see from (54) that $\mathcal{G}$ is diagonal. We have then $\theta = 0$ ($\theta = \pi$) when the atoms are on the same side (opposite sides) of the sphere and the nonadditivity terms are given by

$$E_{na}^{(1)} = \pm\frac{\Lambda_{AB}}{36\pi^2\varepsilon_0^2 R_{AB}^3}\frac{ar_A r_B}{(r_A r_B \pm a^2)^3}, \quad (55)$$

$$E_{na}^{(2)} = -\frac{\Lambda_{AB}}{144\pi^2\varepsilon_0^2}\frac{3a^6 \mp 2a^4 r_A r_B + a^2 r_A^2 r_B^2}{(r_A r_B \pm a^2)^6}, \quad (56)$$

where the upper (lower) sign refers to the $\theta = \pi$ ($\theta = 0$) case. Note that both terms are bigger for $\theta = 0$. Since, by symmetry, we expect the same behavior when we change θ by $2\pi - \theta$, we conclude that $\theta = \pi$ will be a minimum for the nonadditivity interaction energy.
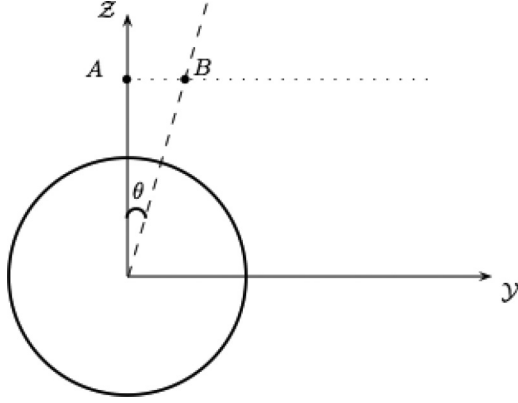
FIG. 5. Two atoms near a conducting sphere. Here $\mathbf{R}_{AB}$ is parallel to the $\mathcal{Y}$ axis.

In this case, in contrast with the capacitor example, nonadditivity effects are practically unnoticeable since the dominant contribution by far comes from the attraction of each atom with the sphere, which contributes in first order of perturbation theory. However, since symmetry considerations ensure that the force exerted by the sphere on each atom is radial, nonadditivity may be relevant for components of the force perpendicular to that direction. Let us then consider the situation where the atom A is at the $\mathcal{Z}$ axis and $\mathbf{R}_{AB}$ is parallel to the $\mathcal{Y}$ direction, as illustrated in Fig. 5.

In this setup, only the London interaction and nonadditivity terms contribute to the y component of the force on atom B. In Fig. 6 we plot this component of the force as a function of the distance from atom B to the center of the sphere, keeping the vector $\mathbf{R}_{AB}$ fixed. We see that the nonadditivity parcel may indeed be comparable to the London interaction for close distances. For a sphere 1 μm in radius and atom B at a distance of 1 nm from the surface of the sphere and separated by a distance 2 nm from atom A, the nonadditivity force is 30% from the London force between the atoms.
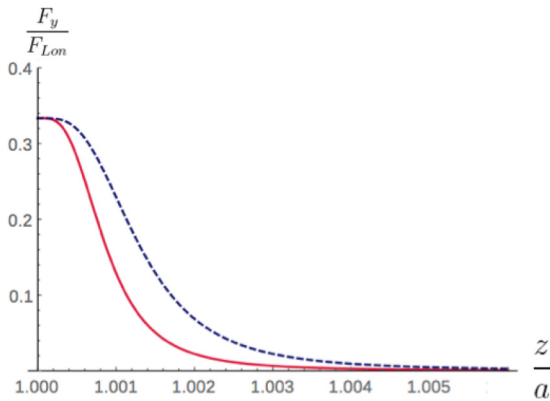

FIG. 6. (Color online) Nonadditive part of the x component of the force exerted on atom B (normalized by the London force) as a function of the distance between atom B and the center of the sphere. Here $\mathbf{R}_{AB}$ remains always perpendicular to $\mathbf{r}_B$. The red solid curve is for two atoms separated by $d_A = 0.002a$ and the blue dashed curve is for $d_A = 0.003a$.

We finish this example with some remarks. First, we recall that the setup of two atoms in the presence of a plane $E_{na}^{(2)}$, given in Eq. (47), could readily be identified as the London interaction between each atom and the image of the other. In this example, $E_{na}^{(2)}$ does not have such a simple interpretation. This is related to the fact that to solve the electrostatic problem of a dipole in the presence of a grounded conducting sphere we must have not only an image dipole but two point charges as well [44]. In addition, as a particular case of our results we may obtain, by substituting Eq. (53) into Eq. (12), the dispersive interaction between one atom and a conducting sphere. In so doing we arrive at the same result obtained in Ref. [45].

D. Two atoms and a conducting isolated neutral sphere

We need only minor modifications to tackle the case where the two atoms are in the presence of an isolated sphere, instead of a grounded one. In the electrostatic case, the isolated sphere interacts more weakly with a charge since in the grounded case the sphere is supplied by the earth with additional charges. Let us see what happens to the nonadditivity effects in this quantum problem.

