The Casimir Problem of Spherical Dielectrics: A Solution in Terms of Quantum Statistical Mechanics

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The Casimir energy for a compact dielectric sphere is considered in a novel way, using the quantum statistical method introduced by Høye and Stell and others. Dilute media are assumed. It turns out that this method is a very powerful one: we are actually able to derive an expression for the Casimir energy that contains also the negative part resulting from the attractive van der Waals forces between the molecules. It is precisely this part of the Casimir energy that has turned out to be so difficult to extract from the formalism when using the conventional field-theoretic methods for a continuous medium. Assuming a frequency cutoff, our results are in agreement with those recently obtained by G. Barton.

1. INTRODUCTION

It is a pleasure to contribute this article to a festschrift volume for Professor Stell. The article is one in a series of articles published by the present authors on the Casimir effect and related topics, using methods of statistical mechanics for quantized systems at thermal equilibrium. Besides contributions from others, these methods were developed by one of the authors (JSH) in cooperation with Professor Stell in their extensive studies

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of polar and polarizable fluids through several years. This long-lasting cooperation, which was initiated in 1972, is still active today.

The Casimir energy problem for a compact spherical ball is a many-facetted problem; the formal solution of it is to an unusual degree dependent on the mathematical method of approach chosen. The Casimir effect as such is now a well-known effect in physics.[1] It is ordinarily examined with the use of field theory in dielectric media, allowing the medium to possess a refractive index n (even dispersive effects can in principle be dealt with in this way, if n is assumed to depend on the frequency). The standard configuration does not involve curved boundaries at all, but consists instead of two plane plates separated by a small gap. In this geometrical configuration the phenomenological electromagnetic theory, as constructed mainly by Lifshitz[2] is fully adequate, and leads to a prediction for the Casimir force between the plates that has recently been verified experimentally to an impressive accuracy of about one per cent.[3, 4]

If we now leave the parallel-plate configuration and consider instead a single dielectric ball, the situation becomes much less clear-cut. The history along this direction of research may be taken to start with the calculation of Boyer on a singular perfectly conducting *shell*:[5] he found the Casimir energy E to be positive, corresponding to an *outward* directed surface force. Later, the dielectric ball was considered by Milton,[6–8] Milton and Ng,[9, 10] Brevik *et al.*[11–14] and several others. Some consensus seems by now to have been reached as regards the Casimir energy E as found by field theoretical methods: this energy is positive, corresponding to a repulsive force, and is given by

$$E = \frac{23}{384} \frac{hc}{\pi a} (n-1)^2 \qquad (1)$$

for a dilute sphere whose radius is a.

Faced with this field theoretical result one becomes however surprised, for the following physical reason: the Casimir energy should be the cooperative result of the van der Waals forces between the molecules in the ball. The van der Waals forces are necessarily *attractive*. How can these forces sum up to give a *repulsive* total surface force? The natural answer to this question is that the field theoretical calculation, based as it is on a continuum model for the dielectric, is unable to cope with the attractive part. In other words, the attractive terms are necessarily lost in the regularization process. An important progress was recently made by Barton;[15] he made use of quantum mechanical perturbation theory to second order, imposed an exponential cutoff in wave numbers, and

arrived at a definite expression for the Casimir energy containing also the cutoff dependent, attractive (and actually also repulsive) terms. Moreover, a cutoff independent, repulsive term was contained in the energy expression, which was in precise agreement with Eq. (1) above. There are actually some indications of the same kind already in the paper of Milton and Ng:[10] they derived the cutoff independent Casimir energy starting from the van der Waals forces, omitting the divergent terms.

And this brings us to the central theme of the present paper, namely to rederive the expression for the Casimir energy using the perhaps somewhat more unconventional quantum statistical methods that were developed by Høye and Stell, and others. Central references for the present work are refs. 16 and 17. Others that also include evaluation of frequency spectra are ref. 18. As we will see, this method is very powerful, and we will be able to establish contact with the results of Barton. The line of development of the application of this method to the Casimir problem is the following: Some years ago Brevik and Høye[19] showed that the Casimir energy between two point particles is the same as the free energy due to two quantized fluctuating dipole moments interacting via the dipolar radiation interaction (zero frequency limit corresponds to the static dipole-dipole interaction). Later, Høye and Brevik[20] extended this method to evaluate the Casimir force between a pair of parallel dielectric plates separated by a small gap. Performing this more complex calculation with the use of statistical mechanics for systems in thermal equilibrium, we were able to rederive the known results.

