

On thermal Casimir force between real metals

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Abstract. The physical reasons why the Drude dielectric function is not compatible with the Lifshitz formula, as opposed to the generalized plasma-like permittivity, are presented. Essentially, the problem is connected with the applicability conditions of the Lifshitz theory. It is shown that the Lifshitz theory combined with the generalized plasma-like permittivity is thermodynamically consistent.

1. Introduction: Casimir free energy and different models of dielectric permittivity

The Casimir effect [1], arising due to the alteration of the spectrum of electromagnetic zero-point oscillations by material boundaries, is one of the most important subjects of interdisciplinary interest. Recent trends go toward complex experimental settings for real materials, including applications to nanotechnology, and their consistent theoretical explanation on the framework of quantum electrodynamics (see, e.g., monographs [2, 3], proceedings [4] and more recent papers [5, 6, 7]) appropriately adapted to real material bodies.

The basic theory of the van der Waals and Casimir forces between dielectric materials in terms of their frequency-dependent dielectric permittivities $\varepsilon(\omega)$ was proposed by Lifshitz [8] for the most simple configuration (two parallel plates/semispaces) and ideal surfaces. In the Lifshitz theory the *free energy* of the fluctuating field between two *infinite, electrically neutral* plane parallel plates of thickness d at temperature T in *thermal equilibrium* is given by [8, 9]

$$\mathcal{F}(a, T) = \frac{k_B T}{2\pi} \sum_{l=0}^{\infty} \left(1 - \frac{1}{2} \delta_{0l}\right) \int_0^{\infty} k_{\perp} dk_{\perp} \left\{ \ln \left[1 - r_{\text{TM}}^2(\xi_l, k_{\perp}) e^{-2aq_l}\right] + \ln \left[1 - r_{\text{TE}}^2(\xi_l, k_{\perp}) e^{-2aq_l}\right] \right\}. \quad (1)$$

Here, a is the separation distance between the plates, k_B is the Boltzmann constant, $\xi_l = 2\pi k_B T l / \hbar$ are the Matsubara frequencies defined for any $l = 0, 1, 2, \dots$, and $k_{\perp} = |\mathbf{k}_{\perp}|$ is the magnitude of the wave vector projection onto the plane of the plates. The reflection coefficients for the two independent polarizations of the electromagnetic field (transverse magnetic, TM, and transverse electric, TE) are expressed [10] in terms of the frequency-dependent dielectric permittivity, $\varepsilon(\omega)$, along the imaginary frequency axis:

$$r_{\text{TM}}(\xi_l, k_{\perp}) = \frac{\varepsilon_l^2 q_l^2 - k_l^2}{\varepsilon_l^2 q_l^2 + k_l^2 + 2q_l k_l \varepsilon_l \coth(k_l d)}, \quad r_{\text{TE}}(\xi_l, k_{\perp}) = \frac{k_l^2 - q_l^2}{q_l^2 + k_l^2 + 2q_l k_l \coth(k_l d)}, \quad (2)$$

where

$$q_l = \sqrt{k_{\perp}^2 + \xi_l^2 / c^2}, \quad k_l = \sqrt{k_{\perp}^2 + \varepsilon_l \xi_l^2 / c^2}, \quad \varepsilon_l = \varepsilon(i\xi_l). \quad (3)$$

The Lifshitz theory remains the basis also for real metals and dielectrics and more complicated experimental settings, usually ignoring the finite extension of the material probes under appropriate conditions. However, depending on theoretical model for their dielectric permittivity this theory meets problems and paradoxes.

Especially, the application of the *Drude model* for metals,

$$\varepsilon_D(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}, \quad (4)$$

where ω_p is the plasma frequency and γ is the relaxation parameter, led to conflict with Nernst's heat theorem for plates with perfect crystal lattice [11, 12], see also [13, 14, 15, 16]. In addition, recent precision measurements of the Casimir force [17, 18, 19, 20] excluded the Drude model at a 99.9% confidence level. Also the application of the Lifshitz theory to dielectrics or semiconductors with not too high density of free charge carriers results in a violation of the Nernst heat theorem if the nonzero dc-conductivity at zero frequency is taken into account [21, 22, 23, 24]. Its inclusion also leads to a contradiction with experiment [25, 26, 27].

