The thermal Casimir–Polder interaction of an atom with a spherical plasma shell

Nail R Khusnutdinov

Institute of Physics, Kazan Federal University, Kremlenskaya 18, Kazan 420008, Russia

E-mail: 7nail7@gmail.com

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Abstract

The van der Waals and Casimir–Polder interaction energy of an atom with an infinitely thin sphere with finite conductivity is investigated in the framework of the hydrodynamic approach at finite temperature. This configuration models the real interaction of an atom with fullerene. The Lifshitz approach is used to find the free energy. We find the explicit expression for the free energy and perform its analysis for (i) high and low temperatures, (ii) large radii of the sphere and (iii) short separation between an atom and sphere. At low temperatures the thermal part of the free energy approaches zero as the fourth power of the temperature, while for high temperatures it is proportional to the first degree of the temperature. The entropy of this system is positive for small radii of the sphere and it becomes negative at low temperatures and for large radii of the sphere.

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(Some figures may appear in colour only in the online journal)

1. Introduction

Van der Waals dispersion forces play an important role in different physical, biological as well as chemical phenomena [1–4]. These forces arise between objects due to correlated quantum fluctuations. They depend on the shape of the body and their structure. The last achievements in Casimir effect and Casimir–Polder interaction have been discussed in great depth in books and reviews (see, for example, the recent book [3]). Of special interest are dispersion forces between microparticles and macroscopic bodies; in the case of interaction between particle and plate, it is commonly referred to as the Casimir–Polder force [5]. At a short range the energy rises as the third power of inverse distance between the microparticle and the plate. The retardation of the interaction should be taken into account at large distances and the interaction energy falls down as the fourth power of distance. At separations larger than a few micrometers, thermal effects become dominant.
Cavity quantum electrodynamic effects inside a spherical cavity have been considered in [6–8]. The non-retarded van der Waals potential for an atom inside and outside a metallic bubble was considered in [9]. The corrections for proximity force approximation of the Casimir interactions of an object inside and outside a spherical metal shell were calculated in [10, 11]. The perfectly conductive sphere with an atom was investigated in [12]. In [13, 14] the Casimir–Polder interaction between an atom and a magnetodielectric sphere was considered on the basis of point-scattering techniques. The consideration was related to results obtained in [15] for interaction of an atom with a curved surface and in [16, 17] for an atom with a dielectric sphere. The Casimir and Casimir–Polder energies were considered in [18, 19] in the framework of the plasma (hydrodynamical) model of the sphere which was developed in Barton’s papers [20, 21] in the context of the fullerene molecule C60. Barton’s approach was based on the Fetter paper [22] which was devoted to the properties of the two-dimensional free-electron gas. This model was exploited in [23] for flat sheet. A complete list of references about particles in spherical geometry may be found in reviews [24, 25].

The spherical plasma shell was considered in the framework of this model in [18] in the context of the Casimir energy and in [19] for the Casimir–Polder interaction of an atom with a spherical plasma surface. In the context of this model, the conductive sphere is represented by the two-dimensional surface with free-electron gas on this surface. The dynamic of this gas is described by hydrodynamics. All information about electron gas is encoded solely by a parameter with dimension of wave number: \( \Omega = \frac{4\pi n c^2/m c^2}{e} \), where \( n \) is the surface density of electron gas, and \( e \) and \( m \) are, respectively, the charge and mass of the electron. It was shown that the interaction energy between an atom and the sphere has the form of the Casimir–Polder energy for an atom and a plate if the atom is situated close to the sphere. Away from the sphere, the energy has the form of the atom–atom interaction energy with an expression for the polarizability of the sphere. The numerical simulations were made for the sphere with parameters of the fullerene molecule C60 and hydrogen atom.

In this paper we consider the thermal Casimir–Polder interaction energy for a system comprising of an atom and a conductive sphere. The sphere is described by the plasma (hydrodynamical) model which was considered in detail in [20, 21] and we use the Lifshitz approach [26, 27] to calculate the free energy of this system. We outline the Lifshitz approach in section 2.

The calculations performed in this paper could find a potential application to the interaction of microparticles with a molecule of fullerene. The Casimir–Polder interaction of the fullerene molecules C60 and C70 with gold and silicon nitride surfaces was considered in detail in [28]. The fullerene molecule is graphene shaped as a sphere and the more realistic model to calculate the Casimir–Polder interaction between an atom and a fullerene is the Dirac model. In this model, the frequency spectrum has a linear form and it is valid for low energies. Some calculations related to the Casimir and Casimir–Polder energies were made in the framework of this model in [29–31]. The frequency spectrum in the hydrodynamic (plasma) model has a quadratic form and the model is the high-frequency approximation of the theory. Both models are in qualitative agreement with respect to the Casimir–Polder interaction. The calculations performed in [31] showed that the Casimir–Polder force calculated in the framework of the Dirac model is smaller than that for the hydrodynamic model.

The rest of this paper is organized as follows. In section 2 we discuss the Lifshitz approach and derive the expression for the free energy and represent it in different forms. Section 3 is devoted to consideration of the specific limit cases. We consider the ideal case with infinite radius of the sphere but finite distance between the surface and the atom and obtain the first correction term over inverse radius of the sphere. We also obtain the low- and high-temperature expansions and consider the case of the short distance from the sphere. The expression for the
entropy is found in section 4 and it is analyzed in the limits of low and high temperatures. We show plots of the numerical calculations for the system comprised of the hydrogen atom and sphere with parameters of the fullerene C60. In section 5 we discuss the results obtained.