Below we will evaluate the free energy in a dielectric dilute medium, again using the same methods of statistical mechanics. Based on this we will make contact with the results of Barton, as mentioned, as well as with the results obtained in field theory. On the basis of our method the physical origin of the divergences is easily understood. The problem, as anticipated above, has its origin in a continuum description of dielectric media, while a realistic system has to have a microscopic structure involving a minimum separation between molecules due to repulsive cores.

2. BASIC FORMALISM

We begin by recapitulating some of the basic formulas from our earlier work.[19] For a pair of polarizable particles the free energy F due to their mutual interaction is

$$-\beta F = \tfrac{3}{2} \sum_K \alpha_K^2 (2\psi_{DK}^2(r) + \psi_{\Delta K}^2(r)) \tag{2}$$

cf. Eq. (5.14) in ref. 19. Here α_K is the frequency dependent polarizability, and

$$K = 2\pi n/\beta \tag{3}$$

is the Matsubara frequency related to the frequency ω via

$$K = -i\hbar\omega \tag{4}$$

Further, n is an integer, $\beta = 1/k_B T$ is the inverse temperature, and ψ_{DK} and ψ_{AK} are the two radial parts of the radiating dipole-dipole interaction as given by Eqs. (5.9) and (5.10) in ref. 19. Performing the sum in (2) for $\beta \to \infty$ (i.e., $T \to 0$), we obtain Eq. (5.16) in ref. 19

$$F = -\frac{23\hbar c \alpha^2}{4\pi r^7} \tag{5}$$

which is the known result for the Casimir effect.

For a low density medium the total free energy Δ can now be obtained by summing or integrating (2) over pairs of particles in a volume V such that

$$\Delta = \tfrac{1}{2}\,\rho^2 \int d\mathbf{r}_1\, d\mathbf{r}_2\, F \tag{6}$$

where ρ is the number density and $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$. At $T = 0$, (5) is to be inserted. Clearly, the integral will diverge, due to the behaviour for small values of r. However by integrating over a small sphere a finite term, which is positive, can be separated out; cf. ref. 10 and 13. This finite term turns out to coincide with the field theoretical result. We will show below that the divergences found using other kinds of approach are connected with this small r behaviour. Equation (2), together with (6), will be shown to lead to results in agreement with those obtained from quantum mechanical perturbation theory to second order [15]. The exponential cutoff used by Barton in Fourier space can be introduced also in our approach; it corresponds to a small "soft" r cut out from the otherwise continuous medium, and will be a rough approximation to real systems. As mentioned above, real systems are not continuous but consist of molecules that have a minimum separation due to hard cores.

3. CALCULATION OF THE FREE ENERGY

Let us now calculate the free energy Δ, as given by (6). In order to establish connection with perturbation theory, we first represent (2) in

terms of Fourier quantities for which, as we will see, a wave vector cutoff can be introduced easily. The radiating dipole-dipole interaction used in (2) can be written as

$$\psi(12) = \psi_{DK}(r)\, D_K(12) + \psi_{\varDelta K}(r)\, \varDelta_K(12) \tag{7}$$

with

$$D_K(12) = 3(\hat{r}\hat{a}_{1K})(\hat{r}\hat{a}_{2K}) - \hat{a}_{1K}\hat{a}_{2K}$$

$$\varDelta_K(12) = \hat{a}_{1K}\hat{a}_{2K}$$

Here the hats denote unit vectors, and $\mathbf{a}_{iK}$ is the Fourier transform of the fluctuating dipole moment of particle number i in imaginary time; cf. Eq. (5.2) in ref. 19. Equation (7) can be Fourier transformed to give

$$\tilde{\psi}(12) = \tilde{\psi}_{\varDelta K}(k)\, \tilde{D}_K(12) + \tilde{\psi}_{\varDelta K}(k)\, \varDelta_K(12) \tag{8}$$

with

$$\tilde{D}_K(12) = 3(\hat{k}\hat{a}_{1K})(\hat{k}\hat{a}_{2K}) - \hat{a}_{1K}\hat{a}_{2K}$$

$$\psi(12) = \frac{1}{(2\pi)^3} \int \tilde{\psi}(12)\, e^{i\mathbf{k}\mathbf{r}}\, d\mathbf{k}$$

