

Nuclear mass corrections to the Casimir-Polder interaction

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We present a derivation of the finite nuclear mass corrections to the Casimir-Polder interaction between two atomic systems in the ground state. Equivalently, we show how the long-range asymptotics of the adiabatic correction is modified due to the finite speed of light. We show that in addition to the contribution resulting from the finite-mass correction to atomic polarizabilities, a further contribution exists.

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I. INTRODUCTION

The finite nuclear mass corrections to the long-range retarded interactions between atomic systems have not yet been studied in the literature. For most atomic systems both the finite speed of light and the finite nuclear mass lead to small corrections to the interaction energy. Their nonadditive combination could be expected to be even smaller. Additionally, there has been no theoretical framework in which retardation and the adiabatic correction could be combined. While the Born-Oppenheimer (BO) potential at large distances is replaced by the Casimir-Polder (CP) interaction [1] which incorporates the retardation effects, it has not been known how the adiabatic correction to the BO potential is modified by these retardation effects. Here we present a complete derivation of the atom-atom interaction potential including both the retardation and finite nuclear mass effects. We demonstrate that besides the Casimir-Polder potential modified by finite-mass corrections to atomic polarizabilities, there exists an additional effect, which leads to a modification of the CP formula. The obtained results, together with the previously obtained relativistic corrections to the Casimir-Polder potential [2], have an application to the accurate description of long-range interactions between atomic systems. While we derive only the lowest-order results, the formalism we present can also be applied to higher orders of perturbation theory and to higher orders of the multipole expansion.

II. SEPARATION OF THE ATOMIC MASS CENTER

In order to derive the finite nuclear mass corrections to the long-range interaction, one has to separate out the total atomic coordinates from the internal degrees of freedom in the presence of the electromagnetic field. We follow here Ref. [3] and consider a general system of N nonrelativistic charged particles, electrons, and nuclei, placed in the electromagnetic field. For consistency we use the natural system of units ($\hbar = c = 1$), and the conversion to the atomic units is done using the Bohr radius $a_0 = 1/(m\alpha)$ and the Hartree energy $E_h = m\alpha^2$. The initial Hamiltonian of a nonrelativistic system of charged particles is

$$H = \sum_a \left(\frac{\vec{\pi}_a^2}{2m_a} + e_a A_a^0 \right) + \sum_{a>b} \frac{e_a e_b}{4\pi r_{ab}}, \quad (1)$$

where $\vec{\pi}_a = \vec{p}_a - e_a A(\vec{r}_a)$, and the summation goes over electrons and nuclei. We now introduce global variables, the

center of mass \vec{R} and the total momentum $\vec{\Pi}$,

$$\vec{R} = \sum_a \frac{m_a}{M} \vec{r}_a, \quad (2)$$

$$\vec{\Pi} = \sum_a [\vec{p}_a - e_a \vec{A}(\vec{R})] = \vec{P} - e_\Sigma \vec{A}(\vec{R}), \quad (3)$$

where $M = \sum_a m_a$ and $e_\Sigma = \sum_a e_a$, and the relative coordinates are

$$\vec{x}_a = \vec{r}_a - \vec{R}, \quad (4)$$

$$\vec{q}_a = \vec{p}_a - \frac{m_a}{M} \vec{P}, \quad (5)$$

such that

$$[x_a^i, q_b^j] = i \delta^{ij} \left(\delta_{ab} - \frac{m_b}{M} \right), \quad (6)$$

$$[R^i, P^j] = i \delta^{ij}, \quad (7)$$

$$[x_a^i, P^j] = [R^i, q_a^j] = 0. \quad (8)$$

Next, we perform a canonical transformation ϕ ,

$$H' = e^{-i\phi} H e^{i\phi} + \partial_t \phi, \quad (9)$$

with

$$\phi = \sum_a e_a \int_0^1 du \vec{x}_a \cdot \vec{A}(\vec{R} + u \vec{x}_a). \quad (10)$$

Assuming that the characteristic wavelength of the electromagnetic field is larger than the size of each of the interacting systems, ϕ can be expanded into multipoles:

$$\phi = \sum_a e_a \left[x_a^i A^i(\vec{R}) + \frac{1}{2!} x_a^i x_a^j \frac{\partial A^i(\vec{r})}{\partial r^j} \Big|_{\vec{r}=\vec{R}} + \dots \right]. \quad (11)$$

