Comment on “Does the transverse electric zero mode contribute to the Casimir effect for a metal?”

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Recently J. S. Høye, I. Brevik, J. B. Aarseth, and K. A. Milton [Phys. Rev. E 67, 056116 (2003)] proposed that if the Lifshitz formula is combined with the Drude model, the transverse electric zero mode does not contribute to the result for real metals and there arises a linear temperature correction to the Casimir force at small temperatures. The authors claim that in spite of the fact that the Casimir entropy in their approach is negative, the Nernst heat theorem is satisfied. In the present Comment we show that the authors’ conclusion regarding the Nernst heat theorem is in error. We demonstrate also the resolution of this thermodynamic puzzle based on the use of the surface impedance instead of the Drude dielectric function. The results of numerical computations obtained by the authors are compared with those from use of the surface impedance approach which are thermodynamically consistent.

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A recent calculation shows that the transverse electric zero mode does not contribute to the Casimir free energy [1]. This results in an observable linear temperature correction to the Casimir force between parallel plates made of real metal [1]. According to Ref. [1], the Casimir free energy increases with increasing temperature in some temperature interval, and the Casimir entropy becomes negative in this interval. From the viewpoint of the authors of Ref. [1], this, however, does not lead to violation of the Nernst heat theorem. Below we show that on the contrary the transverse electric zero mode does contribute to the Casimir free energy which results in the absence of a linear in temperature correction to the Casimir force at small temperatures. In reality, the Casimir free energy is universally a decreasing function of temperature, and the Casimir entropy is always positive. We demonstrate also that if, as is done in Ref. [1], the Drude model and the Bloch-Grüneisen formula are used to calculate the Casimir force, the Nernst heat theorem is necessarily violated.

The correct description of the thermal Casimir force between real metals has assumed great importance due to the recent precision experiments [2–8], followed by the prospective applications of the Casimir effect in nanotechnology [9,10] and also its use as a test for predictions of fundamental physical theories [11–14]. Unexpectadly, it was found that the calculations of the thermal Casimir force on the basis of the Lifshitz formula [15] supplemented by the Drude model ran into serious difficulties [16–29]. The key question of the controversy is whether the transverse electric zero mode contributes to the Casimir effect in the case of real metals. Boström and Sernelius [16] have used the Drude model to compute the contributions of both transverse electric and transverse magnetic modes and found that the transverse electric zero mode does not contribute. Later on it was shown [26,28] that in this approach the Casimir entropy is negative in some temperature interval and the third law of thermodynamics (the Nernst heat theorem) is violated. On the contrary, the authors of Ref. [1] claim that the entropy is zero at zero temperature, i.e. the third law of thermodynamics is not violated.

To prove this statement they present in Sec. IV of Ref. [1] two sets of arguments, analytical and numerical. Both of them start from Eq. (3.4) representing the Lifshitz result for the Casimir free energy per unit area in the configuration of two semispaces separated by a distance $a$:

$$\beta F = \frac{1}{2\pi} \sum_{m=0}^{\infty} \int_{\zeta_m}^{\infty} \left[ \ln \left( 1 - \lambda_{TM}^m \right) + \ln \left( 1 - \lambda_{TE}^m \right) \right] q \, dq$$

(1)

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where
\[ \lambda_m^{TM} = A_m e^{-2qa}, \quad \lambda_m^{TE} = B_m e^{-2qa}, \quad A_m = \frac{\varepsilon_m q - \sqrt{(\varepsilon_m - 1)\zeta_m^2 + q^2}}{\varepsilon_m q + \sqrt{(\varepsilon_m - 1)\zeta_m^2 + q^2}}, \quad B_m = \frac{q - \sqrt{(\varepsilon_m - 1)\zeta_m^2 + q^2}}{q + \sqrt{(\varepsilon_m - 1)\zeta_m^2 + q^2}}. \]

Here the dielectric permittivity \( \varepsilon_m \equiv \varepsilon(i\zeta_m) \) is computed on the imaginary Matsubara frequencies, \( \zeta_m = 2\pi m/\beta, \beta \equiv 1/T, \) and units with \( k_B = \hbar = c = 1 \) are used.