With this Eq. (2) can be rewritten as

$$-\beta F = \frac{3}{2} \frac{1}{(2\pi)^6} \sum_K \int M_K e^{i(\mathbf{k}+\mathbf{k}')\,\mathbf{r}}\, d\mathbf{k}\, d\mathbf{k}' \tag{9}$$

where

$$M_K = \tilde{\psi}_{\varDelta K}(k)\, \tilde{\psi}_{\varDelta K}(k')(3\hat{k}\hat{k}' - 1) + \tilde{\psi}_{\varDelta K}(k)\, \tilde{\psi}_{\varDelta K}(k') \tag{10}$$

Like expression (2), this is obtained after orientational averaging of the products of terms containing $\tilde{D}_K(12)$ and $\varDelta_K(12)$ with respect to $\hat{a}_{iK}$.

To get further the explicit Fourier transformed interaction terms are needed. These follow from the solution of Maxwell's equations, and like the corresponding terms in (2) (Eq. (5.10) in ref. 19) they were used by Høye and Stell when dealing with the refractive index of fluids.[17] Thus from Eq. (7) in ref. 17 we have[3]

$$\tilde{\psi}_{DK}(k) = -\frac{4\pi}{3} \frac{k^2}{k^2 - \omega^2}, \qquad \tilde{\psi}_{\varDelta K}(k) = \frac{4\pi}{3} \left(\frac{2k^2}{k^2 - \omega^2} - (2 + \Theta) \right) \tag{11}$$

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[3] Note here that the Fourier transform in imaginary time on the interval from 0 to β is the same function as the real time transform; cf. the derivation in Appendix B of ref. 19.

with $K = -ihc\omega$. (For simplicity ω is replaced by $c\omega$ where c is the light velocity.) Here the parameter Θ introduced by Høye and Stell[21] is used. A purpose to introduce it was by γ-parametrization of the dipole-dipole interaction to obtain a continuous family of mean field theories ($\gamma \to 0$) of polar fluids. Here γ is the inverse range of $\psi_{AK}(r)$, and for $\psi_{DK}(r)$ it is the inverse range inside which the dipolar $1/r^3$ behaviour is cut or rounded off. As seen from (11) the Θ is thus the integrated amplitude of the ψ_{AK}-term ($\omega = 0$). This parameter was also used in ref. 17, part IV, and its Eq. (57) for the direct correlation function corresponds to Eq. (11) here. With its Eqs. (56) and (59) the dielectric constant ε can then in general be expressed as (for small $\omega/\gamma \to 0$)

$$\frac{\varepsilon - 1}{(1 - \Theta)\,\varepsilon + (2 + \Theta)} = \frac{4\pi}{3}\,\rho\alpha \tag{12}$$

where ρ is the number density of particles. (Here a possible density dependence of $\alpha \to \alpha_{\text{eff}}$ which is proportional to the fluctuating dipole moment squared, is disregarded.)

The separate term $(2 + \Theta)$ at the end of Eq. (11) will necessarily yield infinity when inserted in (9) and summed. In **r**-space it gives a δ-function at $\mathbf{r} = 0$. As pairs of particles in reality are separated, we can simply remove this term here. This amounts to putting $2 + \Theta = 0$, by which

$$\gamma = \frac{\varepsilon - 1}{\varepsilon} = 4\pi\rho\alpha \tag{13}$$

(The quantity γ in Eq. (13) is not the γ used in the γ-parametrization.) Note here that this choice is consistent with the continuum approach ($\gamma \to \infty$), where only a transverse radiating field is implicitly considered. That is, with $\Theta = -2$ the longitudinal part vanishes, as follows from Eq. (28) in ref. 17. Then one has (with the direct correlation function $c \to \psi$) $\tilde{\psi}_1 = \frac{1}{3}(\tilde{\psi}_{AK} + 2\tilde{\psi}_{DK}) = 0$, while the transverse part becomes $\tilde{\psi}_2 = \frac{1}{3}(\tilde{\psi}_{AK} - \tilde{\psi}_{DK}) = (4\pi/3)\,k^2/(k^2 - \omega^2)$.

