On the impact of dissipation on dispersion interactions between two atoms

Pablo Barcellona¹, Roperto Passante², Lucia Rizzuto², and Stefan Yoshi Buhmann¹,³

¹Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany
²Dipartimento di Fisica e Chimica, Università degli Studi di Palermo and CNISM, Via Archirafi 36, I-90123, Palermo, Italy
³Freiburg Institute for Advanced Studies, Albert-Ludwigs-Universität Freiburg, Albertstr. 19, 79104 Freiburg, Germany

Abstract

We consider the dispersion interaction between two neutral, ground-state atoms at finite temperature by means of a dynamical approach. Our result differs from those obtained with time-independent perturbation theory in that it accounts for the influence of dissipation via the atomic decay rates. Modern measurements of Casimir force seem to only be compatible with Lifshitz theory when neglecting dissipation in the medium response function, which has led heated debates. Our new result offers an explanation for the suppressed dependence on dissipation.

We also consider the interaction between a ground-state atom and an excited one. There are discordant results in the literature for the retarded potential: one oscillating and one monotonous. Our dynamical result uniquely leads to the oscillating result when taking into account the decay rates.

1 Introduction

Casimir and Casimir-Polder forces are electromagnetic interactions between neutral macroscopic bodies or atoms due to the quantum fluctuations of the electromagnetic field [1 2 3]. They are responsible for many phenomena in physics, chemistry and biology: the deviation from ideal gas behavior in non-polar gases [4], latent heat of liquids, capillary attraction, physical absorption, and the cell adhesion [5]. Dispersion interactions also contribute to the total binding energy of liquids and solids and they have played an important role during the early stages of planet formation [6]. The dispersion interaction between two ground-state atoms at zero temperature shows a $R^{-6}$ dependence for distances much smaller than the wavelengths of atomic electronic transitions,
and $R^{-7}$ for much larger distances. The interaction energy at arbitrary distances may be expressed as an integral over imaginary frequency $\xi$:

$$U_{T=0} (r_A, r_B) = -\frac{\hbar \mu_0^2}{2\pi} \int_0^\infty d\xi \xi^4 \alpha_A^0 (i\xi) \alpha_B^0 (i\xi) \text{Tr} \{G(r_A, r_B, i\xi) G(r_B, r_A, i\xi)\}$$  \hspace{1cm} (1)$$

where

$$\alpha_A^0 (\omega) = \frac{1}{3\hbar} \lim_{\epsilon \to 0^+} \sum_k \left( \frac{|d_{A0}^A|^2}{\omega_{A0}^k - \omega - i\epsilon} + \frac{|d_{A0}^A|^2}{\omega_{A0}^k + \omega + i\epsilon} \right)$$  \hspace{1cm} (2)$$

is the isotropic ground-state polarizability of a non-absorbing atom, $G$ the Green tensor and Tr the trace. This expression, obtained with time-independent perturbation theory, is not quite satisfactory because the polarizabilities do not depend on the dissipative properties of the atoms. The difficulty was overcome with the help of the linear response theory, where the Casimir interaction between two dielectric half-spaces is evaluated [7]; the two-atom dispersion interaction is obtained in the dilute-gas limit and its expression depends on dissipation via the line-widths or decay rates of the excited states:

$$\alpha_A (\omega) = \frac{1}{3\hbar} \sum_k \left( \frac{|d_{A0}^A|^2}{\omega_{A0}^k - \omega - i\Gamma_k/2} + \frac{|d_{A0}^A|^2}{\omega_{A0}^k + \omega + i\Gamma_k/2} \right)$$  \hspace{1cm} (3)$$

The macroscopic Casimir force between a metal sphere and a metal plate has been measured [8]. Theoretically, this force is traditionally described by Lifshitz theory [9] where the dielectric permittivity plays the role that the atomic polarizability does for the microscopic Casimir–Polder force. Again, one is faced with the question of whether and how to account for dissipation in the response functions. One possibility is the Drude model which accounts includes a finite line width and hence accounts for dissipation in a natural way. However, the experimental measurements seem to agree with the plasma limit of this model where this line width is set to zero. This presented a cause for heated debate, because the plasma model is an approximation of the Drude model which neither takes into account the energy losses of light in the material nor does it exhibit a finite static conductivity. Our new model features a suppressed dependence of the Casimir–Polder force on dissipation and hence, by extrapolation to the macroscopic case, suggests a possible explanation of the agreement of the observed Casimir force with the plasma model. Note that further investigations are needed to include many-body effects.

A number of studies have considered the closely related Casimir-Polder dispersion interaction between a ground-state atom and an excited atom at zero temperature [10, 11, 12]. The interaction in this case is different from the interaction between two ground-state atoms because of the presence of a resonant term which is due to the possible emission of a real photon. As a consequence, the force between the two atoms can be attractive or repulsive in contrast to the force between ground-state atoms which is always attractive. This is easily seen for the well-understood non-retarded regime,
that is, for distances much smaller than the wavelength of atomic electronic transitions:

\[ U_{\text{non-ret}, T=0} (r) = - \frac{1}{24\pi^2 \varepsilon_0 r^6} \sum_{n,j} |d_{0n}|^2 |d_{kl}^B|^2 E_{n0}^A - E_{kl}^B \]

(4)

where \( |k\rangle \) is the state of the excited atom B. For downward transitions, \( E_{n0}^A - E_{kl}^B \) can be negative and the resulting energy positive.

The interaction at larger separations has been subject to controversies. In a first group of works, it was found that the retarded potential oscillates as a function of interatomic distance \[13, 14, 15\]. In a later group of publications it was found that the retarded potential is non-oscillatory and proportional to \( 1/r^2 \) \[11, 16, 17\]. The two conflicting results are due to subtle differences in treating divergent energy denominators. The diverging energy denominators can be treated by using the principal value prescription leading to the oscillating result, or by adding an infinitesimal factor, resulting in the non-oscillating result. The two contradictory results are both valid from a mathematical point of view and the possible oscillatory behavior of the dispersion interaction remains an open question. In our model, the energy denominators are not diverging because of the presence of the finite line widths and our model can give a conclusive answer to these controversies. We will demonstrate agreement with the oscillatory result, which is consistent with recent measurements of the related, spatially oscillating Casimir–Polder between an excited barium ion and a mirror \[18, 19\].

