CASIMIR SURFACE FORCE ON A DILUTE DIELECTRIC BALL

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Abstract

The Casimir surface force density $F$ on a dielectric dilute spherical ball of radius $a$, surrounded by a vacuum, is calculated at zero temperature. We treat $(n - 1)$ ($n$ being the refractive index) as a small parameter. The dispersive properties of the material are taken into account by adopting a simple dispersion relation, involving a sharp high frequency cutoff at $\omega = \omega_0$. For a nondispersive medium there appears (after regularization) a finite, physical, force $F_{\text{nondisp}}$ which is repulsive. By means of a uniform asymptotic expansion of the Riccati-Bessel functions we calculate $F_{\text{nondisp}}$ up to the fourth order in $1/\nu$. For a dispersive medium the main part of the force $F_{\text{disp}}$ is also repulsive. The dominant term in $F_{\text{disp}}$ is proportional to $(n - 1)^2 \omega_0^3/a$, and will under usual physical conditions outweigh $F_{\text{nondisp}}$ by several orders of magnitude.

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1 Introduction

Consider a compact dielectric nonmagnetic ball of radius $a$, surrounded by a vacuum. The permittivity of the material is $\varepsilon = n^2$, $n$ being the refractive index. The purpose of the present work is to calculate the “ordinary” Casimir surface density $F$ on the sphere (i.e. with the exception of the electrostrictive force), at zero temperature, making use of the same calculational technique as in an earlier work [1]. For mathematically simplifying reasons we will be concerned only with the case of dilute media, meaning that $n - 1 \ll 1$. As in [1], we will make use of the uniform asymptotic expansion (also called the Debye expansion) for the modified Bessel functions [2]. Here $1/\nu$ is the expansion parameter, with $\nu = l + 1/2$, $l = 1, 2, 3, \ldots$. In the main body of this paper we will expand the formalism to second order in $(n - 1)$, and to fourth order in $1/\nu$. In Appendix A we carry out the expansion in $(n - 1)$ three orders further, to the fifth order, at the price, however, of retaining the Debye expansion to second order in $1/\nu$. This treatment improves considerably on the theory given in [1]. We use the mathematical package Maple V Release 4 Power Edition. It turns out to be necessary to keep relatively high accuracy in this problem, since subsequent terms in the uniform asymptotic expansion alternate in sign. Thus, if one simply truncates the series after a few terms, without observing this alternating effect, one risks getting even the sign in the final force wrong.

The ultimate goal in a calculation of this type is to get a Casimir force expression that is directly comparable to experiment; if not a real experiment, at least a Gedanken experiment. To achieve this goal, one must observe that there are several contributions to the surface force. Let us try to put the things into perspective, by writing down the general expression for the electromagnetic volume force density $f$ in a charge-free nonmagnetic medium (cf., for instance, the book [3] or the review [4])

$$f = -\frac{1}{2} E^2 \nabla \varepsilon + \frac{1}{2} \nabla \left[ E^2 \rho \left( \frac{\partial \varepsilon}{\partial \rho} \right)_T \right] + \frac{\varepsilon - 1}{c} \frac{\partial}{\partial t} (E \times H). \quad (1)$$

Here $\rho$ is the mass density of the fluid.

Consider the first term in (1). We may call it the Abraham-Minkowski force. It is equal to zero in the interior homogeneous region of the medium and acts in the boundary region around $r = a$ only. From thermodynamic perturbation theory we know in general that $\varepsilon > 1$ for a dielectric at zero frequency [3], Sect.14, and we expect the same to be true for moderate and high frequencies also, except possibly from special frequency bands. That is, since the sphere is surrounded by a vacuum, we would expect the first term in (1) to give a repulsive surface force. This turns out to be true also, in

classical theory. In quantum theory, however, we cannot be sure in advance about the direction of the force; the result depends on the magnitude of the contact term being subtracted off in the regularization procedure. One has to carry out the calculation in detail in order to determine even the sign.

The Abraham - Minkowski force can in turn be divided into two parts:

(1) A nondispersive part. It corresponds to letting the ultraviolet (nondimensional) cutoff frequency $x_0$ go to infinity. The early works on the Casimir effect took into account this part of the force only. The nondispersive force is formally divergent, but can usually be easily regularized. The recipe is to (i) put $x_0 = \infty$; (ii) evaluate the remaining divergent sum over $l$, from $l = 1$ to $l = \infty$, by the Riemann zeta - function method. We stressed the efficiency of this recipe in [1]; cf. also some other related prior papers [5] - [7]. The method gives, in a simple way, results that are in agreement with what can be obtained in other ways. The quantum nondispersive force turns out in our calculation to be repulsive, thus qualitatively in agreement with the classical result noted above.

(2) There is a dispersive part of the Abraham - Minkowski force, caused by the cutoff $x_0$. In an earlier work on a spherical shell [4] we actually found the dispersive force to be attractive. The dispersive force is under usual physical conditions *stronger* than the nondispersive force. Candelas [8] seems to have been the first to emphasize the importance of the dispersive force. His general ideas were later on essentially supported by explicit model calculations [3], [4].

Consider next the second term in (1). This is the *electrostriction* force. It generally acts inwards. Whether this force is detectable in a real experiment or not, depends on the detailed circumstances. Usually, there is established a compensating force from the elastic pressure in the interior of the fluid, so that the effect from electrostriction does not show up explicitly in the measurements. Only under special conditions, such as in the Goetz - Zahn non-equilibrium experiment testing the attractive force between two condenser plates immersed in a dielectric liquid when there is a high-frequency electric field between the plates [11], [12], does one have the possibility to measure the electrostriction force. The point is that the electromagnetic force will then vary so quickly that the elastic pressure does not have sufficient time to build itself up. What determines the time scale here, is obviously the transit time for sound waves in the body. (The Goetz - Zahn experiment, as well as some other related experiments, are discussed in detail in [3].) In (1), we have deliberately written $(\partial \varepsilon/\partial \rho)_T$ as an isothermal partial derivative. This is often appropriate in practical cases, but is an assumption that has to be considered with some care since if the deformation of the body is occurring rapidly it will be more appropriate to take the adiabatic derivative $(\partial \varepsilon/\partial \rho)_S$.
instead. If the fluid is non-polar, as one can usually assume at high frequencies, then one need not distinguish between the isothermal and adiabatic cases since the permittivity depends on density only, though the Clausius-Mossotti relation. The electrostrictive contribution to the Casimir effect has not been paid much attention to in the past. There is a paper by one of us some years ago [13], and recently Milton and Ng have returned to the topic [14]. In the present case, where we are considering the Casimir force on a static spherical boundary, it is clear that a compensating elastic pressure will be built up on the inside. Because of this, most possibilities for measuring the surface force that one may conceive of, would not be able to detect the electrostrictive contribution at all. A measurement of electrostriction would require a measurement of the local pressure in the interior region.

