

## Casimir entropy and nonlocal response functions to the off-shell quantum fluctuations

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The nonlocal response functions to quantum fluctuations are used to find asymptotic expressions for the Casimir free energy and entropy at an arbitrarily low temperature in the configuration of two parallel metallic plates. It is shown that by introducing an alternative nonlocal response to the off-the-mass-shell fluctuations the Lifshitz theory is brought into agreement with the requirements of thermodynamics. According to our results, the Casimir entropy calculated using the nonlocal response functions, which take into account dissipation of conduction electrons, remains positive and monotonously goes to zero with vanishing temperature, i.e., satisfies the Nernst heat theorem. This is true for both plates with perfect crystal lattices and for lattices with defects of structure. The obtained results are discussed in the context of the Casimir puzzle.

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### I. INTRODUCTION

The Casimir effect is the most impressive physical phenomenon demonstrating an existence of the zero-point oscillations of quantum fields. As was shown by H. B. G. Casimir [1], two parallel ideal metal plates at zero temperature placed in vacuum at a distance  $a$  are attracted by the force per unit area  $-\pi^2\hbar c/(240a^4)$ , which is completely determined by the zero-point oscillations of the electromagnetic field. Shortly thereafter E.M. Lifshitz created unified theory of the van der Waals and Casimir forces between two parallel plates made of any materials kept at temperature  $T$  in thermal equilibrium with the environment [2,3]. In the framework of this theory, the Casimir force is caused by the joint action of zero-point and thermal fluctuations. In so doing the response of plate materials to quantum fluctuations is described by the frequency-dependent dielectric permittivities, whereas the van der Waals force proves to be a special case of the Casimir force when separations between the plates are so small that the speed of light  $c$  can be considered as infinitely large. In recent years, the Lifshitz theory was generalized for the case of two compact arbitrarily shaped bodies [4–6]. As a result, it has become possible to express the Casimir force between any two bodies using the formalism of thermal quantum field theory in terms of the reflection amplitudes of quantum fluctuations on their boundary surfaces.

In the early twenty first century, precise measurements of the Casimir interaction between metallic surfaces performed by R. S. Decca *et al.* [7–10] discovered a serious

discrepancy between theoretical Casimir forces calculated using the Lifshitz theory and the measurement data. In calculations, the response of metal to low-frequency quantum fluctuations was described by the dissipative Drude model, i.e., in the same manner as to real electromagnetic fields on the mass shell with nonzero field strengths. The surprising thing is that the Lifshitz theory agreed with the same experimental data if the dissipationless plasma response function was used which should not be applicable at low frequencies. More recently, the results of Refs. [7–10] were conclusively confirmed by many experiments [11–19] (see also reviews [20,21]). This situation has been characterized in the literature as the Casimir puzzle or Casimir conundrum [22–26].

At the same time it was shown that the Casimir entropy calculated within the Lifshitz theory for metals with perfect crystal lattices goes to a negative quantity depending on the plate parameters with vanishing temperature, i.e., violates the third law of thermodynamics, the Nernst heat theorem, if the Drude response function to quantum fluctuations is used [27–31]. What is more, employing the experimentally consistent plasma model brings the Lifshitz theory in agreement with thermodynamics [27–31]. The reasons why the evidently inapplicable at low frequencies response function leads to so good results whereas the well tested Drude response fails to reach an agreement with the measurement data and thermodynamical laws remained unknown.

It should be mentioned that several authors argued for an accord between the Lifshitz theory employing the Drude

response function and thermodynamics [32–34]. In favor of this statement they have shown that if the crystal lattice of a metal contains some fraction of defects the Casimir entropy abruptly jumps to zero at very low temperature starting from negative values. For the perfect crystal lattice, however, it remained impossible to reconcile the Casimir entropy calculated using the Drude model with thermodynamics. Besides, the consideration of imperfect crystal lattices was not helpful to attain an agreement between the experimental data and theoretical predictions. Many attacks to this problem have been undertaken in the literature (see, e.g., Refs. [35–48]) but no wholly satisfactory solution is yet available.

Quite recently a novel approach to the resolution of the Casimir puzzle has been proposed [49]. This approach assumes that the electromagnetic response of a metal to quantum fluctuations is spatially nonlocal. However, unlike the commonly applied nonlocal response functions [36,50–55], the suggested response leads to nearly the same results, as the standard Drude function, for the quantum fluctuations on the mass shell, but to significantly different results for the off-shell fluctuations. The proposed alternative Drude-like transverse and longitudinal dielectric permittivities take a complete account of dissipation at low frequencies, as does the conventional Drude model. It was shown, however, that the Lifshitz theory using the nonlocal response functions to quantum fluctuations predicts values of the Casimir force which agree with the measurement data as well as those predicted using the dissipationless plasma model. It was also shown that the suggested nonlocal dielectric functions describe correctly the response of metal to real electromagnetic fields on the mass shell in optical experiments and obey the Kramers-Kronig relations. In essence, the possibility to introduce the nonlocal Drude-like functions is based on the fact that the electromagnetic response of a metal to quantum fluctuations off the mass shell is not directly measurable. Experimentally, some indirect information can be obtained only concerning the longitudinal dielectric function [55] which, however, does not affect a comparison between theoretical Casimir forces and the measurement data [49].

In this paper, we find the Casimir free energy and entropy given by the Lifshitz theory and the nonlocal response functions introduced in Ref. [49]. We derive an analytic asymptotic expression for the Casimir free energy at arbitrarily low temperature and show that for metals with perfect crystal lattices the respective Casimir entropy is positive and goes to zero with vanishing temperature. Thus, as opposed to the conventional Drude model, the Lifshitz theory using nonlocal response functions is in agreement with the laws of thermodynamics. It is shown also that this result remains valid for crystal lattices with some fraction of defects, i.e., the Casimir entropy remains positive and monotonously goes to zero with decreasing temperature according to the Nernst heat theorem.

The structure of the paper is as follows. In Sec. II, we briefly introduce the nonlocal response functions and basic expressions of the Lifshitz theory for the Casimir free energy in spatially nonlocal case. Section III contains derivation of the low-temperature expansion for the Casimir free energy and entropy using these response functions for metals with perfect crystal lattices. In Sec. IV, the case of crystal lattices with defects of structure is considered. Section V is for our conclusions and a discussion. The Appendix contains some details of mathematical derivations.

## II. THE NONLOCAL RESPONSE FUNCTIONS TO QUANTUM FLUCTUATIONS AND THE CASIMIR FREE ENERGY

We consider two parallel thick metallic plates at temperature  $T$  in thermal equilibrium with the environment. The separation distance between plates is notated  $a$ . For good metals (Au, for instance) the plates of more than 100 nm thickness can be considered as semispaces when calculating the Casimir free energy and pressure [20]. According to the Lifshitz theory, the free energy of the Casimir interaction per unit area of the plates can be presented in the form [2,3] (see also Refs. [20,21] for the current notations),

$$\mathcal{F}(a, T) = \sum_{\alpha} \mathcal{F}_{\alpha}(a, T), \quad (1)$$

where the sum is over two independent polarizations of the electromagnetic field, transverse magnetic ( $\alpha = \text{TM}$ ) and transverse electric ( $\alpha = \text{TE}$ ), and

$$\mathcal{F}_{\alpha}(a, T) = \frac{k_B T}{2\pi} \sum_{l=0}^{\infty}{}' \int_0^{\infty} k_{\perp} dk_{\perp} \times \ln [1 - r_{\alpha}^2(i\xi_l, k_{\perp}, T) e^{-2aq_l}]. \quad (2)$$

Here,  $k_B$  is the Boltzmann constant, the prime on the summation sign in  $l$  divides the term with  $l = 0$  by 2,  $k_{\perp}$  is the magnitude of the projection of wave vector  $\mathbf{k}$  on the plane of plates (this plane is perpendicular to the Casimir force), the Matsubara frequencies are  $\xi_l = 2\pi k_B T l / \hbar$  with  $l = 0, 1, 2, \dots$ , and  $q_l = q_l(k_{\perp}) = (k_{\perp}^2 + \xi_l^2/c^2)^{1/2}$ .

The quantities  $r_{\alpha}$  in Eq. (2) have a meaning of the reflection coefficients calculated at the pure imaginary Matsubara frequencies. In the spatially local case they coincide with the familiar Fresnel coefficients,

$$\begin{aligned} r_{\text{TM}}(i\xi_l, k_{\perp}, T) &= \frac{\varepsilon_l q_l - k_l}{\varepsilon_l q_l + k_l}, \\ r_{\text{TE}}(i\xi_l, k_{\perp}, T) &= \frac{q_l - k_l}{q_l + k_l}, \end{aligned} \quad (3)$$

where  $k_l = k_l(k_{\perp}, T) = (k_{\perp}^2 + \varepsilon_l \xi_l^2/c^2)^{1/2}$  and  $\varepsilon_l = \varepsilon(i\xi_l, T)$  is the dielectric permittivity which describes the local

response of a metal to quantum fluctuations. Our notations underline that the dielectric permittivity of metals explicitly depends on  $T$  through the relaxation parameter (see below). The reflection coefficients (3) also possess an implicit dependence on  $T$  through the Matsubara frequencies.