Our article is organised as follows. In the Section 2, we will consider the dynamics of two atoms and the medium-assisted field, mutually coupled. In the subsequent Section 3, we will consider the Casimir–Polder energy between two atoms at finite temperature. Finally, we will consider the case where an excited atom interacts with a ground-state atom in a zero-temperature environment (Section 4).

2 Atom–field dynamics

We consider the mutually coupled evolution of two atoms and the medium-assisted field at thermal equilibrium (temperature \( T \)). The dynamics of the atoms can be described with time-dependent atomic density matrices \( \sigma^A, \sigma^B \), which can be expanded in terms of internal-energy eigenstates:

\[ \sigma^A = \sum_{m,n} \sigma_{mn}^A |m^A\rangle \langle n^A|. \]  

(5)

The atomic flip operators are defined by \( A_{mn} = |m^A\rangle \langle n^A| \) and \( B_{pq} = |p^B\rangle \langle q^B| \), respectively. The diagonal elements \( \sigma_{nn} \) represent the probabilities of the atom to be in the respective internal state \( |n^A\rangle \).

In order to evaluate the force between the two atoms we must first solve the atom-field dynamics to obtain the flip operators and the field operators in the Heisenberg picture. The total Hamiltonian is the sum of three terms, the atomic and the field Hamiltonian.
and the interaction term \[20]:

\[ H = H_A + H_F + H_{AF}, \]  

\[ H_A = \sum_n E_n^A A_{nn} + \sum_n E_n^B B_{nn} \quad H_F = \sum_{\lambda=e,m} \int d^3 r \int_0^\infty d\omega \omega f_{\lambda}^\dagger (r, \omega) \cdot f_{\lambda} (r, \omega), \]  

\[ H_{AF} = -d^A \cdot E (r_A) - d^B \cdot E (r_B) \]  

where \( f_{\lambda} (r, \omega) \) is the annihilation operator for the elementary electric and magnetic excitations of the system:

\[ E (r) = \int_0^\infty d\omega E (r, \omega) + h.c. \]

\[ E (r, \omega) = \sum_{\lambda=e,m} \int d^3 r' G_{\lambda} (r, r', \omega) \cdot f_{\lambda} (r', \omega) \]

\[ G_e (r, r', \omega) = \frac{i \omega^2}{c^2} \sqrt{\frac{\hbar}{\pi \varepsilon_0}} \text{Im} \varepsilon (r', \omega) \mathbf{G} (r, r', \omega) \]

\[ G_m (r, r', \omega) = \frac{i \omega}{c} \sqrt{\frac{\hbar}{\pi \varepsilon_0}} \text{Im} \mu (r', \omega) \frac{1}{|\mu (r', \omega)|} (\nabla \times \mathbf{G} (r', r, \omega))^T \]

The presence of an arbitrary arrangement of magnetoelectric bodies is taken macroscopically into account via the Green tensor \( \mathbf{G} \).

The Heisenberg equations of motion for the flip operators and the electric field are:

\[ \dot{A}_{mn} (t) = \frac{1}{i\hbar} [A_{mn} (t), H (t)] = i \omega^A_{mn} A_{mn} (t) + \frac{i}{\hbar} K^A_{mn} (t) \cdot \mathbf{E} (r_A, t) \]

\[ \dot{\mathbf{E}} (r, \omega, t) = \frac{1}{i\hbar} [\mathbf{E} (r, \omega, t), H (t)] = -i \omega \mathbf{E} (r, \omega, t) + \frac{i\mu_0}{\pi} \omega^2 \text{Im} \mathbf{G} (r, r_A, \omega) \cdot d^A (t) \]

\[ + \frac{i\mu_0}{\pi} \omega^2 \text{Im} \mathbf{G} (r, r_B, \omega) \cdot d^B (t) \]

where

\[ K^A_{mn} (t) = \sum_k \langle A_{mk} (t) d_{nk} - A_{kn} (t) d_{km} \rangle = [A_{mn} (t), d (t)]. \]

In the atomic Heisenberg equations the first terms describe the free evolution, while the second terms describe the effect of the interaction with the electric field. From these equations is clear that the fluctuating dipoles \( d_A \) and \( d_B \) are the source of the electric field and that the electric field is the source of the fluctuating dipoles.

The electric field at the position of atom A position consists of two terms: the radiation reaction and the field due to the other atom B. As shown in the literature \[21\], the self
term $i\omega^2\text{Im} \mathbf{G}(r_A, r_A, \omega) d^4$, or the radiation reaction, gives rise to frequency shifts and radiative decays for the atom A:

$$\langle A_{mn} \rangle = \left[ i\hat{\omega}_{mn} - \left( \Gamma^A_n + \Gamma^A_m \right) / 2 \right] \langle A_{mn} \rangle + \frac{i}{\hbar} \langle \mathbf{K}^A_{mn} \cdot \mathbf{E}_A(\mathbf{r}_A, t) \rangle$$

where the expectation value is taken over the field thermal state. The Lamb-shift and the decay rates are given for arbitrary environment by these formulas:

$$\tilde{\omega}_{mn} = \omega_{mn} + \delta \omega_m - \delta \omega_n$$

$$\delta \omega_n = -\frac{\mu_0}{\pi \hbar} \sum_k P \int_0^\infty d\omega \frac{1}{\omega - \tilde{\omega}_{mk}} d^A_{mk} \cdot \text{Im} \mathbf{G}(\mathbf{r}_A, \mathbf{r}_A, \omega)$$

$$\Gamma^A_n = 2\frac{\mu_0}{\hbar} \sum_{k<n} \tilde{\omega}_{mk}^2 d^A_{mk} \cdot \text{Im} \mathbf{G}(\mathbf{r}_B, \mathbf{r}_A, \tilde{\omega}_{mk})$$

The Lamb shift is due the emission and absorption of a virtual photon by the same atom and the decay rate is due to the possible emission of a real photon from an excited state.