2. Lifshitz approach for an atom near the plasma spherical shell

We adopt here the approach developed by Lifshitz in [26, 27]. This approach was applied to calculate the Casimir–Polder interaction energy of an atom and plasma sphere at zero temperature in [19]. Let us shortly discuss the approach. We consider a conductive infinitely thin sphere with radius \( R \) inside the vacuum spherical cavity with radius \( L = R + d \) which is into the dielectric media with parameters \( \varepsilon, \mu \neq 1 \) (see figure 1). Due to the spherical symmetry of the problem under consideration, the electromagnetic field may be divided in the two polarizations which are usually called TE and TM modes. We have two concentric spheres and we should consider the boundary conditions on the two spherical boundaries. The full information about the problem is encoded in these boundary conditions. The information about the plasma sphere is encoded in single parameter \( \Omega_1 = \frac{4\pi n e^2}{mc^2} \) with dimension of the wave number, where \( e \) is the charge of the electron, \( m \) is its mass and \( n \) is the surface density of electrons on the sphere. Applications of this model for vacuum fluctuations of field can be found in [20, 21, 23, 18].

The spectrum of the electromagnetic oscillations, \( \omega = k c \), in this configuration may be found from these boundary conditions (see [19] for more details):

\[
\Sigma_{TE} = -i \left\{ H' (z_{e}) \Psi_{TE} - \frac{1}{\sqrt{\varepsilon}} H (z_{e}) \Psi_{TE}' \right\} = 0, \tag{1a}
\]

\[
\Sigma_{TM} = -iz^2 \left\{ H (z_{e}) \Psi_{TM}' - \frac{1}{\sqrt{\varepsilon}} H' (z_{e}) \Psi_{TM} \right\} = 0, \tag{1b}
\]

where

\[
\Psi_{TE}(z) = J(z) + \frac{Q}{x} J(x)[J(x)Y_{1} (z) - J(z)Y_{1}(x)], \tag{1c}
\]

\[
\Psi_{TM}(z) = J(z) + \frac{Q}{x} J'(x)[J'(x)Y_{1}(z) - J(z)Y_{1}'(x)], \tag{1d}
\]

and \( J(x) = x j (x) \), \( Y(x) = x y (x) \) and \( H (x) = x h_{11} (x) \) are the Riccati–Bessel functions. Here \( x = k R, Q = \Omega R \) and \( z_{e} = z \sqrt{\varepsilon}, z = kL = \omega L / c \).
The free energy, \( F \), of the system is expressed in terms of these frequencies in the following form [3]:

\[
F = k_B T \sum_{l=1}^{\infty} (2l + 1) \sum_{n=1}^{\infty} \left[ \ln \left( 2 \sinh \frac{\hbar \omega_{n,l}^{TM}}{2k_B T} \right) + \ln \left( 2 \sinh \frac{\hbar \omega_{n,l}^{TE}}{2k_B T} \right) \right],
\]

where index \( n \) numbers the solutions of the boundary condition equations (2). Then we convert the sum over \( n \) to the contour integral

\[
F = k_B T \sum_{l=1}^{\infty} (2l + 1) \frac{1}{2\pi i} \oint_{\gamma} \ln \left( 2 \sinh \frac{\hbar \omega}{2k_B T} \right) d(\ln \Sigma_{TM} + \ln \Sigma_{TE}),
\]

where the contour \( \gamma \) counter-clockwise encloses all real positive solutions of equation (2). To put the contour to the imaginary axes, we should clear up the zero-frequency behavior of functions \( \Sigma \). If \( Q \neq 0 \), we obtain (\( \nu = l + 1/2 \))

\[
\Sigma_{TE}|_{\omega \to 0} = \left( 1 + \frac{Q}{2\nu} \right) e^{-\frac{i\pi}{2}},
\]

\[
\Sigma_{TM}|_{\omega \to 0} = \frac{Q l(l+1)(1+\nu^2) + a^{l+1+1}(l+\nu^2(l+1))}{4\nu^2} e^{-\frac{i\pi}{2}},
\]

where \( \chi = L/R = 1 + d/R, \) \( Q = \Omega R \) and \( \epsilon \) is the dielectric permittivity. In the case \( Q = 0 \), we have

\[
\Sigma_{TE}|_{\omega \to 0} = e^{-\frac{i\pi}{2}},
\]

\[
\Sigma_{TM}|_{\omega \to 0} = \frac{l + \epsilon^2(l+1)}{2\nu} \chi^2 e^{-\frac{i\pi}{2}}.
\]

Therefore, no poles appear at the point \( \omega = 0 \) and we may shift the contour \( \gamma \) to the imaginary axes.

In the imaginary axes, the function under logarithm has zeros at the Matsubara frequencies

\[
\omega_n = i\xi_n = \frac{2\pi nk_B T}{\hbar}, \quad n = 0, \pm 1, \pm 2, \ldots
\]

We put the contour to the right of the imaginary axes,

\[
F = k_B T \sum_{l=1}^{\infty} (2l + 1) \frac{1}{2\pi i} \int_{\epsilon+i\infty}^{\epsilon-i\infty} \ln \left( 2 \sinh \frac{\hbar \omega}{2k_B T} \right) d(\ln \Sigma_{TM} + \ln \Sigma_{TE}),
\]

and then take the limit \( \epsilon \to 0 \). Integrating by part, we obtain

\[
F = -k_B T \sum_{l=1}^{\infty} (2l + 1) \frac{1}{2\pi i} \int_{\epsilon+i\infty}^{\epsilon-i\infty} \frac{\hbar}{2k_B T} \coth \frac{\hbar \omega}{2k_B T} (\ln \Sigma_{TM} + \ln \Sigma_{TE}) d\omega.
\]

Then we shift the contour \( \gamma \) to the imaginary axes. We obtain infinite sum of integrals in the imaginary axes between the Matsubara frequencies and integrals over semi-spheres around these frequencies. The integrals between \( \xi_n \) and \( \xi_{n+1} \) and between \( \xi_{-n} \) and \( \xi_{-n-1} \) cancel each other and we obtain (\( \nu = l + 1/2 \))

\[
F = k_B T \sum_{l=1}^{\infty} \sum_{n=\infty}^{\infty} (\ln \Sigma_{TM} + \ln \Sigma_{TE})|_{\omega=\pm i\xi_n}.
\]