In our case the dielectric response of a metal is spatially nonlocal and, thus, is described by two independent permittivities, the transverse one,  $\varepsilon^{\text{Tr}}(\omega, \mathbf{k}, T)$ , and the longitudinal one,  $\varepsilon^{\text{L}}(\omega, \mathbf{k}, T)$ , where the wave vector  $\mathbf{k} = (k_{\perp}, k^3)$  [55,56]. We recall that  $\varepsilon^{\text{Tr}}$  and  $\varepsilon^{\text{L}}$  describe the response of a metal to the transverse electric field which is perpendicular to  $\mathbf{k}$  and to the longitudinal one which is parallel to  $\mathbf{k}$ , respectively. As was argued in Ref. [49], in the plane-parallel Casimir geometries the nonlocal permittivities should depend only on  $k_{\perp}$ . In this case it was proven [49] that the reflection coefficients in Eq. (2) are given by

$$\begin{aligned} r_{\text{TM}}(i\xi_l, k_{\perp}, T) &= \frac{\varepsilon_l^{\text{Tr}} q_l - k_l^{\text{Tr}} - k_{\perp}(\varepsilon_l^{\text{Tr}} - \varepsilon_l^{\text{L}})(\varepsilon_l^{\text{L}})^{-1}}{\varepsilon_l^{\text{Tr}} q_l + k_l^{\text{Tr}} + k_{\perp}(\varepsilon_l^{\text{Tr}} - \varepsilon_l^{\text{L}})(\varepsilon_l^{\text{L}})^{-1}}, \\ r_{\text{TE}}(i\xi_l, k_{\perp}, T) &= \frac{q_l - k_l^{\text{Tr}}}{q_l + k_l^{\text{Tr}}}, \end{aligned} \quad (4)$$

where  $\varepsilon_l^{\text{Tr}} = \varepsilon^{\text{Tr}}(i\xi_l, k_{\perp}, T)$ ,  $\varepsilon_l^{\text{L}} = \varepsilon^{\text{L}}(i\xi_l, k_{\perp}, T)$  and

$$k_l^{\text{Tr}} = k_l^{\text{Tr}}(k_{\perp}, T) = \left[ k_{\perp}^2 + \varepsilon_l^{\text{Tr}} \frac{\xi_l^2}{c^2} \right]^{1/2}. \quad (5)$$

The alternative Drude-like nonlocal response functions suggested in Ref. [49] take the form,

$$\begin{aligned} \tilde{\varepsilon}_D^{\text{Tr}}(\omega, k_{\perp}, T) &= 1 - \frac{\omega_p^2}{\omega[\omega + i\gamma(T)]} \left( 1 + i \frac{v^{\text{Tr}} k_{\perp}}{\omega} \right), \\ \tilde{\varepsilon}_D^{\text{L}}(\omega, k_{\perp}, T) &= 1 - \frac{\omega_p^2}{\omega[\omega + i\gamma(T)]} \left( 1 + i \frac{v^{\text{L}} k_{\perp}}{\omega} \right)^{-1}, \end{aligned} \quad (6)$$

where  $\omega_p$  is the plasma frequency,  $\gamma(T)$  is the relaxation parameter, and  $v^{\text{Tr}}, v^{\text{L}}$  are the constants of the order of Fermi velocity  $v_F \sim 0.01c$ . In the local limit  $k_{\perp} = 0$  and the permittivities (6) reduce to the standard permittivity of the Drude model,

$$\begin{aligned} \tilde{\varepsilon}_D^{\text{Tr}}(\omega, 0, T) &= \tilde{\varepsilon}_D^{\text{L}}(\omega, 0, T) = \varepsilon_D(\omega, T) \\ &= 1 - \frac{\omega_p^2}{\omega[\omega + i\gamma(T)]}. \end{aligned} \quad (7)$$

For the electromagnetic fields on the mass shell it holds  $k_{\perp} \leq \omega/c$ . As a result one obtains

$$\frac{v^{\text{Tr,L}} k_{\perp}}{\omega} \sim \frac{v_F c k_{\perp}}{c \omega} \leq \frac{v_F}{c} \ll 1. \quad (8)$$

Because of this, for the on-shell fields the dielectric permittivities (6) lead to nearly the same results as the

commonly used local Drude permittivity (7). As to the off-shell fluctuations, the quantity  $v^{\text{Tr,L}} k_{\perp} / \omega$  can be of the order of and even larger than unity depending on the value of  $k_{\perp}$ . Thus, by using the permittivities (6) at the pure imaginary Matsubara frequencies,

$$\begin{aligned} \tilde{\varepsilon}_{D,l}^{\text{Tr}} &= 1 + \frac{\omega_p^2}{\xi_l[\xi_l + \gamma(T)]} \left( 1 + \frac{v^{\text{Tr}} k_{\perp}}{\xi_l} \right), \\ \tilde{\varepsilon}_{D,l}^{\text{L}} &= 1 + \frac{\omega_p^2}{\xi_l[\xi_l + \gamma(T)]} \left( 1 + \frac{v^{\text{L}} k_{\perp}}{\xi_l} \right)^{-1}, \end{aligned} \quad (9)$$

one can restore an agreement between theoretical predictions of the Lifshitz theory with taken into account dissipation properties of conduction electrons and the measurement data [49].

In the next section, we consider the asymptotic behavior of the Casimir free energy (1), (2) with the reflection coefficients (4) and dielectric permittivities (9) at arbitrarily low temperature. For this purpose it is convenient to present both contributions to the free energy (1) as the sum of the zero-temperature terms and the thermal corrections to them,

$$\mathcal{F}_{\alpha}(a, T) = E_{\alpha}(a) + \Delta_T \mathcal{F}_{\alpha}(a, T). \quad (10)$$

The quantity  $E_{\alpha}(a)$  is obtained from  $\mathcal{F}_{\alpha}$  defined in Eq. (2) by putting  $T = 0$  and replacing the discrete Matsubara frequencies  $\xi_l$  with a continuous variable  $\xi$ . In so doing the sum in  $l$  is replaced with an integral,

$$k_B T \sum_{l=0}^{\infty} \rightarrow \frac{\hbar}{2\pi} \int_0^{\infty} d\xi, \quad (11)$$

and one obtains

$$\begin{aligned} E_{\alpha}(a) &= \frac{\hbar}{4\pi^2} \int_0^{\infty} d\xi \int_0^{\infty} k_{\perp} dk_{\perp} \\ &\times \ln [1 - r_{\alpha}^2(i\xi, k_{\perp}, 0) e^{-2a q}]. \end{aligned} \quad (12)$$

Here, the reflection coefficients are given by

$$\begin{aligned} r_{\text{TM}}(i\xi, k_{\perp}, 0) &= \frac{\varepsilon^{\text{Tr}(0)} q - k^{\text{Tr}(0)} - k_{\perp}(\varepsilon^{\text{Tr}(0)} - \varepsilon^{\text{L}(0)})(\varepsilon^{\text{L}(0)})^{-1}}{\varepsilon^{\text{Tr}(0)} q + k^{\text{Tr}(0)} + k_{\perp}(\varepsilon^{\text{Tr}(0)} - \varepsilon^{\text{L}(0)})(\varepsilon^{\text{L}(0)})^{-1}}, \\ r_{\text{TE}}(i\xi, k_{\perp}, 0) &= \frac{q - k^{\text{Tr}(0)}}{q + k^{\text{Tr}(0)}}, \end{aligned} \quad (13)$$

where  $\varepsilon^{\text{Tr}(0)} = \varepsilon^{\text{Tr}}(i\xi, k_{\perp}, 0)$ ,  $\varepsilon^{\text{L}(0)} = \varepsilon^{\text{L}}(i\xi, k_{\perp}, 0)$  and

$$q = q(k_{\perp}) = \left[ k_{\perp}^2 + \varepsilon^{\text{Tr}(0)} \frac{\xi^2}{c^2} \right]^{1/2},$$

$$k^{\text{Tr}(0)} = k^{\text{Tr}}(k_{\perp}, 0) = \left[ k_{\perp}^2 + \varepsilon^{\text{Tr}(0)} \frac{\xi^2}{c^2} \right]^{1/2}. \quad (14)$$

Equation (10) can be considered as a definition of the thermal correction which presents a simple and straightforward way for its calculation in the limiting case of low temperature.

### III. LOW-TEMPERATURE BEHAVIOR OF THE CASIMIR FREE ENERGY AND ENTROPY USING THE NONLOCAL RESPONSE FUNCTIONS FOR PERFECT CRYSTAL LATTICES

Now we consider asymptotic behavior of the thermal correction  $\Delta_T \mathcal{F}_\alpha$  defined in Eq. (10) at low temperature where the Casimir free energy  $\mathcal{F}_\alpha$  and energy  $E_\alpha$  are defined in Eqs. (2) and (12), respectively. It is convenient to perform the asymptotic expansion using the dimensionless variables,

$$\zeta_l = \frac{2a\xi_l}{c}, \quad y = 2aq_l. \quad (15)$$

In terms of these variables the thermal correction takes the form,

$$\Delta_T \mathcal{F}_\alpha(a, T) = \frac{k_B T}{8\pi a^2} \sum_{l=0}^{\infty} \int_{\zeta_l}^{\infty} y dy \ln [1 - r_\alpha^2(i\zeta_l, y, T) e^{-y}]$$

$$- \frac{\hbar c}{32\pi^2 a^3} \int_0^{\infty} d\zeta$$

$$\times \int_{\zeta}^{\infty} y dy \ln [1 - r_\alpha^2(i\zeta, y, 0) e^{-y}], \quad (16)$$

where  $\zeta = 2a\xi/c$  similar to (15).