In this case, G_H may be obtained from (53) by simply adding one term [31], yielding

$$G_H(\mathbf{r},\mathbf{r}') = -\frac{a}{4\pi \sqrt{r^2 r'^2 - 2\mathbf{r}\cdot\mathbf{r}' a^2 + a^4}} + \frac{a}{4\pi r r'}. \quad (57)$$

Substituting Eq. (57) into Eq. (36) and using Eq. (54), we obtain

$$\mathcal{G}_{ij}^H = -\frac{3a\left(x_i^A r_B^2 - x_i^B a^2\right)\left(x_j^B r_A^2 - x_j^A a^2\right)}{4\pi\left[r_A^2 r_B^2 - 2\mathbf{r}_A\cdot\mathbf{r}_B a^2 + a^4\right]^{5/2}}$$
$$+ \frac{a\left(2x_i^A x_j^B - \delta_{ij} a^2\right)}{4\pi\left[r_A^2 r_B^2 - 2\mathbf{r}_A\cdot\mathbf{r}_B a^2 + a^4\right]^{3/2}} + \frac{a x_i^A x_j^B}{4\pi r_A^3 r_B^3}. \quad (58)$$

Once more, we may evaluate the nonadditivity contribution to the interaction energy of the system in any of its configurations. As before, we write explicitly only the case when the atoms are collinear with the center of the sphere. Using Eq. (56) we obtain

$$E_{na}^{(1)} = \pm\frac{\Lambda_{AB}}{36\pi^2 \varepsilon_0^2 R_{AB}^3}\left[\frac{a r_A r_B}{(r_A r_B \pm a^2)^3} - \frac{a}{r_A^2 r_B^2}\right], \quad (59)$$

$$E_{na}^{(2)} = -\frac{\Lambda_{AB}}{144\pi^2 \varepsilon_0^2}\left[\left(\frac{a r_A r_B \mp a^3}{(r_A r_B \pm a^2)^3} - \frac{a}{r_A^2 r_B^2}\right)^2\right.$$
$$\left. + \frac{2a^6}{(r_A r_B \pm a^2)^6}\right]. \quad (60)$$

It can be seen in Fig. 7 that the nonadditivity effects are greatly diminished in comparison with the previous case, where the sphere was grounded. Letting $a \to 0$ in Eqs. (59) and (60), we see that the first contribution to $E_{na}^{(1)}$ ($E_{na}^{(2)}$) is of third (fourth) order in a. Therefore, keeping only the lowest-order contribution, we may write for the nonadditivity term of two atoms plus a very small sphere the expression

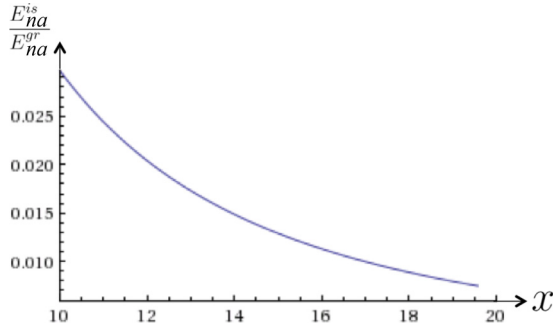$$E_{na} = -\frac{K\Lambda_{AB}}{R_{AB}^3 r_A^3 r_B^3}a^3 + O(a^4), \quad (61)$$

FIG. 7. (Color online) Ratio between the isolated sphere's and the grounded sphere's nonadditive contributions for the interaction energy. The horizontal axis stands for the distance of atom A to the center of the sphere (in units of the radius of the sphere). Atom B is farther from the sphere, separated by a distance $R_{AB} = 0.002a$ from atom A and collinear with it and the center of the sphere.

where K is a positive constant. In this limit we may identify the shrunk sphere as an atom with polarizability $\alpha(C)$ proportional to a^3. Also, r_A and r_B become the respective distances from atoms A and B to atom C. Therefore, we see that our result is compatible with the well-known Axilrod-Teller potential for three atoms, with the expected negative sign [6]. Note that such compatibility follows from the absence of the powers a^0, a, and a^2 in the above expansion, which did not happen for the grounded sphere in the previous section. This could be expected, since this limit on the grounded sphere can by no means be thought of as an atom. We may recast Eq. (61) into a more familiar form in the particular case of two-level atoms, where [39] Λ_{AB} is proportional to $\alpha(A)\alpha(B)$, leaving Eq. (61) as

$$E_{na} = -\frac{K\alpha(A)\alpha(B)\alpha(C)}{R^3 r_A^3 r_B^3} + O(a^4). \qquad (62)$$

V. CONCLUSION

We have dealt with two atoms in the presence of a conducting surface of an arbitrary shape. In systems composed of three bodies it is well known that nonadditivity effects must be taken into account. In the case of three atoms the effects

are rather small since they are of third order in perturbation theory while the additivity terms are of second order. When a conducting surface is present, however, the nonadditivity term is also of second order and may be relevant to the interaction between the atoms.

By mapping our quantum two-atom problems onto simpler electrostatic problems involving only one charge, we have obtained explicit analytical expressions for the influence of a conducting surface in the dispersion interaction of two atoms. We checked the self-consistency of our results by reobtaining London formula. In addition, for the case of two atoms and a conducting plane we also reobtained the result displayed in literature. Then we discussed our most important example of two atoms inside a plate capacitor, where the nonadditivity cannot be neglected. We showed that the nonadditivity shields one atom from the other, making the interaction between them fall exponentially with the distance. This effect is present also for N atoms between two infinite planes up to second order in perturbation theory. In such a way we concluded that a gas is closer to an ideal one between conducting planes, leading us to conjecture that the gas-liquid transition takes place at lower temperatures inside a plate capacitor than inside nonconducting plates. As a last example we treated two atoms in the presence of a conducting sphere, both grounded and isolated. We demonstrated that when the sphere is isolated the nonadditivity is much smaller than in the grounded case. The isolated case, however, has a nice particular limit, namely, the three-atom configuration, obtained when we let the radius of the sphere go to zero. We expect that the general and simple nature of the results enlisted in this paper allows for a broader understanding of nonadditivity effects in situations where the distances involved are small enough for a nonretarded treatment to be appropriate.

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