The scalar potential is transformed to

$$\sum_a e_a A_a^0 + \partial_t \phi = e_\Sigma A^0 - d^i E^i, \quad (12)$$

where d is the dipole moment

$$d^i = \sum_a e_a x_a^i, \quad (13)$$

$A^0 \equiv A^0(\vec{R})$, $\vec{E} \equiv \vec{E}(\vec{R})$, and $\vec{B} \equiv \vec{B}(\vec{R})$. The kinetic momentum is transformed to

$$e^{-i\phi} \pi_a^j e^{i\phi} = \tilde{\pi}_a^j + \frac{m_a}{M} \Pi^j, \quad (14)$$

where

$$\tilde{\pi}_a = \vec{q}_a + \frac{1}{2} \left(e_a \vec{x}_a + \frac{m_a}{M} \vec{d} \right) \times \vec{B}, \quad (15)$$

and the kinetic energy is

$$e^{-i\phi} \sum_a \frac{\pi_a^2}{2m_a} e^{i\phi} = \frac{\Pi^2}{2M} + \frac{\vec{\Pi}}{M} \cdot \vec{d} \times \vec{B} + \sum_a \frac{\tilde{\pi}_a^2}{2m_a}. \quad (16)$$

The total Hamiltonian after the transformation ϕ takes the form

$$H' = \sum_a \frac{\tilde{\pi}_a^2}{2m_a} + \sum_{a>b} \frac{e_a e_b}{4\pi r_{ab}} + \frac{\vec{\Pi}^2}{2M} + e_\Sigma A^0 - \vec{d} \cdot \vec{E} + \vec{d} \cdot \vec{B} \times \frac{\vec{\Pi}}{M}. \quad (17)$$

From now on we assume that the external magnetic field and the total charge both vanish ($\vec{B} = 0$, $e_\Sigma = 0$), whereupon the

transformed Hamiltonian simplifies to

$$H' = \sum_a \frac{\vec{q}_a^2}{2m_a} + \sum_{a>b} \frac{e_a e_b}{4\pi r_{ab}} + \frac{\vec{P}^2}{2M} - \vec{d} \cdot \vec{E}. \quad (18)$$

III. EFFECTIVE HAMILTONIAN APPROACH

Let's assume for a moment that this atomic system is placed in the inhomogeneous electric field $\vec{E}(\vec{R})$, and we would like to find out an effective wave function for the center-of-mass motion \vec{R} . Because of the $\vec{d} \cdot \vec{E}(\vec{R})$ coupling, the center-of-mass motion cannot be completely separated from internal degrees of freedom, and therefore one cannot uniquely define this effective wave function. Consequently, the effective Hamiltonian is not unique, and we will use this freedom to transform it to its simplest possible form. Let us return now to the main topic of this work, which is the long-range interaction between atomic systems. The effective Hamiltonian H_{eff} is obtained by taking the matrix element of the resolvent between the ground atomic states,

$$\frac{1}{E - H_{\text{eff}}} = \langle \phi_A, \phi_B, 0 | \frac{1}{E + E_A + E_B - H_A - \frac{\vec{p}_A^2}{2M_A} - H_B - \frac{\vec{p}_B^2}{2M_B} - \vec{d}_A \cdot \vec{E}(\vec{r}_A) - \vec{d}_B \cdot \vec{E}(\vec{r}_B) - H_{EM}} | 0, \phi_A, \phi_B \rangle, \quad (19)$$

where H_{EM} is the Hamiltonian for the electromagnetic field and $|0\rangle$ is the electromagnetic vacuum state. E_A , E_B are the ground-state energies, ϕ_A , ϕ_B are the corresponding wave functions, and M_A , M_B are the total masses of atoms A and B . H_A , H_B are the internal Hamiltonians of atoms A and B , respectively. The first two terms of Eq. (18) are

$$H_A = \sum_a \frac{\vec{q}_a^2}{2m_a} + \sum_{a>b} \frac{e_a e_b}{4\pi r_{ab}} \quad (20)$$

$$= \sum_a' \frac{\vec{q}_a^2}{2m} + \frac{(\sum_a' \vec{q}_a)^2}{2m_A} + \sum_{a>b} \frac{e_a e_b}{4\pi r_{ab}}, \quad (21)$$

where \sum_a' denotes the sum over only electrons and m_A and m_B are masses of nuclei A and B , respectively.

