Low-temperature behavior of the Casimir-Polder free energy and entropy for an atom interacting with graphene

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The analytic expressions for the free energy and entropy of the Casimir-Polder interaction between a polarizable and magnetizable atom and a graphene sheet are found in the limiting case of low temperature. In so doing, the response of graphene to electromagnetic fluctuations is described in the framework of the Dirac model by means of the polarization tensor in (2+1)-dimensional space-time. It is shown that the dominant contribution to the low-temperature behavior is given by an explicit dependence of the polarization tensor on temperature as a parameter. We demonstrate that the Lifshitz theory of atom-graphene interaction satisfies the Nernst heat theorem, i.e., is thermodynamically consistent. On this basis possible reasons of thermodynamic inconsistency arising for the Casimir-Polder and Casimir interactions in the case of Drude metals are discussed. The conclusion is made that, although large thermal effect arising in the Casimir interaction between Drude metals at short separations should be considered as an artifact, the giant thermal effect predicted for graphene systems is an important physical phenomenon which awaits its experimental observation.

I. INTRODUCTION

Physical phenomena known under a generic name of the Casimir-Polder interaction refer to the fluctuation-induced forces acting between polarizable and (or) magnetizable atoms (atomic systems) and material surfaces. These forces depend on the atomic and material properties, on the atom-surface separation, and on the temperature. At the shortest separations below a few nanometers they are of nonrelativistic character and are often called the van der Waals forces [1], whereas the relativistic generalization was obtained by Casimir and Polder [2] for the case of an ideal-metal plane surface. Independent of separation, the Casimir-Polder interaction is of an entirely quantum nature. Similar to the Casimir interaction, which refers to two macroscopic bodies separated by a narrow gap, it is described by the Lifshitz theory [3]. In the framework of this theory, the Casimir-Polder interaction was investigated for different atoms and surface materials [4–11]. Calculations of this kind are useful for interpretation of experiments on Bose-Einstein condensation [12–14], quantum reflection [15–17], and, e.g., for understanding of the resonance interaction of two atoms near a boundary surface [18].

In the past few years much attention has been focused on the Casimir-Polder interaction of different atoms with graphene and graphene-coated substrates [19–29]. Graphene is a two-dimensional sheet of carbon atoms packed in a hexagonal lattice which possesses unusual electrical, optical, and mechanical properties [30,31]. At low energies it is well described by the Dirac model which assumes that graphene quasiparticles are massless, obey a linear dispersion relation, but move at the Fermi velocity $v_F \approx c/300$ in place of the speed of light. As a result, the Casimir-Polder interaction of

atoms with a graphene sheet possesses the giant thermal effect at short separations [20] predicted earlier for the Casimir force between two graphene sheets [32].

It has been known that large thermal effect at short separations arises also in the Casimir interaction between two metallic plates if the dielectric properties of metal at low frequencies are described by the Drude model [33,34]. A similar effect arises in the Casimir-Polder force acting on an atom possessing both electric polarizability and magnetic susceptibility when it interacts with a metallic plate described by the Drude model. For the case of both nonmagnetic and magnetic metallic plates described by the Drude model an existence of large thermal effect in the Casimir force at short separations was unambiguously excluded by many experiments [35–43]. On the theoretical side, it was shown that the Lifshitz theory comes into conflict with the Nernst heat theorem when the response of metals with perfect crystal lattices to low-frequency electromagnetic fluctuations is described by the Drude model. This was proven in different geometries for the Casimir interaction between nonmagnetic [44–48] and magnetic [49] metals and, very recently, for the Casimir-Polder interaction of both polarizable and magnetizable atoms interacting with metallic plate [50]. This raises a question of whether the theoretical description of the Casimir-Polder interaction of atoms with graphene is thermodynamically consistent. For the Casimir interaction between two graphene sheets this fundamental question was solved positively [51], but for an atom possessing both the electric polarizability and magnetic susceptibility it still remains unsolved.