On the other hand the usual, non-dissipative *plasma model*

$$\varepsilon_p(\omega) = 1 - \frac{\omega_p^2}{\omega^2}, \quad (5)$$

resulting from (4) for $\gamma = 0$, is in agreement with thermodynamics and longer separation experiments [17, 18, 19, 20], but is in contradiction with the experiment performed at short separations [28, 29]. Also the Leontovich surface impedance approach [35] was found to be in agreement with the longer separation experiments just mentioned but, unfortunately, is simply not applicable at shorter separations.

Recently, in order to overcome these difficulties, for real metals the *generalized plasma-like dielectric permittivity* has been proposed [30] as follows

$$\varepsilon_{gp}(\omega) = 1 - \frac{\omega_p^2}{\omega^2} + \sum_{j=1}^K \frac{f_j}{\omega_j^2 - \omega^2 - ig_j\omega}, \quad (6)$$

where $\omega_j \neq 0$ are the resonant frequencies of oscillators describing the core electrons, g_j are the respective relaxation parameters, f_j are the oscillator strengths and K is the number of oscillators; the values of oscillator parameters for different materials can be found in [31]. That model includes dissipation processes due to the interband transitions of core electrons but disregards dissipation due to scattering processes of free electrons. As was shown in [30], the Lifshitz formula combined with the generalized plasma-like dielectric permittivity is consistent with both short and long separation experiments. It also exactly satisfies the Kramers-Kronig relations [30]. In [32] it has been shown that this model also satisfies the Nernst heat theorem and, therefore, is compatible with thermodynamics.

Here, following the lines of Ref. [32] and adopting the point of view of Ref. [33], we first show why the Drude dielectric function is incompatible with the Lifshitz formula. Then, we derive an analytic asymptotic expression for the Casimir free energy in the limit of low temperatures. Finally, we show the validity of the Nernst heat theorem in the Lifshitz theory combined with the generalized plasma-like dielectric permittivity.

2. Incompatibility of Drude model with the Lifshitz formula for metallic plates

Despite originally derived for dielectric plates of infinite area, Eq. (1) is correctly used [9, 34] also for both dielectric and ideal metal plates of finite area S under the condition $a \ll \sqrt{S}$. However,

when plates of real metal are described by the Drude dielectric function (4), the appearance of a drift current of conduction electrons leads to a crucially new physical situation.

To gain a better understanding of this statement, let us derive Eq. (4) starting from Maxwell equations in an unbounded nonmagnetic metallic medium,

$$\begin{aligned}\operatorname{rot}\mathbf{B} &= \frac{1}{c}\frac{\partial\mathbf{E}}{\partial t} + \frac{4\pi}{c}\sigma_0\mathbf{E}, & \operatorname{div}\mathbf{B} &= 0, \\ \operatorname{rot}\mathbf{E} &= -\frac{1}{c}\frac{\partial\mathbf{B}}{\partial t}, & \operatorname{div}\mathbf{E} &= 0.\end{aligned}\quad (7)$$

Here, the electric current density $\mathbf{j} = \sigma_0\mathbf{E}$ is induced in a metal under the influence of external sources, and σ_0 is the conductivity at zero frequency. Physically the demand that the medium is unbounded means that it should be much larger than the extension of the wave fronts of electromagnetic waves coming from external sources (i.e., of zero-point oscillations and thermal photons).

Solving Eqs. (7) by monochromatic waves, $\mathbf{E} = \operatorname{Re}[\mathbf{E}_0(\mathbf{r})e^{-i\omega t}]$ and $\mathbf{B} = \operatorname{Re}[\mathbf{B}_0(\mathbf{r})e^{-i\omega t}]$ with $\mathbf{E}_0(\mathbf{r})$ and $\mathbf{B}_0(\mathbf{r})$ satisfying the well known wave equations, the wave vector \mathbf{k} obeys,

$$k^2 = \frac{\omega^2}{c^2} + i\frac{4\pi\sigma_0\omega}{c^2} \equiv \frac{\varepsilon_n(\omega)\omega^2}{c^2}. \quad (8)$$

Here, the dielectric permittivity of the *normal skin effect* is introduced being defined as

$$\varepsilon_n(\omega) = 1 + i\frac{4\pi\sigma_0}{\omega} \quad \text{with} \quad \sigma_0 = \omega_p^2/(4\pi\gamma). \quad (9)$$

This equation is applicable at not too high frequencies (the region of the normal skin effect) where the relation $\mathbf{j} = \sigma_0\mathbf{E}$ is valid.