To calculate the free energy per unit atom, \( F' \), let us rarefy the media with \( \epsilon(i\omega) = 1 + 4\pi N_\alpha(i\omega) + O(N^2) \), where \( \alpha \) is the polarizability of the atom and the density of the
dielectric matter $N \rightarrow 0$ (see figure 1). In this case, the free energy, $F$, is expressed in terms of the energy per unit atom $F^a$ by relation

$$F = N \int_0^\infty d \tau \left( F^a 4\pi (R + \tau)^2 + O(N^2) \right).$$

(10)

From this expression, it follows that

$$F^a = - \lim_{N \to 0} \frac{\partial_d F}{4\pi N(R + d)^2}. $$

(11)

After straightforward calculations, we obtain

$$F^a = \frac{kB T}{(R + d)^2} \sum_{l=1}^\infty \sum_{n=-\infty}^{+\infty} \left( \frac{k\alpha(i\omega)}{G_{TE}} + \frac{k\alpha(i\omega)}{G_{TM}} \right)_{\omega=\xi_n},$$

(12)

where

$$G_{TE}^{-1} = \frac{x}{s} \frac{s' \xi(z)}{s \xi(z)},$$

$$G_{TM}^{-1} = -\frac{t'}{s} \xi(z) - \frac{t}{s} \xi(z) \xi(z) - \frac{Q}{x} \frac{s' \xi(z)}{s \xi(z)} + \frac{Q}{x} \frac{s' \xi(z)}{s \xi(z)} \frac{\xi(z)}{x},$$

and

$$s_1(x) = \sqrt{\frac{\pi x}{2}} L_{\nu+1/2}(x), \quad \nu = \sqrt{\frac{2x}{\pi} K_{\nu+1/2}(x)}$$

(13)

are the modified Riccati–Bessel spherical functions.

For renormalization, we throw away terms which survive in the limit $Q \to 0$ and arrive with formula

$$F^a = - \frac{2kB T \Omega}{(R + d)^2} \sum_{l=1}^\infty \sum_{n=0}^{+\infty} \alpha(i\xi_n) \left\{ \frac{s^2(x) \xi(z)}{f_{TE}(ix)} + \frac{s' \xi(z)}{f_{TM}(ix)} + \frac{1}{x} \frac{s' \xi(z)}{s \xi(z)} \frac{\xi(z)}{x} \right\}_{\omega=\xi_n},$$

(14)

where $x = \omega R/c$ and $\omega = \omega L/c = \omega(R + d)/c$,

$$f_{TE}(ix) = 1 + \frac{Q}{x} s_1(x) e_1(x),$$

$$f_{TM}(ix) = 1 - \frac{Q}{x} s'(x) e_1(x)$$

are the Jost functions on the imaginary axes, and the sum with prime means that the zero term has factor 1/2. Formula (14) has the same form as for the zero-temperature case obtained in [19] but with summation over Matsubara frequencies instead of integration.

Now we use single-oscillatory approximation for $\alpha$:

$$\alpha(i\omega) = \frac{\frac{\xi^2}{\omega^2 + \omega^2}}{\omega^2 + \omega^2}. $$

(15)

At Matsubara frequencies, we have

$$\alpha(i\xi_n) = \frac{\frac{\xi^2}{\omega^2} + \frac{\xi^2}{\omega^2} + 1}{\frac{\xi^2}{\omega^2} + 1} = \frac{\alpha(0)}{\frac{\xi^2}{\omega^2} + 1}. $$

(16)

In the problem under consideration, the three different effective temperatures appear

$$T_\omega = \frac{\hbar \omega}{2\pi k_B}, \quad T_R = \frac{\hbar c}{2\pi k_B R}, \quad T_d = \frac{\hbar c}{2\pi k_B d}.$$  

(17)
The variables are expressed in terms of these temperatures by relations
\[
\frac{\xi_n}{\omega_n} = \frac{2\pi k_B T n}{\hbar \omega_n} = n \frac{T}{T_R}, \tag{18a}
\]
\[
x = \frac{\xi_n R}{c} = \frac{2\pi k_B T R n}{\hbar c} = n \frac{T}{T_R}, \tag{18b}
\]
\[
z = \frac{\xi_n L}{c} = \frac{2\pi k_B T L n}{\hbar c} = n \left( \frac{T}{T_R} + \frac{T}{T_d} \right). \tag{18c}
\]

Let us estimate these temperatures for molecule C\(_{60}\) and the hydrogen atom. For molecule C\(_{60}\) \cite{20}, we have \(R = 0.342\) nm and the frequency \(\omega_n\) for the hydrogen atom reads \(32–34\) \(\omega_n = 11.65\) eV = \(17.698 \times 10^{15}\) Hz. Taking into account these parameters, we obtain \(T_R = 2.15 \times 10^4\) K, \(T_d = 1.06 \times 10^8\) K.

Now we obtain another representation of formula (14). We rewrite equation (14) in the following form:
\[
\mathcal{F}^a = -\frac{2k_BT Qa(0)a^2}{R^3(1 + r)^2} \sum_{n=0}^{+\infty} \sum_{l=1}^{\infty} F_l(n), \tag{19a}
\]
where
\[
F_l(n) = \frac{g_l(n)}{n^2 + a^2}, \tag{19b}
\]
\[
g_l(n) = \left\{ \begin{array}{ll}
\frac{s_l^2(x)e_l^2(z)}{f_{TE}(ix)} + \frac{s_l^2(x)e_l^2(z) + s_l^2(x)e_l^2(z) \pi^2}{f_{TM}(ix)} & \text{for } l = \text{TE} \\
0 & \text{for } l = \text{TM}
\end{array} \right\}. \tag{19c}
\]

Here \(a = T/R\) and \(r = d/R\). The function \(g_l\) consists of two parts which comes from TE and TM polarizations of electromagnetic field, correspondingly. Hereafter, we will denote these contributions as \(s_l^{TE}\) and \(s_l^{TM}\).