Inserting (11) into (10) we obtain

$$M_K = \frac{(4\pi)^2}{3} \frac{k^2}{k^2 - \omega^2} \frac{k'^2}{k'^2 - \omega^2} \left[(\hat{k}\hat{k}')^2 + 1 \right]$$

$$= \frac{(4\pi)^2}{3} \frac{k^2 k'^2}{k^2 - k'^2} \left[\frac{1}{k'^2 - \omega^2} - \frac{1}{k^2 - \omega^2} \right] \left[(\hat{k}\hat{k}')^2 + 1 \right] \tag{14}$$

Now the summation in (9) can be considered, and by restricting ourselves to frequency independent polarizability $\alpha_K = \alpha$ we can easily perform the

sum since (14) is of the standard form for simple harmonic oscillators.[16] We have $(K = 2\pi n/\beta)$

$$\sum_K \frac{(\hbar\omega_0)^2}{(\hbar\omega_0)^2 + K^2} = \frac{1}{2} \beta\hbar\omega_0 \frac{\cosh(\frac{1}{2}\beta\hbar\omega_0)}{\sinh(\frac{1}{2}\beta\hbar\omega_0)} \xrightarrow{\beta \to \infty} \frac{1}{2} \beta\hbar\omega_0 \qquad (15)$$

With $\alpha_K = \alpha$, use of (15), and (14) inserted for M_K, we now easily find at $T = 0$ $(K = -i\hbar c\omega, \hbar\omega_0 \to \hbar ck,$ and $\hbar ck')$

$$\sum_K M_K = \frac{(4\pi)^2}{3} \frac{1}{k^2 - k'^2} \frac{1}{2} \beta\hbar c(k'k^2 - kk'^2)[(\hat{k}\hat{k}')^2 + 1]$$

$$= \beta\hbar c \frac{(4\pi)^2}{6} \frac{kk'}{k+k'} [(\hat{k}\hat{k}')^2 + 1] \qquad (16)$$

When inserting this into (9), and further inserting into (6), we see that the result will diverge due to the small r (or large k) behaviour. This divergence can be avoided by introducing a large wave number cutoff, as Barton did.[15] Thus we incorporate a factor $\exp(-\lambda k)$ in the interaction terms (11), which implies a factor $\exp(-\lambda(k+k'))$ in (16). Regarding the electromagnetic field as a set of harmonic oscillators that mediate the interaction between the particles, the effect of this cutoff is to remove the high frequency oscillators. With (16) and (9) inserted into (6) we then obtain

$$\Delta = -\frac{\gamma^2}{2(16\pi^3)^2} \int d\mathbf{r}_1 \, d\mathbf{r}_2 \int d\mathbf{k} \, d\mathbf{k}' \, kk' e^{i(\mathbf{k}+\mathbf{k}')\mathbf{r}} \frac{e^{-\lambda(k+k')}}{k+k'} [(\hat{k}\hat{k}')^2 + 1] \qquad (17)$$

where γ is given by (13).

With $\Delta = \gamma^2 \Delta_2$ this is precisely the result obtained by Barton[15] applying quantum mechanical perturbation theory to the dielectric continuum. That is, we have recovered the second order contribution which in the case of a scalar field is given by Eq. (4.2) in ref. 15 where the equality

$$\frac{\exp(-\lambda(k+k'))}{k+k'} = \int_\lambda^\infty d\xi \exp(-\xi(k+k')) \qquad (18)$$

has been used. When dealing with the electromagnetic field, as we do here, the result in ref. 15 is modified with the factor $[(\hat{k}\hat{k}')^2 + 1]$ as given by Eq. (B.2) in ref. 15. With this, it is seen that our results are identical with those of Barton. Thus, further considerations based upon this basic agreement will necessarily be the same, and will not be repeated here.