We introduce in the second line of Eq. (16) the integration variable  $t = \zeta/\tau$ , where  $\tau = 4\pi k_B T a / (\hbar c)$ , and add and subtract on the right-hand side of Eq. (16) the following quantity:

$$\frac{k_B T}{8\pi a^2} \sum_{l=0}^{\infty} \int_{\zeta_l}^{\infty} y dy \ln [1 - r_\alpha^2(i\zeta_l, y, 0) e^{-y}]. \quad (17)$$

Then, taking into account, that  $\zeta_l = \tau l$ , one can identically rewrite Eq. (16) as

$$\Delta_T \mathcal{F}_\alpha(a, T) = \Delta_T^{\text{expl}} \mathcal{F}_\alpha(a, T) + \Delta_T^{\text{impl}} \mathcal{F}_\alpha(a, T), \quad (18)$$

where

$$\Delta_T^{\text{expl}} \mathcal{F}_\alpha(a, T) = \frac{k_B T}{8\pi a^2} \sum_{l=0}^{\infty} \int_{\tau l}^{\infty} y dy \ln \frac{1 - r_\alpha^2(i\tau l, y, T) e^{-y}}{1 - r_\alpha^2(i\tau l, y, 0) e^{-y}} \quad (19)$$

and

$$\Delta_T^{\text{impl}} \mathcal{F}_\alpha(a, T) = \frac{k_B T}{8\pi a^2} \left[ \sum_{l=0}^{\infty} \Phi_\alpha(\tau l) - \int_0^{\infty} dt \Phi_\alpha(\tau t) \right], \quad (20)$$

where

$$\Phi_\alpha(x) \equiv \int_x^{\infty} y dy \ln [1 - r_\alpha^2(ix, y, 0) e^{-y}]. \quad (21)$$

The quantity  $\Delta_T^{\text{expl}} \mathcal{F}$  is called the explicit thermal correction. It vanishes for the response functions and hence for the reflection coefficients which do not possess an explicit dependence on temperature as a parameter. As to the quantity  $\Delta_T^{\text{impl}} \mathcal{F}$ , which is called the implicit thermal correction, it depends on temperature only through the Matsubara frequencies.

We start with the implicit thermal correction which, as shown below, provides the dominant contribution to the low-temperature dependence of the Casimir free energy. Using the Abel-Plana formula for the difference between the sum and the integral [57], one can identically rearrange Eqs. (1) and (20) to

$$\Delta_T^{\text{impl}} \mathcal{F}(a, T) = \frac{ik_B T}{8\pi a^2} \int_0^{\infty} \frac{dt}{e^{2\pi t} - 1}$$

$$\times \sum_{\alpha} [\Phi_\alpha(i\tau t) - \Phi_\alpha(-i\tau t)]. \quad (22)$$

In subsequent derivations one should take into account the dependence of the relaxation parameter  $\gamma$  on  $T$ . In this section, we consider metals with perfect crystal lattices. In this case, the relation  $\gamma(T) = bT^2$ , where  $b$  is some constant coefficient, is followed starting from the liquid helium temperature down to zero temperature owing to the electron-electron scattering [58]. Rewriting Eq. (9) at  $T = 0$  (where  $\xi_l$  is replaced with  $\xi$ ) in terms of the dimensionless variables introduced above, one obtains

$$\tilde{\varepsilon}_D^{\text{Tr}(0)} = \tilde{\varepsilon}_D^{\text{Tr}}(ix, y, 0)$$

$$= 1 + \frac{\tilde{\omega}_p^2}{x^2} \left( 1 + \tilde{v}^{\text{Tr}} \frac{\sqrt{y^2 - x^2}}{x} \right),$$

$$\tilde{\varepsilon}_D^{\text{L}(0)} = \tilde{\varepsilon}_D^{\text{L}}(ix, y, 0)$$

$$= 1 + \frac{\tilde{\omega}_p^2}{x^2} \left( 1 + \tilde{v}^{\text{L}} \frac{\sqrt{y^2 - x^2}}{x} \right)^{-1}, \quad (23)$$

where  $\tilde{\omega}_p = 2a\omega_p/c$ ,  $\tilde{v}^{\text{Tr}} = v^{\text{Tr}}/c$ , and  $\tilde{v}^{\text{L}} = v^{\text{L}}/c$  are the dimensionless plasma frequency and respective velocities.

As a result, the reflection coefficients  $r_\alpha(ix, y, 0)$ , entering Eq. (21), take the form,

$$r_{\text{TM}}(ix, y, 0) = \frac{\tilde{\epsilon}_D^{\text{Tr}(0)} y - \sqrt{y^2 + (\tilde{\epsilon}_D^{\text{Tr}(0)} - 1)x^2} - [\tilde{\epsilon}_D^{\text{Tr}(0)} - \tilde{\epsilon}_D^{\text{L}(0)}](\tilde{\epsilon}_D^{\text{L}(0)})^{-1} \sqrt{y^2 - x^2}}{\tilde{\epsilon}_D^{\text{Tr}(0)} y + \sqrt{y^2 + (\tilde{\epsilon}_D^{\text{Tr}(0)} - 1)x^2} + [\tilde{\epsilon}_D^{\text{Tr}(0)} - \tilde{\epsilon}_D^{\text{L}(0)}](\tilde{\epsilon}_D^{\text{L}(0)})^{-1} \sqrt{y^2 - x^2}},$$

$$r_{\text{TE}}(ix, y, 0) = \frac{y - \sqrt{y^2 + (\tilde{\epsilon}_D^{\text{Tr}(0)} - 1)x^2}}{y + \sqrt{y^2 + (\tilde{\epsilon}_D^{\text{Tr}(0)} - 1)x^2}}. \quad (24)$$

From Eqs. (23) and (24) it is easily seen that in the limiting case of zero temperature, i.e., for  $x = \tau t \rightarrow 0$ , it holds

$$\lim_{x \rightarrow 0} r_{\text{TM}}(ix, y, 0) = 1, \quad \lim_{x \rightarrow 0} r_{\text{TE}}(ix, y, 0) = -1. \quad (25)$$

Now we substitute Eqs. (23) and (24) in Eq. (21) and find the first expansion terms in the powers of small  $x$ ,

$$\Phi_{\text{TM}}(x) = \int_0^\infty dy y \ln(1 - e^{-y}) + \frac{4\tilde{v}^{\text{L}}}{\tilde{\omega}_p^2} x \int_0^\infty dy \frac{y^2}{e^y - 1} + O(x^2 \ln x),$$

$$\Phi_{\text{TE}}(x) = \int_0^\infty dy y \ln(1 - e^{-y}) + \frac{4}{\tilde{\omega}_p \sqrt{\tilde{v}^{\text{Tr}}}} \sqrt{x} \int_0^\infty dy \frac{y^{3/2}}{e^y - 1} + O(x). \quad (26)$$

From Eq. (26) it is seen that the leading contribution to the thermal correction at a vanishing temperature is given by the TE mode,

$$\begin{aligned} \Phi(i\tau t) - \Phi(-i\tau t) &= \Phi_{\text{TE}}(i\tau t) - \Phi_{\text{TE}}(-i\tau t) \\ &= \frac{3i}{\tilde{\omega}_p \sqrt{\tilde{v}^{\text{Tr}}}} \sqrt{2\pi\tau t} \zeta\left(\frac{5}{2}\right) + O(\tau) \\ &= \frac{3i\pi c}{\omega_p} \sqrt{\frac{2k_B T t}{av^{\text{Tr}}\hbar}} \zeta\left(\frac{5}{2}\right) + O(T), \end{aligned} \quad (27)$$

where  $\zeta(z)$  is the Riemann zeta function.

Substituting Eq. (27) in Eq. (22) and integrating with respect to  $t$ , one arrives at

$$\Delta_T^{\text{impl}} \mathcal{F}(a, T) = -\frac{3c\zeta(3/2)\zeta(5/2)}{32\pi\omega_p a^{5/2} \sqrt{\tilde{v}^{\text{Tr}}\hbar}} (k_B T)^{3/2} + O(T^2). \quad (28)$$

Now we consider the explicit thermal correction defined in Eq. (19). In terms of dimensionless variables, the dielectric permittivities entering the reflection coefficients  $r_\alpha(i\tau l, y, T)$  are obtained from Eq. (9),

$$\begin{aligned} \tilde{\epsilon}_{D,l}^{\text{Tr}} &= \tilde{\epsilon}_D^{\text{Tr}}(i\tau l, y, T) \\ &= 1 + \frac{\tilde{\omega}_p^2}{\tau l(\tau l + \tilde{b}T^2)} \left( 1 + \tilde{v}^{\text{Tr}} \frac{\sqrt{y^2 - \tau^2 l^2}}{\tau l} \right), \\ \tilde{\epsilon}_{D,l}^{\text{L}} &= \tilde{\epsilon}_D^{\text{L}}(i\tau l, y, T) \\ &= 1 + \frac{\tilde{\omega}_p^2}{\tau l(\tau l + \tilde{b}T^2)} \left( 1 + \tilde{v}^{\text{L}} \frac{\sqrt{y^2 - \tau^2 l^2}}{\tau l} \right)^{-1}. \end{aligned} \quad (29)$$

Here, the dimensionless relaxation parameter  $\tilde{\gamma}(T) = 2a\gamma(T)/c = \tilde{b}T^2$ . As to the permittivities entering the reflection coefficients  $r_\alpha(i\tau l, y, 0)$ , they are obtained from Eq. (29) by putting  $T = 0$ .