According to the Drude model
\[ \varepsilon(i\zeta) = 1 + \frac{\omega_p^2}{\zeta(\zeta + \nu)}, \]
where \( \omega_p \) is the plasma frequency, \( \nu = \nu(T) \) is the relaxation parameter. Substituting Eq. (3) into Eq. (2) one obtains \( B_0(0) = 0 \), i.e. the transverse electric zero mode does not contribute in the case of the Drude metal in accordance with Refs. [1,16].

The authors advance three analytical arguments in support of the validity of the Nernst heat theorem in Ref. [1], but no direct proof is given. As the starting point, two simple systems containing three harmonic oscillators each are considered. It is shown that for some “subsystem” the entropy may be negative. Needless to say, systems of this type are not realistic models of full-scale Quantum Electrodynamical interaction between the plates made of real metal as they do not reflect the main features of this interaction. Next, as a “real metal” the case where \( \nu \) is independent of \( \zeta \) is considered. Up to order of magnitude estimation for the entropy is obtained consistent with the Nernst heat theorem. This estimation, however, is not relevant to the Drude metal because the dielectric permittivity (3) depends on frequency. The final analytical argument of Ref. [1] is for the frequency-dependent \( \varepsilon \). In this case, however, no result is obtained. The authors speculate that “For a real metal obeying the Drude dispersion relation (3) (with \( \nu \neq 0 \)),... we expect a \( T^3 \) (or \( T^4 \)) correction to the free energy at sufficiently low temperature.” If this were the case, the Nernst heat theorem would be satisfied. We show below that this is, however, not the case.

To find the asymptotic behavior of the free energy, given by Eqs. (1)-(3), on temperature in the region of low temperatures, \( T \ll T_{eff} = 1/(2a) \), one needs the explicit dependence of the relaxation parameter \( \nu \) on temperature. This is given by the Bloch-Grüneisen law through the linear connection between the relaxation parameter and the static resistivity. As a result, \( \nu \sim T^\alpha \) where at low temperatures \( \alpha \approx 5 \) whereas at high temperatures \( \alpha = 1 \) [1,26,28] (the tabulated data for different metals can be found in Ref. [30]). Note that \( \nu(T) \neq 0 \) except of one point \( T = 0 \). Performing the perturbation expansion in Eqs. (1)-(3) in powers of three small parameters, \( T/T_{eff}, \nu/\omega_p \) and \( \lambda_p/(2\pi a) \) (we consider separation distances \( a \geq \lambda_p \) where \( \lambda_p \) is the plasma wavelength), the following result for the free energy is obtained
\[ F = -\frac{\pi^2}{720a^3} \left[ 1 - 2\frac{\lambda_p}{\pi a} + \frac{18}{5}\frac{\lambda_p^2}{\pi^2 a^2} \right] - \frac{\zeta(3)}{16\pi a^3} \left[ \left( 1 + \frac{\lambda_p}{\pi a} \right) \left( T/T_{eff} \right)^3 - \frac{\pi^3}{45\zeta(3)} \left( 1 + 2\frac{\lambda_p}{\pi a} \right) \left( T/T_{eff} \right)^4 \right] \]
\[ + \frac{\zeta(3) T}{16\pi a^2} \left[ 1 - 2\frac{\lambda_p}{\pi a} + \frac{3\lambda_p^2}{\pi^2 a^2} \right] + \nu \frac{T}{\omega_p 4\pi a^2} \sum_{m=1}^{\infty} \left[ \frac{\zeta_m}{e^y - 1} + \frac{1}{\zeta_m} \int_{\zeta_m}^{\infty} \frac{y^2 dy}{e^y - 1} \right]. \]

Here the nondimensional Matsubara frequencies are defined as \( \tilde{\zeta}_m = 2a\zeta_m = 4\pi ma/\beta \). All the details of derivation of perturbation Eq. (4) can be found in Ref. [26].

It is quite clear from Eq. (4) that the expectations of Ref. [1] are not borne out. In addition to the higher-order terms in temperature, there is a linear term which leads to a nonzero value of entropy at zero temperature.
Thus, we confirm the conclusion of Refs. [26,28] that in the framework of the Lifshitz formula supplemented by the Drude model with a temperature-dependent relaxation parameter (see Fig. 6 of Ref. [1]) the Nernst heat theorem is in fact violated.