We define $\mathbf{E}_A(\mathbf{r}_A, \omega, t)$ to be the electric field without self interactions due to atom A; it is due only to the fluctuating dipole B and has two contributions: the free field and the source field of the atom B. We can formally integrate the Heisenberg equations, from the initial time $t_0$ at which the atoms and the field are uncoupled:

$$\langle A_{mn} (t) \rangle = f^A_{mn} (t - t_0) \langle A_{mn} (t_0) \rangle + \frac{i}{\hbar} \int_{t_0}^t dt_2 f^A_{mn} (t - t_2) \langle \mathbf{K}^A_{mn} (t_2) \cdot \mathbf{E}_A(\mathbf{r}_A, t_2) \rangle$$

$$\mathbf{E}_A(\mathbf{r}_A, \omega, t) = e^{-i\omega(t-t_0)} \mathbf{E}^{(0)}(\mathbf{r}_A, \omega, t_0) + \frac{i\mu_0}{\pi} \omega^2 \int_{t_0}^t dt_2 e^{-i\omega(t-t_2)} \text{Im} \mathbf{G}(\mathbf{r}_A, \mathbf{r}_B, \omega) \cdot d^B(t_2)$$

$$\mathbf{E}_B(\mathbf{r}_B, \omega, t) = e^{-i\omega(t-t_0)} \mathbf{E}^{(0)}(\mathbf{r}_B, \omega, t_0) + \frac{i\mu_0}{\pi} \omega^2 \int_{t_0}^t dt_2 e^{-i\omega(t-t_2)} \text{Im} \mathbf{G}(\mathbf{r}_B, \mathbf{r}_A, \omega) \cdot d^A(t_2)$$

where:

$$f^A_{mn} (t) = e^{[i\omega_{mn} - (\Gamma^A_n + \Gamma^A_m)/2]t}$$

The flip operator is the sum of two terms: the free flip operator and the induced flip operator due to $\mathbf{E}_A(\mathbf{r}_A, t_2)$. Similarly, the electric field is the sum of the vacuum field, as it would be in absence of the atoms, and the induced field due to both atoms. The atomic and field systems depend on their history because of their coupling.

Let us consider in detail the equation of the electric field. We assume that the dynamics of the atomic flip operators is dominated by oscillations with frequencies $\omega_{mn}$: $d^A(t_2) = $
This means that the following memory kernel appears in the electric-field expression:

$$K(t - t_2) = \int_0^\infty d\omega \omega^2 \text{Im} G(r_A, r_B, \omega) e^{-i(\omega - \omega_A)(t - t_2)}$$

(23)

The factor $1/2$ arises because the dipole moment is induced. This expression can be generalised to an atom with its frequency-dependent polarisability [22]. The interaction energy between one non-absorbing atom $A$ and the medium, without any other atom, is described by the Casimir-Polder energy:

$$U(r_A) = -\frac{1}{2} \int_0^\infty \langle \left( E^{(0)}(r_A, \omega, t) + E^{(0)\dagger}(r_A, \omega, t) \right) \cdot \alpha_0^0(\omega) \cdot E^{(0)}(r_A, t) \rangle$$

(25)

where the expectation value is taken over the field thermal state and $E^{(0)}(r_A, t)$ is the free field.

The energy is proportional to the electric-field fluctuations in the thermal state. $\alpha_0^0$ is the thermal polarizability of the non-absorbing atom:

$$\alpha_0^0(\omega) = \frac{1}{\hbar} \lim_{\epsilon \to 0^+} \sum_n p_n A \sum_k \left( \frac{d^A_{nk} \otimes d^A_{kn}}{\tilde{\omega}_{kn}^{A} + \omega + i\epsilon} + \frac{d^A_{nk} \otimes d^A_{kn}}{\tilde{\omega}_{kn}^{A} - \omega - i\epsilon} \right)$$

In this formula $p_n A = \frac{e^{-E_n^A/kBT}}{Z}$ is the thermal population of the state $|n^A\rangle$, and $\tilde{\omega}_{kn}^{A}$ is the shifted frequency because of the Lamb-shift. We also assumed the atomic Hamiltonian...
to be time-reversal invariant, so that the dipole-matrix elements are real \(d_{kn} = d_{nk}\).

Using the free field fluctuation equations \[49\], we obtain the known Casimir-Polder energy:

\[
U(r_A) = \mu_0 k_B T \sum_{j=0}^{\infty} \xi_j^2 \text{Tr} \left\{ \alpha_0^0 (i\xi_j) G(r_A, r_A, i\xi_j) \right\}
\]

(27)

where \(\xi_j = \frac{2\pi k_B T}{\hbar} j\), \(j = 0, 1, 2, \ldots\) are the Matsubara frequencies. The prime at the Matsubara sum indicates the term \(j = 0\) carries half-weight. This formula agrees with the literature result of the CP-energy of an atom in the thermal state. Our approach hence generalizes the effective Hamiltonian from the literature to finite temperature \[23\].

We now introduce a second atom B and calculate the Casimir-Polder energy in the presence of the atom B. In order to obtain the Casimir interaction between the two atoms, we first determine the electromagnetic field emitted by the atom B, \(E_A(r_A, t)\), and then we evaluate the interaction energy of the atom A with this field:

\[
U = U(r_A) + U(r_A, r_B)
\]

\[
= -\frac{1}{2} \int_0^\infty \left\langle \left( E_A(r_A, \omega, t) + E_A^\dagger(r_A, \omega, t) \right) \cdot \alpha_A^0(\omega) \cdot E_A(r_A, t) \right\rangle
\]

(28)

where the expectation value is taken over atomic and field thermal states.