To calculate the quantity (19) at low temperature, we present the reflection coefficients  $r_\alpha(i\tau l, y, T)$  as the zero-temperature contributions and the thermal corrections to them

$$r_\alpha(i\tau l, y, T) = r_\alpha(i\tau l, y, 0) + \Delta_T r_\alpha(i\tau l, y, T). \quad (30)$$

It is evident that the thermal corrections  $\Delta_T r_\alpha$  go to zero with vanishing temperature.

We substitute Eq. (30) in Eq. (19) and expand Eq. (19) up to the first order in the small parameter,

$$\frac{\Delta_T r_\alpha(i\tau l, y, T)}{r_\alpha(i\tau l, y, 0)} \ll 1, \quad (31)$$

like this was done in the literature for the case of graphene [59]. The result is

$$\begin{aligned} \Delta_T^{\text{expl}} \mathcal{F}_\alpha(a, T) &= -\frac{k_B T}{4\pi a^2} \sum_{l=0}^{\infty} \int_{\tau l}^{\infty} y dy r_\alpha(i\tau l, y, 0) \\ &\quad \times \frac{\Delta_T r_\alpha(i\tau l, y, T)}{e^y - r_\alpha^2(i\tau l, y, 0)}. \end{aligned} \quad (32)$$

It is convenient to consider separately the term of Eq. (32) with  $l = 0$ ,  $\Delta_{T,l=0}^{\text{expl}} \mathcal{F}_\alpha$ , and the sum of all terms with  $l \geq 1$ ,  $\Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_\alpha$ . The explicit form of the reflection coefficients  $r_\alpha(i\tau l, y, T)$  in terms of the dimensionless variables is given by Eq. (24) where  $x$  should be replaced with  $\tau l$  and  $\tilde{\epsilon}_D^{\text{Tr}(0)}$ ,  $\tilde{\epsilon}_D^{\text{L}(0)}$  with  $\tilde{\epsilon}_{D,l}^{\text{Tr}}$ ,  $\tilde{\epsilon}_{D,l}^{\text{L}}$  defined in Eq. (29). As a result, for  $\alpha = \text{TM}$ ,  $l = 0$  one obtains

$$r_{\text{TM}}(0, y, T) = 1 - \frac{2\tilde{b}T^2\tilde{v}^L y}{\tilde{\omega}_p^2 + 2\tilde{b}T^2\tilde{v}^L y}. \quad (33)$$

Expanding this quantity up to the first order in a small parameter,

$$\beta(T) = \frac{\tilde{b}T^2\tilde{v}^L}{\tilde{\omega}_p^2} \ll 1, \quad (34)$$

one arrives at

$$r_{\text{TM}}(0, y, T) = 1 - 2\beta(T)y. \quad (35)$$

From Eqs. (33) and (35), taking into account Eq. (30), we find

$$r_{\text{TM}}(0, y, 0) = 1, \quad \Delta_T r_{\text{TM}}(0, y, T) = -2\beta(T)y. \quad (36)$$

Finally, substituting Eq. (36) in Eq. (32), we obtain

$$\Delta_{T,l=0}^{\text{expl}} \mathcal{F}_{\text{TM}}(a, T) = \frac{k_B T}{4\pi a^2} \beta(T) \int_0^\infty dy \frac{y^2}{e^y - 1}. \quad (37)$$

By integrating and returning to dimensional quantities in Eq. (34), the result (37) leads to

$$\Delta_{T,l=0}^{\text{expl}} \mathcal{F}_{\text{TM}}(a, T) = \frac{k_B b v^L \zeta(3)}{4\pi a^3 \omega_p^2} T^3. \quad (38)$$

One can see that it is a correction of the higher order in  $T$  than in Eq. (28).

Now we turn our attention to the case  $l = 0$ ,  $\alpha = \text{TE}$ . Using the dielectric permittivity (29), the reflection coefficient  $r_{\text{TE}}(0, y, T)$  takes the form,

$$r_{\text{TE}}(0, y, T) = -\frac{\sqrt{1 + \delta(T)y} - \sqrt{\delta(T)y}}{\sqrt{1 + \delta(T)y} + \sqrt{\delta(T)y}}, \quad (39)$$

where

$$\delta(T) = \frac{\tilde{b}T^2}{\tilde{v}^{\text{Tr}} \tilde{\omega}_p^2} \ll 1. \quad (40)$$

Expanding Eq. (39) up to the lowest order in  $\delta(T)$ , we have

$$r_{\text{TE}}(0, y, T) = -1 + 2\sqrt{\delta(T)y}. \quad (41)$$

Then from Eqs. (39) and (41) one obtains

$$r_{\text{TE}}(0, y, 0) = -1, \quad \Delta_T r_{\text{TE}}(0, y, T) = 2\sqrt{\delta(T)y}. \quad (42)$$

Substituting Eq. (42) in Eq. (32) for  $\alpha = \text{TE}$ , we arrive at

$$\begin{aligned} \Delta_{T,l=0}^{\text{expl}} \mathcal{F}_{\text{TE}}(a, T) &= \frac{k_B T}{4\pi a^2} \sqrt{\delta(T)} \int_0^\infty dy \frac{y^{3/2}}{e^y - 1} \\ &= \frac{3k_B \sqrt{bc} \zeta(5/2)}{16\sqrt{2}\pi a^{5/2} \omega_p \sqrt{v^{\text{Tr}}}} T^2. \end{aligned} \quad (43)$$

One can see that although  $\Delta_{T,l=0}^{\text{expl}} \mathcal{F}_{\text{TE}}$  is of the lower order than  $\Delta_{T,l=0}^{\text{expl}} \mathcal{F}_{\text{TM}}$  defined in Eq. (38), it is of the higher order in  $T$  than  $\Delta_T^{\text{impl}} \mathcal{F}$  defined in Eq. (28).

The sum of all terms with  $l \geq 1$  in Eq. (32) results in the same temperature dependence at low  $T$  as in Eq. (43), i.e.,

$$\Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_{\text{TM}}(a, T) \sim \Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_{\text{TE}}(a, T) \sim T^2, \quad (44)$$

(see the Appendix). Thus, by comparing Eqs. (28), (38), (43), and (44), we conclude that the leading low-temperature behavior of the thermal correction to the Casimir energy between metallic plates with perfect crystal lattices found within the Lifshitz theory using the alternative Drude-like response functions is determined by the implicit term in Eq. (18) and takes the form,

$$\Delta_T \mathcal{F}(a, T) = -\frac{3c\zeta(3/2)\zeta(5/2)}{32\pi\omega_p a^{5/2} \sqrt{v^{\text{Tr}} \hbar}} (k_B T)^{3/2}. \quad (45)$$

From Eq. (45) it is easy to evaluate the dominant contribution to the Casimir entropy at arbitrarily low temperature,

$$\begin{aligned} S(a, T) &= -\frac{\partial \Delta_T \mathcal{F}(a, T)}{\partial T} \\ &= \frac{9k_B c \zeta(3/2) \zeta(5/2)}{64\pi\omega_p a^{5/2} \sqrt{v^{\text{Tr}} \hbar}} \sqrt{k_B T}. \end{aligned} \quad (46)$$

It is seen that the Casimir entropy is positive and goes to zero with vanishing temperature as it should be in accordance to the third law of thermodynamics, the Nernst heat theorem [60]. This makes the nonlocal Drude-like response function (6) preferable as compared to the conventional Drude response (7). For metals with perfect crystal lattices the latter leads to the negative Casimir entropy at zero temperature [27],

$$S_D(a, T) = -\frac{k_B \zeta(3)}{16\pi a^2} [1 - 4\kappa + 12\kappa^2 - \dots] < 0, \quad (47)$$

where  $\kappa \equiv c/(\omega_p a)$ . This entropy depends on the parameters of a system, such as the separation distance between the plates and the plasma frequency of a metal, and, thus, violates the Nernst heat theorem [60,61].