Consider finally the third term in (1). This is the Abraham term, detectable under special circumstances at low frequencies [4], but fluctuating out at higher frequencies, especially at optical frequencies. In our case, the Abraham term does not contribute. It fluctuates out.

Summing up so far, we see that it is the Abraham-Minkowski term in (1) that gives the most important contribution to the surface force. We will be concerned with this term in the following, and will consider both subclasses (1) and (2) listed above. In the next section we highlight the general Green-function formalism, and explain the regularization method that we use. Section 3 is devoted to a study of the nondispersive force. By using the expansion technique mentioned above we derive the expression for $F_{\text{nondisp}}$, to order $1/\nu^4$, in (27). This force is repulsive. Some earlier results, derived by purely analytical means, are corrected. As regards the dispersive force, considered in Section 4, we restrict ourselves to giving essentially estimates. For high ultraviolet nondimensional cutoff $x_0$ the dispersive force is given as the sum of expressions (39) and (46). The dispersive force for a compact ball is strong, and it is repulsive. It may be as large as about $10^6 - 10^9$ times the nondispersive force. This fact does not seem to have been universally recognized before.

We put henceforth $\hbar = c = 1$. Electromagnetic Heaviside-Lorentz units are employed.

2 Basic formalism. Regularization

The general formalism was developed in our earlier paper [1], but for the benefit of the reader we will recapitulate some of the essential points.

The dispersion of the material is accounted for in a simple way, by taking the permittivity $\varepsilon(i\hat{\omega})$ as a function of imaginary frequencies $\hat{\omega}$ to be a step
function: \( \varepsilon(i\hat{\omega}) = n^2 = \text{constant} > 1 \) for \( \hat{\omega} < \omega_0 \), \( \varepsilon(i\hat{\omega}) = 1 \) for \( \hat{\omega} > \omega_0 \). Thus there is one single "absorption" frequency \( \omega_0 \), serving as an ultraviolet cutoff.

When considering the two-point functions, we let the two spatial points \( r \) and \( r' \) be separated in the radial direction. There are two scalar Green functions in the problem, \( F_l(r, r') \) and \( G_l(r, r') \), defined by

\[
\begin{align}
& r, r' < a : \quad F_l, G_l = \text{ink} j_l(nkr_) [h_l^{(1)}(nkr_) - \tilde{A}_{F,G}(ka)j_l(nkr_)] \quad (2) \\
& r, r' > a : \quad F_l, G_l = ik \left[ j_l(kr_) - \tilde{B}_{F,G}(ka)h_l^{(1)}(kr_) \right] h_l^{(1)}(kr_). \quad (3)
\end{align}
\]

Here \( k = |\omega|, j_l \) is the spherical Bessel function, and \( h_l^{(1)} \) the spherical Hankel function of the first kind. The coefficients \( \tilde{A}_{F,G} \) and \( \tilde{B}_{F,G} \) are related to the boundary.

We shall work with the two-point functions that contain the surface-induced contributions only, and thus have to subtract off the contact terms corresponding to a uniform medium. It means that the two-point functions refer to "disturbed" quantities caused by the boundaries. Specifically, we let (i) the contact force on the inner side \( r = a- \) be calculated as if the inner medium be filling all space; (ii) the contact force on the outer side \( r = a+ \) be calculated as if the outer medium be filling all space. This means that the first term in (2) has to be subtracted off as a contact term. Similarly the first term in (3) has to be subtracted off.

We stress that this method of regularization is not an arbitrary choice. It is the only kind of regularization that separates off the volume terms, and it is the only method that permits us to write the relation between the surface force \( F \) and the Casimir energy \( E \) as \( F = -(1/4\pi a^2)\partial E/\partial a \). It implies, in particular, that both the surface force and the Casimir energy goes to zero when the radius goes to infinity, what seems to us a most natural result. Our method of regularization is the same as used by Milton \[15\], \[16\], \[18\]; cf. also \[14\], \[17\]. It should be mentioned, however, that this method is different from the one advocated recently by Carlson \textit{et al.} \[19\], \[20\]. We shall call the modified two-point functions, constructed by the boundary-related parts of the Green functions only, the "effective" two-point functions, and write them as \( < >_{\text{eff}} \). We introduce the nondimensional cutoff frequency

\[
x_0 = \omega_0 a, \quad (4)
\]

and perform a complex frequency rotation

\[
k \rightarrow ik = i\hat{\omega}, \quad ka \rightarrow i\hat{\omega}a \equiv ix, \quad (5)
\]
whereby the Riccati-Bessel functions $s_l$ and $e_l$ defined by

$$s_l(x) = \sqrt{\frac{\pi x}{2}} I_\nu(x), \quad e_l(x) = \sqrt{\frac{2x}{\pi}} K_\nu(x)$$

(6)
correspond to the Wronskian $W\{s_l, e_l\} = -1$. Here $\nu = l + 1/2$, $I_\nu$ and $K_\nu$ denoting modified Bessel functions.

The effective electric field products in the limit $r' \to r$ in the interior region $r < a$ are

$$\langle E_r(r)E_r(r') \rangle_{\text{eff}} = -\frac{1}{\pi r^4} \int_0^{x_0} \frac{dx}{n^2 x} \sum_{l=1}^{\infty} \frac{2l+1}{4\pi} l(l+1) A_G(x) s_l^2(n x \tilde{r}),$$

(7)

$$\langle E_\perp(r)E_\perp(r') \rangle_{\text{eff}} = -\frac{1}{\pi r^2 a^2} \int_0^{x_0} \frac{dx}{n} \sum_{l=1}^{\infty} \frac{2l+1}{4\pi} \{ A_F(x) s_l^2(n x \tilde{r}) - A_G(x)[s_l(n x \tilde{r})]^2 \},$$

(8)

with $\tilde{r} = r/a$, $n = n(ix)$, prime meaning differentiation with respect to the whole argument. We need the expressions for the frequency-rotated coefficients:

$$A_F(x) = \frac{e_l(nx)e_l(x) - ne_l(x)e_l(nx)}{s_l(nx)e_l(x) - ne_l(x)s_l(nx)},$$

(9)

$$A_G(x) = \frac{ne_l(nx)e_l(x) - e_l(x)e_l(nx)}{ns_l(nx)e_l(x) - e_l(x)s_l(nx)}$$

(10)

(it may be noted that $\tilde{A}_{F,G}(ix) = (-1)^{l+1} A_{F,G}(x)$). Similarly in the exterior region $r > a$ we have

$$\langle E_r(r)E_r(r') \rangle_{\text{eff}} = -\frac{1}{\pi r^4} \int_0^{x_0} \frac{dx}{x} \sum_{l=1}^{\infty} \frac{2l+1}{4\pi} l(l+1) B_G(x) e_l^2(x \tilde{r}),$$