The emission of a photon by one of the atoms is necessarily accompanied by a transition to a new state; each atom must absorb one photon and emit another photon to return in the same state; the leading order approximation involves four interaction vertices or the exchange of two photons. Exchange of more than two photons leads to many-atom forces which we neglect here. The final expression of the energy must contain two Green tensors of different frequencies, because involves the exchange of only
two photons. The resulting two-atom energy, for the assumed weak coupling between atoms and field, is

\[ U(\mathbf{r}_A, \mathbf{r}_B) = -\frac{\mu_0^2}{2\pi^2} \sum_n \sum_k p_n^B \int_0^\infty d\omega' \int_0^\infty d\omega'' \omega'' \omega' \times \]

\[ \times d_{nk}^B \cdot \text{Im} \mathbf{G} (\mathbf{r}_B, \mathbf{r}_A, \omega) \cdot \alpha_A^0 (\omega) \cdot \text{Im} \mathbf{G} (\mathbf{r}_A, \mathbf{r}_B, \omega') \cdot d_{nk}^B \times \]

\[ \times \left\{ \sum_{i=1}^{16} \frac{1}{D_i} + \left[ \sum_{i=1}^{4} \frac{1}{D_i} n (\omega) + \sum_{i=5}^{8} \frac{1}{D_i} n (\omega') + \text{c.c.} (\omega \leftrightarrow \omega') \right] \right\} \tag{29} \]

where \( n (\omega) \) is the Bose-Einstein distribution and c.c. \((\omega \leftrightarrow \omega')\) indicates the complex conjugate of the terms inside the square bracket after exchange between \( \omega \) and \( \omega' \).

| Energy denominators | Energy denominators |
|---------------------|---------------------|
| \( D_1 \) | \( (\omega + \omega) (\omega + \omega B(+) \) |
| \( D_2 \) | \( (\omega + \omega) (\omega - \omega B(+) \) |
| \( D_3 \) | \( (\omega - \omega) (\omega + \omega B(+) \) |
| \( D_4 \) | \( (\omega - \omega) (\omega + \omega B(+) \) |
| \( D_5 \) | \( (\omega + \omega) (\omega + \omega B(+) \) |
| \( D_6 \) | \( (\omega + \omega) (\omega - \omega B(+) \) |
| \( D_7 \) | \( (\omega - \omega) (\omega + \omega B(+) \) |
| \( D_8 \) | \( (\omega - \omega) (\omega - \omega B(+) \) |

Table 1: Energy denominators. In this table \( D_{kn}^B = \rho_{kn}^B + i (\Gamma_n^B + \Gamma_k^B) / 2 \) and \( D_{kn}^B = \rho_{kn}^B - i (\Gamma_n^B + \Gamma_k^B) / 2 \)

As expected the final expression of the energy contains two Green tensors at different frequencies, because it involves the exchange of two photons. The energy denominators as resulting from time integrals (see Appendix) are listed in the Table. They account contain the finite line-widths of the atomic energy levels.

After symmetrization we get the following interaction between two absorbing atoms (see Appendix):

\[ U(\mathbf{r}_A, \mathbf{r}_B) = -\frac{\mu_0^2}{2\pi^2} T \sum_{j=0}^{\infty} \xi_j^4 \times \]

\[ \times \text{Tr} \left\{ \frac{\alpha_A (i \xi_j) + \alpha_A (-i \xi_j)}{2} \mathbf{G} (\mathbf{r}_A, \mathbf{r}_B, i \xi_j) \frac{\alpha_B (i \xi_j) + \alpha_B (-i \xi_j)}{2} \mathbf{G} (\mathbf{r}_B, \mathbf{r}_A, i \xi_j) \right\} \tag{30} \]

It depends on the thermal polarizability of both atoms:

\[ \alpha_A (\omega) = \frac{1}{\hbar} \sum_n p_n^A \sum_k \left( \frac{d_{nk}^A \otimes d_{nk}^A}{\omega_{kn}^A + \omega + i (\Gamma_k^A + \Gamma_n^A) / 2} + \frac{d_{nk}^A \otimes d_{nk}^A}{\omega_{kn}^A - \omega - i (\Gamma_k^A + \Gamma_n^A) / 2} \right) \tag{31} \]
The energy \( E \) depends on the real combination \( \alpha(i\xi) + \alpha(-i\xi) \) rather than \( \alpha(i\xi) \) alone, as predicted by linear response theory. In the limit of small line widths \( \Gamma_n \ll \tilde{\omega}_{kn} \) the linear order in \( \Gamma \) is absent. This suppressed dependence of our result on the line widths means that our predicted Casimir–Polder potential closely resembles the perturbative (or plasma-model) result that one would obtain when neglecting the influence of line width altogether. In contra-distinction, the linear-response result predicts a linear leading-order dependence on the line widths.

As an example, we consider the interaction between two isotropic fullerene atom in vacuum and at \( T = 0 \) (ground-state atoms). The atomic frequencies and the dipole matrix elements are given in literature [24]. We choose fullerene atoms because they have large line-widths and can hence give us a clear distinction between our model and the previous theories. The interaction force is obtained substituting the vacuum Green tensor:

\[
F_{T=0}(r) = -\frac{\hbar}{8\pi^3 \varepsilon_0 r^4} \int_0^\infty d\xi \frac{\alpha_A(i\xi) + \alpha_A(-i\xi)}{2} \frac{\alpha_B(i\xi) + \alpha_B(-i\xi)}{2} g\left(\frac{\xi r}{c}\right) \hat{r} \tag{32}
\]

where:

\[
g(x) = e^{-2x} \left(9 + 18x + 16x^2 + 8x^3 + 3x^4 + x^5\right) \tag{33}
\]

We compare our result with two different model: the plasma model or perturbation theory, where the line-widths are zero, and the linear response theory:

\[
F_{\text{lin},T=0}(r) = -\frac{\hbar}{8\pi^3 \varepsilon_0 r^4} \int_0^\infty d\xi \alpha_A(i\xi) \alpha_B(i\xi) g\left(\frac{\xi r}{c}\right) \hat{r} \tag{34}
\]

This figure shows the Casimir interaction between two fullerene molecules in three different case: our model, the plasma model and the linear response theory.

Our model shows a close agreement with the plasma model. The Casimir interaction between two ground-state atoms, which is due to exchange of virtual photons, is not affected in a large measure by the dissipation of the atomic system: virtual photons exist for periods of time short compared to decay-time. Our model predicts a force smaller than the force of perturbation theory: the dissipation makes the force sightly weaker. Furthermore, the force is always attractive.