#### IV. LOW-TEMPERATURE BEHAVIOR OF THE CASIMIR FREE ENERGY AND ENTROPY IN THE PRESENCE OF DEFECTS OF A CRYSTAL LATTICE

Crystal lattices of real metallic samples unavoidably contain some small fractions of defects (e.g., atoms which are different from the native atoms of the lattice, vacancies etc.). In this case, with decreasing temperature the relaxation parameter  $\gamma(T)$  reaches at  $T = T_0$  some minimum value  $\gamma_0 > 0$  and remains unchanged at  $T < T_0$  [58]. Thus, for typical Au samples  $\gamma_0 \approx 5.3 \times 10^{10}$  rad/s. As discussed in Sec. I, this fact was used in the literature [32–34] in an attempt to reconcile the Lifshitz theory using the Drude model with the Nernst heat theorem. It was shown, however, that with decreasing temperature the Casimir entropy takes a negative constant value over the wide temperature interval and abruptly jumps to zero only at  $T < 10^{-3}$  K which is somewhat nonphysical [20]. Because of this, it is interesting to find the low-temperature behavior of the Casimir entropy calculated using the nonlocal Drude-like response functions for metals with defects of a crystal structure.

Below we consider the temperature interval  $0 \leq T < T_0$  where  $\gamma(T) = \gamma_0 = \text{const}$ . In this interval, the nonlocal Drude-like permittivities are given by Eq. (29) where one should replace the term  $\tilde{b}T^2$  with  $\tilde{\gamma}_0 = 2a\gamma_0/c$ . These permittivities do not possess an explicit dependence on  $T$ :  $\tilde{\epsilon}_{D,l}^{\text{Tr}} = \tilde{\epsilon}_D^{\text{Tr}}(i\tau l, y)$  and  $\tilde{\epsilon}_{D,l}^{\text{L}} = \tilde{\epsilon}_D^{\text{L}}(i\tau l, y)$ . As a result, the thermal correction (10) takes the same form as an implicit thermal correction (20),

$$\Delta_T \mathcal{F}_\alpha(a, T) = \frac{k_B T}{8\pi a^2} \left[ \sum_{l=0}^{\infty} \Phi_\alpha(\tau l) - \int_0^\infty dt \Phi_\alpha(\tau t) \right], \quad (48)$$

where now

$$\Phi_\alpha(x) \equiv \int_x^\infty y dy \ln [1 - r_\alpha^2(ix, y)e^{-y}]. \quad (49)$$

The reflection coefficients  $r_\alpha(ix, y)$  are given by the right-hand sides of Eq. (24) where  $\tilde{\epsilon}_D^{\text{Tr}(0)}$ ,  $\tilde{\epsilon}_D^{\text{L}(0)}$  should be replaced with  $\tilde{\epsilon}_{D,l}^{\text{Tr}}$ ,  $\tilde{\epsilon}_{D,l}^{\text{L}}$  specified above.

Using the Abel-Plana formula, Eq. (48) can be rewritten similar to Eq. (22),

$$\Delta_T \mathcal{F}_\alpha(a, T) = \frac{ik_B T}{8\pi a^2} \int_0^\infty dt \frac{\Phi_\alpha(i\tau t) - \Phi_\alpha(-i\tau t)}{e^{2\pi t} - 1}. \quad (50)$$

In order to obtain the behavior of  $\Delta_T \mathcal{F}_\alpha$  at low  $T$ , it is convenient to expand  $\Phi_\alpha$  up to the first power in  $x$ ,

$$\Phi_\alpha(x) = \Phi_\alpha(0) + x\Phi'_\alpha(0), \quad (51)$$

where

$$\Phi'_\alpha(0) = -2 \int_0^\infty y dy \frac{r_\alpha(0, y)r'_\alpha(0, y)e^{-y}}{1 - r_\alpha^2(0, y)e^{-y}}. \quad (52)$$

We start with the transverse magnetic polarization  $\alpha = \text{TM}$ . In this case, we use the parameter (34) which becomes temperature-independent because  $\tilde{\gamma}(T) = \tilde{b}T^2$  is now equal to  $\tilde{\gamma}_0$ ,

$$\beta_0 = \frac{\tilde{\gamma}_0 \tilde{v}^{\text{L}}}{\tilde{\omega}_p^2} \ll 1. \quad (53)$$

An extreme smallness of this parameter is evident because  $\tilde{\gamma}_0/\tilde{\omega}_p \sim 10^{-6}$ ,  $\tilde{v}^{\text{L}} \sim 10^{-2}$ , whereas  $\tilde{\omega}_p \sim 1$  at  $a = 10$  nm and increases with increasing separation between the plates.

Expanding the reflection coefficient  $r_{\text{TM}}(0, y)$  up to the first power of small parameter (53), we find

$$r_{\text{TM}}(0, y) = 1 - 2\beta_0 y. \quad (54)$$

In a similar way,

$$r'_{\text{TM}}(0, y) = \left. \frac{\partial r_{\text{TM}}(x, y)}{\partial x} \right|_{x=0} = -2\beta_0 \left( \frac{1}{\tilde{v}^{\text{L}}} + \frac{y}{\tilde{\gamma}_0} \right). \quad (55)$$

Substituting Eqs. (54) and (55) in Eq. (52), in the lowest order of the small parameter  $\beta_0$  one obtains

$$\begin{aligned} \Phi'_{\text{TM}}(0) &= 4\beta_0 \int_0^\infty y dy \frac{(\tilde{v}^{\text{L}})^{-1} + \tilde{\gamma}_0^{-1} y}{e^y - 1} \\ &= 4\beta_0 \left[ \frac{\pi^2}{6\tilde{v}^{\text{L}}} + \frac{2\zeta(3)}{\tilde{\gamma}_0} \right]. \end{aligned} \quad (56)$$

Using Eqs. (51) and (56), we have

$$\Phi_{\text{TM}}(i\tau t) - \Phi_{\text{TM}}(-i\tau t) = 8i\beta_0 \tau t \left[ \frac{\pi^2}{6\tilde{v}^{\text{L}}} + \frac{2\zeta(3)}{\tilde{\gamma}_0} \right]. \quad (57)$$

Then we substitute Eq. (57) in Eq. (50) and arrive at

$$\begin{aligned} \Delta_T \mathcal{F}_{\text{TM}}(a, T) &= -\frac{k_B T}{\pi a^2} \beta_0 \tau \left[ \frac{\pi^2}{6\tilde{v}^{\text{L}}} + \frac{2\zeta(3)}{\tilde{\gamma}_0} \right] \\ &\quad \times \int_0^\infty \frac{t dt}{e^{2\pi t} - 1}. \end{aligned} \quad (58)$$

After integration and returning to the dimensional quantities, we finally find

$$\Delta_T \mathcal{F}_{\text{TM}}(a, T) = -\frac{(k_B T)^2}{12a^2 \hbar \omega_p^2} \left[ \gamma_0 \frac{\pi^2}{6} + \frac{v^{\text{L}}}{a} \zeta(3) \right]. \quad (59)$$

Now we continue with the transverse electric polarization,  $\alpha = \text{TE}$ . The parameter (40) used in this case in Sec. III also becomes temperature-independent,

$$\delta_0 = \frac{\tilde{\gamma}_0}{\tilde{v}^{\text{Tr}} \tilde{\omega}_p^2} \ll 1. \quad (60)$$

This parameter takes the maximum value  $\approx 4 \times 10^{-4}$  at  $a = 10$  nm and further decreases with increasing separation between the plates. The reflection coefficient  $r_{\text{TE}}(0, y)$  takes the same form as in Eq. (39),

$$r_{\text{TE}}(0, y) = -\frac{\sqrt{1 + \delta_0 y} - \sqrt{\delta_0 y}}{\sqrt{1 + \delta_0 y} + \sqrt{\delta_0 y}}. \quad (61)$$

Expanding in Eq. (61) up to the lowest order of the parameter (60), one obtains

$$r_{\text{TE}}(0, y) = -1 + 2\sqrt{\delta_0 y}. \quad (62)$$

Then, by expanding the derivative of  $r_{\text{TE}}(x, y)$  with respect to  $x$  at  $x = 0$ , we have

$$r'_{\text{TE}}(0, y) = \frac{\sqrt{\delta_0}}{\tilde{\gamma}_0} \left( -\frac{\tilde{\gamma}_0}{\tilde{v}^{\text{Tr}} \sqrt{y}} + \sqrt{y} \right). \quad (63)$$

Substituting Eqs. (62) and (63) in Eq. (52), in the lowest order of the parameter (60), one finds

$$\begin{aligned} \Phi'_{\text{TE}}(0) &= 2 \frac{\sqrt{\delta_0}}{\tilde{\gamma}_0} \left( -\frac{\tilde{\gamma}_0}{\tilde{v}^{\text{Tr}}} \int_0^\infty \frac{\sqrt{y} dy}{e^y - 1} + \int_0^\infty \frac{y \sqrt{y} dy}{e^y - 1} \right) \\ &= \frac{\sqrt{\pi \delta_0}}{\tilde{\gamma}_0} \left[ \frac{3}{2} \zeta\left(\frac{5}{2}\right) - \frac{\tilde{\gamma}_0}{\tilde{v}^{\text{Tr}}} \zeta\left(\frac{3}{2}\right) \right]. \end{aligned} \quad (64)$$