4. FURTHER REMARKS

Let us make a few remarks on the resulting free energy (or internal energy at $T = 0$) for a spherical body of radius a where a positive cutoff independent term going like $1/a$ shows up also within the field theoretical approach. This is commonly interpreted as equivalent to a repulsive Casimir surface force. From our approach it is now obvious that this term reflects the large r-behaviour of the free energy (5) of particle pairs. For a finite system such as a sphere the resulting free energy will in general be *larger* than its bulk value since there is no material outside with which it can interact. This missing interation first of all manifests itself in a positive term that reflects the surface tension and is proportional to the surface area. This term is present in the final result given in the Abstract in ref. 15. In additon, there is the $1/a$ term which can be associated with the $1/r^7$-tail in connection with the missing material away from the surface.

As another point, let us note that in ref. 15 there is an additional leading term due to first order perturbation theory. This term is not present in our derivation above. However it can be identified in a straightforward manner from the earlier work of Høye and Stell.[17] It represents a self-energy of the electromagnetic field attached to the polarizable particles. As such it is just part of the properties of isolated single particles, and should accordingly not be included in the free energy density above. In fact, it is part of the radiation reaction of accelerated charges and is incorporated in the resulting physical momentum. Unfortunately this momentum correction for a classical particle is infinite, so the "bare" mass of particles is negative resulting in the well known "runaway" problem in classical electromagnetism.[22] This also reflects itself in the refractive index problem considered in ref. 17. Further, Høye and Lomba made numerical calculations to obtain a minor non-causal tail in the dielectric response of the fluid (i.e., a minor response will appear before an electric field is applied to avoid exponential growth or runaway for increasing time).[23]

With cutoff in wave vectors the above mass correction can be made finite. The term of interest is then the component $\widetilde{\psi}_{\varLambda K}(k)$ of Eq. (8). Including the shielding factor in (11) we then have (with $2 + \varTheta = 0$ as above)

$$\widetilde{\psi}_{\varLambda K}(k) = \frac{4\pi}{3} \frac{2k^2}{k^2 - \omega^2} e^{-\lambda k} \qquad (19)$$

As shown by Eqs. (12)–(15) of ref. 17, this gives rise to a self-interaction (for given K, $s \to a_K$)

$$\varDelta\phi(a_K) = -\tfrac{1}{2} a_K^2 \psi_{\varLambda K}(0) \qquad (20)$$

where the minus sign is due to the definition used. The total internal energy contribution that follows from this is

$$\Delta_1 = \sum_K \rho V \langle \Delta \phi(a_K) \rangle = -\tfrac{1}{2} \rho V \sum_K \langle a_K^2 \rangle \psi_{\Delta K}(0) \tag{21}$$

where ρV is the number of particles in a volume V.

Now $\langle a_K^2 \rangle = 3\alpha_k/\beta$; cf. ref. 19. So with $\alpha_K = \alpha$ we obtain by first using (15) for $\beta \to \infty$

$$\sum_K \tilde{\psi}_{\Delta K}(k) = \frac{4\pi}{3} \beta h c k e^{-\lambda k}$$

or

$$\sum_K \psi_{\Delta K}(0) = \frac{4\pi}{3} \frac{\beta h c}{(2\pi)^3} \int k e^{-\lambda k} \, d\mathbf{k} = \frac{4}{\pi} \frac{\beta h c}{\lambda^4} \tag{22}$$

which together with (13) and (21) yield

$$\Delta_1 = -\frac{1}{2} \rho V \frac{3\alpha}{\beta} \frac{4}{\pi} \frac{\beta h c}{\lambda^4} = -\gamma \frac{3}{2\pi^2} \frac{h c}{\lambda^4} V \tag{23}$$

This is precisely the first order contribution obtained by Barton.[15] As mentioned above, this energy is a quantity that is part of the free particles themselves, and can thus not be separated out. So the free energy of the interacting system of polarizable particles will not include this term.

After the above was written, we have become aware that the recent paper of Bordag, Kirsten, and Vassilevich[24] arrives at results closely related to those obtained by Barton.[15] The authors of ref. 24 make use of field theoretical path integral methods which are in themselves quite different from those used in ref. 15 as well as in the present paper, but the expression for the Casimir energy obtained in Section IV in ref. 24 nevertheless parallels that obtained in ref. 15. (There are some differences in numerical coefficients of the divergent terms, due to different regularization methods employed.) We thank the authors of ref. 24 for making us aware of this correspondence.

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