In order to extend the applicability of (9) to higher frequencies, up to the plasma frequency, the following replacement is made,

$$\sigma_0 \rightarrow \sigma_D(\omega) = \frac{\sigma_0\left(1 + \frac{i\omega}{\gamma}\right)}{1 + \frac{\omega^2}{\gamma^2}}, \quad (10)$$

herewith obtaining the (complex) conductivity of the Drude model; namely, substituting (10) in (9) we recover the dielectric permittivity (4).

Obviously, in the limit $\omega \ll \gamma$, in both the numerator and the denominator of (10), unity dominates over ω/γ and ω^2/γ^2 leading back to the *real conductivity* of conduction electrons σ_0 . This converts the dielectric permittivity of the Drude model (4) in the dielectric permittivity of the normal skin effect (9). Contrary, at sufficiently high frequencies $\gamma \ll \omega < \omega_p$ (the region of infrared optics) one can neglect unity as compared to ω/γ and ω^2/γ^2 . Then (4) and (10) lead to the plasma model with dielectric permittivity (5) and *purely imaginary conductivity*,

$$\sigma_P(\omega) = i\frac{\sigma_0\gamma}{\omega}. \quad (11)$$

The total current in the framework of the Drude model (4) is given by

$$\begin{aligned}\mathbf{j}_{\text{tot}}(\mathbf{r}, t) &= \operatorname{Re}\left[-\frac{i\omega}{4\pi}\varepsilon_D(\omega)\mathbf{E}_0(\mathbf{r})e^{-i\omega t}\right] \\ &= \frac{\omega}{4\pi}\left(1 - \frac{\omega_p^2}{\omega^2 + \gamma^2}\right)\operatorname{Im}\left[\mathbf{E}_0(\mathbf{r})e^{-i\omega t}\right] + \frac{\sigma_0\gamma^2}{\omega^2 + \gamma^2}\operatorname{Re}\left[\mathbf{E}_0(\mathbf{r})e^{-i\omega t}\right].\end{aligned}\quad (12)$$

The first term on the right-hand side of this equation has the meaning of the *displacement current*, whereas the second term is proportional to the physical electric field $\mathbf{E} = \mathbf{E}(\mathbf{r}, t)$ and describes the *real current of conduction electrons*. Under the condition $\gamma \ll \omega < \omega_p$, i.e., in the region of infrared optics, the first term on the right-hand side of (12), i.e., the displacement current of the plasma model with pure imaginary conductivity dominates. Under the opposite condition $\omega \ll \gamma$, i.e., in the region of the normal skin effect, the second term, i.e., the real physical current of conduction electrons dominates.

By the foregoing discussion we found that the Drude dielectric function (4) is obtained from the Maxwell equations with, besides of the displacement current, also a real drift current of conducting electrons initiated by the external electric field \mathbf{E} . That current unavoidably leads to a Joule heating of the Casimir plates and an irreversible process driving the system out of thermal equilibrium [35]. In order to held the temperature constant it is necessary to admit an unidirectional flux of heat from the system to the heat bath [36]. However, this is prohibited by the definition of thermal equilibrium which requires not only that $T = \text{const}$, but also that all irreversible processes of dissipation of energy into heat have been terminated [37, 38]. As a consequence, this means that the substitution of the Drude dielectric permittivity into the Lifshitz formula violates at least one of the suppositions of its derivation and, therefore, may lead to a violation of Nernst's theorem and to contradictions with experimental data.

On the other hand, the dielectric permittivity of the plasma model is connected with only the displacement current which does not lead to Joule's heating and, therefore, no violation of thermodynamics should appear. Consequently, the free electron plasma model in combination with the Lifshitz formula satisfies the Nernst heat theorem [11, 12]. However, since it is in disagreement with the measurement of the Casimir force in short separation experiments it should be extended in such a way that additional dispersion forces do not lead to real drift currents of conducting electrons. This was achieved by the approach using the generalized plasma-like permittivity [30].

