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Universal behaviour of dispersion forces between two dielectric plates in the low-temperature limit

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Abstract

The universal analytic expressions in the limit of low temperatures (short separations) are obtained for the free energy, entropy and pressure between the two parallel plates made of any dielectric. The analytical proof of the Nernst heat theorem in the case of dispersion forces acting between dielectrics is provided. This permitted us to formulate the stringent thermodynamical requirement that must be satisfied in all models used in the Casimir physics.

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It is common knowledge that dispersion force is a quantum phenomenon which results from fluctuating (thermal) electromagnetic fields [1]. The most well-known examples of dispersion interaction are the van der Waals [1] and the Casimir [2–4] forces. Dispersion forces play a very important role in surface phenomena, layered structures, colloid-substrate interactions, adhesion, foam formation and in stability of microelectromechanical systems [5–8]. Recently they were found to be of considerable significance in experiments on quantum reflection and Bose–Einstein condensation of ultracold atoms near different surfaces [9, 10]. Dispersion forces create a free-energy difference between materials in the normal and the superconducting phase which may influence the value of the critical magnetic field [11]. They are responsible for the interaction of atoms and molecules with nanostructures such as carbon nanotubes [12].

The theoretical description of all above-listed phenomena is based on the Lifshitz theory [13]. This theory presents the dispersion force, free energy and entropy between the two plates in terms of their dielectric permittivity $\varepsilon(\mathrm{i}\xi)$ along the entire imaginary frequency axis including zero frequency. $\varepsilon(\mathrm{i}\xi)$ is found by means of the dispersion relation using the experimental optical data for the complex refractive index [14]. As these data are available only within a restricted frequency region, the use of some theoretical models of dielectric response becomes unavoidable. Different extrapolations of data outside the regions where they are measured (e.g., to low frequencies) may lead, however, to very different theoretical predictions. This places strong emphasis on the extrapolation problem in applications of the Lifshitz theory.

In this paper we present an analytical derivation for the low-temperature (short-separation) behaviour of the Lifshitz entropy and thermal corrections to the energy and pressure between two thick dielectric plates (semispaces). It is shown to be the same as for metals, i.e., universal. We demonstrate that if the plate material at low frequencies is described by the static dielectric permittivity, the entropy goes to zero in the limit of zero temperature in accordance with the Nernst heat theorem. Alternatively, if one includes the nonzero dc conductivity of the dielectric material into the model of the dielectric response, the entropy goes to a nonzero positive value when the temperature T goes to zero (i.e., the Nernst heat theorem is violated). Finally, we formulate the thermodynamic constraint on the extrapolations of the optical data to low frequencies and apply it to the topical problem of noncontact atomic friction [15–18].

The Lifshitz formula for the free energy of dispersion interaction between two thick plates in thermal equilibrium, written in terms of dimensionless variables, is

$$\mathcal{F}(a,T)=\frac{\hbar c\tau}{32\pi^2a^3}\sum_{l=0}^{\infty}\left(1-\frac{\delta_{l0}}{2}\right)\int_{\tau l}^{\infty}y\,\mathrm{d}y\left[\ln\left(1-r_{\parallel}^2\,\mathrm{e}^{-y}\right)+\ln\left(1-r_{\perp}^2\,\mathrm{e}^{-y}\right)\right]. \tag{1}$$

Here, a is the separation between the plates and we use the dimensionless variable $\tau=4\pi k_BaT/(\hbar c)$ (k_B is the Boltzmann constant). The reflection coefficients for the two independent polarizations of the electromagnetic field are given by

$$r_{\parallel}=\frac{\varepsilon_l y-\sqrt{y^2+\zeta_l^2(\varepsilon_l-1)}}{\varepsilon_l y+\sqrt{y^2+\zeta_l^2(\varepsilon_l-1)}},\qquad r_{\perp}=\frac{\sqrt{y^2+\zeta_l^2(\varepsilon_l-1)}-y}{\sqrt{y^2+\zeta_l^2(\varepsilon_l-1)}+y}. \tag{2}$$

The dimensionless Matsubara frequencies are $\zeta_l=\xi_l/\xi_c=l\tau$ where the dimensional ones are $\xi_l=2\pi k_BTl/\hbar$ and $\xi_c=c/(2a)$. The dielectric permittivity is computed at imaginary Matsubara frequencies $\varepsilon_l=\varepsilon(\mathrm{i}\xi_l)=\varepsilon(\mathrm{i}\zeta_l\xi_c)$.