In this paper, we investigate the low-temperature behavior of the Casimir-Polder free energy and entropy for a

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polarizable and magnetizable atom interacting with a graphene sheet. All derivations are made in the framework of the Lifshitz theory and graphene is described by the Dirac model. The response of graphene to electromagnetic fluctuations is found on the basis of first principles of quantum electrodynamics at nonzero temperature using the polarization tensor in (2+1)-dimensional space-time. It is shown that both a summation over the discrete Matsubara frequencies and an explicit temperature dependence of the polarization tensor contribute to the Casimir-Polder free energy and entropy. The behaviors of both the free energy and entropy at low temperature are found analytically. In so doing, the dominant contribution to them originates from an explicit dependence of the polarization tensor on temperature. We demonstrate that the Lifshitz theory of atom-graphene interaction is in agreement with the Nernst heat theorem and, thus, is thermodynamically consistent. On this basis some conjectures concerning the reasons of inconsistency arising when the Drude model is used are inferred. Specifically, it is concluded that, although large thermal effect arising in the Casimir interaction between Drude metals should be considered as an artifact, the giant thermal effect for graphene is an important physical phenomenon which awaits its experimental observation.

The paper is organized as follows. In Sec. II the general formalism for the free energy of an atom-graphene interaction at low temperature is presented. Section III contains calculation of the contribution to the Casimir-Polder free energy due to an implicit temperature dependence. The contribution due to an explicit temperature dependence is found in Sec. IV. In Sec. V the Nernst heat theorem for an atom interacting with graphene is proven and some relevant problems are touched on. Section VI contains our conclusions and a discussion.

II. CASIMIR-POLDER FREE ENERGY FOR A POLARIZABLE AND MAGNETIZABLE ATOM INTERACTING WITH GRAPHENE AT LOW TEMPERATURE

We consider an atom characterized by the dynamic electric polarizability $\alpha(\omega)$ and magnetic susceptibility $\beta(\omega)$ separated by a distance a from a graphene sheet in thermal equilibrium at temperature T. In this case the free energy is given by the Lifshitz formula [7,11]

$$
\mathcal{F}(a, T) = -k_B T \sum_{l=0}^{\infty}{}' \int_0^{\infty} k_{\perp} dk_{\perp} q_l e^{-2aq_l}
$$
$$
\times \left\{ 2[\alpha_l r_{\mathrm{TM}}(i\xi_l, k_{\perp}) + \beta_l r_{\mathrm{TE}}(i\xi_l, k_{\perp})] \right.
$$
$$
\left. - \frac{\xi_l^2}{q_l^2 c^2}(\alpha_l + \beta_l)[r_{\mathrm{TM}}(i\xi_l, k_{\perp}) + r_{\mathrm{TE}}(i\xi_l, k_{\perp})] \right\}.
$$
$$(1)$$

Here, k_B is the Boltzmann constant, $k_{\perp}$ is the magnitude of the projection of the wave vector on the plane of graphene, $q_l^2 = k_{\perp}^2 + \xi_l^2/c^2$, $\xi_l = 2\pi k_B T l/\hbar$ with $l = 0, 1, 2, \ldots$ are the Matsubara frequencies, $\alpha_l = \alpha(i\xi_l)$, $\beta_l = \beta(i\xi_l)$, and the prime on the summation sign means that the term with $l = 0$ has to be multiplied by $1/2$. The quantities r_{TM} and r_{TE} are

the reflection coefficients of electromagnetic fluctuations on graphene with the transverse magnetic (TM) and transverse electric (TE) polarizations. Their explicit form is specified below.

Note that the free energy (1) is an approximate expression obtained as the first perturbation order in the small parameters α_l and β_l. The nonperturbative generalization of the zero-temperature Casimir-Polder force between an atom and an ideal-metal plane to the case $T \neq 0$ was obtained only a few years ago [52]. Very recently, the nonperturbative generalization of Eq. (1) was also derived for the case of any material plate [53]. It was shown, however, that the exact and perturbative free energies may differ for no more than 1% and only at $a < 1$ nm [53]. Taking into account that the Dirac model of graphene is applicable at frequencies below approximately $2 \, \mathrm{eV} \approx 3.05 \times 10^{15}$ rad/s, the formalism developed in this section works well at atom-graphene separations $a > 50$ nm. In this separation region the perturbative free energy (1) is indistinguishable from the exact one.