(11)

$$\langle E_\perp(r)E_\perp(r') \rangle_{\text{eff}} = -\frac{1}{\pi r^2 a^2} \int_0^{x_0} \frac{dx}{x} \sum_{l=1}^{\infty} \frac{2l+1}{4\pi} \{ B_F(x) e_l^2(x \tilde{r}) - B_G(x)[e_l(x \tilde{r})]^2 \},$$

(12)

with

$$B_F(x) = \frac{s_l(nx)s_l(x) - ns_l(x)s_l(nx)}{s_l(nx)e_l(x) - ne_l(x)s_l(nx)},$$

(13)
\[ B_G(x) = \frac{ns_l(nx)s_l'(x) - s_l(x)s_l(nx)}{ns_l(nx)e_l'(x) - e_l(x)s_l(nx)}. \]  

(14)

The surface force density, as calculated by use of the Maxwell stress tensor, is \( F = F_{\text{int}} + F_{\text{ext}} \), where the interior force is

\[ F_{\text{int}} = \frac{1}{2\pi a^4} \int_0^{x_0} n x dx \sum_{l=1}^{\infty} \frac{2l + 1}{4\pi} \Lambda_l^{\text{int}} \left[ \ln \left( \frac{s_l'(nx)}{s_l(nx)} \right) \right]', \]  

(15)

with

\[ \Lambda_l^{\text{int}} = [A_F(x) + A_G(x)]s_l(nx)s_l'(nx). \]  

(16)

Similarly the exterior force is

\[ F_{\text{ext}} = -\frac{1}{2\pi a^4} \int_0^{x_0} x dx \sum_{l=1}^{\infty} \frac{2l + 1}{4\pi} \Lambda_l^{\text{ext}} \left[ \ln \left( \frac{-e_l'(x)}{e_l(x)} \right) \right]', \]  

(17)

with

\[ \Lambda_l^{\text{ext}} = [B_F(x) + B_G(x)]e_l(x)e_l'(x). \]  

(18)

These expressions assume the step-function dispersion relation for \( n(ix) \), as explained above. The value of the constant \( n \) for \( x < x_0 \) is however at this stage arbitrary; \( (n - 1) \) is not necessarily small.

The natural question is now: are the force expressions (15) and (17) finite? The answer is no. This can be seen by means of the uniform asymptotic series to be employed below, or by direct machine calculation. Thus regularization is called for. The regularization procedure is quite straightforward, at least for the case of dilute media, the case to which we now turn.

3 Nondispersive force

We assume henceforth \( (n - 1) \) to be a small quantity, and expand the formalism, by means of Maple, up to order \( (n - 1)^2 \). We make use of the uniform asymptotic expansions for the Riccati-Bessel functions, as explained in \[\text{[1]}\]. As mentioned above, we insert these expansions up to order \( 1/\nu^4 \).

As we mentioned in Sect.1, the nondispersive case can be treated by setting \( x_0 = \infty \). We introduce the symbols

\[ z = x/\nu, \quad t(z) = (1 + z^2)^{-1/2}, \]  

(19)
and calculate in the interior region the integrand of Eq. (15), using the Debye expansion. Taking the integral from $z = 0$ to $z = \infty$ we obtain

$$\int_0^{\infty} dz \nu^3 z \Lambda^\text{int}_l \left[ \ln \left( \frac{s_l f(nx)}{s_l (nx)} \right) \right]' = -\frac{(n - 1) \pi \nu^2}{8} \left[ 1 - \frac{41}{128 \nu^2} + \frac{711}{16384 \nu^4} \right]$$

$$+ \frac{5(n - 1)^2 \pi \nu^2}{32} \left[ 1 - \frac{592}{525 \pi \nu} - \frac{229}{640 \nu^2} + \frac{29504}{75075 \pi \nu^3} + \frac{3771}{81920 \nu^4} \right].$$

(20)

This is to be inserted into the expression for the interior nondispersive surface force density $F^\text{nondisp}_\text{int}$ which, according to (15), can be written as

$$F^\text{nondisp}_\text{int} = \frac{1}{4 \pi^2 a^4} \sum_{l=1}^{\infty} \int_0^{\infty} dz \nu^3 z \Lambda^\text{int}_l \left[ \ln \left( \frac{s_l f(nx)}{s_l (nx)} \right) \right]' .$$

(21)

In the exterior region we calculate similarly

$$-\int_0^{\infty} dz \nu^3 z \Lambda^\text{ext}_l \left[ \ln \left( \frac{-e_l f(x)}{e_l (x)} \right) \right]' = \frac{(n - 1) \pi \nu^2}{8} \left[ 1 - \frac{41}{128 \nu^2} + \frac{711}{16384 \nu^4} \right] -$$

$$- \frac{3(n - 1)^2 \pi \nu^2}{32} \left[ 1 - \frac{592}{315 \pi \nu} - \frac{33}{128 \nu^2} + \frac{29504}{45045 \pi \nu^3} + \frac{639}{16384 \nu^4} \right],$$

(22)

and the exterior nondispersive surface force density $F^\text{nondisp}_\text{ext}$ is

$$F^\text{nondisp}_\text{ext} = -\frac{1}{4 \pi^2 a^4} \sum_{l=1}^{\infty} \int_0^{\infty} dz \nu^3 z \Lambda^\text{ext}_l \left[ \ln \left( \frac{-e_l f(x)}{e_l (x)} \right) \right]' .$$

(23)

When constructing the total nondispersive surface force density as $F^\text{nondisp} = F^\text{nondisp}_\text{int} + F^\text{nondisp}_\text{ext}$, we see from (20) and (22) that the terms of order $(n - 1)$ compensate each other. The surface force accordingly starts with terms containing $(n - 1)^2$.

(The application of the general formalism developed in [1] to dilute balls contains a calculational error; Eq.(94) should have been multiplied by a factor 1/2. Therefore the final conclusion of Sect. 4 in [1] is incorrect. We thank K. A. Milton for informing us about this point.)
Adding (21) and (23) we now get

\[
F_{\text{nondisp}} = \frac{(n - 1)^2}{64\pi a^4} \sum_{l=1}^{\infty} \left( \nu^2 - \frac{65}{128} + \frac{927}{16384\nu^2} \right)
\]  

(24)

Here the divergence mentioned above is made explicit; regularization is needed in the first two terms. We shall use the Riemann zeta function regularization. This method has in recent years become a standard tool in all areas of quantum field theory, and is found to be in agreement with what one can derive by other means, such as the dimensional regularization, cutoff regularization, etc. (cf, for instance, the monograph of Elizalde et al. [21]). Zeta function regularization has been applied in the context of spherical dielectrics in [1] and [7]. An interesting contribution to this technique is further provided by the recent paper of Lambiase et al. [22]. Here, no formal divergences appear; an extra parameter \( s \) is introduced whereby all expressions are retained finite. At the end, an analytical continuation of \( s \) is performed. The final results are in agreement with what one finds by a straightforward application of the Riemann zeta function technique.