Using Eq. (64), we have from Eq. (51),

$$\begin{aligned} \Phi_{\text{TE}}(i\tau t) - \Phi_{\text{TE}}(-i\tau t) &= 2i\tau t \frac{\sqrt{\pi \delta_0}}{\tilde{\gamma}_0} \\ &\times \left[ \frac{3}{2} \zeta\left(\frac{5}{2}\right) - \frac{\tilde{\gamma}_0}{\tilde{v}^{\text{Tr}}} \zeta\left(\frac{3}{2}\right) \right], \end{aligned} \quad (65)$$

and substituting this in Eq. (50) arrive at

$$\begin{aligned} \Delta_T \mathcal{F}_{\text{TE}}(a, T) &= -\frac{k_B T}{4\sqrt{\pi} a^2} \frac{\tau \sqrt{\delta_0}}{\tilde{\gamma}_0} \\ &\times \left[ \frac{3}{2} \zeta\left(\frac{5}{2}\right) - \frac{\tilde{\gamma}_0}{\tilde{v}^{\text{Tr}}} \zeta\left(\frac{3}{2}\right) \right] \int_0^\infty \frac{tdt}{e^{2\pi t} - 1}. \end{aligned} \quad (66)$$

Integrating in Eq. (66) and returning to the dimensional quantities, we finally obtain

$$\begin{aligned} \Delta_T \mathcal{F}_{\text{TE}}(a, T) &= -\frac{(k_B T)^2 \sqrt{\pi} c}{48 a^{5/2} \hbar \omega_p \sqrt{2\gamma_0 v^{\text{Tr}}}} \\ &\times \left[ \frac{3}{2} \zeta\left(\frac{5}{2}\right) - \frac{2a\gamma_0}{v^{\text{Tr}}} \zeta\left(\frac{3}{2}\right) \right]. \end{aligned} \quad (67)$$

As can be seen from Eqs. (59) and (67), the dominant contribution to the thermal correction is given by the TE mode due to the much larger coefficient near the same power in  $T$ . This is ultimately determined by the fact that  $\beta_0 \sim 10^{-8}$  whereas  $\delta_0 \sim 10^{-4}$ . Because of this, the leading low-temperature behavior of the thermal correction to the Casimir energy between metallic plates with typical concentration of impurities found using the nonlocal Drude-like response functions is given by

$$\begin{aligned} \Delta_T \mathcal{F}(a, T) &= -\frac{\sqrt{\pi} c}{48 a^{5/2} \hbar \omega_p \sqrt{2\gamma_0 v^{\text{Tr}}}} (k_B T)^2 \\ &\times \left[ \frac{3}{2} \zeta\left(\frac{5}{2}\right) - \frac{2a\gamma_0}{v^{\text{Tr}}} \zeta\left(\frac{3}{2}\right) \right]. \end{aligned} \quad (68)$$

The respective Casimir entropy at low temperature takes the form,

$$\begin{aligned} S(a, T) &= \frac{\sqrt{\pi} k_B c}{24 a^{5/2} \hbar \omega_p \sqrt{2\gamma_0 v^{\text{Tr}}}} k_B T \\ &\times \left[ \frac{3}{2} \zeta\left(\frac{5}{2}\right) - \frac{2a\gamma_0}{v^{\text{Tr}}} \zeta\left(\frac{3}{2}\right) \right]. \end{aligned} \quad (69)$$

Thus, for metals with impurities the Casimir entropy calculated using the alternative Drude-like response functions monotonously decreases to zero with vanishing temperature in accordance with the Nernst heat theorem. Note also that the entropy (69) remains positive at all separations below approximately  $20 \mu\text{m}$ , i.e., in the region related to the Casimir effect. These properties of Eq. (69) are advantageous in comparison with the conventional Drude model. The latter leads to the negative Casimir entropy which abruptly jumps to zero for metals with defects of a crystal lattice only at very low temperature.

## V. CONCLUSIONS AND DISCUSSION

In the foregoing, we have elucidated thermodynamic properties of the Casimir interaction calculated in the framework of the Lifshitz theory using the spatially non-local Drude-like response functions to quantum fluctuations. Although the Lifshitz theory is a well-studied branch of thermal quantum field theory, it suffers from a serious flaw known as the Casimir puzzle. The problem lies in the fact that theory is in disagreement with the measurement data and thermodynamic constraints when using the conventional Drude response function to quantum fluctuations. Recently it was shown, however, that an agreement of the



Lifshitz theory with the measurement data can be restored when using alternative nonlocal response functions which ensure almost the same response, as the standard Drude model, to the on-shell fluctuations but respond differently to quantum fluctuations off the mass shell [49]. However, the thermodynamic problem in the Lifshitz theory remained unresolved.

According to the above results, the Lifshitz theory using the suggested nonlocal Drude-like response functions is in complete agreement with the requirements of thermodynamics. Using rigorous analytic methods, it is shown that for metals with perfect crystal lattices the Casimir entropy calculated using these functions is positive and monotonously goes to zero as the square root of temperature when the temperature vanishes. The same characteristic properties are preserved for crystal lattices possessing some fraction of defects with the only difference that the Casimir entropy vanishes linearly in temperature. This means that the Lifshitz theory combined with nonlocal Drude-like response functions satisfies the third law of thermodynamics, the Nernst heat theorem.

Taking into consideration that the suggested nonlocal response functions to quantum fluctuations not only make the theoretical predictions consistent with the measurement data but also secure an agreement between two fundamental theories, they constitute a serious alternative to the commonly accepted Drude model and deserve further investigation.

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### APPENDIX: CONTRIBUTION FROM NONZERO MATSUBARA FREQUENCIES

Here, we derive low-temperature asymptotic behavior of the contribution to explicit thermal correction (32) from all Matsubara terms with  $l \geq 1$ . For this purpose, in each term of Eq. (32) with  $l \geq 1$  we introduce the new integration variable  $z = y - \tau l$  and obtain

$$\Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_\alpha(a, T) = -\frac{k_B T}{4\pi a^2} \sum_{l=1}^{\infty} e^{-\tau l} X_{\alpha,l}(\tau), \quad (\text{A1})$$

where

$$X_{\alpha,l}(\tau) = \int_0^\infty dz (z + \tau l) \times \frac{r_\alpha(i\tau l, z + \tau l, 0) \Delta_T r_\alpha(i\tau l, z + \tau l, \tau)}{e^z - r_\alpha^2(i\tau l, z + \tau l, 0) e^{-\tau l}}. \quad (\text{A2})$$

Note that in Eq. (A2) we have replaced an explicit dependence of  $\Delta_T r_\alpha$  on  $T$  with a dependence on  $\tau$  using the fact that  $T = \hbar c \tau / (4\pi a k_B)$ . The thermal correction  $\Delta_T r_\alpha$  on  $T$  depends on  $T$  as a parameter owing to the temperature-dependent relaxation parameter. As discussed in Sec. III, at temperatures below the liquid helium temperature the dimensionless relaxation parameter is of the order of  $T^2$ . Below we present it in the form,

$$\tilde{\gamma}(T) = \frac{2a\gamma(T)}{c} = \tilde{b}T^2 = \tilde{\tilde{b}}\tau^2, \quad (\text{A3})$$

where

$$\tilde{b} = \frac{2ab}{c}, \quad \tilde{\tilde{b}} = \frac{c\hbar^2 b}{8\pi^2 k_B^2 a}. \quad (\text{A4})$$

The reflection coefficients  $r_\alpha(i\tau l, z + \tau l, 0)$  are given by Eq. (24) where  $x$  should be replaced with  $\tau l$  and  $y$  with  $z + \tau l$ . To obtain the coefficients  $r_\alpha(i\tau l, z + \tau l, \tau)$  from Eq. (24), one should also replace  $\tilde{\epsilon}_D^{\text{Tr}(0)}$ ,  $\tilde{\epsilon}_D^{\text{L}(0)}$  with  $\tilde{\epsilon}_{D,l}^{\text{Tr}}$ ,  $\tilde{\epsilon}_{D,l}^{\text{L}}$  defined in Eq. (29) taking into account Eq. (A3).