It is convenient to introduce the dimensionless variables

$$
y = 2q_l a, \quad \zeta_l = \frac{2a\xi_l}{c} \equiv \tau l, \qquad (2)
$$

where

$$
\tau = 4\pi \frac{a k_B T}{\hbar c} = 2\pi \frac{T}{T_{\mathrm{eff}}} \qquad (3)
$$

and the effective temperature for the Casimir effect is defined as $k_B T_{\mathrm{eff}} = \hbar c/(2a)$. In terms of these variables the free energy (1) takes the form

$$
\mathcal{F}(a, T) = -\frac{k_B T}{8a^3} \sum_{l=0}^{\infty}{}' \int_{\zeta_l}^{\infty} dy \, e^{-y} \{ 2y^2[\alpha_l r_{\mathrm{TM}}(i\zeta_l, y)
$$
$$
+ \beta_l r_{\mathrm{TE}}(i\zeta_l, y)] - \zeta_l^2(\alpha_l + \beta_l)[r_{\mathrm{TM}}(i\zeta_l, y)
$$
$$
+ r_{\mathrm{TE}}(i\zeta_l, y)] \}.
$$
$$(4)$$

The reflection coefficients r_{TM} and r_{TE} on a graphene sheet have been expressed via its polarization tensor in Ref. [54]. Here we use an equivalent form for the reflection coefficients

$$
r_{\mathrm{TM}}(i\zeta_l, y) = \frac{y\tilde{\Pi}_{00}(i\zeta_l, y)}{y\tilde{\Pi}_{00}(i\zeta_l, y) + 2(y^2 - \zeta_l^2)},
$$
$$
r_{\mathrm{TE}}(i\zeta_l, y) = -\frac{\tilde{\Pi}(i\zeta_l, y)}{\tilde{\Pi}(i\zeta_l, y) + 2y(y^2 - \zeta_l^2)},
$$
$$(5)$$

where $\tilde{\Pi}_{nm}$ with n, $m = 0$, 1, 2 is the dimensionless polarization tensor of graphene connected with the dimensional one, Π_{nm}, by $\tilde{\Pi}_{nm} = 2a\Pi_{nm}/\hbar$, the quantity $\tilde{\Pi}$ is defined as

$$
\tilde{\Pi}(i\zeta_l, y) = (y^2 - \zeta_l^2)\mathrm{tr}\tilde{\Pi}_{nm} - y^2\tilde{\Pi}_{00}, \qquad (6)
$$

and $\mathrm{tr}\tilde{\Pi}_{nm} = \tilde{\Pi}_n^n$ is the trace of the polarization tensor.

It is convenient to present the polarization tensor in the form

$$
\tilde{\Pi}_{00}(i\zeta_l, y) = \tilde{\Pi}_{00}^{(0)}(i\zeta_l, y) + \Delta_T \tilde{\Pi}_{00}(i\zeta_l, y),
$$
$$
\tilde{\Pi}(i\zeta_l, y) = \tilde{\Pi}^{(0)}(i\zeta_l, y) + \Delta_T \tilde{\Pi}(i\zeta_l, y),
$$
$$(7)$$

where $\tilde{\Pi}_{00}^{(0)}$ and $\tilde{\Pi}^{(0)}$ are found at $T = 0$ but with continuous dimensionless frequencies ζ replaced by the discrete Matsubara frequencies ζ_l and $\Delta_T \tilde{\Pi}_{00}$, $\Delta_T \tilde{\Pi}$ have the meaning of

thermal corrections. The polarization tensor at $T = 0$ has a very simple form [54,55]

$$\tilde{\Pi}_{00}^{(0)}(i\zeta_l, y) = \frac{\pi\alpha(y^2 - \zeta_l^2)}{\tilde{g}_l},$$

$$\tilde{\Pi}^{(0)}(i\zeta_l, y) = \pi\alpha(y^2 - \zeta_l^2)\tilde{g}_l, \tag{8}$$

where $\alpha = e^2/(\hbar c)$ is the fine-structure constant,

$$\tilde{g}_l = \tilde{g}_l(y) = \left[\tilde{v}_F^2 y^2 + (1 - \tilde{v}_F^2)\zeta_l^2\right]^{1/2} \tag{9}$$

and $\tilde{v}_F = v_F/c \approx 1/300$. Taking this into account, one can safely put

$$\tilde{g}_l \approx \left[\tilde{v}_F^2 y^2 + \zeta_l^2\right]^{1/2}. \tag{10}$$