To effectuate this kind of regularization we need in practice only the formula

\[
\sum_{l=0}^{\infty} \nu^s = (2^{-s} - 1)\zeta(-s),
\]

(25)

according to which

\[
\sum_{l=1}^{\infty} \nu^2 = -\frac{1}{4}, \quad \sum_{l=1}^{\infty} \nu^0 = -1.
\]

(26)

The third sum in (24) is finite and is known exactly: \( \sum_{l=1}^{\infty} \nu^{-2} = (\pi^2/2) - 4 \). From (24) we then get, when writing the force as \( F_{\text{nondisp}}|_{1/\nu^4} \) to emphasize that it is based upon a uniform asymptotic expansion to order \( 1/\nu^4 \):

\[
F_{\text{nondisp}}|_{1/\nu^4} = \frac{(n - 1)^2}{4\pi a^4} \left[ \frac{129}{65536} + \frac{927}{524288}\pi^2 \right].
\]

(27)

It is of interest to compare this expression with the expression \( F_{\text{nondisp}}|_{1/\nu^2} \) following from a uniform asymptotic expansion to order \( 1/\nu^2 \) only:

\[
F_{\text{nondisp}}|_{1/\nu^2} = \frac{(n - 1)^2}{4\pi a^4} \frac{33}{2048}.
\]

(28)

The reason for this comparison is the following: the combination of terms occurring in the surface force density (24) involves even powers of \( 1/\nu \) only.

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The structure of terms is such that the $O(1/\nu^4)$ force is somewhat larger than the correct force, whereas the $O(1/\nu^2)$ force is somewhat smaller. From (27) and (28) we are therefore able to give quite accurate bounds for the ”exact” nondispersive surface force density $F_{\text{nondisp}}$:

$$F_{\text{nondisp}}|_{1/\nu^2} < F_{\text{nondisp}} < F_{\text{nondisp}}|_{1/\nu^4}. \quad (29)$$

(The term ”exact” here refers to the uniform asymptotic expansion only; we are of course concerned with a dilute-medium theory of order $(n-1)^2$.) Numerically,

$$\frac{(n-1)^2}{4\pi a^4} 0.0161 < F_{\text{nondisp}} < \frac{(n-1)^2}{4\pi a^4} 0.0194. \quad (30)$$

The nondispersive part of the surface force is thus repulsive. The quantum mechanical result is in this sense in agreement with the prediction of the Abraham-Minkowski term in (1).

We note that some care has to be taken to include a sufficient number of terms of the uniform asymptotic expansion when constructing the expression for $F_{\text{nondisp}}$. To illustrate this, let us see what becomes the result from including only the first term in (24) (this corresponds to including only the zeroth order of the uniform asymptotic expansion). Making use of the first regularizing equation in (26) we obtain

$$F_{\text{nondisp}}|_{1/\nu^0} = -\frac{(n-1)^2}{4\pi a^4} \frac{1}{64}. \quad (31)$$

Since $1/64 = 0.0156$ we see that the magnitude of (31) is not very different from the second order approximation in (30), but the sign is wrong. We conclude that it is necessary to go to at least the second order in the uniform asymptotic expansion to get the sign right. The expression (31) is actually in precise agreement with the nondispersive force as calculated by Milton [15], [16], and Milton and Ng [14]. The reason why these authors got a discrepancy with (30) is simply that they included an insufficient number of terms. In [17] and [18] the following accurate value is given, with the assistance of numerical methods,

$$F_{\text{nondisp}} = \frac{(n-1)^2}{4\pi a^4} 0.01907 \quad (32)$$

The fourth order result given in (27) is about 1.7 per cent high.
4 Dispersive force

This case is more difficult to handle than the nondispersive case, and we will essentially be able to give only order-of-magnitude estimates. We restrict henceforth the uniform asymptotic expansion to the second order. Inclusion of higher order terms would here be pointless. The ball is assumed to be dilute, as before.

We now have to keep the nondimensional ultraviolet cutoff \( x_0 = \omega_0 a \) as a finite quantity in the formalism. One may ask: what are the physically reasonable magnitudes of \( x_0 \)? We note that \( x_0 \) is here an "external" parameter that has to be inserted into the formalism by hand. Its value is given by the cutoff frequency \( \omega_0 \), which is determined by the molecular structure of the medium, and by the radius \( a \). We will take \( \omega_0 \) to lie in the ultraviolet region. It seems reasonable, by comparison with the Lorentz dispersion model (cf. \[23\], for instance), to put \( \omega_0 = 3 \times 10^{16} \text{s}^{-1} \). Thus in dimensional units, assuming a small ball with radius \( a = 1 \mu m \), we get \( x_0 = \omega_0 a/c = 100 \). Balls with somewhat larger radii can also be actual. Accordingly, \( 100 \leq x_0 \leq 1000 \) seems to be a reasonable range for \( x_0 \).

Another question is: what is the upper physical limit \( l = l_0 \) to be taken in the summations? This point becomes accentuated by the circumstance that some of the sums are formally divergent. Obviously, this behaviour is a spurious effect. Our dispersion relation, as explained in the beginning of Sect. 2, implies that photons having frequencies \( \omega \) higher than \( \omega_0 \) do not "see" the medium at all. If a photon of limiting frequency \( \omega_0 \) just touches the surface of the sphere, its angular momentum is equal to \( \omega_0 a \), i.e., equal to \( x_0 \). We thus expect, on physical grounds, that \( l_0 \) is of the same order as \( x_0 \). This kind of argument has repeatedly been used in Casimir calculations \[8\], \[7\], \[24\]. It means that one confines oneself to giving order-of-magnitude estimates rather than exact numbers. The physical force, of course, is a definite number in a given case, but to evaluate it one needs detailed information about the dispersive structure of the medium. That lies outside the scope of the present paper.