The function \(g_l(n)\) has no poles in the imaginary axes. Taking into account the Abel–Plana formula, we obtain
\[
\sum_{n=0}^{+\infty} F_l(n) = \int_0^{+\infty} \frac{g_l(t)}{t^2 + a^2} \, dt + \frac{\pi}{2a} \frac{g_l(ia) + g_l(-ia)}{e^{2\pi a} - 1} - \frac{i}{2a} \int_0^{+\infty} \left( \frac{g_l(it) - g_l(-it)}{e^{2\pi t} - 1} \right) \ln \frac{a + t}{a - t} \, dt. \tag{20}
\]

In the first term, we change integrand variable \(t = ak/(c\omega_n)\) and the contribution of this term is exactly the same as was obtained in \cite{19} for zero temperature
\[
E_0 = -\frac{\hbar c}{\pi R^3(1 + r)^2} \int_0^{+\infty} dk \, a(\omega)e_0(k), \tag{21}
\]
where
\[
e_0(t) = Q \sum_{l=1}^{+\infty} v g_l(t). \tag{22}
\]

The second and third terms give the temperature corrections
\[
\mathcal{F}_1^T = -\frac{\alpha(0)e_1(a)}{R^3(1 + r)^2} \frac{\hbar \omega_n}{\varphi^{2\pi a - 1}},
\]
\[
\mathcal{F}_2^T = -\frac{\alpha(0)h \omega_n}{\pi R^3(1 + r)^2} \int_0^{+\infty} \left( \frac{e_2(t)}{\varphi^{2\pi t} - 1} \right) \ln \frac{1 + t}{1 - t} \, dt,
\]
\[
= -\frac{\alpha(0)h \omega_n}{\pi R^3(1 + r)^2} \int_0^{+\infty} \left( \frac{e_2(at)}{\varphi^{2\pi at} - 1} \right) \ln \frac{1 + t}{1 - t} \, dt. \tag{23}
\]
where

\[
e_1(t) = \frac{1}{2} Q \sum_{l=1}^{\infty} v [g_l(i\tau) + g_l(-i\tau)] = \Re e_0(i\tau),
\]

\[
e_2(t) = \frac{-i}{2} Q \sum_{l=1}^{\infty} v [g_l(i\tau) - g_l(-i\tau)] = \Im e_0(i\tau).
\]

We note that the functions \(g_l(\pm ia)\) and \(g_l(\pm i\tau a)\) (also \(e_1(a)\) and \(e_2(a\tau)\)) have no dependence on the temperature. Indeed, at the beginning (2), they depend on \(n \equiv nT/T_R\) and \(z = nT/T_R(1 + r)\), only. After applying the Abel–Plana formula, we should replace \(n \to \pm ia\) (or \(n \to \pm i\tau a\)). Therefore, we obtain that \(x \to i\tau a\) and \(z \to i\tau a(1 + r)\) and whole dependence on the temperature is accumulated in the exponential factor equation (23) via the single parameter \(a = T_0/T\).

3. Limit cases

3.1. An atom near the ideal plate: \(\Omega \to \infty\) and \(R \to \infty\)

First of all, we have to compare our formulas with the well-known case of an atom near the flat surface. There is a need to take the limit of infinite conductivity \(\Omega \to \infty\) to obtain an ideal surface case and then take the limit of infinite radius of the sphere, providing finite distance, \(d\), between an atom and the surface. To take limits, we use the expression for free energy in the form of equation (14). Taking the limit of infinite conductivity \(\Omega \to \infty\), we obtain the following expression for the free energy:

\[
\mathcal{F}^a = -\frac{2\kappa T}{R^2 \chi^2} \sum_{n=0}^{+\infty} \alpha(i\xi_n) \sum_{l=1}^{\infty} v_x \left\{ \frac{s_l^2(x)e_l^2(z)}{s_l(x)e_l(x)} - \frac{s_l^2(x)e_l^2(z) + s_l^2(x)e_l^2(z)v^2l^2}{s_l(x)e_l(x)} \right\}_{\alpha=\xi_n},
\]

where \(x = \xi_n/R; z = x\chi\) and \(\chi = 1 + r = 1 + d/R\).

To obtain expansion over \(r = d/R \ll 1\), we use the uniform Debye expansions for modified Bessel functions from textbook [35]. Taking into account these expansions, we obtain

\[
\mathcal{F}^a = -\frac{2\kappa T}{R^2 \chi^2} \sum_{n=0}^{+\infty} \alpha(i\xi_n) \sum_{l=1}^{\infty} v_x e^{-2v[\eta(z) - \eta(x)]} \times \left\{ 1 + \frac{1}{12v} \left( 3[t(x) - t(z)] + [t^3(x) - t^3(z)] + 6t^4(z) \right) + \cdots \right\}_{\nu = \frac{i}{\xi_n}},
\]

where \(t(x) = 1/\sqrt{1 + x^2}\) and \(\eta(x) = 1/t(x) + \ln \frac{\sqrt{1 + x^2} + x}{\sqrt{1 + x^2} - x}\). Now we replace summation over \(v = l + 1/2\) to integration over \(v\) and change the integrand variable \(v\) to \(k_\perp = vR\). We observe that the uniform expansion over \(1/v\) corresponds to the expansion over \(1/R\). For this reason, in the second term of expansion, we may set \(z = x\) up to the first power of \(1/R\) and the expression in braces takes the form \(1 + t^3(z)\xi_n/c_{k_\perp}/2k_\perp R\). We expand the exponent in the above formula over \(d/R \ll 1\) and obtain

\[
e^{-2v[\eta(z) - \eta(x)]} = e^{-2dk_\perp \sqrt{1 + \frac{k^2}{2Rd^2}}} \left\{ 1 + \frac{d^2k_\perp}{R} - \frac{\xi_n}{ck_\perp} + \cdots \right\}.
\]

Taking into account these expansions, we obtain the following expansion the free energy up to \(R^{-1}\):

\[
\mathcal{F}^a = -2\kappa T \sum_{n=0}^{+\infty} \alpha(i\xi_n) \int_{0}^{d_\perp} dk_\perp k_\perp q_\perp e^{-2dk_\perp} \left\{ 1 - \frac{3d}{R} - \frac{k^4}{2Rd^2} + \frac{d^2k^2}{Rd^3} \right\},
\]

\[
\text{(27)}
\]
where \( q_n^2 = k_n^2 + \xi_n^2/c^2 \). The first term of this expansion corresponds to the case of an atom near flat ideal surface and it has the same form as in [36]. The next terms give corrections due to finite radius of the sphere.