3. Thermodynamic test for the generalized plasma-like dielectric permittivity

Now, we demonstrate that the generalized plasma-like permittivity (6), which is in agreement with all experiments performed up to date also satisfies the requirements of thermodynamics. The generalized plasma-like dielectric permittivity takes into account the interband transitions of core electrons but does not include relaxation of the free conduction electrons. The latter are described by an oscillator with zero resonant frequency, $\omega_0 = 0$, which is explicitly included in by the second term of (6) with $g_0 = 0$ and $f_0 = \omega_p^2$. Thus, similar to the usual plasma model, the generalized plasma-like permittivity admits only the displacement current and does not allow for a heat transfer. Because of this, the generalized plasma-like permittivity is compatible with the Lifshitz formula. Reviewing shortly the proof of Ref. [32] we show that the Lifshitz formula combined with the generalized plasma-like permittivity is in agreement with Nernst's heat theorem and thus withstands the thermodynamic test.

To find the asymptotic behavior of the Casimir free energy and entropy at low temperature, we first present equations (1), (2) and (6) in terms of the following dimensionless parameters:

$$\begin{aligned} \tilde{\omega}_p &= \frac{\omega_p}{\omega_c} \equiv \frac{1}{\alpha}, & \zeta_l &= \frac{\xi l}{\omega_c} \equiv \tau l, & y &= \sqrt{4a^2 k_{\perp}^2 + \zeta_l^2}, \\ \gamma_j &= \frac{\omega_c^2}{\omega_j^2}, & \delta_j &= \frac{\omega_c g_j}{\omega_j^2}, & C_j &= \frac{f_j}{\omega_j^2}, \end{aligned} \quad (13)$$

where $\omega_c \equiv c/(2a)$ is the so-called characteristic frequency of the Casimir effect and the parameter τ is represented as $\tau = 2\pi T/T_{\text{eff}}$ with $k_B T_{\text{eff}} \equiv \hbar c/2a$; here, T_{eff} is the so-called effective temperature which, for example, at separation $a = 1 \mu\text{m}$ is equal $T_{\text{eff}} \approx 1145 \text{ K}$.

In terms of these new variables, the Lifshitz formula (1) takes the form

$$\mathcal{F}(a, T) = \frac{\hbar c \tau}{32\pi^2 a^3} \sum_{l=0}^{\infty} \left(1 - \frac{1}{2} \delta_{0l}\right) \int_{\zeta_l}^{\infty} dy f(\zeta_l, y) \quad \text{with} \quad (14)$$

$$f(\zeta_l, y) = y \left\{ \ln \left[1 - r_{\text{TM}}^2(\zeta_l, y) e^{-y}\right] + \ln \left[1 - r_{\text{TE}}^2(\zeta_l, y) e^{-y}\right] \right\}. \quad (15)$$

The reflection coefficients (2) are given by

$$r_{\text{TM}}(\zeta_l, y) = \frac{(\varepsilon_l^2 - 1)(y^2 - \zeta_l^2)}{(\varepsilon_l + 1)y^2 + (\varepsilon_l - 1)\zeta_l^2 + 2\varepsilon_l y h_l(y) \coth [(d/2a)h_l(y)]},$$

$$r_{\text{TE}}(\zeta_l, y) = \frac{(\varepsilon_l - 1)\zeta_l^2}{2y^2 + (\varepsilon_l - 1)\zeta_l^2 + 2y h_l(y) \coth [(d/2a)h_l(y)]}, \quad (16)$$

where

$$h_l(y) = \sqrt{y^2 + (\varepsilon_l - 1)\zeta_l^2}. \quad (17)$$

The generalized plasma-like dielectric permittivity along the imaginary frequency axis can be presented as

$$\varepsilon_l = \varepsilon(i\zeta_l) = 1 + \frac{1}{\alpha^2 \zeta_l^2} + \sum_{j=1}^K \frac{C_j}{1 + \gamma_j \zeta_l^2 + \delta_j \zeta_l}. \quad (18)$$