Applying the Abel-Plana formula [3, 14]

$$\sum_{l=0}^{\infty}\left(1-\frac{1}{2}\delta_{l0}\right)F(l)=\int_0^{\infty}F(t)\,\mathrm{d}t+\mathrm{i}\int_0^{\infty}\mathrm{d}t\,\frac{F(\mathrm{i}t)-F(-\mathrm{i}t)}{\mathrm{e}^{2\pi t}-1}, \tag{3}$$

we can rearrange equation (1) to the form $\mathcal{F}(a,T)=E(a)+\Delta\mathcal{F}(a,T)$ where

$$\begin{aligned}E(a)&=\frac{\hbar c}{32\pi^2a^3}\int_0^{\infty}\mathrm{d}\zeta\int_{\zeta}^{\infty}\mathrm{d}y\,f(\zeta,y),\\ f(\zeta,y)&=y\left\{\ln\left[1-r_{\parallel}^2(\zeta,y)\,\mathrm{e}^{-y}\right]+\ln\left[1-r_{\perp}^2(\zeta,y)\,\mathrm{e}^{-y}\right]\right\}\end{aligned} \tag{4}$$

is the energy of dispersion interaction at zero temperature, and

$$\Delta\mathcal{F}(a,T)=\frac{\mathrm{i}\hbar c\tau}{32\pi^2a^3}\int_0^{\infty}\mathrm{d}t\,\frac{F(\mathrm{i}t\tau)-F(-\mathrm{i}t\tau)}{\mathrm{e}^{2\pi t}-1},\qquad F(x)\equiv\int_x^{\infty}\mathrm{d}y\,f(x,y) \tag{5}$$

is the thermal correction to it. The asymptotic expansions of the energy (4) at both short separations and large separations are well known [13, 14]. Here we obtain the low-temperature (short-separation) behaviour of the thermal correction (5) for the case of dielectric plates.

To solve this problem, it is sufficient to describe the dielectric by its static dielectric permittivity $\varepsilon_0=\varepsilon(0)$. The reason is that for dielectrics at sufficiently low temperatures the Matsubara frequencies giving the leading contribution to equation (5) belong to the region where ε practically does not depend on the frequency and is equal to ε_0 (this is true for $\Delta\mathcal{F}$ but not for $E(a)$). To obtain the asymptotic behaviour of $\Delta\mathcal{F}(a,T)$ at $\tau\ll 1$ we, first, expand the function $f(x,y)$, defined in equation (4), in powers of $x=t\tau$. Then we introduce the new variable $\tilde{y} = y - x$ to exclude x from the lower integration limit in equation (5). The subsequent integration of the obtained expansion with respect to $\tilde{y}$ from 0 to infinity leads to

$$F(\mathrm{i}x) - F(-\mathrm{i}x) = \mathrm{i}\pi \frac{(\varepsilon_0 - 1)^2}{2(\varepsilon_0 + 1)} x^2 - \mathrm{i}\alpha x^3 + O(x^4), \qquad (6)$$

where α is real and remains unknown at this stage because all powers of the expansion of $f(x, y)$ contribute to its value. Next, we substitute equation (6) in equation (5) with the result

$$\mathcal{F}(a, T) = E(a) - \frac{\hbar c}{32\pi^2 a^3} \left[\frac{\zeta(3)(\varepsilon_0 - 1)^2}{8\pi^2(\varepsilon_0 + 1)} \tau^3 - C_4 \tau^4 + O(\tau^5) \right], \qquad (7)$$

where $C_4 = \alpha/240$ and $\zeta(z)$ is the Riemann zeta-function. Note that this equation (and respective equations for a pressure and entropy) does not allow a limiting transition $\varepsilon_0 \to \infty$ in order to obtain the case of ideal metals. The mathematical reason is that in our perturbation theory it is impermissible to interchange the limits $\tau \to 0$ and $\varepsilon_0 \to \infty$ in the power expansions of functions depending on ε_0 as a parameter.