Let us discuss an error which appears when we replace summation by integration. The precision of calculations is down to \( 1/R \). The second term of expansion over \( 1/v \) in equation (25) gives contribution \( \sim R^{-1} \) and when we replace summation by integration, the error will be outside the considered precision. Let us consider the first term in equation (25):

\[
\sum_{l=1}^{\infty} v^2\sqrt{v^2 + z^2}e^{-2v[n(l)-\eta(l)]}.
\]

To estimate error, we use the Abel–Plana formula:

\[
\sum_{l=1}^{\infty} f(v) = -f\left(\frac{1}{2}\right) + \int_{0}^{\infty} f(v) dv - i \int_{0}^{\infty} \frac{f(it) - f(-it)}{\xi_{2\pi t} + 1} dt
\]

with \( f(v) = v^2\sqrt{v^2 + z^2}e^{-2v[n(l)-\eta(l)]} \). It is easy to show that the term \( f(\frac{1}{2}) \) gives contribution \( \sim R^{-2} \) which is outside of the precision and we drop this term. The domain of the integration in the last term is to be divided for three intervals, namely \( 0 < t < x, x < t < z = \chi x \) and \( t > z \). Considering each interval separately, we obtain

\[
\int_{0}^{x} \frac{2t\sqrt{z^2 - t^2}e^{-2f_1}}{\xi_{2\pi t} + 1} dt + \int_{x}^{z} \frac{2t\sqrt{z^2 - t^2}e^{-2f_1/2}cos(2v_1)}{\xi_{2\pi t} + 1} dt + \int_{z}^{\infty} \frac{2t\sqrt{t^2 - z^2}e^{-2f_1/2}sin(2v_1)}{\xi_{2\pi t} + 1} dt,
\]

where the functions read

\[
\begin{align*}
    f_1(t) &= \sqrt{z^2 - t^2} - \sqrt{x^2 - t^2} - t \left( \arctan \frac{t}{\sqrt{x^2 - t^2}} - \arctan \frac{t}{\sqrt{z^2 - t^2}} \right), \\
    f_2(t) &= \sqrt{z^2 - t^2} - t \arctan \frac{\sqrt{z^2 - t^2}}{t}, \\
    v_2(t) &= t \ln \frac{t + \sqrt{t^2 - x^2}}{x} - \sqrt{t^2 - x^2}, \\
    v_3(t) &= \sqrt{t^2 - z^2} - \sqrt{t^2 - x^2} + t \ln \left[ \frac{zt + \sqrt{t^2 - x^2}}{zt + \sqrt{t^2 - z^2}} \right].
\end{align*}
\]

Now we take the limit \( R \to \infty \) and expand above integrals over \( 1/R \). The expansion of the first integral reads: \( x e^{-x/24} \), where \( \tau = 4\pi k_B dT/\hbar^2 \) and \( x = \xi_n R/c \). Because of the factor \( R^{-3} \) in the free energy (25), the contribution of this integral is \( \sim R^{-2} \). Both rest integrals in (30) give exponentially small contributions \( \sim e^{-2x_t} \). Therefore, the error which appears when we replace summation to integration is order \( R^{-2} \) and it is outside the precision under consideration.

### 3.2. Low-temperature expansion

Let us consider now the low-temperature expansion: \( T \ll T_w, T_R \) and \( T \ll T_{R+\delta} \). The last relation means that we consider distances \( r = \frac{d}{R} \ll \frac{h}{R} \). The first temperature correction given by equation (23) gives exponentially small contribution, \( \mathcal{F}_1(t) \sim e^{-2\pi R/T} \). To find expansion of the second temperature correction (23), we take into account that the function \( v_2(t) \) depends on the \( TR/R \) and \( TTR/(R+\delta) \). Owing to the exponential factor, we have to take into account only a small domain of integration \( t \ll 1 \) in the integral. Let us find the power expansion of the function \( g_r(t) \) over \( t \) and keep the main terms.
(1) TE contribution. The expansion begins with $s^2$:

$$e_2^{TE}(t) = -s^2 \frac{Q - 2(3 + Q)x^3}{6(3 + Q)^2 x^2} + \ldots,$$

where $s = tT/T_R$ and $x = 1 + r = 1 + d/R$.

(2) TM contribution. The expansion begins with $s^3$:

$$e_2^{TM}(t) = -s^3 \frac{2}{x^4} - s^3 \frac{15Q + 72(5 + Q)x^2 + 40Qx^4 + 88Qx^7}{60Qx^6} + \ldots.$$

This expression holds for $T/T_R \ll Q$. The case $Q = 0$ should be considered separately. We set $Q = 0$ at the beginning and obtain

$$e_2^{TM}_{|Q=0} = Q \left\{ 3, \frac{44}{45} - s^3 \frac{107 + 184x^2}{315} \right\}_{Q \to 0} = 0.$$

Therefore, the main contribution for the case of the finite conductivity, $Q > 0$, comes from TM mode and it reads

$$F^T_2 = \frac{2\alpha(0)\hbar\omega_l}{\pi x^6 R^3} \int_0^\infty \left( \frac{t^3}{e^{2\pi t} - 1} \right) \ln \left| \frac{a + t}{a - t} \right| dt.$$  

Because of $a = T_\omega/T \gg 1$, we expand logarithm over $t \ll a$ and obtain

$$\int_0^\infty \left( \frac{t^3}{e^{2\pi t} - 1} \right) \ln \left| \frac{a + t}{a - t} \right| dt = \int_0^\infty \left( \frac{t^3}{e^{2\pi t} - 1} \right) \frac{2t}{a} dt = -\frac{1}{120a}.$$  

Therefore,

$$F^T_2 = -\frac{\hbar\alpha(0)}{60\pi R^4(1 + r)^6} \frac{T^4}{T_R^2} = -\frac{4\pi^3}{15} \frac{\alpha(0)}{(1 + r)^6} \frac{(k_BT)^4}{(\hbar c)^3},$$

and

$$F^a = E_{CP} \left\{ S_\Omega + S_F \left( \frac{T}{T_R} \right)^4 \right\},$$

where

$$S_F = \frac{2r^4}{45(1 + r)^6},$$

and $E_{CP} = -3\hbar\alpha(0)/8\pi d^4$ is the Casimir–Polder energy. The plots of $S_\Omega$ and $S_F$ are shown in figure 2 for the case of the molecule C$_{60}$ and hydrogen atom.