The thermal corrections to the polarization tensor can be presented in the form valid only at the pure imaginary Matsubara frequencies [54] and over the entire plane of complex frequencies [55,56]. The latter form is used below. It is given by

$$\Delta_T\tilde{\Pi}_{00}(i\zeta_l, y) = \frac{8\alpha\tilde{g}_l}{\tilde{v}_F^2}\int_0^\infty \frac{du}{e^{B_l u} + 1}\left\{1 - \frac{1}{\sqrt{2}}\left[\sqrt{(1 + u^2)^2 - 4\frac{\tilde{v}_F^2(y^2 - \zeta_l^2)u^2}{\tilde{g}_l^2}} + 1 - u^2\right]^{1/2}\right\},$$

$$\Delta_T\tilde{\Pi}(i\zeta_l, y) = \frac{8\alpha\tilde{g}_l}{\tilde{v}_F^2}\int_0^\infty \frac{du}{e^{B_l u} + 1}\left\{-\zeta_l^2 + \frac{\tilde{g}_l^2}{\sqrt{2}}\left[\sqrt{(1 + u^2)^2 - 4\frac{\tilde{v}_F^2(y^2 - \zeta_l^2)u^2}{\tilde{g}_l^2}} + 1 - u^2\right]^{1/2}\right.$$

$$\left. \times \left[1 - \frac{\tilde{v}_F^2(y^2 - \zeta_l^2)}{\tilde{g}_l^2\sqrt{(1 + u^2)^2 - 4\frac{\tilde{v}_F^2(y^2 - \zeta_l^2)u^2}{\tilde{g}_l^2}}}\right]\right\}, \tag{11}$$

where $B_l = \pi\tilde{g}_l/\tau$.

From Eq. (11) it is seen that $B_l \to \infty$ when $\tau \to 0$ and, thus,

$$\lim_{T\to 0}\Delta_T\tilde{\Pi}_{00}(i\zeta_l, y) = \lim_{T\to 0}\Delta_T\tilde{\Pi}(i\zeta_l, y) = 0, \tag{12}$$

whereas, according to Eq. (8),

$$\lim_{T\to 0}\tilde{\Pi}_{00}^{(0)}(i\zeta_l, y) = \frac{\pi\alpha y}{\tilde{v}_F} \neq 0,$$

$$\lim_{T\to 0}\tilde{\Pi}^{(0)}(i\zeta_l, y) = \pi\alpha y^3\tilde{v}_F \neq 0. \tag{13}$$

Therefore, at sufficiently low T, one obtains

$$\frac{\Delta_T\tilde{\Pi}_{00}(i\zeta_l, y)}{\tilde{\Pi}_{00}^{(0)}(i\zeta_l, y)} \ll 1, \quad \frac{\Delta_T\tilde{\Pi}(i\zeta_l, y)}{\tilde{\Pi}^{(0)}(i\zeta_l, y)} \ll 1. \tag{14}$$

Substituting Eq. (7) in Eq. (5) and expanding up to the first power in small parameters (14), we find

$$r_{\text{TM}}(i\zeta_l, y) = r_{\text{TM}}^{(0)}(i\zeta_l, y) + \Delta_T r_{\text{TM}}(i\zeta_l, y),$$

$$r_{\text{TE}}(i\zeta_l, y) = r_{\text{TE}}^{(0)}(i\zeta_l, y) + \Delta_T r_{\text{TE}}(i\zeta_l, y). \tag{15}$$

Here, $r_{\text{TM(TE)}}^{(0)}$ are the reflection coefficients at zero temperature calculated at the pure imaginary Matsubara frequencies [55]

$$r_{\text{TM}}^{(0)}(i\zeta_l, y) = \frac{\alpha\pi y}{\alpha\pi y + 2\tilde{g}_l},$$

$$r_{\text{TE}}^{(0)}(i\zeta_l, y) = -\frac{\alpha\pi\tilde{g}_l}{\alpha\pi\tilde{g}_l + 2y}. \tag{16}$$

They are obtained by the substitution of Eq. (8) in place of Eq. (7) in Eq. (5). The quantities $\Delta_T r_{\text{TM(TE)}}$ in Eq. (15) have the meaning of the thermal corrections to the reflection

coefficients calculated up to the first order in parameters (14). They are given by

$$\Delta_T r_{\text{TM}}(i\zeta_l, y) = \frac{2\alpha\pi y\tilde{g}_l}{(\alpha\pi y + 2\tilde{g}_l)^2}\frac{\Delta_T\tilde{\Pi}_{00}(i\zeta_l, y)}{\tilde{\Pi}_{00}^{(0)}(i\zeta_l, y)},$$

$$\Delta_T r_{\text{TE}}(i\zeta_l, y) = -\frac{2\alpha\pi y\tilde{g}_l}{(\alpha\pi\tilde{g}_l + 2y)^2}\frac{\Delta_T\tilde{\Pi}(i\zeta_l, y)}{\tilde{\Pi}^{(0)}(i\zeta_l, y)}. \tag{17}$$