Using the Abel-Plana formula [2, 9]

$$\sum_{l=0}^{\infty} \left(1 - \frac{1}{2} \delta_{0l}\right) F(l) = \int_0^{\infty} F(t) dt + i \int_0^{\infty} dt \frac{F(it) - F(-it)}{e^{2\pi t} - 1}, \quad (19)$$

where $F(z)$ is an analytic function in the right half of the complex plane, we can rearrange (14) to the form

$$\mathcal{F}(a, T) = E(a) + \Delta\mathcal{F}(a, T). \quad (20)$$

Here, the energy of the Casimir interaction at zero temperature is given by

$$E(a) = \frac{\hbar c}{32\pi^2 a^3} \int_0^{\infty} d\zeta \int_{\zeta}^{\infty} f(\zeta, y) dy, \quad (21)$$

and the thermal correction to the Casimir energy is expressed as follows:

$$\Delta\mathcal{F}(a, T) = \frac{i\hbar c \tau}{32\pi^2 a^3} \int_0^{\infty} dt \frac{F(i\tau t) - F(-i\tau t)}{e^{2\pi t} - 1}, \quad (22)$$

where

$$F(x) = \int_x^{\infty} dy f(x, y). \quad (23)$$

The behavior of the thermal correction (22), (23) at low temperature will be the subject of our further consideration.

Perturbation expansion can be performed in analogy to papers [11, 12, 39, 40]. At first, we expand the reflection coefficients $r_{\text{TM}}(\zeta, y)$ and $r_{\text{TE}}(\zeta, y)$ in powers of the parameter α preserving all powers up to the fourth inclusive. Since α can be identically presented as $\alpha = \lambda_p/(4\pi a)$, where λ_p is the plasma wavelength, this means that $\alpha \ll 1$ at all separation distances between the plates larger than λ_p . Furthermore, an expansion at low τ is performed up to fifth order.

As is seen from (15), it is more convenient to expand the logarithmic functions containing the reflection coefficients being multiplied by the variable y . The details of these expansion are contained in Ref. [32] and will not be repeated here.

It is remarkable that these expansions effectively do not depend on d , the thickness of the plates, contained in (16). This is because asymptotically, when α goes to zero, the factor containing the thickness d behaves as $\coth [(d/2a)h_l(y)] \approx 1 + 2 \exp[-d/(a\alpha)]$. Thus, this factor could only contribute exponentially small terms in these expansions providing the plate thickness d is much larger than the penetration depth of electromagnetic oscillations into the metal [recall that $2a\alpha = \lambda_p/(2\pi)$]. Under this condition the perturbation expansions are common for two semispaces and for two plates of finite thickness.

In [32] it was shown that up to second power in α there appear no contributions of the core electrons, i.e., that contributions coincide with those of the free electron plasma model [39, 40],

$$\Delta\mathcal{F}_p(a, T) = -\frac{\hbar c \tau^3}{32\pi^2 a^3} \left\{ \frac{\zeta(3)}{4\pi^2} - \frac{\tau}{360} + \alpha \left(\frac{\zeta(3)}{\pi^2} - \frac{\tau}{45} \right) - \alpha^2 \frac{\zeta(5)}{2\pi^4} \tau^2 \right\}, \quad (24)$$

where $\zeta(z)$ is the Riemann zeta function. As was shown in [40], the terms in (24) of order α^0 and α do not contain corrections of order τ^n with $n \geq 5$; they contain only exponentially small corrections of order $\exp(-2\pi/\tau)$.

Now, let us consider the contributions from the core electrons which are contained only in the terms of order α^3 and α^4 . Their contribution to the thermal correction is as follows:

$$\Delta\mathcal{F}_g(a, T) = \frac{\hbar c \alpha^3 \tau^4}{32\pi^2 a^3} \left\{ \left[\frac{\zeta(3)}{60} \sum_{j=1}^K C_j \delta_j + \tau \frac{3\zeta(5)}{2\pi^4} \left(\sum_{j=1}^K C_j + 2 \right) \right] - \alpha \left[\frac{4\zeta(3)}{15} \sum_{j=1}^K C_j \delta_j + \tau \frac{6\zeta(5)}{\pi^4} \right] \right\}. \quad (25)$$

The total Casimir free energy computed using the generalized plasma-like permittivity can be now found from (20), (24) and (25) as follows:

$$\mathcal{F}(a, T) = E(a) + \Delta\mathcal{F}_p(a, T) + \Delta\mathcal{F}_g(a, T). \quad (26)$$