### 3.3. High-temperature expansion

In this case, it is better to use equation (14):

$$F^a = -\frac{2k_BT\Omega}{(R + d)^2} \sum_{l=1}^{\infty} \sum_{n=0}^{\infty} \alpha(\xi_n) \left\{ \frac{s_1^2(x)e_1^2(z)}{f_{TE}(ix)} + \frac{s_1^2(x)e_1^2(z) + s_2^2(x)e_2^2(z)\sqrt{\frac{2}{\pi}}}{f_{TM}(ix)} \right\} e_{\xi_n},$$

where $x = kR$ and $z = kL = k(R + d)$, and sum with prime means that the zero term has factor 1/2.

At high temperatures, the Matsubara frequencies $\xi_n \sim nT \to \infty$ for $n > 0$ and these terms in sum over $n$ give exponentially small contribution $\sim e^{-2\xi_n}$. The main contribution
Figure 2. The plot of the functions $S_\Omega$ and $S_T$ from equation (37) calculated for molecule C$_{60}$ and hydrogen atom. The function $S_\Omega$ is the zero-temperature contribution and depends on the parameters of the sphere and the atom. The function $S_T$ is universal and depends on distance $r$, only.

comes from zero mode, $n = 0$, and it may be calculated in the manifest form. Indeed, taking the limit $n \to 0$ in the above expression, we obtain the following contribution:

$$F_a = -k_B \alpha(0) \frac{6r^4 + 24r^3 + 33r^2 + 18r + 4}{2r^3(r+1)^2(r+2)^3R^3},$$

(40)

where $\chi = 1 + d/R$ and $r = d/R$. We have the following asymptotic for short and large separations between an atom and the sphere:

$$F_a = - \frac{k_B \alpha(0)}{4d^3}, \quad \frac{d}{R} \ll 1,$$

$$F_a = - \frac{3k_B \alpha(0)R^3}{d^6}, \quad \frac{d}{R} \gg 1.$$

Extracting the Casimir–Polder energy, we obtain

$$F_a = E_{CP} \overline{S}_T \frac{T}{T_R},$$

(41)

where

$$\overline{S}_T = \frac{2r(6r^4 + 24r^3 + 33r^2 + 18r + 4)}{3(r+1)^2(r+2)^3},$$

(42)

3.4. Short distance behavior, $r = \frac{d}{R} \ll 1$

Let us consider equation (23). In the limit $r \to 0$, the series is divergent for great $l$. For this reason, we use standard formulas (9.3.1) from [35]:

$$J_v(z) \approx \frac{1}{\sqrt{2\pi v}} \left( \frac{ez}{2v} \right)^v,$$

(43a)

$$Y_v(z) \approx - \frac{2}{\sqrt{2\pi v}} \left( \frac{ez}{2v} \right)^{-v},$$

(43b)
which are fulfilled for $\nu \to \infty$. Taking into account these formulas in the expression for $g_i(i\alpha)$, we obtain

$$
\nu \left[ g_i^{TM}(ia) + g_i^{TM}(-ia) \right] \approx \frac{8\nu^2 \chi^{-1-2\nu}}{4\nu^2 - 1} Q - 8\nu q_a^2 \begin{cases} \frac{2\nu^2 \chi^{-1-2\nu}}{Q}, & Q > 0, r < Q, \\ \frac{\nu^2 \chi^{-1-2\nu}}{q_a^2}, & Q = 0, r < q_a^2, \end{cases}
$$

for TM contribution, and

$$
\nu \left[ g_i^{TE}(ia) + g_i^{TE}(-ia) \right] \approx -\frac{q_a^2 \chi^{-1-2\nu}}{2\nu + Q} \begin{cases} \frac{q_a^2 \chi^{-1-2\nu}}{2\nu}, & Q \geq 0, \\ -\frac{\nu^2 \chi^{-1-2\nu}}{q_a^2}, & Q \to \infty, \end{cases}
$$

for TE contribution. We make summation for finite $Q > 0$ and $r < Q$ and obtain

$$
e_1^{TM}(a) \approx \frac{1 - 2\chi^2 + 9\chi^4}{4\chi^2(\chi^2 - 1)^3} \approx \frac{1}{4r^3},
$$

(45a)

$$
e_1^{TE}(a) \approx \frac{Q q_a^2}{2} \left( 1 - \chi \arctanh \frac{1}{\chi} \right) \approx \frac{Q q_a^2 \ln r}{4 q_a^2}.
$$

(45b)

For the case of the ideal surface, $Q \to \infty$, the TM contribution will be the same, but the contribution of TE mode changes from logarithm to first inverse power

$$
e_1^{TE}(a) \approx -\frac{q_a^2}{2(1 - \chi^2)} \approx -\frac{q_a^2}{8r}.
$$

(45c)

Therefore, for short distances $d \ll R$ and finite conductivity $Q > 0$ (or $Q \to \infty$), the main contribution comes from TM polarization and it reads

$$
\tilde{f}_1^{TM} \approx -\frac{a(0)}{4d^2} \frac{h_{0k}}{e^{\nu k} - 1}.
$$

(46)

If $Q = 0$, we have

$$
e_1^{TM}(a) \approx \frac{1 - 5\chi^2 - 17\chi^4 - 27\chi^6}{8q_a^2 \chi^2(\chi^2 - 1)^4} \approx -\frac{3}{8q_a^2 r^4},
$$

(47)

and therefore for $Q \to 0$, we obtain

$$
\tilde{f}_1^{TM} \approx \frac{3Qa(0)R}{16q_a^2 d^4} \frac{h_{0k}}{e^{\nu k} - 1} \to 0
$$

(48)

as should be the case.