Adding (15) and (17), making use of the second order approximation, we
get

\[ F = F_{\text{int}} + F_{\text{ext}} \]

\[ = \frac{(n - 1)^2}{4\pi^2a^4} \sum_{l=1}^{\infty} \nu^2 \int_0^{\infty} z^2t^6(z)dz + \frac{1}{4\nu^2} \int_0^{x_0/\nu} z^2t^{12}(z)(3 - 42z^2 + 18z^4 + 2z^6)dz + O\left(\frac{1}{\nu^4}\right) \] (33)

Here the first and the second integral stem respectively from the zeroth, and the second order terms in the uniform asymptotic expansion. We write \( F \) as a sum of two terms, one zeroth order term \( A \) and one second order term \( B \), and consider \( B \) first. This term is more easy to handle than the term \( A \), since the sums over \( l \) in \( B \) are convergent and can be calculated by means of the Euler-Maclaurin formula \[2\]. It is convenient to write \( B \) in the form

\[ B = \frac{(n - 1)^2}{4\pi^2a^4} \frac{1}{4} \sum_{l=1}^{\infty} \int_0^{x_0/\nu} z^2t^{12}(z)(3 - 42z^2 + 18z^4 + 2z^6)dz, \] (34)

and to use the Euler-Maclaurin formula in the following version:

\[ \sum_{l=1}^{\infty} f(l) = \sum_{l=1}^{L} f(l) + \int_{L+1}^{\infty} df(l) + \frac{1}{2} [f(\infty) + f(L + 1)] + \frac{1}{12} [f'(\infty) - f'(L + 1)] + ... \] (35)

Here \( L \) is an auxiliary integer, the use of which may improve the accuracy of the series. Usually it suffices to let the magnitude of \( L \) be moderate (\( L \leq 10 \); cf. \[2\] for instance, where we made an analogous calculation). We write the expression (34) as \( B = \sum_{l=1}^{\infty} f(l) \), and obtain for the first term in (35) to a good accuracy

\[ \sum_{l=1}^{L} f(l) = \frac{(n - 1)^2}{4\pi^2a^4} \frac{L}{4} \int_0^{\infty} z^2t^{12}(z)(3 - 42z^2 + 18z^4 + 2z^6)dz = \]

\[ = -\frac{(n - 1)^2}{4\pi a^4} \frac{65}{2048} L. \] (36)

Here we could set the upper limit of the integral equal to infinity, since the main contribution comes from \( z \) close to the lower limit. The second term in (35) yields after some algebra, when introducing the abbreviation
\[ w = L + 3/2, \]
\[
\int_{L+1}^{\infty} df(l) = \frac{(n-1)^2}{4\pi^2a^4} \left[ \frac{x_0}{8} + \frac{w^8x_0}{64(w^2 + x_0^2)^4} \right. \\
- \frac{11}{128} \frac{w^6x_0}{(w^2 + x_0^2)^3} + \frac{105}{512} \frac{w^4x_0}{(w^2 + x_0^2)^2} \\
- \frac{325}{1024} \frac{w^2x_0}{w^2 + x_0^2} + \frac{315}{1024} w \arctan \frac{x_0}{w} - \frac{125}{1024} \frac{w\pi}{w} \right] \\
\rightarrow \frac{(n-1)^2}{4\pi^2a^4} \left( -\frac{x_0}{8} + \frac{65}{2048} w\pi \right), \quad x_0 \rightarrow \infty. \tag{37}
\]

The last term between square parentheses in (37) was evaluated putting \( x_0 = \infty \). Similarly we put \( x_0 = \infty \) when evaluating the last nonvanishing terms in (35):

\[
\frac{1}{2} f(L + 1) - \frac{1}{12} f'(L + 1) = -\frac{(n-1)^2}{4\pi^2a^4} \frac{65}{4096} \pi. \tag{38}
\]

Altogether, for large cutoffs, we get the \( L \)-independent result

\[
B = \frac{(n-1)^2}{4\pi^2a^4} \left( -\frac{x_0}{8} + \frac{65}{2048} w\pi \right), \quad x_0 \rightarrow \infty. \tag{39}
\]

We extract the nondispersive part of this expression:

\[
B(\text{nondispersive part}) = \frac{(n-1)^2}{4\pi a^4} \frac{65}{2048}. \tag{40}
\]

This is compared with the second order contribution to the nondispersive force obtained previously in Eq.(24):

\[
F_{\text{nondisp}(2nd \ order)} = \frac{(n-1)^2}{64\pi a^4} \left( -\frac{65}{128} \sum_{l=1}^{\infty} \nu^0 = \frac{(n-1)^2}{4\pi a^4} \frac{65}{2048}. \tag{41}
\]

The expressions (40) and (41) are seen to be equal. This brings us to the following important conclusion: If we perform a dispersive calculation of the surface force, to second order, we find a term that is independent of the cutoff. This term is precisely equal to the second order contribution within a calculation that is nondispersive from the outset. In the second case zeta-function regularization is being employed, while in the first case it is not.
Consider next the zeroth order term $A$ in (33). Performing the integration over $z$ we get

$$A \equiv \frac{(n-1)^2}{4\pi^2a^4} \sum_{l=1}^{\infty} \nu^2 \int_0^{x_0/\nu} z^2t^6(z)dz$$

$$= \frac{(n-1)^2}{4\pi^2a^4} \sum_{l=1}^{\infty} \left[ -\frac{1}{4} \frac{\nu^5x_0}{(\nu^2+x_0^2)^2} + \frac{1}{8} \frac{\nu^3x_0}{\nu^2+x_0^2} + \frac{\nu^2}{8} \arctan\frac{x_0}{\nu} \right].$$

(42)

We will only be able to give an order-of-magnitude analysis of the physically meaningful part of this divergent expression. Note first, however, the following point: take the nondispersive limit, $x_0 = \infty$. Then only the last term in (42) survives and we get, again employing zeta function regularization,

$$A(\text{nondispersive part}) = \frac{(n-1)^2}{4\pi^2a^4} \frac{1}{16} \sum_{l=1}^{\infty} \nu^2 = -\frac{(n-1)^2}{4\pi^2a^4} \frac{1}{64}.$$  (43)

This is precisely the same as the expression (31), and serves as a corollary of our dispersive calculation.