One calculates H_{eff} by taking the power series of both sides of Eq. (19) in the fine-structure constant $\alpha = e^2/(4\pi)$. In the zeroth order

$$H_{\text{eff}}^{(0)} = \frac{\vec{P}_A^2}{2M_A} + \frac{\vec{P}_B^2}{2M_B}. \quad (22)$$

Neglecting the self-interaction, the leading correction comes from the two-photon exchange,

$$\frac{1}{E - H_{\text{eff}}} = \frac{1}{E - H_{\text{eff}}^{(0)}} + \frac{1}{E - H_{\text{eff}}^{(0)}} H_{\text{eff}}^{(2)}(E) \frac{1}{E - H_{\text{eff}}^{(0)}}, \quad (23)$$

where

$$H_{\text{eff}}^{(2)}(E) = \langle \phi_A, \phi_B, 0 | [\vec{d}_A \cdot \vec{E}(\vec{r}_A) + \vec{d}_B \cdot \vec{E}(\vec{r}_B)] \frac{1}{E - H_0} [\vec{d}_A \cdot \vec{E}(\vec{r}_A) + \vec{d}_B \cdot \vec{E}(\vec{r}_B)] \frac{1}{E - H_0} [\vec{d}_A \cdot \vec{E}(\vec{r}_A) + \vec{d}_B \cdot \vec{E}(\vec{r}_B)] \times \frac{1}{E - H_0} [\vec{d}_A \cdot \vec{E}(\vec{r}_A) + \vec{d}_B \cdot \vec{E}(\vec{r}_B)] | 0, \phi_A, \phi_B \rangle \quad (24)$$

and where

$$H_0 = H_A - E_A + \frac{\vec{P}_A^2}{2M_A} + H_B - E_B + \frac{\vec{P}_B^2}{2M_B} + H_{EM}. \quad (25)$$

The main issue is the elimination of dependence of H_{eff} on E . This is achieved as follows. The atomic excitation energy is the largest among all the energy scales, so one can expand the matrix element in Eq. (24) in powers of $E - P_A^2/(2M_A) - P_B^2/(2M_B) = E - H_{\text{eff}}^{(0)}$. At the zeroth order, it is the well-known Casimir-Polder interaction V_{CP} . We will be interested in the first-order term in this expansion, which can be represented as

$$H_{\text{eff}}^{(2)}(E) = V_{\text{CP}} + V'_{\text{CP}} + (E - H_{\text{eff}}^{(0)}) Q + Q (E - H_{\text{eff}}^{(0)}), \quad (26)$$

with some Hermitian operator Q . The last two terms do not change the position of a pole of the resolvent of the effective Hamiltonian

$$\frac{1}{E - H_{\text{eff}}} = \frac{1}{E - H_{\text{eff}}^{(0)}} + Q \frac{1}{E - H_{\text{eff}}^{(0)}} + \frac{1}{E - H_{\text{eff}}^{(0)}} Q \quad (27)$$

and only change the residuum, namely, $|\phi\rangle\langle\phi|$, where ϕ is the effective (unnormalized) wave function for the center of mass of each atom A and B . As we have already emphasized, this wave function is not a well-defined quantity, so we can change its definition to simplify the effective Hamiltonian. We therefore define a new effective wave function

$$\phi' = (1 - Q)\phi, \quad (28)$$

for which the stationary Schrödinger equation takes the form

$$(E - H'_{\text{eff}})\phi' = 0, \quad (29)$$

with the new effective Hamiltonian

$$H'_{\text{eff}} = \frac{P_A^2}{2M_A} + \frac{P_B^2}{2M_B} + V_{\text{CP}} + V'_{\text{CP}}, \quad (30)$$

which does not depend on energy and includes the Casimir-Polder potential V_{CP} and an additional correction V'_{CP} due to the finite atomic masses.