Let us consider equation (23). By using the same asymptotic as above, we observe that the series over $l$ in function $e_2(\alpha l)$ is convergent for any $r$ including $r = 0$. Indeed, in the limit $l \to \infty$, we have

$$
-\nu \left[ g_i^{TE}(ial) - g_i^{TE}(-ial) \right] \approx \frac{q_a^2 \chi l^2}{2\nu} \left( \frac{et q_a}{2\nu} \right)^{2\nu},
$$

(49)

$$
-\nu \left[ g_i^{TM}(ial) - g_i^{TM}(-ial) \right] \approx \frac{\nu^2}{Q \chi} \left( \frac{et q_a}{2\nu \chi} \right)^{2\nu}.
$$

(50)

Therefore, the series is convergent as $l^{-l}$ even for $r = 0$ ($\chi = 1$) and this term gives a finite contribution and we may throw it away comparing with terms considered above.
The term with zero-temperature contribution (21) may be considered in the same way. In
the limit \( r \to 0 \), the series is divergent for large \( l \). For this reason, we use the Debye uniform
expansion formulas from [35] in which we change \( z \to z/\nu \) and make the limit \( \nu \to \infty \):

\[
I_\nu(z) \approx \frac{1}{2\pi \nu} \left( \frac{\nu z}{2} \right)^\nu, \tag{51a}
\]

\[
K_\nu(z) \approx \frac{\pi}{2\pi \nu} \left( \frac{\nu z}{2} \right)^{-\nu}. \tag{51b}
\]

Taking into account these formulas in the expression for \( g_l(k) \), we obtain

\[
\nu^3 \chi - 1 - 2\nu + 2 \nu Q + 2(kR)^2 \approx \frac{\pi}{2\nu} \sqrt{\frac{Q}{\nu}} + \sqrt{2\nu} q_a, \tag{52}
\]

and obtain series

\[
\sum_{l=1}^{\infty} \frac{\pi}{2\nu} \sqrt{\frac{Q}{\nu}} (2) = \frac{\pi}{2\nu} \sqrt{\frac{Q}{\nu}} + \sqrt{2\nu} q_a, \tag{53}
\]

in which we take limit of short distances, \( \chi = 1 + r \to 1 \). If \( Q \neq 0 \), we obtain

\[
\sum_{l=1}^{\infty} \frac{\pi}{2\nu} v^2 \chi (1 - 2\nu) \approx \frac{\pi}{8\nu} \frac{1}{\chi^2} (\chi^2 - 1)^3 \approx \frac{\pi}{8\nu} q_a. \tag{54}
\]

Therefore, we arrive with expression

\[
E_0 = -\frac{\hbar \omega a(0)}{8d^3}, \tag{55}
\]

which is valid for \( r = d/R \ll Q = \Omega R \). In the case of \( Q = 0 \), we obtain from equation (54)
that \( E_0 = 0 \).

Therefore, for \( Q > 0 \) and \( r = d/R \ll Q \) we obtain free energy

\[
F_a = -\frac{\hbar \omega a(0)}{8d^3} + \frac{\hbar \omega}{\nu} - 1, \tag{56}
\]

This expression is in agreement with high- and low-temperature expansions. Indeed,
taking the corresponding limits in (57), we obtain

\[
F_a = -\frac{\hbar \omega a(0)}{4d^3}, \quad T \to \infty, \tag{57a}
\]

\[
F_a = -\frac{\hbar \omega a(0)}{8d^3}, \quad T \to 0, \tag{57b}
\]

which coincide with equations (41) and (37).

4. The entropy

Taking the minus derivative with respect to the temperature (with \( k_B \)) from the free energy
given by equations (23), we obtain expression for entropy \( S = S_1 + S_2 \) (\( a = T_m/T \)):

\[
S_1 = \frac{\alpha(0)}{R^3} \frac{e_1(a)}{(1 + r)^2} \left( \frac{\pi a}{\sinh \pi a} \right)^2, \tag{58a}
\]

(58a)
Figure 3. The plot of the entropy $S$ calculated for the hydrogen atom and the sphere with parameters of the fullerene C$_{60}$ and distance from fullerene $d = R/2$ ($r = 0.5$). The entropy for large interval of temperature is shown in (a). In (b) we show the entropy for low temperature. The exact numerical calculation is upper curve and lower curve is an approximate expression (60b).

$$S_2 = \frac{\alpha(0)}{R^3} \frac{1}{\pi(1+r)^2} \int_0^{\infty} \left\{ t e'_2 + e_2 (1 - 2 \pi at \coth \pi at) \right\} \ln \left| \frac{1+t}{1-t} \left( \frac{\pi a}{\sinh \pi a} \right)^2 \right| dt. \quad (59b)$$

The entropy at low and high temperatures may be found from equations (36) and (40), namely

$$S = 16\pi^3 \frac{\alpha(0)}{15} \left\{ k_B T \right\}^3 = \frac{2}{15} \frac{\alpha(0) k_0^3}{(1+r)^6} \left( \frac{T}{T_0} \right)^3, \quad T \ll T_R, T_\omega, T_{R+d}, \quad (60a)$$

$$S = \frac{\alpha(0)}{R^3} \frac{6r^4 + 24r^3 + 33r^2 + 18r + 4}{2r^3(r+1)^4(r+2)^3}, \quad T \gg T_R, T_\omega, T_{R+d}. \quad (60b)$$

The typical behavior of the entropy is shown in figure 3 for sphere with parameters of the fullerene C$_{60}$ and hydrogen atom with distance between them $d = R/2$, ($r = 0.5$). For this situation, we have the following numbers: $R = 3.42$ Å, $Q = \Omega R = 4.94 \times 10^{-2}$, $\alpha(0) = 0.667$ Å$^3$, $q_o = \omega_o R/c = 0.0202$.