Now return to the dispersive case, and estimate the contribution to the sum in (42) from low and moderate values of $l$, such that $\nu \leq x_0$. When $\nu \ll x_0$, we see that the two first terms in (42) are negligible in comparison to the third term. Moreover, when $\nu = x_0$ the two first terms compensate each other. Consequently we may write

$$A|_{\nu \leq x_0} \simeq \frac{(n-1)^2}{4\pi^2a^4} \sum_{\nu \leq x_0} \frac{\nu^2}{8} \arctan\frac{x_0}{\nu}.$$  (44)

Here the variation in the factor $\arctan(x_0/\nu)$ is moderate; it decreases from $\pi/2$ for small $\nu$, to $\pi/4$ when $x_0 = \nu$. For estimate purposes we may replace $\arctan(x_0/\nu)$ by a constant $C$, where $C$ lies in the interval $\pi/4 \leq C \leq \pi/2$. The sum in (44) then becomes simple to evaluate, since

$$\sum_{l=1}^{x_0^{-1/2}} \nu^2 = \frac{1}{3} x_0^3 + \frac{1}{2} x_0^2 + \frac{1}{6} x_0 - \frac{1}{4},$$  (45)

and so we get

$$A|_{\nu \leq x_0} = \left(\frac{n-1)^2}{4\pi^2a^4}\right) \left(\frac{C}{8} \left(\frac{1}{3} x_0^3 + \frac{1}{2} x_0^2 + \frac{1}{6} x_0 - \frac{1}{4}\right) \right).$$  (46)

It is here natural to argue such as we did above, in connection with (40) and (41). We may thus associate the $x_0$-independent part of (46) with the
nondispersive surface force, calculated according to the zeroth order uniform asymptotic expansion. Actually, if we equate the \( x_0 \)-independent part (arising from the last term in (46)) to the expression (31), we obtain \( C = \pi/2 \). This is precisely the value of \( C \) corresponding to the small values of \( \nu \), according to our discussion above. We may thus conclude that the nondispersive part of the force, at least as far as the leading term is concerned, is associated with the low angular momenta \( l \).

When \( x_0 \) is included, (46) predicts under usual physical circumstances a strong, repulsive force. For instance, if we take \( C = \pi/2 \) and \( x_0 = 100 \), we find that (46) is about \( 10^6 \) as large as the nondispersive second order (or fourth order) force given in (30). This large difference can be expected to be quite important, in practice. An eminent example to think about is the sonoluminescence effect \([14]\). We will briefly return to this point in the Conclusion section.

The result (46) was derived under the assumption of low and moderate nondimensional frequencies, \( \nu \leq x_0 \). Is there an appreciable contribution to \( A \) from higher frequencies also? We think that this is not so, the reason being, as explained above, that photons with \( \omega > \omega_0 \) do not "see" the medium. It is physically speaking most safe to truncate the \( l \) summation at \( \nu \approx x_0 \) and adopt (46) as a reasonable order-of-magnitude estimate of the leading dispersive term. The total surface force density \( F \) is thus estimated adding the expressions (46) and (39).

Finally we mention the following alternative method of handling the divergent sum \( A \), which may appear mathematically convenient. The method consists in adding and subtracting the weakly divergent sum representing \( A \) in the frequency region \( \nu > x_0 \). Starting from (42) we see that this amounts to writing

\[
A = \frac{(n - 1)^2}{4\pi^2 a^4} \sum_{l=1}^{\infty} \left[ -\frac{1}{4} \frac{\nu^5 x_0}{(\nu^2 + x_0^2)^2} + \frac{1}{8} \frac{\nu^3 x_0}{\nu^2 + x_0^2} + \frac{\nu^2}{8} \arctan \frac{x_0}{\nu} - \frac{x_0^3}{3\nu} \right] +
\frac{(n - 1)^2 x_0^3}{3\nu^3} \sum_{l=1}^{\infty} \frac{1}{\nu},
\]  

(47)

where we have chosen to let the sums over \( l \) in the extra terms run from 1 to infinity. The advantage of writing \( A \) in this way is that the first of the two sums in (47) is finite, and that we have explicit control over the divergence in the second sum. Let the first sum be denoted by \( A(\text{finite}) \). Making a numerical calculation of this expression at the lower and upper
limits \( x_0 = 100 \) and \( x_0 = 1000 \), we get approximately

\[
A(\text{finite}) \approx \frac{(n - 1)^2}{4\pi^2a^4} \left\{ -1.7 \times 10^6, \quad x_0 = 100 \\
-2.4 \times 10^9, \quad x_0 = 1000 \right\}
\]

(48)

A graphical representation of \( A(\text{finite}) \) versus \( x_0 \) shows that the variation is roughly linear in the actual frequency region. Exploiting this fact, and truncating the second sum in (47) at \( l = x_0 \), we arrive at the following estimate for \( A \):

\[
A \approx \frac{(n - 1)^2}{4\pi^2a^4} \left[ \frac{x_0^3}{3} \sum_{l=1}^{x_0} \frac{1}{\nu} - (266.5 \frac{x_0}{100} - 264.8)10^6 \right],
\]

(49)

valid for \( 100 \leq x_0 \leq 1000 \). It is seen, in fact, that the structure of (49) is not very different from the structure of our previous estimate (46). The advantage of (49) in comparison with (46), of course, is that there appears no extra constant \( C \) to be estimated separately.

5 Conclusion and final remarks

The physical model considered in this paper is that of a compact dilute spherical ball, surrounded by a vacuum. The dispersion relation of the material is such that \( n(i\hat{\omega}) = n = \text{const} \) when the imaginary frequency \( \hat{\omega} \) is lower than the ultraviolet cutoff frequency \( \omega_0 \) whereas \( n(i\hat{\omega}) = 1 \) for higher \( \hat{\omega} \). This is a very simple dispersion relation: we know on general grounds that \( n(i\hat{\omega}) \), being closely related to a so-called generalized susceptibility, is real on the positive imaginary frequency axis and decreases monotonically towards 1 when \( \hat{\omega} \to \infty \) (cf., for instance, [25]). However, this is not a serious restriction here as we may well imagine that \( n(i\hat{\omega}) \) instead is endowed with a small negative slope for \( \hat{\omega} < \omega_0 \).

The nondimensional cutoff is \( x_0 = \omega_0a \), \( a \) being the radius of the ball. Under typical physical conditions, if the radius lies in the interval \( 1\mu m \leq a \leq 10\mu m \), we expect that \( 100 \leq x_0 \leq 1000 \).

Our basic Green function formalism (Sect. 2) is Taylor expanded in \( (n - 1) \) up to the second order, and is expanded in \( 1/\nu \) up to the fourth order \( (\nu = l + 1/2) \). The Maple V program here proves to be an effective tool.

The surface force density \( F \) is constructed by taking the difference between the radial Maxwell stress tensor components on the two sides of the boundary \( r = a \). Alternatively, \( F \) can be found as the integral of the radial component of the volume force density \( f \) across the boundary. The general
expression for $f$ is given in (1), and the meaning of the various terms is discussed. Only the first, and under usual conditions most important, term (the Abraham - Minkowski term) is further considered in this paper.

The nondispersive force is calculated in Sect.3. Formally, this case is treated by setting $x_0 = \infty$, and regularizing the divergent sums using the Riemann zeta function. Working to the order $1/\nu^4$ in the uniform asymptotic expansion, the nondispersive force is given by (27) and (30). A slightly more accurate value is given by (32). This force is repulsive.