One may mention, as the authors of Ref. [1] do in the end of Appendix D, that their dependence of $\nu$ on $T$ neglects the effect of impurities, which give rise to some nonzero residual resistivity at zero temperature [31]. This, however, cannot remedy the situation concerning the inconsistency with the Nernst heat theorem. In fact, the resistivity ratio of a sample can be defined as the ratio of its resistivity at room temperature to its residual resistivity. For pure samples the resistivity ratio may be as high as $10^6$ [31]. As an example let us consider Au with $\nu(T = 300\,K) = 5.32 \times 10^{13}\,\text{rad/s}$ [30]. In this case for the residual value of the relaxation parameter one obtains $\nu_{res} = 5.32 \times 10^7\,\text{rad/s}$. The asymptotic expression (4) is applicable under the condition $\nu \ll \zeta = 2\pi T$ (this condition is automatically fulfilled for the above used dependence $\nu(T) \sim T^\alpha$ with $\alpha \geq 1$). Thus, with allowance made for impurities, the asymptotic (4) is applicable at temperatures $T \gg \nu_{res}/(2\pi) = 6.5 \times 10^{-5}\,\text{K}$. What this means is that the entropy at temperature $T = 5 \times 10^{-4}\,\text{K}$ has a nonzero negative value given by Eq. (5). Physically this is equivalent to the violation of the Nernst heat theorem. We would like to point out also that the usual theory of the electron-phonon interaction, describing the electrons interacting with the elementary excitations of a perfect lattice with no impurities, must satisfy and does satisfy all the requirements of thermodynamics. That is why, the attempt to remedy the violation of the Nernst heat theorem at the expense of impurities is meaningless.

We would now like to address the numerical arguments of Ref. [1] in support of the validity of the Nernst heat theorem in the Lifshitz theory combined with the Drude model. All the numerical computations in Ref. [1] are performed for Au with $\omega_p = 9.0\,\text{eV}$, $\nu(T = 300\,K) = 35\,\text{meV}$. Once again, no direct computations by Eqs. (1)-(3) with $\nu(T)$ given by Fig. 6 of Ref. [1] are done. Instead, when calculating force-temperature relation, the room temperature data for $\varepsilon(i\zeta)$ or the above value of $\nu(T = 300\,K)$ were used ignoring the correct temperature dependence [e.g. our Eq. (3) combined with Fig. 6 of Ref. [1] presenting $T$-dependence of $\nu$]. All computed force-temperature and force-distance curves demonstrate nonmonotonous behavior (see Figs. 2-4 of Ref. [1]). This is a counterintuitive effect as the authors of Ref. [1] themselves recognize. In fact, with an increase of temperature the population of modes and, consequently, force modulus should increase. The nonmonotonous character of the force curves is an artifact. It is accounted for by the absence of the transverse electric zero mode contribution for metals in the formalism under discussion (see below).

In an effort to confirm the validity of the Nernst heat theorem in their formalism, the authors of Ref. [1] compute numerically the force-temperature dependence for dielectrics with constant $\varepsilon = 10^2, 10^3, 10^4$, and $\varepsilon = \infty$ (Fig. 5 of Ref. [1]). They consider the horizontal slope near the point $T = 0$, which is present in all curves, except for $\varepsilon = \infty$ and for Au as a desirable property. (“If the force had a linear dependence on $T$ for small $T$ so would the free energy $F$, in contradiction with the requirement that the entropy $S = -\partial F/\partial T$ has to go to zero as $T \rightarrow 0$.”) The absence of a desirable slope for Au is considered as a lack of resolution on the scale of the Figure. It is noted also in [1] that not only dispersive but also nondispersive curves are nonmonotonous.