From Figure 3 we observe that the Casimir–Polder entropy tends to zero as the third power of the temperature and it is zero at zero temperature according the Nernst heat theorem. It is positive for arbitrary temperatures. In [36] we observed the negative sign of the entropy for short separations, $r \to 0$, and low temperatures in the case of the atom with plate. We note that the low-temperature dependence given by equation (60a) is valid for $T \ll T_R = h c / 2 \pi k_B R$.

To compare with the case of atom near the flat boundary considered in [36], we should take the limit $R \to \infty$ and in this case the relation $T \ll T_R = h c / 2 \pi k_B R$ is no longer valid.

To show the appearance of the positive entropy for finite radius of sphere and negative sign of entropy for infinite radius of sphere, we use expansion over $1/R$ obtained above (27). For the case of static polarizability, $\alpha(\omega) = \alpha(0)$, we obtain from equation (27)

$$\mathcal{F} = E_{CP} \left( \eta_0 + \frac{d}{R} \eta_1 \right),$$

where

$$\eta_0 = \frac{\tau}{6} \left\{ 1 + \frac{2}{e^\tau - 1} + \frac{2\pi e^\tau}{(e^\tau - 1)^2} + \frac{\tau^2 e^\tau (e^\tau + 1)}{(e^\tau - 1)^3} \right\},$$
The plot of the function $\sigma$ calculated for different values of $r = d/R = 0, 0.05, 0.1$. Plot (a) shows the function for large domain of $\tau = 4\pi k_B T / \hbar c$. In (b) we show the function $\sigma$ close to the origin. Starting with $r = d/R = 0.085$, the entropy becomes completely positive.

$$\eta_1 = -\frac{\tau}{6} \left\{ 1 + \frac{2}{\tau} \left( e^{\tau} - 1 \right) + \frac{3\tau e^\tau (e^\tau + 1)}{2(\tau^2 - 1)^2} + \frac{\tau^3 e^\tau (e^\tau + 4e^\tau + 1)}{2(\tau^2 - 1)^4} \right\}$$

$$+ \frac{\tau^5}{16} \int_{\tau/2}^{\infty} \frac{dr}{r \sinh^2 r} + \frac{\tau^3 (\tau^2 - 4)}{48} \int_{\tau/2}^{\infty} \frac{dt \cosh t}{t \sinh^3 t}$$

and $\tau = 2T / T_d = 4\pi k_B T / \hbar c$. The function $\eta_0$ was found in [36]; the function $\eta_1$ gives the correction due to finite radius of sphere. Now we use these expressions to calculate the entropy. Taking the minus derivative with respect to the $k_B T$, we obtain the following expression for entropy:

$$S = \frac{3\alpha(0)}{2d^3} - \sigma,$$

where $\sigma = \eta_{0,\tau} + r\eta_1$. The numerical calculations of the entropy for large $R$ by using formula (61) for different values of $r = d/R = 0, 0.05, 0.1$ are shown in figure 4. Numerically, we found that for $r = d/R > 0.085$ the entropy becomes completely positive.

5. Conclusion

In the above sections we considered in detail the thermal Casimir–Polder interaction of an atom with the spherical conductive surface which models fullerene. For a sphere, we use the plasma (hydrodynamic) model [20, 21]: the sphere is a two-dimensional conductive surface with finite conductivity. All information about the sphere is encoded in one parameter $\Omega = 4\pi n e^2 / mc^2$ with dimension of wave number. To obtain the free energy, we adopt the Lifshitz approach in the framework of which the spherical shell is situated inside the spherical cavity into the infinite dielectric media. Rarefying this media, we obtain the free energy per unit atom. The renormalization procedure is simple—we throw away all terms which survive in the limit $\Omega \to 0$ because this case corresponds to the case without a sphere and the energy should be zero. The opposite case $\Omega \to \infty$ corresponds to the ideal case. The general expression for the free energy is given by equation (14) or equation (23).

We made analysis of this expression for different cases. In the limit of infinite radius, $R$, of the sphere with finite distance, $d$, between an atom and sphere, we obtain the expected expression for the energy for the case of the atom close to the plate. We also found the next term
of expansion $\sim d/R$. In the problem under consideration, we have three different parameters with dimension of the temperature:

$$T_\omega = \frac{\hbar \omega_a}{2\pi k_B}, \quad T_R = \frac{\hbar c}{2\pi k_B R}, \quad T_d = \frac{\hbar c}{2\pi k_B d}.$$  \hfill (63)

Each temperature connects with a specific scale, namely first connects with the scale of atom, the second—with the scale of sphere and the third connects with the distance between an atom and the sphere. For a molecule of fullerene $C_{60}$, we have $T_\omega = 2.15 \times 10^4$ K, $T_R = 1.06 \times 10^6$ K.

For low temperatures, $T \ll T_\omega, T_R, T_d$ we obtain (see equation (37)) that the free energy has a temperature correction proportional to the fourth power of the temperature $\sim T^4$. The high-temperature expansion, $T \gg T_\omega, T_R, T_d$ given by equation (40) reveals the first power of the temperature. Nearby to the sphere, the energy has the form given by equation (57). It has the form of the sum of the zero-temperature term and the Plank form temperature correction.

The entropy of this system shows interesting behavior (see figure 3). The entropy is positive for any distance between an atom and sphere of small radius. It tends to zero for low temperatures according to the Nernst heat theorem and it tends to constant for high temperatures. It has been shown in [36] that the entropy is negative for short distances between an atom and the flat boundary. To reveal the sign changing, we found expansion of the entropy over $d/R$ up to the first power of $d/R$. The analytical and numerical analysis shows that the entropy indeed has the region of the negative sign for large radii of the sphere and becomes completely positive beginning with some definite value of $d/R$ (see figure 4).

As noted in the introduction, the hydrodynamical (plasma) approach has limited applications and the more realistic model to describe the interaction between an atom with fullerene is the Dirac model. It is based on the $(2 + 1)$-dimensional Dirac equation with a very small parameter of the mass and the Fermi velocity instead of the velocity of light. The future aim of our consideration is the Casimir–Polder interaction in the framework of the Dirac model.

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