Taking into account the definition of τ , it can be represented in the form

$$\begin{aligned} \mathcal{F}(a, T) = & E(a) - \frac{\hbar c \zeta(3)}{16\pi a^3} \left(\frac{T}{T_{\text{eff}}} \right)^3 \left\{ 1 + 4\alpha \right. \\ & - \frac{\pi^3}{45\zeta(3)} \frac{T}{T_{\text{eff}}} \left(1 + 8\alpha + 6\zeta(3)\alpha^3 \sum_{j=1}^K C_j \delta_j - 96\zeta(3)\alpha^4 \sum_{j=1}^K C_j \delta_j \right) \\ & \left. - \frac{8\zeta(5)}{\zeta(3)} \left(\frac{T}{T_{\text{eff}}} \right)^2 \alpha^2 \left[1 + 3\alpha \left(\sum_{j=1}^K C_j + 2 \right) - 12\alpha^2 \right] \right\}. \quad (27) \end{aligned}$$

Here one can see that the free energy calculated using the generalized plasma-like permittivity contains the correction of order $(T/T_{\text{eff}})^4$ not only in the terms of order α^0 and α (as in the usual plasma model) but also in the third and fourth order expansion terms in α . In the usual plasma model the terms of order α^3 and α^4 contain the thermal corrections only of order of $(T/T_{\text{eff}})^5$ and higher [39].

To estimate the relative role of the additional terms arising due to the use of the generalized plasma-like permittivity, one can use the parameters of oscillator terms in (18) for Au [20]. This results in

$$\sum_{j=1}^6 C_j = 6.3175, \quad \sum_{j=1}^6 C_j \delta_j = \begin{cases} 0.272, & a = 200 \text{ nm}, \\ 0.109, & a = 500 \text{ nm}. \end{cases} \quad (28)$$

From (27) it is easy to find the asymptotic behavior of the Casimir entropy

$$S(a, T) = -\frac{\partial \mathcal{F}(a, T)}{\partial T} \quad (29)$$

at low temperature. Without performing the simple differentiation it is immediately seen from (27) that the entropy goes to zero (and remains positive) when the temperature vanishes,

$$S(a, T) \rightarrow 0 \quad \text{when} \quad T \rightarrow 0. \quad (30)$$

This means that the Nernst heat theorem is satisfied and the Lifshitz theory combined with the generalized plasma-like dielectric permittivity withstands the thermodynamic test.

4. Conclusions and discussion

In the foregoing we have discussed the physical reasons why the Drude dielectric function is not applicable in the case of finite plates made of real metal. It was shown that for the validity of the Drude model a nonzero current of conduction electrons exists, which leads to a Joule's heating of the Casimir plates. As a consequence, one of the suppositions for the derivation of the Lifshitz formula, namely, thermal equilibrium is violated. Of course, any dielectric permittivity being related to a drift current of conducting electrons is ruled out by that consideration.

On the contrary, not only the free electron plasma dielectric permittivity but also the generalized plasma-like permittivity leads to only a displacement current and does not result to a dissipation of energy into heat. From this consideration it becomes clear why the generalized plasma-like permittivity, which does not include the relaxation processes of conduction electrons, is consistent with all available measurements of the Casimir force at both short and large separations.

To conclusively establish the applicability of the generalized plasma-like permittivity in the theory of the thermal Casimir force between real metals we reviewed the thermodynamic test of this model. Studying the asymptotic behavior of both the Casimir free energy and Casimir entropy at low temperature it is shown that the latter is positive and takes zero value at zero temperature. Thus, the Nernst heat theorem is satisfied. Of course, when the oscillator parameters describing the core electrons go to zero, the newly obtained expressions for the Casimir free energy and entropy go into the ones found for the usual plasma model.

From this we conclude that no problems with the Lifshitz theory occur if it is applied under observation of its region of applicability. Similar conclusion is made in Ref. [33] agreeing "with the fact that the conductivity processes in real materials connected with the drift current of conduction electrons violate the applicability conditions of the Lifshitz theory and, thus, are not described by this theory." Since these processes, however, exist in reality, one must ask if they are to "be taken into account in some future general theory of dispersion forces."

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