It was shown [51] that for sufficiently low temperatures, satisfying the condition

$$k_B T \ll \frac{\hbar v_F}{2a} \equiv k_B T_{\text{eff}}^{(g)}, \tag{18}$$

where $T_{\text{eff}}^{(g)} \neq T_{\text{eff}}$ is one more effective temperature for the Casimir effect in graphene systems, the dominant contributions to the parameters (14) at $l \geqslant 1$ take the form

$$\frac{\Delta_T\tilde{\Pi}_{00}(i\zeta_l, y)}{\tilde{\Pi}_{00}^{(0)}(i\zeta_l, y)} = \frac{48\zeta(3)}{\pi\tilde{g}_l^3}\left(\frac{T}{T_{\text{eff}}}\right)^3,$$

$$\frac{\Delta_T\tilde{\Pi}(i\zeta_l, y)}{\tilde{\Pi}^{(0)}(i\zeta_l, y)} = \frac{96\zeta(3)}{\pi\tilde{g}_l^3}\left(\frac{T}{T_{\text{eff}}}\right)^3\left(\frac{3\zeta_l^2}{2\tilde{g}_l^2} - 1\right). \tag{19}$$

Here, $\zeta(z)$ is the Riemann ζ function.

Finally, substitution of Eq. (19) in Eq. (17) results in the formulas

$$\Delta_T r_{\text{TM}}(i\zeta_l, y) = \frac{92\zeta(3)\alpha y}{\tilde{g}_l^2(\alpha\pi y + 2\tilde{g}_l)^2}\left(\frac{T}{T_{\text{eff}}}\right)^3,$$

$$\Delta_T r_{\text{TE}}(i\zeta_l, y) = -\frac{192\zeta(3)\alpha y}{\tilde{g}_l^2(\alpha\pi\tilde{g}_l + 2y)^2}\left(\frac{T}{T_{\text{eff}}}\right)^3\left(\frac{3\zeta_l^2}{2\tilde{g}_l^2} - 1\right), \tag{20}$$

which are valid for any $l \geqslant 1$. Equations (4), (15), (16), and (20) are used below to find the low-temperature behavior of

the Casimir-Polder free energy and entropy (special attention will be given to the case $l = 0$).

III. CONTRIBUTION TO THE FREE ENERGY DUE TO IMPLICIT TEMPERATURE DEPENDENCE

Substituting Eq. (15) in Eq. (4), one can present the Casimir-Polder free energy as a sum of two contributions

$$\mathcal{F}(a, T) = \mathcal{F}^{(1)}(a, T) + \Delta_T^{(2)} \mathcal{F}(a, T), \quad (21)$$

where both $\mathcal{F}^{(1)}$ and $\Delta_T^{(2)} \mathcal{F}$ have the same form as Eq. (4), but the reflection coefficients $r_{\mathrm{TM(TE)}}$ are replaced with $r_{\mathrm{TM(TE)}}^{(0)}$ and $\Delta_T r_{\mathrm{TM(TE)}}$, respectively.

In the lowest order with respect to the parameter $\zeta_l = \tau l$, we can restrict our attention to the static electric polarizability and magnetic susceptibility (note that the latter is essentially independent of frequency for many atoms [57]). If this is the case, using Eq. (4), the quantity $\mathcal{F}^{(1)}$ can be written in the form

$$\mathcal{F}^{(1)}(a, T) = -\frac{k_B T}{8a^3} \sum_{l=0}^{\infty} {}' \Phi(\tau l), \quad (22)$$

where

$$\Phi(\tau l) = \int_{\tau l}^{\infty} dy \, e^{-y} \{[2y^2 \alpha_0 - (\tau l)^2(\alpha_0 + \beta_0)] r_{\mathrm{TM}}^{(0)}(i\tau l, y)$$
$$+ [2y^2 \beta_0 - (\tau l)^2(\alpha_0 + \beta_0)] r_{\mathrm{TE}}^{(0)}(i\tau l, y)\} \quad (23)$$

and $\alpha_0 = \alpha(0)$, $\beta_0 = \beta(0)$.