The distance dependence can be reduced to simple power laws in the retarded and non-retarded limits. We consider a typical time of evolution of the atomic system \( T_0 \), and with \( \omega_0, \lambda_0 \) the associated frequency and wavelength. For an atom \( \omega_0 \) can be the relevant atomic transition frequency. The electronic polarizability decays to zero for frequencies much larger than \( \omega_0 \). In the non-retarded regime, or when the distances between the two atoms is much smaller that \( \lambda_0 \) the polarizabilities restrict the \( \xi \) integral in a region where \( g(\xi r/c) \approx g(0) \) so the potential reduces to

\[
F_{\text{non-ret}}(r) = -\frac{9\hbar}{8\pi^3 \varepsilon_0} \int_0^\infty d\xi \frac{\alpha_A(i\xi) + \alpha_A(-i\xi)}{2} \frac{\alpha_B(i\xi) + \alpha_B(-i\xi)}{2} \frac{1}{r^7} = \frac{C_7}{r^7} \hat{r} \tag{35}
\]
Figure 3: The Casimir-Polder interaction between two fullerene molecules at zero temperature in three different case: our model, the plasma model and the linear response theory. The unit for the force is the femto-Newton $f N = 10^{-15} N$.

In the non-retarded limit there are deviations between our model, the plasma model and the linear response model. The following tabular gives the value of $C_7$ for two fullerene molecules at zero temperature, according to the three models:

|            | $C_7$ (SI-units) |
|------------|------------------|
| Our model  | $-9.58 \times 10^{-75}$ |
| Plasma model | $-1.00 \times 10^{-74}$ |
| Linear response | $-6.75 \times 10^{-75}$ |

In the retarded limit or when the distance $r$ is much larger than $\lambda_0$ the exponential restricts the $\xi$ integral to a range where the atomic polarizabilities are well approximated by their static values $\alpha_A (i\xi) \simeq \alpha_A (0)$:

$$F_{ret} (r) = -\frac{161hc}{64\pi^3\varepsilon_0^2} \alpha_A (0) \alpha_B (0) \frac{1}{r^8} \hat{r}$$

(36)

In this retarded limit, all three models coincide.
4 Interaction between a ground-state atom and an excited atom at zero temperature

We consider now the interaction between a ground-state atom and an excited atom at zero temperature. In order to describe such interaction we use equation (56) where \( \alpha_0^A \) is the polarizability of the ground-state atom A, and \( P_n^B = \delta_{nm} \) where \(|m\rangle\) is the excited state of atom B:

\[
U(r_A, r_B) = \frac{\mu_0^2}{4\pi i} \sum_{k} \int_{0}^{\infty} d\omega \omega^4 \left( \frac{1}{\omega_{mk}^{B(+)}} - \omega \right) \left( \frac{1}{\omega_{mk}^{B(–)}} - \omega \right) \times \left\{ d_{mk}^B \cdot G(r_B, r_A, \omega) \cdot \alpha_A^0(\omega) \cdot G(r_A, r_B, \omega) \right\}
\]

With atom B being excited, a pole appears in the above expression, leading to a resonant term. The interaction then contains two terms: the non-resonant term (which has the same structure as the Casimir-Polder potential for ground-state atoms) resulting from virtual photons exchange and the resonant term from the possible emission of resonant real photons. Because of the presence of line widths of atom B in our expression, there are no divergent denominators and we can obtain an unique resonant contribution.

The resonant term arises from the residues at \( \omega = \omega_{mk}^{B(+)}, -\omega_{mk}^{B(–)} \) (there are no residues for \( \omega = -\omega_{mk}^{B(+)}, \omega_{mk}^{B(–)} \)):

\[
U_{res}^{(2)}(r_A, r_B) = -\frac{\mu_0^2}{2} \sum_{k<m} \left( \omega_{mk}^{B(+)} \right)^4 \times \left\{ d_{mk}^B \cdot G(r_B, r_A, \omega_{mk}^{B(+)}) \cdot \alpha_A^0(\omega_{mk}^{B(+)}) \cdot G(r_A, r_B, \omega_{mk}^{B(+)}) \right\} + c.c.
\]

In order to compare with the discordant perturbative results we neglect the decay rates for both atoms, and describe the atoms by isotropic polarizabilities:

\[
U_{res}(r_A, r_B) = -\frac{\mu_0^2}{3} \sum_{k<m} \left( \omega_{mk}^{B} \right)^4 \left| d_{mk}^B \right|^2 \alpha_A^0(\omega_{mk}^{B}) \times \right\{ \text{Re} \left( G(r_A, r_B, \omega_{mk}^{B}) G(r_B, r_A, \omega_{mk}^{B}) \right) \right\}
\]

If the atoms are in vacuum we obtain the following energy:

\[
U_{res-vac}(r) = -\frac{1}{24\pi \varepsilon_0^2 \varepsilon_r} \sum_{k<m} \left| d_{mk}^B \right|^2 \alpha_A^0(\omega_{mk}^{B}) \times \left\{ \left( 3 - 5x_{mk}^2 + x_{mk}^4 \right) \cos(2x_{mk}) + (6x_{mk} - 2x_{mk}^3) \sin(2x_{mk}) \right\}
\]

where \( x_{mk} = r\omega_{mk}^{B}/c \).

For large distances, where the resonant term is dominant, we obtain a Casimir force
oscillating with respect to the distance. The time-dependent evaluation of Rizzuto and Passante \[10\], and the experimental observed force between an excited atom and a mirror \[18, 19\], are in agreement with our oscillating result.

The two conflicting perturbative results in the literature, as mentioned in the introduction, are due to differing convention in treating divergent energy denominators. Our model, on the other hand contains the finite line widths which remove the divergences in the energy denominators and can give a conclusive answer.

Figure 4: Casimir force between a Cesium in its ground-state and excited Rubidium at \(T = 0K\), according our and other models.