IV. CP INTERACTION WITH THE FINITE NUCLEAR MASS

The leading Casimir-Polder interaction V_{CP} comes from the two-photon exchange with the interaction $\vec{d} \cdot \vec{E}$. It is given by Eq. (24), but it can also be derived from the scattering amplitude [4,5] using Feynman diagrams. Two diagrams for two-photon exchange (Fig. 1) contribute, and

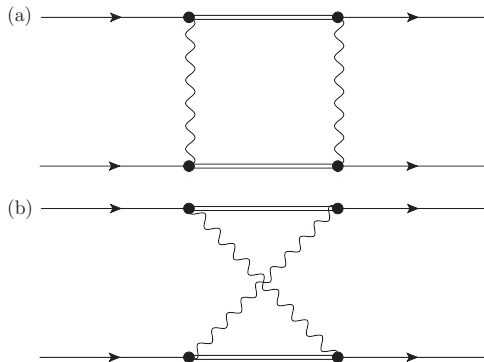


FIG. 1. The only Feynman diagrams contributing, in the lowest order, to the atom-atom interaction energy.

the corresponding expression for the energy is

$$V_{\text{CP}}(R) = -\frac{1}{2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \alpha_A^{ik}(\omega) \alpha_B^{jl}(\omega) g^{ij}(\vec{R}) g^{kl}(\vec{R}), \quad (31)$$

where

$$g^{ij}(\vec{R}) = \int \frac{d^3k}{(2\pi)^3} e^{i\vec{k}\cdot\vec{R}} \frac{(\omega^2 \delta^{ik} - k^i k^k)}{\omega^2 - k^2}, \quad (32)$$

$$\alpha_A^{ij}(\omega) = -\left\langle d^i \frac{1}{E_A - H_A + \omega} d^j + d^j \frac{1}{E_A - H_A - \omega} d^i \right\rangle, \quad (33)$$

where R is the distance between mass centers of atoms A and B and H is the Hamiltonian from Eq. (21).

The ω integration in Eq. (31) is assumed along the Feynman contour. If this integration is performed, the resulting expression corresponds to the standard Rayleigh-Schrödinger perturbation theory but becomes much more lengthy. Instead, this integration contour is deformed to the imaginary axis by the replacement $\omega = i\lambda$. The k integral leads to

$$\begin{aligned} g^{ij}(\vec{R}) &= \left(\lambda^2 \delta^{ij} - \frac{\partial^2}{\partial R^i \partial R^j} \right) \frac{e^{-\lambda R}}{4\pi R} \\ &= \frac{e^{-\lambda R}}{4\pi R^3} \left[\delta^{ij} (\lambda^2 R^2 + \lambda R + 1) \right. \\ &\quad \left. - \frac{R^i R^j}{R^2} (\lambda^2 R^2 + 3\lambda R + 3) \right]. \end{aligned} \quad (34)$$

For atomic and molecular states with spherically symmetric polarizability tensors $\alpha_A^{ij} = \delta^{ij} \alpha_A$, $\alpha_B^{ij} = \delta^{ij} \alpha_B$ the CP formula simplifies to

$$\begin{aligned} V_{\text{CP}}(R) &= -\frac{1}{16\pi^3} \int_0^\infty d\lambda \alpha_A(i\lambda) \alpha_B(i\lambda) \frac{\lambda^4 e^{-2\lambda R}}{R^2} \\ &\quad \times \left(1 + \frac{2}{\lambda R} + \frac{5}{(\lambda R)^2} + \frac{6}{(\lambda R)^3} + \frac{3}{(\lambda R)^4} \right). \end{aligned} \quad (35)$$

This result for infinite nuclear masses has been derived by Casimir and Polder [1]. At short distances, the potential simplifies to $-C_6/R^6$,

$$V_{\text{CP}}(R) \approx -E_h \left(\frac{a_0}{R} \right)^6 C_6, \quad (36)$$

$$\begin{aligned} C_6 &= \frac{3}{\pi} \frac{1}{(4\pi a_0^3)^2} \int_0^\infty d\lambda \alpha_A(i\lambda) \alpha_B(i\lambda) \\ &= \frac{2}{3} \frac{E_h}{a_0^4} \left\langle d_A^i d_B^j \frac{1}{H_A - E_A + H_B - E_B} d_A^i d_B^j \right\rangle. \end{aligned} \quad (37)$$