Now we begin with the transverse magnetic polarization,  $\alpha = \text{TM}$ , and expand both reflection coefficients  $r_{\text{TM}}(i\tau l, z + \tau l, 0)$  and  $r_{\text{TM}}(i\tau l, z + \tau l, \tau)$  in the powers of small  $\tau$ ,

$$\begin{aligned} r_{\text{TM}}(i\tau l, z + \tau l, 0) &= 1 - \frac{2\tilde{v}^{\text{L}} l z}{\tilde{\omega}_p^2} \\ &+ \frac{2l^2}{\tilde{\omega}_p^4} [-\tilde{\omega}_p^2 (1 + \tilde{v}^{\text{L}}) + 2(\tilde{v}^{\text{L}})^2 z^2] \tau^2 + O(\tau^5/2), \\ r_{\text{TM}}(i\tau l, z + \tau l, \tau) &= 1 - \frac{2\tilde{v}^{\text{L}} l z}{\tilde{\omega}_p^2} \tau \\ &- \frac{2l^2}{\tilde{\omega}_p^4} \left[ \tilde{\omega}_p^2 \left( 1 + \tilde{v}^{\text{L}} + \frac{\tilde{\tilde{b}} \tilde{v}^{\text{L}}}{l^2} z \right) - 2(\tilde{v}^{\text{L}})^2 z^2 \right] \tau^2 + O(\tau^5/2). \end{aligned} \quad (\text{A5})$$

Then from Eq. (30) one obtains

$$\Delta_T r_{\text{TM}}(i\tau l, z + \tau l, \tau) = -\frac{2\tilde{\tilde{b}} \tilde{v}^{\text{L}} z}{\tilde{\omega}_p^2} \tau^2. \quad (\text{A6})$$

Substituting the first line of Eqs. (A5) and (A6) in Eq. (A2), we find in the lowest order,

$$X_{\text{TM},l}(\tau) = -\frac{2\tilde{b}\tilde{v}^L}{\tilde{\omega}_p^2}\tau^2 \int_0^\infty \frac{z^2 dz}{e^z - 1} = -\frac{4\tilde{b}\tilde{v}^L}{\tilde{\omega}_p^2}\zeta(3)\tau^2. \quad (\text{A7})$$

Then from Eq. (A1) one arrives at

$$\Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_{\text{TM}}(a, T) = \frac{k_B T \tilde{b} \tilde{v}^L \tau^2}{\pi a^2 \tilde{\omega}_p^2} \zeta(3) \sum_{l=1}^{\infty} e^{-\tau l}. \quad (\text{A8})$$

Performing the summation in  $l$  and returning to the dimensional quantities with account of Eq. (A4), in the lowest order we finally find

$$\begin{aligned} \Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_{\text{TM}}(a, T) &= \frac{k_B T \tilde{b} \tilde{v}^L \tau^2}{\pi a^2 \tilde{\omega}_p^2} \frac{\zeta(3)}{e^\tau - 1} \\ &= \frac{\hbar c b v^L \zeta(3)}{8\pi^2 a^4 \omega_p^2} T^2. \end{aligned} \quad (\text{A9})$$

This result is in accordance with Eq. (44).

We continue with the transverse electric polarization,  $\alpha = \text{TE}$ , and expand the coefficients  $r_{\text{TE}}(i\tau l, z + \tau l, 0)$  and  $r_{\text{TE}}(i\tau l, z + \tau l, \tau)$  in the powers of  $\tau$ ,

$$\begin{aligned} r_{\text{TE}}(i\tau l, z + \tau l, 0) &= -1 + \frac{2\sqrt{lz}}{\tilde{\omega}_p \sqrt{\tilde{v}^{\text{Tr}}}} \sqrt{\tau} - \frac{2zl}{\tilde{v}^{\text{Tr}} \tilde{\omega}_p^2} \tau \\ &\quad + \frac{l^{3/2}(\tilde{v}^{\text{Tr}} \tilde{\omega}_p^2 - \tilde{\omega}_p^2 + z^2)}{(\tilde{v}^{\text{Tr}})^{3/2} \tilde{\omega}_p^3 \sqrt{z}} \tau^{3/2} + O(\tau^2), \\ r_{\text{TE}}(i\tau l, z + \tau l, \tau) &= -1 + \frac{2\sqrt{lz}}{\tilde{\omega}_p \sqrt{\tilde{v}^{\text{Tr}}}} \sqrt{\tau} - \frac{2zl}{\tilde{v}^{\text{Tr}} \tilde{\omega}_p^2} \tau \\ &\quad + \frac{l^{3/2}[\tilde{v}^{\text{Tr}} \tilde{\omega}_p^2 (1 + \tilde{b} l^{-2} z) - \tilde{\omega}_p^2 + z^2]}{(\tilde{v}^{\text{Tr}})^{3/2} \tilde{\omega}_p^3 \sqrt{z}} \\ &\quad \times \tau^{3/2} + O(\tau^2). \end{aligned} \quad (\text{A10})$$

Using Eq. (30) for the thermal correction, we obtain

$$\Delta_T r_{\text{TE}}(i\tau l, z + \tau l, \tau) = \frac{\tilde{b} \sqrt{z}}{\sqrt{\tilde{v}^{\text{Tr}}} \tilde{\omega}_p \sqrt{l}} \tau^{3/2}. \quad (\text{A11})$$

Substituting Eq. (A11) and the first line of Eq. (A10) in Eq. (A2), one finds

$$\begin{aligned} X_{\text{TE},l}(\tau) &= -\frac{\tilde{b}}{\sqrt{\tilde{v}^{\text{Tr}}} \tilde{\omega}_p \sqrt{l}} \tau^{3/2} \int_0^\infty \frac{z^{3/2}}{e^z - 1} dz \\ &= -\frac{3\sqrt{\pi} \tilde{b}}{4\sqrt{\tilde{v}^{\text{Tr}}} \tilde{\omega}_p \sqrt{l}} \zeta\left(\frac{5}{2}\right) \tau^{3/2}. \end{aligned} \quad (\text{A12})$$

Then, the substitution of Eq. (A12) in Eq. (A1) and summation in  $l$  lead to

$$\Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_{\text{TE}}(a, T) = \frac{3k_B T \tilde{b} \tau^{3/2}}{16\sqrt{\pi} a^2 \sqrt{\tilde{v}^{\text{Tr}}} \tilde{\omega}_p} \zeta\left(\frac{5}{2}\right) \text{Li}_{1/2}(e^{-\tau}), \quad (\text{A13})$$

where  $\text{Li}_\beta(z)$  is the polylogarithm function.

Taking into account that at small  $\tau$  it holds [62]

$$\text{Li}_{1/2}(e^{-\tau}) \approx \frac{\sqrt{\pi}}{\sqrt{\tau}}, \quad (\text{A14})$$

and returning to the dimensional quantities, we obtain

$$\Delta_{T,l \geq 1}^{\text{expl}} \mathcal{F}_{\text{TE}}(a, T) = \frac{3\hbar c^{3/2} b \zeta(5/2)}{64\pi a^3 \sqrt{\tilde{v}^{\text{Tr}}} \tilde{\omega}_p} T^2; \quad (\text{A15})$$

i.e., Eq. (44) is finally proven.

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- [1] H. B. G. Casimir, On the attraction between two perfectly conducting plates, *Proc. Kon. Ned. Akad. Wet. B* **51**, 793 (1948).
- [2] E. M. Lifshitz, The theory of molecular attractive forces between solids, *Zh. Eksp. Teor. Fiz.* **29**, 94 (1955) [*Sov. Phys. JETP* **2**, 73 (1956)].
- [3] E. M. Lifshitz and L. P. Pitaevskii, *Statistical Physics, Part II* (Pergamon, Oxford, 1980).
- [4] T. Emig, N. Graham, R. L. Jaffe, and M. Kardar, Casimir Forces between Arbitrary Compact Objects, *Phys. Rev. Lett.* **99**, 170403 (2007).
- [5] O. Kenneth and I. Klich, Casimir forces in a T-operator approach, *Phys. Rev. B* **78**, 014103 (2008).
- [6] S. J. Rahi, T. Emig, N. Graham, R. L. Jaffe, and M. Kardar, Scattering theory approach to electrodynamic Casimir forces, *Phys. Rev. D* **80**, 085021 (2009).
- [7] R. S. Decca, E. Fischbach, G. L. Klimchitskaya, D. E. Krause, D. López, and V. M. Mostepanenko, Improved tests of extra-dimensional physics and thermal quantum field theory from new Casimir force measurements, *Phys. Rev. D* **68**, 116003 (2003).
- [8] R. S. Decca, D. López, E. Fischbach, G. L. Klimchitskaya, D. E. Krause, and V. M. Mostepanenko, Precise comparison of theory and new experiment for the Casimir force leads to stronger constraints on thermal quantum effects and long-range interactions, *Ann. Phys. (NY)* **318**, 37 (2005).