All these conclusions depend on the authors’ physically incorrect assumption that there exist dielectrics with arbitrary large constant $\varepsilon$ and that metals may be considered as a limiting case of such kind dielectrics when $\varepsilon \rightarrow \infty$ on the imaginary frequency axis. In actual truth, $\varepsilon$ can be assumed to be frequency- and temperature-independent only in the case of so called non-polar dielectrics whose atoms or molecules do not have their own dipole moments. The electric susceptibility of non-polar dielectrics arises due to the electronic polarization of atoms and molecules. The values
of $\varepsilon$ for non-polar dielectrics are of order of one only [30,32,33]. Large values of $\varepsilon$ can exist only for polar dielectrics where the partial orientation of permanent dipole moments occurs. But for polar dielectrics $\varepsilon$ depends strongly on the frequency and temperature. Specifically, their $\varepsilon$ quickly decreases with the increase of frequency. As a result, at optical and infrared frequencies, which are characteristic for the Casimir effect, the values of $\varepsilon$ are determined by the electronic polarizability [30] and cannot exceed several units. Thus, the nonmonotonous force-temperature curves presented in Fig. 5 of Ref. [1] are not relevant to the resolution of the above puzzle with the Nernst heat theorem.

The reference to the experiment by Bressi et al [8] where, supposedly, “the observed Casimir forces were lower than those predicted by the traditional (SDM) theory for conducting plates, in cases where the distances were low, $a \leq 0.5\mu m$” is a misunderstanding. According to Ref. [1], this reduction effect is apparent from Fig. 4 of [8] and could experimentally confirm the characteristic temperature variations predicted by the authors. In Fig. 4 of [8], however, all data are presented only for $a > 0.5\mu m$ and there is no evidence that these data support expectations of [1].

In view of the above, we arrive at the conclusion that the arguments presented in Ref. [1] are not correct. The use of the Drude model in combination with the Lifshitz formula actually leads to the violation of the Nernst heat theorem and the other nonphysical features such as a linear temperature correction to the Casimir force at small separations, negative values of entropy and nonmonotonous force-temperature and force-distance relations.

Recently the resolution of these complicated problems was obtained [29] using an alternative approach to the description of real metals based on the concept of the surface impedance. This approach offers a fundamental understanding of the reason why the Drude model is not compatible with the theory of the thermal Casimir force between real metals. This is due to the fact that the Drude dielectric function (3) is obtained for the frequency region of the normal skin effect, where the electromagnetic oscillations penetrate through the skin layer, and lead to a real current of the conduction electrons. The interaction of the conduction electrons with the elementary excitations of the crystal lattice (phonons) leads to the occurrence of electric resistance and heating of a metal. By contrast, the thermal photons in thermal equilibrium with a metal plate or, much less, the virtual photons giving rise to the Casimir effect cannot, under any circumstances, lead to the initiation of a real current and heating of a metal. Of course, this is strictly prohibited by thermodynamics. Hence the standard concept of a fluctuating electromagnetic field penetrating inside a metal described by the Drude dielectric function (3) fails to describe virtual and thermal photons. As a consequence, the Lifshitz formula in combination with the Drude model leads to the above contradictions with thermodynamics. Note that the dielectric permittivity depending only on frequency is also inapplicable in the frequency domain of the anomalous skin effect (see Ref. [29] for details).

In contrast to the dielectric permittivity, the surface impedance $Z(\omega)$ is defined at all frequencies. The impedance boundary conditions are

$$E_t = Z(\omega) [B_t \times \mathbf{n}],$$

where $E_t, B_t$ are the tangential components of electric and magnetic fields, $\mathbf{n}$ is the internal normal vector to the surface. They take into account the reflection properties of real metal with no consideration of the electromagnetic fluctuations inside of it.

By the use of the surface impedance instead of the Drude model (3), the Lifshitz formula (1) is preserved, but the coefficients $A_m, B_m$ from Eq. (2) should be replaced by [29]

$$A_{m}^{imp} = \left( \frac{q - Z_m \zeta_m}{q + Z_m \zeta_m} \right)^2, \quad B_{m}^{imp} = \left( \frac{\zeta_m - Z_m q}{\zeta_m + Z_m q} \right)^2,$$

(7)
where the impedance is computed on the Matsubara frequencies. Substituting in Eq. (7) the impedance function of the normal skin effect or the anomalous skin effect, one finds $B_{imp}^0(0) = 1$ \[29\]. In the region of the infrared optics it follows \[29\] $B_{imp}^0(0) = (\omega_p - q)^2/(\omega_p + q)^2$.