In the above figure we plot the Casimir force between a Cesium in its ground-state and excited Rubidium in vacuum at \(T = 0K\) (the Rubidium can be in the state \(5^{2}P_{1/2}\) or \(4^{2}P_{3/2}\)). At large distances the resonant term dominates and the force shows an oscillating behavior with an amplitude \(r^{-2}\), being attractive or repulsive. There are stable equilibrium regions in which the force is vanishing. At small distances the non-resonant term dominates and we have a non-oscillating repulsive force. The force differs from the non-oscillating result of Power et al. (also displayed).

5 Conclusions

We have derived the microscopic dispersion interaction between two atoms on the basis of an effective atom-field interaction energy, evaluated by means of a dynamical solution to the coupled atom-field equations. In particular, our approach gives a unique answer to the question regarding the impact of the line widths of atomic transitions.

We find that for two atoms at equilibrium with a finite-temperature environment, the impact of line dissipation is suppressed at linear order: The interaction according to our model with line widths considered is almost the same as the perturbative result which
neglects the line widths from the outset. Further to correcting microscopic Casimir–Polder potentials between two atoms, we believe that our result will have an impact on the ongoing Drude-plasma debate in Casimir theory: When extrapolating to the Casimir force, one may conjecture that a similar mechanism is at work, suppressing the influence of the Drude absorption parameter and hence leading to de facto agreement of a conjectured microscopic alternative to Lifshitz theory for absorbing media with the plasma limit of Lifshitz theory. A confirmation of this conjecture will in principle require extending our work to include many-atom interactions and summing these over the atoms contained in two plates to give the total Casimir energy. In addition our microscopic model, appropriate for dielectrics, would have to be generalised to metals.

We have further shown that an inclusion of finite line widths can provide a unique answer to the question whether the interaction between an excited atom and a ground-state one is oscillating. We find that the force is indeed oscillating, in agreement with observed oscillations of the atom–plate force.

6 Appendix

6.1 Lax-regression theorem

We recall the Lax-regression theorem because we will use it for the evaluation of the interaction energy. Let us suppose that the expectation values of the flip operators at different times are related according to

\[ \langle A_{mn}(t) \rangle = f^{A}_{mn}(t-t_1) \langle A_{mn}(t_1) \rangle \]  

(41)

The quantum regression theorem [25, 26] states that two-time correlation functions are given by:

\[ \langle A_{mn}(t) A_{pq}(t_1) \rangle = \begin{cases} 
    f^{A}_{mn}(t-t_1) \langle A_{mn}(t_1) A_{pq}(t_1) \rangle & t \geq t_1 \\
    f^{A}_{pq}(t_1-t) \langle A_{mn}(t) A_{pq}(t) \rangle & t \leq t_1 
\end{cases} \]  

(42)

6.2 Evaluation of the Casimir energy between two-atoms.

We consider the interaction energy of the atom A with the field of the atom B, Eq. (28). The final expression of the energy must contain two Green tensors of different frequencies, because involves the exchange of only two virtual photons. For this purpose, we express the electric field of the atom B in terms of the free electric field and the source field due to the atom B. Using (21) with starting point \( t_0 \):

\[ U = U(r_A) + U(r_A, r_B) \]  

(43)

where:

\[ U(r_A) = -\frac{1}{2} \int_0^\infty \left\langle \left( E^{(0)}(r_A, \omega, t) + E^{(0)\dagger}(r_A, \omega, t) \right) \cdot \alpha_0^A(\omega) \cdot E^{(0)}(r_A, t) \right\rangle \]  

(44)
operator gives a vanishing contribution. For the third term, we split the interval $t$ in terms of the free flip operator and the induced flip operator (19); the free flip atom $B$. The relevant term for the two-atom interaction is

$$U = \left\langle E^{(0)} (r_A, \omega, t) \cdot \alpha_A (\omega) \cdot \text{Im} G (r_A, r_B, \omega') \cdot d^B_{pq} B_{pq} (t_2) \right\rangle + \text{c.c.} (\omega \leftrightarrow \omega')$$

The term

$$\frac{-i\mu_0}{2\pi} \int_0^\infty d\omega \int_0^\infty d\omega' \left[ \sum_{p,q} \int_{t_0}^t dt_2 \left( e^{-i\omega'(t-t_2)} - e^{i\omega(t-t_2)} \right) \omega^2 \times \right.$$  

$$\left. \times \left\langle E^{(0)} (r_A, \omega, t) \cdot \alpha_A (\omega) \cdot \text{Im} G (r_A, r_B, \omega') \cdot d^B_{pq} B_{pq} (t_2) \right\rangle + \text{c.c.} (\omega \leftrightarrow \omega') \right] +$$

$$\frac{-i\mu_0}{2\pi} \int_0^\infty d\omega \int_0^\infty d\omega' \left[ \sum_{p,q} \int_{t_0}^t dt_2 \left( e^{-i\omega(t-t_2)} - e^{i\omega(t-t_2)} \right) \omega^2 \times \right.$$  

$$\left. \times \left\langle d^B_{pq} \cdot \text{Im} G (r_B, r_A, \omega) B_{pq} (t_2) \cdot \alpha_A (\omega) \cdot E^{(0)} (r_A, \omega', t) \right\rangle + \text{c.c.} (\omega \leftrightarrow \omega') \right] +$$

$$+ \frac{\mu_0^2}{2\pi^2} \int_0^\infty d\omega' \sum_{m,n} \int_{t_0}^t dt_1 \left[ e^{-i\omega(t-t_1)} - e^{i\omega(t-t_1)} \right] \int_{t_0}^t dt_2 \left[ e^{-i\omega'(t-t_2)} - e^{i\omega'(t-t_2)} \right] \times$$

$$\times \omega^2 \omega^2 d^B_{mn} \cdot \text{Im} G (r_B, r_A, \omega) \cdot \alpha_A (\omega) \cdot \text{Im} G (r_A, r_B, \omega') \cdot d^B_{pq} \langle B_{mn} (t_1) B_{pq} (t_2) \rangle$$

where c.c. $(\omega \leftrightarrow \omega')$ indicates the complex conjugate of the terms inside the square bracket after exchange between $\omega$ and $\omega'$. We have used the Onsager reciprocity of the Green tensor:

$$G^T (r, r', \omega) = G (r', r, \omega)$$

The term $U (r_A)$ gives the Casimir–Polder energy for the atom A in the absence of atom B. The relevant term for the two-atom interaction is $U (r_A, r_B)$, which we will now evaluate.