For large distances, the potential depends on only the static polarizabilities,

$$V_{\text{CP}}(R) \approx -E_h \left(\frac{a_0}{R} \right)^7 K_7, \quad (38)$$

$$K_7 = \frac{27}{4\pi\alpha} \frac{\alpha_A(0) \alpha_B(0)}{(4\pi a_0^3)^2}. \quad (39)$$

The finite nuclear masses enter through the second term in Eq. (21). For the pair of hydrogen atoms, this term leads to

the reduced-mass ($1/\mu = 1/m_e + 1/M$) scaling of the atomic polarizability,

$$\alpha_H(\omega) = \left(\frac{m_e}{\mu}\right)^3 \alpha_H\left(\frac{\mu}{m_e}\omega\right)\Big|_{M=\infty}, \quad (40)$$

and the leading-order coefficient of the small- R and large- R asymptotic expansions,

$$C_6^H = \left(\frac{m_e}{\mu}\right)^5 C_6^H\Big|_{M=\infty}, \quad K_7^H = \left(\frac{m_e}{\mu}\right)^6 K_7^H\Big|_{M=\infty}.$$

This result was first obtained by Dalgarno and McCarroll [6]. The next terms in the $1/R$ expansion of the adiabatic correction to Born-Oppenheimer energies were considered by Marinescu and Dalgarno [7], but in their derivation, only part of the higher-order terms has been included. The complete expressions for the coefficients of the $1/R^6$, $1/R^8$, and $1/R^{10}$ terms have been derived by Przybytek and Jeziorski [8], and their results are in agreement with the direct numerical calculation of the adiabatic correction performed in Ref. [9]. It should be pointed out that the adiabatic correction is usually computed as a function of the distance between nuclei and not of the distance between atomic mass centers. This difference leads to different finite-mass corrections for higher-order terms in the $1/R$ expansion of the adiabatic energy, like that for the A_8 coefficient [8].

The finite nuclear mass correction due to the atomic kinetic energy can be obtained in an analogous way. Let us include at the beginning the finite mass of only atom A and consider the corresponding scattering amplitude

$$V'_{\text{CP}}(R) = -\frac{1}{2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \beta_A^{ik}(\omega) \alpha_B^{jl}(\omega) \times g^{ij}(\vec{R}) \left(E_A^{\text{kin}} - \frac{P_A^2}{2M_A}\right) g^{kl}(\vec{R}), \quad (41)$$

where E_A^{kin} is the kinetic energy, \vec{P}_A is the momentum operator of atom A , and

$$\beta_A^{ij}(\omega) = \left\langle d^i \frac{1}{(E_A - H_A + \omega)^2} d^j + d^j \frac{1}{(E_A - H_A - \omega)^2} d^i \right\rangle. \quad (42)$$

Due to the symmetry of α^{ij} and β^{ij} in Cartesian indices only the symmetric (in the simultaneous interchange of i, k and j, l indices) component of the integrand contributes to the integral (41), and thus we can use the following identity:

$$\begin{aligned} & g^{ij}(\vec{R}) \left(E_A^{\text{kin}} - \frac{P_A^2}{2M_A}\right) g^{kl}(\vec{R}) + \left(\begin{matrix} i \leftrightarrow k \\ j \leftrightarrow l \end{matrix}\right) \\ &= \left[g^{ij}(\vec{R}), \left[E_A^{\text{kin}} - \frac{P_A^2}{2M_A}, g^{kl}(\vec{R})\right] \right] \\ &+ \left(E_A^{\text{kin}} - \frac{P_A^2}{2M_A}\right) g^{ij}(\vec{R}) g^{kl}(\vec{R}) \\ &+ g^{ij}(\vec{R}) g^{kl}(\vec{R}) \left(E_A^{\text{kin}} - \frac{P_A^2}{2M_A}\right). \end{aligned}$$

Since it is the on-shell scattering amplitude, the last two terms in Eq. (43) automatically vanish, and the remainder becomes

$$\left[g^{ij}(\vec{R}), \left[E_A^{\text{kin}} - \frac{P_A^2}{2M_A}, g^{kl}(\vec{R})\right] \right] = -\frac{1}{M_A} \frac{\partial g^{ij}}{\partial R^n} \frac{\partial g^{kl}}{\partial R^n}, \quad (43)$$

and V'_{CP} becomes

$$V'_{\text{CP}}(R) = \frac{1}{4} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \frac{\partial g^{ij}}{\partial R^n} \frac{\partial g^{kl}}{\partial R^n} \times \left(\frac{\beta_A^{ik}(\omega) \alpha_B^{jl}(\omega)}{M_A} + \frac{\alpha_A^{ik}(\omega) \beta_B^{jl}(\omega)}{M_B} \right). \quad (44)$$