We emphasize that $F_{\text{nondisp}}$ is a physical force; the expression is not merely a formal outcome of a more or less arbitrary regularization procedure. It is here of interest to note that, in addition to the Green function approach above, there has recently been developed also two other alternative procedures which lead to results supporting our statement:

First, one may consider the problem from a microscopic point of view and calculate the mutual van der Waals energy for the molecules in the compact sphere. This has been done by Milton and Ng [26]. The van der Waals energy for two molecules is $V = -B/r^7$, with $B$ a positive constant. Integrating this over the sphere one finds a divergent expression which has to be regularized, conveniently by means of dimensional regularization. The finite remaining part of the energy becomes

$$E = \frac{(\varepsilon - 1)^2}{\pi a^2} \frac{23}{1536}$$  \hspace{1cm} (50)

Associating the van der Waals energy $E$ with the surface force $F$ via the relation $F = -(1/4\pi a^2)\partial E/\partial a$, we find the same numerical result for $F = F_{\text{nondisp}}$ as in Eq. (32).

Second, one can calculate the mutual energy between the molecules in the sphere by employing quantum mechanical perturbation theory. This has been done by Barton [27], for a dilute sphere. He used an exponential cutoff for the wave numbers. Again, the formal expression for $E$ was found to diverge, but after omission of the cutoff dependent terms, the result was found to be precisely in agreement with the expression (50).

These two recent alternative developments are welcome, since they signify that a consensus between various approaches to the difficult Casimir surface force problem is finally in sight.

We emphasize that the repulsive force $F_{\text{nondisp}}$, although being a physical force and thus measurable in principle, is not the total surface force, not even for a nondispersive medium. The total force is necessarily attractive, since it results from the attractive van der Waals force between molecules. (This point was apparently recognized very early, by Davies in his classic paper [28].) At first sight it may perhaps be surprising that we operate with
a measurable force component which is only a part of the total force. This kind of situation is however known also from other areas in electrodynamics, and it by be instructive in the present context to discuss briefly the following two examples:

1) Consider the situation where a high intensity laser beam falls vertically from above on a liquid (water) surface. The surface bulges out, corresponding to an outward-directed force. The elevation is slight, of the order of 1 µm, but is clearly detectable. This is the famous Ashkin-Dziedzic experiment \[29\]. The elevation can be described completely, using only the Abraham-Minkowski force term, \(i.e.,\) the first term in (1).

The Abraham-Minkowski force is however not the only force acting on the water surface. There is an additional force, arising from electrostriction, and this force is stronger than the first-mentioned one such that the total force on the surface is actually compressive. Electrostriction does not contribute to the elevating force, as mentioned earlier, since electrostriction merely compresses the column of liquid. The theory of the Ashkin-Dziedic experiment has been given in \[30\] and \[4\].

2) A second, related, situation is the one discussed in many textbooks (cf. \[3\], for instance), namely two parallel condenser plates partly immersed in a dielectric liquid. When a strong static electric field is applied between the plates, the liquid is observed to rise slightly in the interior region. The situation is more involved than one might be inclined to think. Again, the observable elevation can be described entirely with use of the Abraham-Minkowski force only. There is in addition the compressive electrostriction force acting, so that the total surface force density is compressive. The electrostriction force has to be the stronger member of the two forces, since otherwise the column of liquid would not be kept together as a whole. The mathematical reason why the electrostriction force is the stronger one, is that the relative permittivity is larger than 1. This is a thermodynamical result. Seen from this perspective, the deepest reason why the column of liquid is kept together, is neither hydrodynamics nor electrodynamics, but rather thermodynamics. The theory of this situation is discussed, among other places, in \[4\] and \[31\].

The lesson to be learned from the above two examples is fairly obvious: it is physically quite legitimate to consider a part of the complete surface force, and discuss its experimental consequences, without drawing the complete force into consideration at all. This is precisely what we do in principle when we discuss the Casimir force \(F^{\text{nondisp}}\). We are not aware of any concrete operational argument so far to really measure \(F^{\text{nondisp}}\) experimentally, however.

Consider next the dispersive force. It was calculated in Sect. 4, to second order in the asymptotic expansion. The force is written as a sum of two
terms, $F = A + B$, where the second order term $B$ is given in (39). This term, although strictly speaking derived for high cutoffs only, $x_0 \to \infty$, is expected to be fairly accurate for all values of $x_0$ that are actual in practice. The term $B$ has the same basic structure as the surface force calculated earlier on a spherical shell [7]: there is an attractive part linear in $x_0$, which is due to dispersion, and in addition a repulsive part which is dispersion independent. Under usual physical conditions the first of these two parts is the stronger one.

As for the leading term $A$, of zeroth order, we have only been able to give an order-of-magnitude analysis, based upon physical arguments allowing us to truncate the divergent sums over $l$ at an upper limit approximately equal to $x_0$. We gave two estimates for $A$, in equations (46) and (49). The structures of these two expressions are seen to be roughly similar. Under usual circumstances also the force $A$ is a repulsive one.

To get an idea about the magnitudes involved, let us limit ourselves to the dominant first term in (46), putting for definiteness $C = \pi/2$. Then

$$F_{\text{disp}} \simeq \frac{(n - 1)^2 x_0^3}{4\pi a^4 \cdot 48} = \frac{(n - 1)^2 \omega_0^3}{4\pi a^4 \cdot 48}. \quad (51)$$

This force may easily be about $10^6$ as strong as the nondispersive force (27). The force (50) corresponds to the Casimir energy (integrating while keeping $\omega_0$ constant)

$$E_{\text{disp}} \simeq -\frac{(n - 1)^2 \omega_0^3}{96} a^2. \quad (52)$$

This energy is always negative, and has its maximum when the radius is at minimum. Moreover it is independent of the substitution $n \to 1/n$, when $n$ is close to unity. One would therefore think that the formula is equally well applicable to the case when there is a vacuum bubble immersed in a dilute dielectric medium.

One may wonder whether it is possible to apply the present kind of theory to the phenomenon of sonoluminescence (an instructive review of this phenomenon has recently been given by Cheeke [32]). It is well known that the energies involved in the nondispersive Casimir effect are far too small to account for the sonoluminescence effect. Can the very large magnitude of (52) "save" the situation? We see that by taking $(n - 1)^2 \simeq 0.1, \; x_0 \simeq 1000$ (corresponding to $a \simeq 10\mu m$), we obtain from (52) $E_{\text{disp}} \simeq -2 \times 10^4 eV$. The magnitude of this energy is probably acceptable, but the sign is apparently wrong [10], [14]. At least the sign is in conflict with energy emission during the collapse. Consider, however, the following argument: an experimental
fact is that the photon flash does not occur during the whole collapse period but instead precisely at the end of that period, when $a = a_{\text{min}} \simeq 1 \mu m$. Imagine now that the energy of the burst of photons were really taken from the Casimir energy at this particular instant. Consequently there would occur an energy imbalance in the system. The balance of Casimir energy would be restored if the bubble were increasing its radius out to $a \simeq 10 \mu m$.