To evaluate the first and second terms in $U^{(2)} (r_A, r_B)$, we express the flip operator $B$ in terms of the free flip operator and the induced flip operator [19]; the free flip operator gives a vanishing contribution. For the third term, we split the interval $[t_0, t]$ of $t_2$ into two subintervals $[t_0, t_1], [t_1, t]$. In this subintervals the correlation function of the flip operators can be simply determined using the Lax-regression theorem:

$$\int_{t_0}^t dt_2 k (t_1, t_2) \langle B_{mn} (t_1) B_{pq} (t_2) \rangle = \int_{t_0}^{t_1} dt_2 k (t_1, t_2) f^B_{mn} (t_1 - t_2) \langle B_{mn} (t_2) B_{pq} (t_2) \rangle +$$

$$+ \int_{t_1}^t dt_2 k (t_1, t_2) f^B_{pq} (t_2 - t_1) \langle B_{mn} (t_1) B_{pq} (t_1) \rangle$$

(47)
Using these steps we find:

\[
U^{(2)}(r_A, r_B) = \frac{\mu_0}{2\pi\hbar} \int_0^\infty dw \int_0^\infty dw' \int_0^\infty dw'' \sum_{p,q} \sum_{m,n} \omega^2 \omega'^2 d_{mn}^{B} \cdot \text{Im} G(r_B, r_A, \omega) \cdot \alpha_A^0(\omega) \cdot \text{Im} G(r_B, r_A, \omega') \cdot d_{pq}^{B} \left( \mathbf{E}^{(0)}(r_A, \omega, t) \cdot \mathbf{E}^{(0)\dagger}(r_A, \omega, t) \right) - \frac{\mu_0}{\pi} \left( \frac{1}{1 + n(\omega)} \right) \right]
\]

where terms containing three Green tensors have been excluded.

Thermal averages of the free-field fluctuations can be evaluated by means of [20, 21]:

\[
\left\langle \mathbf{E}^{(0)}(r_A, \omega, t) \mathbf{E}^{(0)\dagger}(r', \omega', t) \right\rangle = \frac{\hbar \mu_0}{\pi} (1 + n(\omega)) \text{Im} G(r, r', \omega) \omega^2 \delta(\omega - \omega')
\]

\[
\left\langle \mathbf{E}^{(0)\dagger}(r, \omega, t) \mathbf{E}^{(0)}(r', \omega', t) \right\rangle = \frac{\hbar \mu_0}{\pi} n(\omega) \text{Im} G(r, r', \omega) \omega^2 \delta(\omega - \omega')
\]

where \(n(\omega)\) is the Bose-Einstein distribution:

\[
n(\omega) = \frac{1}{e^{\hbar\omega/k_B T} - 1}
\]
Now we can impose the atom $B$ to be in its thermal state, and use the Markov approximation $t_0 = -\infty$:

$$U^{(2)}(r_A, r_B) = \frac{\mu^2_0}{2\pi^2} \int_0^\infty d\omega \int_0^\infty d\omega' \sum_n p_n^B \sum_k \omega^2 \omega'^2 \times$$

$$\times d_{nk}^B \cdot \text{Im}G(r_B, r_A, \omega) \cdot \alpha_A^0(\omega) \cdot \text{Im}G(r_A, r_B, \omega') \cdot d_{nk}^B \times$$

$$\times \left\{ \frac{1}{1 + n(\omega)} \int_{-\infty}^t dt_2 \left( e^{-i\omega'(t-t_2)} - e^{i\omega'(t-t_2)} \right) \right.$$\n
$$\times \int_{-\infty}^{t_2} dt_3 \left( f_{nk}^B(t_2 - t_3) - f_{kn}^B(t_2 - t_3) \right) e^{i\omega(t_3-t)} +$$

$$+ n(\omega') \int_{-\infty}^t dt_2 \left( e^{-i\omega(t-t_2)} - e^{i\omega(t-t_2)} \right) \times$$

$$\times \int_{-\infty}^{t_2} dt_3 \left( f_{nk}^B(t_2 - t_3) - f_{kn}^B(t_2 - t_3) \right) e^{i\omega'(t_3-t)} + \text{c.c.}(\omega \leftrightarrow \omega') \right\} +$$

$$+ \int_{-\infty}^t dt_1 \left( e^{-i\omega(t-t_1)} - e^{i\omega(t-t_1)} \right) \times$$

$$\times \left( \int_{-\infty}^{t_1} dt_2 f_{nk}^B(t_1 - t_2) + \int_{t_1}^t dt_2 f_{kn}^B(t_2 - t_1) \right) \left( e^{-i\omega'(t-t_2)} - e^{i\omega'(t-t_2)} \right) \right\} \quad (51)$$

Time integration gives the energy $^{(29)}$.

After some algebra the denominators can be rearranged:

$$\sum_{i=1}^{16} D_i = \left( \frac{1}{\omega' + \omega} + \frac{1}{\omega' - \omega} \right) \left( \frac{1}{\omega + \omega_k^{B(+)} + \omega + \omega_k^{B(-)}} \right) +$$

$$+ \left( \frac{1}{\omega + \omega'} + \frac{1}{\omega - \omega'} \right) \left( \frac{1}{\omega' + \omega_k^{B(+)} + \omega' + \omega_k^{B(-)}} \right) \quad (52)$$

We integrate over one of the two frequencies. We use the following prescription: if there is the term $\left( \frac{1}{\omega' + \omega} + \frac{1}{\omega' - \omega} \right)$ we integrate first over $\omega'$, if there is the term $\left( \frac{1}{\omega + \omega'} + \frac{1}{\omega - \omega'} \right)$ we integrate first over $\omega$. 