The pressure of the dispersion interaction is given by

$$P(a, T) = -\frac{\partial \mathcal{F}(a, T)}{\partial a} = P_0(a) - \frac{\hbar c}{32\pi^2 a^4} [C_4 \tau^4 + O(\tau^5)], \qquad (8)$$

where $P_0 = -\partial E/\partial a$ is the pressure at $T = 0$ and only the fourth-power term on the right-hand side of equation (7) contributes to the thermal correction. At low temperatures this analytical result agrees with the behaviour of the Casimir pressure for nondispersive dielectrics calculated numerically in [19].

Alternatively, the pressure can be found directly from the Lifshitz formula

$$P(a, T) = -\frac{\hbar c \tau}{32\pi^2 a^4} \sum_{l=0}^{\infty} \left(1 - \frac{1}{2}\delta_{l0}\right) \int_{\tau l}^{\infty} y^2 \, \mathrm{d}y \left[\frac{r_\parallel^2}{\mathrm{e}^y - r_\parallel^2} + \frac{r_\perp^2}{\mathrm{e}^y - r_\perp^2} \right]. \qquad (9)$$

Applying the Abel–Plana formula (3) in equation (9), we get $P(a, T) = P_0(a) + \Delta P(a, T)$ where the thermal correction to the pressure is

$$\Delta P(a, T) = -\frac{\mathrm{i}\hbar c \tau}{32\pi^2 a^4} \int_0^{\infty} \mathrm{d}t \, \frac{\Phi(\mathrm{i}t\tau) - \Phi(-\mathrm{i}t\tau)}{\mathrm{e}^{2\pi t} - 1} \qquad (10)$$

and the function $\Phi(x) = \Phi_\parallel(x) + \Phi_\perp(x)$ is defined by

$$\Phi_{\parallel,\perp}(x) = \int_x^{\infty} \frac{y^2 \, \mathrm{d}y r_{\parallel,\perp}^2(y, x)}{\mathrm{e}^y - r_{\parallel,\perp}^2(y, x)}. \qquad (11)$$

By finding the leading term of the expansion of $\Phi(x)$ in powers of x, one arrives at

$$\Phi(\mathrm{i}x) - \Phi(-\mathrm{i}x) = -\mathrm{i}\frac{x^3}{3}(\sqrt{\varepsilon_0} - 1)\left(\varepsilon_0^2 + \varepsilon_0\sqrt{\varepsilon_0} - 2\right) + O(x^5). \qquad (12)$$

Substitution of equation (12) into equation (10) leads to the result

$$P(a, T) = P_0(a) - \frac{\hbar c}{32\pi^2 a^4} \left[\frac{(\sqrt{\varepsilon_0} - 1)\left(\varepsilon_0^2 + \varepsilon_0\sqrt{\varepsilon_0} - 2\right)}{720} \tau^4 + O(\tau^5) \right]. \qquad (13)$$

Comparing equations (8) and (13) we find the value of so long unknown coefficient

$$C_4 = \frac{1}{720}(\sqrt{\varepsilon_0} - 1)\left(\varepsilon_0^2 + \varepsilon_0\sqrt{\varepsilon_0} - 2\right). \qquad (14)$$

Thus, the low-temperature (short-separation) behaviour of both the free energy and the pressure is given by equations (7), (13), (14). By using these results, the asymptotic behaviour of the entropy of dispersion interaction is described by the expression

$$S(a,T) = -\frac{\partial\mathcal{F}(a,T)}{\partial T} = \frac{3k_B\zeta(3)(\varepsilon_0-1)^2}{64\pi^3a^2(\varepsilon_0+1)}\tau^2 \\ \times\left[1-\frac{2\pi^2(\varepsilon_0+1)(\varepsilon_0\sqrt{\varepsilon_0}+2\varepsilon_0+2\sqrt{\varepsilon_0}+2)}{135\zeta(3)(\sqrt{\varepsilon_0}+1)^2}\tau\right]. \qquad (15)$$

We see from equation (15) that in the limit $\tau\to 0$ ($T\to 0$) the entropy of both the van der Waals and Casimir interactions goes to zero following the same universal law which was previously found for ideal and for real metals [14]. We have proved that the use of the Ninham-Parsegian representation [1] for $\varepsilon(\mathrm{i}\xi_l)$ instead of ε_0 modifies only the terms of order $O(\tau^5)$ in equations (7), (13). The comparison with the results of numerical computations for real dielectrics demonstrates that at separations 100–500 nm our asymptotic expressions are applicable at $T < 60$–70 K.