Assuming the spherical symmetry of the α^{ij} and β^{ij} tensors of both atoms, the potential $V'_{\text{CP}}(R)$ simplifies to

$$\begin{aligned} V'_{\text{CP}}(R) &= -\frac{1}{32\pi^3} \int_0^\infty d\lambda \left[\frac{1}{M_A} \beta_A(i\lambda) \alpha_B(i\lambda) \right. \\ &+ \left. \frac{1}{M_B} \alpha_A(i\lambda) \beta_B(i\lambda) \right] \\ &\times \frac{\lambda^6 e^{-2\lambda R}}{R^2} \left(1 + \frac{4}{\lambda R} + \frac{14}{(\lambda R)^2} + \frac{42}{(\lambda R)^3} \right. \\ &+ \left. \frac{81}{(\lambda R)^4} + \frac{90}{(\lambda R)^5} + \frac{45}{(\lambda R)^6} \right). \quad (45) \end{aligned}$$

The leading term in the small- R expansion starts with R^{-8} and takes on a form dependent on the reduced mass of the nuclei of both interaction atoms,

$$V'_{\text{CP}}(R) \approx -E_h \left(\frac{m_e}{M_A} + \frac{m_e}{M_B} \right) \left(\frac{a_0}{R} \right)^8 C'_8, \quad (46)$$

$$\begin{aligned} C'_8 &= \frac{45}{\pi} \frac{1}{(4\pi a_0^3)^2} \int_0^\infty d\lambda \alpha_A(i\lambda) \beta_B(i\lambda) \\ &= 5 \frac{E_h^2}{a_0^4} \left\langle d_A^i d_B^j \frac{1}{(H_A - E_A + H_B - E_B)^2} d_A^i d_B^j \right\rangle. \quad (47) \end{aligned}$$

This result differs from A_8 in Ref. [8], which is a consequence of not including the higher-order (dipole-quadrupole) term and of a different definition of R . In our work, the interaction potential is a function of the distance between the atomic mass centers, while in Ref. [8], it is a function of the distance between the atomic nuclei. In the large- R limit V'_{CP} decays as R^{-9} ,

$$V'_{\text{CP}}(R) \approx -\frac{837}{16\pi\alpha} \left(\frac{m_e}{M_A} \right) \frac{\beta_A(0) \alpha_B(0)}{(4\pi a_0^3)^2} \left(\frac{a_0}{R} \right)^9 - (A \leftrightarrow B). \quad (48)$$

One may observe that for atomic systems, where the typical excitation energy is of the order of E_h and the typical dipole moment matrix elements are of the order of ea_0 , $V'_{\text{CP}}(R)$ is smaller than $V_{\text{CP}}(R)$ by a factor of the order of

$$V'_{\text{CP}}(R) \sim \frac{m_e}{M} \left(\frac{a_0}{R} \right)^2 V_{\text{CP}}(R), \quad (49)$$

so it is not expected to be a significant correction. For example, in the case of two interacting hydrogen atoms the ratio between

$V'_{\text{CP}}(R)$ and a finite-mass correction to $V_{\text{CP}}(R)$ (both being of the order m_e/M) varies from $1.707630747(a_0/R)^2$ for small distances to $12.34259259(a_0/R)^2$ in the retardation limit.

V. SUMMARY

This work resolves the problem of the retardation in the adiabatic correction for diatomic molecules. Namely, the adiabatic correction to the Born-Oppenheimer energy, at large distances, is being replaced by the sum of two terms: the finite nuclear mass corrections in the Casimir-Polder potential $V_{\text{CP}}(R)$ through reduced mass and mass polarization

corrections in the atomic matrix elements and an additional contribution $V'_{\text{CP}}(R)$. We also demonstrate that in typical cases $V'_{\text{CP}}(R)$ is much smaller than the finite-mass correction to $V_{\text{CP}}(R)$ and the ratio of these two contributions behaves as $1/R^2$ for large interatomic distances.

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