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The integral over $\omega'$ can be performed as follows. We use the identity $\text{Im} G = \frac{(G - G^*)}{i2}$ and the Schwarz reflection principle:

$$\int_{0}^{+\infty} d\omega' \omega'^2 \left( \frac{1}{\omega' + \omega} + \frac{1}{\omega' - \omega} \right) \text{Im} G(r_B, r_A, \omega') =$$

$$= \frac{1}{2i} \int_{-\infty}^{+\infty} d\omega' \omega'^2 \left( \frac{1}{\omega' + \omega} + \frac{1}{\omega' - \omega} \right) G(r_B, r_A, \omega')$$

(53)

The Green tensor is analytic in the upper half of the complex plane, including the real axis, and it also finite at the origin. Therefore we can replace the integral by contour integrals along infinitely small half-circles surrounding $\pm \omega$ and an infinitely large half-circle in the upper complex half-plane. The integral along the infinite semi-circle vanishes for $r_A \neq r_B$ because:

$$\lim_{|\omega| \to +\infty} \omega^2 G(r_B, r_A, \omega) |_{r_A \neq r_B} = 0$$

(54)

We thus find:

$$\int_{0}^{+\infty} d\omega' \omega'^2 \left( \frac{1}{\omega' + \omega} + \frac{1}{\omega' - \omega} \right) \text{Im} G(r_B, r_A, \omega') = \frac{\pi}{2} \omega^2 [G(r_B, r_A, \omega) + G(r_A, r_B, -\omega)].$$

(55)

Summing all contributions we obtain:

$$U^{(2)}(r_A, r_B) = -\frac{\mu_0^2}{4\pi i} \sum_n p_n^B \sum_k \int_0^{+\infty} d\omega \omega^4 \times$$

$$\times \left\{ d_{nk}^B \cdot G(r_B, r_A, \omega) \cdot \alpha_A^0(\omega) \cdot G(r_A, r_B, \omega) \cdot d_{nk}^B$$

$$- d_{nk}^B \cdot G(r_B, r_A, -\omega) \cdot \alpha_A^0(\omega) \cdot G(r_A, r_B, -\omega) \cdot d_{nk}^B \right\} \times$$

$$\times \left\{ (1 + n(\omega)) \left( \frac{1}{\omega_{kn}^{B(+)} + \omega} + \frac{1}{\omega_{kn}^{B(-)} + \omega} \right) + n(\omega) \left( \frac{1}{\omega_{kn}^{B(+)} - \omega} + \frac{1}{\omega_{kn}^{B(-)} - \omega} \right) \right\}$$

(56)

where $\text{Tr}$ is the trace. The $n$-independent term represents the Casimir-Polder energy for two atoms at $T = 0$, and the terms proportional to $n$ are the corrections due to the interaction of the atoms with the thermal field.

We can transform the integral from $0$ to $\infty$ to an integral from $-\infty$ to $\infty$. Using the property $n(-\omega) = -n(\omega) - 1$ we obtain:

$$U^{(2)}(r_A, r_B) = -\frac{\mu_0^2}{4\pi i} \sum_n p_n^B \sum_k \int_{-\infty}^{+\infty} d\omega \omega^4 d_{nk}^B \cdot G(r_B, r_A, \omega) \cdot \alpha_A^0(\omega) \cdot G(r_A, r_B, \omega) \cdot d_{nk}^B \times$$

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\[ \times \left\{ (1 + n(\omega)) \left( \frac{1}{\omega_{kn}^{B(+)} + \omega} + \frac{1}{\omega_{kn}^{B(-)} + \omega} \right) + n(\omega) \left( \frac{1}{\omega_{kn}^{B(+)} - \omega} + \frac{1}{\omega_{kn}^{B(-)} - \omega} \right) \right\} \tag{57} \]

For the integral from \(-\infty\) to \(+\infty\) we enclose the integration with an infinite semi-circle in the upper complex half-plane, which has no contribution. We take the residuum for \(\omega = \omega_{kn}^{B(+)} , \omega_{nk}^{B(+)} , -\omega_{kn}^{B(-)} , -\omega_{nk}^{B(-)}\). Furthermore the thermal photon number \(n(\omega)\) exhibits poles in the upper half plane for purely imaginary frequencies \(\omega = i\xi_j\),

\[ \xi_j = \frac{2\pi k_B T j}{\hbar} \quad j = 0, 1, 2, \ldots \tag{58} \]

which are commonly known as the Matsubara frequencies. In the vicinity of the Matsubara frequencies, we have:

\[ n(\omega) \simeq \frac{k_B T}{\hbar} \frac{1}{\omega - i\xi_j} \tag{59} \]

Taking all the residuum we get:

\[ U^{(2)}(r_A, r_B) = -\mu_0^2 k_B T \sum_{j=0}^{\infty} i^4 \xi_j^4 \times \]

\[ \times \text{Tr} \left\{ \alpha_A^0(i\xi_j) G(r_A, r_B, i\xi_j) \frac{\alpha_B(i\xi_j) + \alpha_B(-i\xi_j)}{2} G(r_B, r_A, i\xi_j) \right\} \tag{60} \]

The resonant contribution is zero because the atom \(B\) is a thermal state: upward and downward transitions mutually cancel.

This result describes the Casimir interaction between an absorbing atom \(B\) and a non-absorbing atom \(A\) (zero line-widths). We postulate the more general interaction between two absorbing atoms to be of the form

\[ U^{(2)}(r_A, r_B) = -\mu_0^2 k_B T \sum_{j=0}^{\infty} i^4 \xi_j^4 \times \]

\[ \times \text{Tr} \left\{ f^A(\omega_{p'}^A, \Gamma_{p'}^A) G(r_A, r_B, i\xi_j) \frac{\alpha_B(i\xi_j) + \alpha_B(-i\xi_j)}{2} G(r_B, r_A, i\xi_j) \right\} \tag{61} \]

in which the variables of \(A\) and \(B\) appear in a factorized way. The Casimir–Polder interaction between two atoms at temperature \(T\) should be symmetric under the interchange of \(A\) and \(B\). This requirement leads to a unique interaction: Eq. (30).

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