Let us now present several computational results obtained by Eqs. (1), (7) in the framework of the impedance approach in comparison with the results of Ref. [1]. In Fig. 1, the magnitude of the Casimir surface force density $\mathcal{F}^T = -\partial F/\partial T$ for gold is computed, in the temperature interval $1 \, \text{K} \leq T \leq 1200 \, \text{K}$ at a separation distance $a = 1 \, \mu m$. Solid line is calculated in the framework of the impedance approach (separation of 1 $\mu m$ corresponds to the domain of the infrared optics), and dashed line is obtained by the approach of Ref. [1] (i.e. via the Lifshitz formula supplemented by the Drude model with a temperature dependent relaxation parameter). It is clearly seen that the dashed line is nonmonotonous demonstrating the existence of a wide temperature region where force modulus decreases with an increase of temperature (like in Figs. 2, 3 of Ref. [1]). At the same time, solid line demonstrates the monotonous increase of the modulus of the Casimir force with temperature which is consistent with our expectations on the basis of thermodynamics.

In Fig. 2, the magnitude of the Casimir force density between dielectrics with $\varepsilon = const$ is computed at different temperatures at a separation of $a = 1 \, \mu m$. Both lines were obtained by the usual Lifshitz formula (1), (2) with $\varepsilon_m = \varepsilon = const$. Solid line is for mica with $\varepsilon = 7$; dashed line faithfully copies the line of Fig. 5 of Ref. [1] with $\varepsilon = 100$. Solid line demonstrates the monotonous increase of the Casimir force with increasing temperature as is expected from thermodynamics. Dashed line is nonmonotonous because non-polar dielectrics with so high $\varepsilon$ do not exist in nature due to bounds on the possible values of electronic polarization. From this figure it is clear that the Lifshitz formula for dielectrics is consistent with thermodynamics when the correct input data for $\varepsilon$ are substituted.

In Fig. 3, the Casimir entropy for gold is plotted as a function of temperature at a separation distance between plates of $a = 1 \, \mu m$. Solid line is computed in the framework of the impedance approach. Dashed line is obtained by the approach of Ref. [1], i.e. by the usual Lifshitz formula and Drude model (1)-(3) with a relaxation parameter depending on temperature according to Fig. 6 of Ref. [1]. Evidently, the solid line satisfies all conditions, i.e. positive values of entropy at nonzero temperatures, and the validity of the Nernst heat theorem. By contrast, the dashed line presents negative values of entropy and the violation of the Nernst heat theorem (for two semispaces separated by a gap there is no possibility to introduce some “composite system” whose “subsystem” with a negative entropy these semispaces would be; for this reason the Casimir entropy must be positive which is in fact the case in the impedance approach). The analytical proof of the validity of the Nernst heat theorem in the impedance approach can be found in Ref. [29].

Now we are in a position to answer the questions raised in the introductory note to this Comment. The answer to the question on whether the transverse electric zero mode contributes to the Casimir effect is yes. As a consequence, there is no linear temperature correction to the Casimir force between metallic plates at small temperatures. The Casimir free energy is always decreasing and the force magnitude increases with increase of temperature. As to the Casimir entropy, it is always positive and obeys Nernst’s theorem as it must be in accordance with thermodynamics. Regarding the doubts not only on the applicability of the Drude model as such, but even more, doubt on the applicability of the fundamental Lifshitz formula, they are carried too far. Although, as discussed above, the Drude model is in fact not appropriate to describe the thermal Casimir effect in the case of real metals, the Lifshitz formula (1) with coefficients (7) in terms of the surface impedance is perfectly well suited to calculate all the quantities of physical interest.
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FIG. 1. Magnitude of surface force density for gold versus temperature when $a = 1 \mu m$. The solid line is the physical result calculated in the framework of the impedance approach. The dashed line is obtained by the use of the Drude model like in Ref. [1].
FIG. 2. Magnitude of surface force density for nondispersive dielectrics versus temperature when $a = 1 \mu m$. The solid line is the physical result calculated by the usual Lifshitz formula for mica. The dashed line is obtained for non-existent non-polar dielectric with $\varepsilon = 100$ used in Ref. [1].
FIG. 3. Casimir entropy for two gold semispaces versus temperature when $a = 1 \mu m$. The solid line is the physical result calculated in the framework of the impedance approach. The dashed line is obtained by the use of the Drude model like in Ref. [1].