Such a movement actually happens. Evidently, we do not "explain" the sonoluminescence effect by this kind of argument. Our aim is more modest, viz. to suggest that sonoluminescence is perhaps not totally unrelated to the Casimir effect after all.

The last point that we shall dwell on in this paper, is the influence from absorption in the medium. This is an important point since it shows the physical limitations of our adopted material model, and the directions for further research. At first sight, it would seem as if absorption does not create any problems at all in the theory: we are after all adopting a model in which the quantity $n(i\omega)$ is real and shows a simple though physically acceptable behaviour along the positive imaginary frequency axis. Absorption should accordingly be taken into account implicitly in the theory, through the Kramers-Kronig relations, which lead to a complex behaviour of $n(\omega)$ along the real frequency axis. The problem waiting for a complete solution lies at another place, namely in our adoption of formulas (7) and (8) for the effective products. These are the same formulas as for a nondispersive medium. A more complete theory would have to take into account the coupling of the molecules to a heat reservoir, with a corresponding quantization of the source currents. This would in turn imply the presence of Langevin terms in the effective products. Interesting developments of the theory of absorptive media, implying a quantization of the complete system (medium plus reservoir) have been given in [33], [34], and most recently, in [35] and [36]. We only point to this problem here, without going into any detail.

For the convenience of the reader, we summarize in Appendix B the basic assumptions leading to our formulas for the effective products.

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A  Nondispersive force expanded to the fifth order in \((n - 1)\)

Our calculation of the nondispersive force in Sect.3 was based on a dilute-ball expansion up to order \((n - 1)^2\), and on an asymptotic expansion up to order \(1/\nu^4\). A characteristic property was found to be that terms of order \((n - 1)\) in \(F_nondisp_{\text{int}}\) and \(F_nondisp_{\text{ext}}\) compensated each other, so that the leading term in \(F_{\text{nondisp}} = F_{\text{int}} + F_{\text{ext}}\) would go as \((n - 1)^2\); cf. (20) and (22). One may ask: what happens if one makes a higher order expansion in \((n - 1)\), will then terms containing odd powers in \((n - 1)\) continue to compensate each other? We shall investigate this point, giving the expansions for \(F_{\text{int}}\) and \(F_{\text{ext}}\) up to order \((n - 1)^5\). However, we will now have to truncate the uniform asymptotic expansion at order \(1/\nu^2\). (A combined set of higher order expansions in \((n - 1)\), as well as \(1/\nu\), turned out to be beyond the capacity of our Maple program.)

We abstain here from giving the explicit expressions for \(F_{\text{int}}\) and \(F_{\text{ext}}\), but write down their sum:

\[
F_{\text{nondisp}} = \left( \frac{(n - 1)^2}{4\pi a^4} \right) \left[ \frac{33}{2084} - (n - 1) \left( \frac{99}{4096} + \frac{11}{210 \pi} \right) \right] + (n - 1)^2 \left( \frac{1213}{65536} + \frac{11}{105 \pi} \right) - (n - 1)^3 \left( \frac{785}{131072} + \frac{29}{216 \pi} \right) \]

The first term in this expression agrees with our previous second order result (28). We see that odd powers in \((n - 1)\) generally do not compensate each other in the surface force. All powers in \((n - 1)\) are present, with the exception of the first, linear, term. The expansion given above may be useful also in cases when \((n - 1)\) is not so small, perhaps up to \((n - 1) \approx 0.3 - 0.4\).

B  The effective products derived from statistical mechanics

Assuming for definiteness Schwinger’s source theory, we relate the electric field components \(E_i(x)\) to the polarization components \(P_k(x')\) via a tensor \(\Gamma_{ik}(x, x')\):

\[E_i(x) = \int d^4x' \Gamma_{ik}(x, x') P_k(x') \quad (54)\]
Stationarity of the system means that $\Gamma_{ik}$ depends on time only through the difference $\tau = t - t'$. Causality implies that the integration over $t'$ is limited to $t' \leq t$. From a statistical mechanical viewpoint, $\Gamma_{ik}(x, x')$ is a generalized susceptibility \[25].

Introduce the Fourier transform $\Gamma_{ik}(r, r', \omega)$ via

$$\Gamma_{ik}(x, x') = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega \tau} \Gamma_{ik}(r, r', \omega) \tag{55}$$

The basic equation (54), in combination with the causality principle, are thus the necessary conditions for causing the function $\Gamma_{ik}(r, r', \omega)$ to satisfy the following properties:

(i) It is a regular, one-valued function in the upper half frequency plane. At infinity ($|\omega| \to \infty$), it goes to zero.

(ii) There is no singularity on the real axis (except at the origin, in the case of a metal).

(iii) The function does not take real values at any finite point in the upper half plane except on the imaginary axis.

Now invoke Kubo’s formula from statistical mechanics: it states that the spectral generalized susceptibility is related to the commutator between field components as

$$\langle E_i(x) E_k(x') \rangle = \left(\frac{i}{2\pi} \right) \int_0^\infty d\tau e^{i\omega \tau} \langle [E_i(x), E_k(x')] \rangle \tag{56}$$

(cf. for instance, \[25\]). This means that the generalized susceptibility is identifiable with the retarded Green function: $\Gamma_{ik}(x, x') = G_{ik}^R(x, x')$. For $t < t'$ both $\Gamma_{ik}(x, x')$ and $G_{ik}^R(x, x')$ vanish, the first because of causality, the second because of the definition of the retarded Green function.

Consider now the two-point function $\langle E_i(x) E_k(x') \rangle$. Its Fourier transform $\langle E_i(r, \omega) E_k(r', \omega') \rangle$ can be expressed in terms of the quantity $\langle E_i(r) E_k(r') \rangle_\omega$ (the spectral correlation tensor) as

$$\langle E_i(r, \omega) E_k(r', \omega') \rangle = 2\pi \langle E_i(r) E_k(r') \rangle_\omega \delta(\omega + \omega') \tag{57}$$

The crucial step now is to apply the fluctuation-dissipation theorem \[25], according to which

$$\langle E_i(r) E_k(r') \rangle_\omega = Im \, G_{ik}^R(r, r', \omega) \coth\left(\frac{1}{2} \beta \omega\right), \tag{58}$$

with $\beta = 1/(k_B T)$. At zero temperature, $\coth(\frac{1}{2} \beta \omega) \to \text{sgn}(\omega)$. Choosing the causal integration contour in the complex frequency plane we obtain, after a complex frequency rotation, the effective products as given in Eqs.(7) and (8).
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