We now turn to a problem of major importance which arises when one includes the dc conductivity of the dielectric plates into the model of the dielectric response, $\tilde{\varepsilon}_l = \varepsilon_l + 4\pi\sigma_0/\xi_l = \varepsilon_l + \beta(T)/l$. Here σ_0 is the dc conductivity of the dielectric and $\beta = 2\hbar\sigma_0/(k_BT)$. The conductivity depends on T according to $\sigma_0 \sim \exp(-b/T)$ where b is different for different dielectrics. It is significant that for dielectrics the additional Drude term is very small for all $\xi_l \neq 0$. For example, $\beta \sim 10^{-12}$ for SiO_2 at $T = 300$ K and, thus, it is certainly negligible for all $l \geqslant 1$.

One might believe, however, that this term plays a role in the zero-frequency contribution in equation (1). To test this conjecture we substitute $\tilde{\varepsilon}_l$ in equation (1) and arrive at

$$\tilde{\mathcal{F}}(a,T) = \mathcal{F}(a,T) - \frac{k_BT}{16\pi a^2}\left\{\zeta(3) - \mathrm{Li}_3\left[\left(\frac{\varepsilon_0-1}{\varepsilon_0+1}\right)^2\right] + R(\tau)\right\}, \qquad (16)$$

where $\mathrm{Li}_3(z)$ is the polylogarithm function, the asymptotic behaviour of $\mathcal{F}$ is given by equations (7), (14), and R decreases exponentionally when $T\to 0$. As a result the entropy of the dispersion interaction at $T = 0$,

$$\tilde{S}(a,0) = \frac{k_B}{16\pi a^2}\left\{\zeta(3) - \mathrm{Li}_3\left[\left(\frac{\varepsilon_0-1}{\varepsilon_0+1}\right)^2\right]\right\} > 0, \qquad (17)$$

in violation of the Nernst heat theorem. Thus, the dc conductivity of a dielectric must not be included in the models of dielectric response. This should be compared with the case of plates made of real metal (see [19, 20] and review [21] for details), where different opinions on the validity of the Nernst heat theorem were proposed. In fact, the mechanisms for the violation of this theorem in some models of metals and dielectrics are quite different. In metals, the validity of the Nernst heat theorem depends on the scattering processes of free charge carriers on phonons, defects (impurities) etc. For the Drude metals with impurities (like in [19]) the residual relaxation at $T = 0$ is not equal to zero and the Nernst heat theorem is satisfied. The same takes place in the case of metals described by the plasma model. For perfect crystal lattices of the Drude metals with no impurities, relaxation at zero temperature is absent and the Nernst heat theorem is violated [20]. All these cases are discussed in [22] devoted to metals. For dielectrics, the validity of the Nernst heat theorem does not depend on the scattering processes due to quick vanishing of the concentration of carriers when the temperature vanishes. Here, the violation occurs due to the inclusion of infinitely large dielectric permittivity at zero frequency. Even the sign of the entropy at zero temperature for metals and dielectrics is opposite (negative for perfect crystal lattices of the Drude metals and positive for dielectrics with the included dc conductivity). For a complete discussion of this subject, containing all mathematical details, see [23] where equation (17) is re-derived in the framework of a more general case of two dissimilar dielectrics.

The above results are important for many applications of dispersion forces. As an example we refer to the problem of a noncontact atomic friction where the discrepancy between experiment and theory is very large [17, 18]. In [18] it has been proposed that the friction observed in the experiment of [17] could be due to the dc conductivity of an underlying SiO_2 plate described by $\tilde{\varepsilon}_l$. From the preceding discussion, it can be seen that such a proposition would not be in agreement with the thermodynamic constraint. Further applications of this constraint in the theory of dispersion forces are under way (see [23] related to the case of dissimilar dielectrics).

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