- [9] R. S. Decca, D. López, E. Fischbach, G. L. Klimchitskaya, D. E. Krause, and V. M. Mostepanenko, Tests of new physics from precise measurements of the Casimir pressure between two gold-coated plates, *Phys. Rev. D* **75**, 077101 (2007).
- [10] R. S. Decca, D. López, E. Fischbach, G. L. Klimchitskaya, D. E. Krause, and V. M. Mostepanenko, Novel constraints on light elementary particles and extra-dimensional physics from the Casimir effect, *Eur. Phys. J. C* **51**, 963 (2007).
- [11] C.-C. Chang, A. A. Banishev, R. Castillo-Garza, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, Gradient of the Casimir force between Au surfaces of a sphere and a plate measured using an atomic force microscope in a frequency-shift technique, *Phys. Rev. B* **85**, 165443 (2012).
- [12] A. A. Banishev, C.-C. Chang, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, Measurement of the gradient of the Casimir force between a nonmagnetic gold sphere and a magnetic nickel plate, *Phys. Rev. B* **85**, 195422 (2012).
- [13] A. A. Banishev, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, Demonstration of the Casimir Force between Ferromagnetic Surfaces of a Ni-Coated Sphere and a Ni-Coated Plate, *Phys. Rev. Lett.* **110**, 137401 (2013).
- [14] A. A. Banishev, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, Casimir force between two magnetic metals in comparison with nonmagnetic test bodies, *Phys. Rev. B* **88**, 155410 (2013).
- [15] G. Bimonte, D. López, and R. S. Decca, Isoelectronic determination of the thermal Casimir force, *Phys. Rev. B* **93**, 184434 (2016).
- [16] J. Xu, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, Reducing detrimental electrostatic effects in Casimir-force measurements and Casimir-force-based microdevices, *Phys. Rev. A* **97**, 032501 (2018).
- [17] M. Liu, J. Xu, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, Examining the Casimir puzzle with an upgraded AFM-based technique and advanced surface cleaning, *Phys. Rev. B* **100**, 081406(R) (2019).
- [18] M. Liu, J. Xu, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, Precision measurements of the gradient of the Casimir force between ultraclean metallic surfaces at larger separations, *Phys. Rev. A* **100**, 052511 (2019).
- [19] G. Bimonte, B. Spreng, P. A. Maia Neto, G.-L. Ingold, G. L. Klimchitskaya, V. M. Mostepanenko, and R. S. Decca, Measurement of the Casimir force between 0.2 and 8  $\mu\text{m}$ : Experimental procedures and comparison with theory, *Universe* **7**, 93 (2021).
- [20] G. L. Klimchitskaya, U. Mohideen, and V. M. Mostepanenko, The Casimir force between real materials: Experiment and theory, *Rev. Mod. Phys.* **81**, 1827 (2009).
- [21] L. M. Woods, D. A. R. Dalvit, A. Tkatchenko, P. Rodriguez-Lopez, A. W. Rodriguez, and R. Podgornik, Materials perspective on Casimir and van der Waals interactions, *Rev. Mod. Phys.* **88**, 045003 (2016).
- [22] J. S. Høye, I. Brevik, J. B. Aarseth, and K. A. Milton, What is the temperature dependence of the Casimir effect?, *J. Phys. A* **39**, 6031 (2006).
- [23] G. L. Klimchitskaya and V. M. Mostepanenko, Experiment and theory in the Casimir effect, *Contemp. Phys.* **47**, 131 (2006).
- [24] K. A. Milton, Y. Li, P. Kalauni, P. Parashar, R. Guérout, G.-L. Ingold, A. Lambrecht, and S. Reynaud, Negative entropies in Casimir and Casimir-Polder interactions, *Fortschr. Phys.* **65**, 1600047 (2017).
- [25] G. Bimonte, T. Emig, M. Kardar, and M. Krüger, Non-equilibrium fluctuational quantum electrodynamics: Heat radiation, heat transfer, and force, *Annu. Rev. Condens. Matter Phys.* **8**, 119 (2017).
- [26] V. M. Mostepanenko, Casimir puzzle and conundrum: Discovery and search for resolution, *Universe* **7**, 84 (2021).
- [27] V. B. Bezerra, G. L. Klimchitskaya, V. M. Mostepanenko, and C. Romero, Violation of the Nernst heat theorem in the theory of thermal Casimir force between Drude metals, *Phys. Rev. A* **69**, 022119 (2004).
- [28] M. Bordag and I. Pirozhenko, Casimir entropy for a ball in front of a plane, *Phys. Rev. D* **82**, 125016 (2010).
- [29] M. Bordag, Low temperature expansion in the Lifshitz formula, *Adv. Math. Phys.* **2014**, 981586 (2014).
- [30] G. L. Klimchitskaya and V. M. Mostepanenko, Low-temperature behavior of the Casimir free energy and entropy of metallic films, *Phys. Rev. A* **95**, 012130 (2017).
- [31] G. L. Klimchitskaya and C. C. Korikov, Analytic results for the Casimir free energy between ferromagnetic metals, *Phys. Rev. A* **91**, 032119 (2015).
- [32] S. Boström and B. E. Sernelius, Entropy of the Casimir effect between real metal plates, *Physica (Amsterdam)* **339A**, 53 (2004).
- [33] I. Brevik, J. B. Aarseth, J. S. Høye, and K. A. Milton, Temperature dependence of the Casimir effect, *Phys. Rev. E* **71**, 056101 (2005).
- [34] J. S. Høye, I. Brevik, S. A. Ellingsen, and J. B. Aarseth, Analytical and numerical verification of the Nernst theorem for metals, *Phys. Rev. E* **75**, 051127 (2007).
- [35] J. S. Høye, I. Brevik, J. B. Aarseth, and K. A. Milton, Does the transverse electric zero mode contribute to the Casimir effect for a metal?, *Phys. Rev. E* **67**, 056116 (2003).
- [36] B. E. Sernelius, Effects of spatial dispersion on electromagnetic surface modes and on modes associated with a gap between two half spaces, *Phys. Rev. B* **71**, 235114 (2005).
- [37] P. A. Maia Neto, A. Lambrecht, and S. Reynaud, Casimir effect with rough metallic mirrors, *Phys. Rev. A* **72**, 012115 (2005).
- [38] V. B. Svetovoy, P. J. van Zwol, G. Palasantzas, and J. Th. M. De Hosson, Optical properties of gold films and the Casimir force, *Phys. Rev. B* **77**, 035439 (2008).
- [39] P. J. van Zwol, G. Palasantzas, and J. Th. M. De Hosson, Influence of random roughness on the Casimir force at small separations, *Phys. Rev. B* **77**, 075412 (2008).
- [40] G.-L. Ingold, A. Lambrecht, and S. Reynaud, Quantum dissipative Brownian motion and the Casimir effect, *Phys. Rev. E* **80**, 041113 (2009).
- [41] G. Bimonte, Bohr—van Leeuwen theorem and the thermal Casimir effect for conductors, *Phys. Rev. A* **79**, 042107 (2009).
- [42] B. E. Sernelius, Casimir experiments showing saturation effects, *Phys. Rev. A* **80**, 043828 (2009).

- [43] R. O. Behunin, F. Intravaia, D. A. R. Dalvit, P. A. Maia Neto, and S. Reynaud, Modeling electrostatic patch effects in Casimir force measurements, *Phys. Rev. A* **85**, 012504 (2012).
- [44] R. O. Behunin, D. A. R. Dalvit, R. S. Decca, C. Genet, I. W. Jung, A. Lambrecht, A. Liscio, D. López, S. Reynaud, G. Schnoering, G. Voisin, and Y. Zeng, Kelvin probe force microscopy of metallic surfaces used in Casimir force measurements, *Phys. Rev. A* **90**, 062115 (2014).
- [45] G. Bimonte, Hide It to See It Better: A Robust Setup to Probe the Thermal Casimir Effect, *Phys. Rev. Lett.* **112**, 240401 (2014).
- [46] M. Hartmann, G.-L. Ingold, and P. A. Maia Neto, Plasma versus Drude Modeling of the Casimir Force: Beyond the Proximity Force Approximation, *Phys. Rev. Lett.* **119**, 043901 (2017).
- [47] M. Bordag, Casimir and Casimir-Polder forces with dissipation from first principles, *Phys. Rev. A* **96**, 062504 (2017).
- [48] G. Bimonte, Beyond-proximity-force-approximation Casimir force between two spheres at finite temperature. II. Plasma versus Drude modeling, grounded versus isolated spheres, *Phys. Rev. D* **98**, 105004 (2018).
- [49] G. L. Klimchitskaya and V. M. Mostepanenko, An alternative response to the off-shell quantum fluctuations: A step forward in resolution of the Casimir puzzle, *Eur. Phys. J. C* **80**, 900 (2020).
- [50] J. Lindhard, On the properties of a gas of charged particles, *Dan. Mat. Fys. Medd.* **28**, 1 (1954).
- [51] K. L. Kliewer and R. Fuchs, Anomalous skin effect for specular electron scattering and optical experiments at non-normal angles of incidence, *Phys. Rev.* **172**, 607 (1968).
- [52] N. D. Mermin, Lindhard dielectric function in the relaxation time approximation, *Phys. Rev. B* **1**, 2362 (1970).
- [53] E. I. Kats, Influence of nonlocality effects on van der Waals interaction, *Zh. Eksp. Teor. Fiz.* **73**, 212 (1977) [*Sov. Phys. JETP* **46**, 109 (1977)].
- [54] R. Esquivel and V. B. Svetovoy, Correction to the Casimir force due to the anomalous skin effect, *Phys. Rev. A* **69**, 062102 (2004).
- [55] M. Dressel and G. Grüner, *Electrodynamics of Solids: Optical Properties of Electrons in Metals* (Cambridge University Press, Cambridge, England, 2003).
- [56] L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1984).
- [57] A. Erdélyi, W. Magnus, F. Oberhettinger, and F. G. Tricomi, *Higher Transcendental Functions*, Vol. I (Krieger, New York, 1981).
- [58] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1996).
- [59] G. L. Klimchitskaya and V. M. Mostepanenko, Quantum field theoretical description of the Casimir effect between two real graphene sheets and thermodynamics, *Phys. Rev. D* **102**, 016006 (2020).
- [60] L. D. Landau and E. M. Lifshitz, *Statistical Physics*, Part I (Pergamon, Oxford, 1980).
- [61] Yu. B. Rumer and M. S. Ryvkin, *Thermodynamics, Statistical Physics, and Kinetics* (Mir, Moscow, 1980).
- [62] D. C. Wood, The Computation of Polylogarithms, University of Kent, Computing Laboratory, 1992; <http://www.cs.kent.ac.uk/pubs/1992/110>.