Note that the Casimir-Polder energy at zero temperature can be represented in the same form [34]

$$E(a) = -\frac{\hbar c}{32\pi a^4} \int_0^{\infty} d\zeta \, \Phi(\zeta), \quad (24)$$

where

$$\Phi(\zeta) = \int_{\zeta}^{\infty} dy \, e^{-y} \{[2y^2 \alpha_0 - \zeta^2(\alpha_0 + \beta_0)] r_{\mathrm{TM}}^{(0)}(i\zeta, y)$$
$$+ [2y^2 \beta_0 - \zeta^2(\alpha_0 + \beta_0)] r_{\mathrm{TE}}^{(0)}(i\zeta, y)\}. \quad (25)$$

It is seen that Eq. (24) is obtainable from Eq. (22) in the case that a summation over the discrete Matsubara frequencies is replaced with an integration along the imaginary frequency axis in accordance to the rule

$$k_B T \sum_{l=0}^{\infty} {}' \rightarrow \frac{\hbar c}{4\pi a} \int_0^{\infty} d\zeta. \quad (26)$$

Thus, by applying the Abel-Plana formula [34]

$$\sum_{l=0}^{\infty} {}' \Phi(l) = \int_0^{\infty} \Phi(t) dt + i \int_0^{\infty} \frac{\Phi(it) - \Phi(-it)}{e^{2\pi t} - 1} dt, \quad (27)$$

which is valid for any function analytic in the right half-plane, one can represent the quantity $\mathcal{F}^{(1)}$ as

$$\mathcal{F}^{(1)}(a, T) = E(a) + \Delta_T^{(1)} \mathcal{F}(a, T), \quad (28)$$

where

$$\Delta_T^{(1)} \mathcal{F}(a, T) = -i \frac{k_B T}{8a^3} \int_0^{\infty} \frac{\Phi(i\tau t) - \Phi(-i\tau t)}{e^{2\pi t} - 1} dt. \quad (29)$$

From Eqs. (28) and (29) it is apparent that the thermal correction $\Delta_T^{(1)} \mathcal{F}$ represents an implicit temperature dependence

of the free energy which arises from a summation over the Matsubara frequencies in the contribution calculated with the zero-temperature reflection coefficients.

Direct calculation using Eqs. (25), (16), and (10) results in

$$\Phi(i\tau t) - \Phi(-i\tau t) = -2i\tau^3 t^3 (C_{\mathrm{TM}} + C_{\mathrm{TE}}), \quad (30)$$

where

$$C_{\mathrm{TM}} = \frac{2\alpha\pi\alpha_0}{\tilde{v}_F(\alpha\pi + 2\tilde{v}_F)^3} + \frac{\alpha_0 + 3\beta_0}{3(\alpha\pi + 2\tilde{v}_F)},$$
$$C_{\mathrm{TE}} = \frac{\alpha\pi\tilde{v}_F\alpha_0}{2} - \frac{\alpha\pi\beta_0}{2\tilde{v}_F}. \quad (31)$$

Note that, when calculating C_{TE}, we neglect by not only $\tilde{v}_F^2$, but also by $\alpha\pi\tilde{v}_F$, as compared to unity. Note also that the next contribution on the right-hand side of Eq. (30) is of the order of $\tau^4 \ln \tau$.

Substituting Eqs. (30) and (31) in Eq. (29), one obtains

$$\Delta_T^{(1)} \mathcal{F}(a, T) = -\frac{k_B T}{4a^3} \tau^3 (C_{\mathrm{TM}} + C_{\mathrm{TE}}) \int_0^{\infty} \frac{t^3 \, dt}{e^{2\pi t} - 1}$$
$$= -\frac{\pi^3 (k_B T)^4}{15(\hbar c)^3} (C_{\mathrm{TM}} + C_{\mathrm{TE}}). \quad (32)$$

This result is of the same order in T as for an atom interacting with a dielectric plate [34].

The main contribution to Eq. (32) is given by the first term in the coefficient C_{TM} defined in Eq. (31). Thus, if we assume that $\alpha_0 \sim \beta_0$, the first term in C_{TM} is more than the second one and than $|C_{\mathrm{TE}}|$ by the factors of 5×10^5 and 1.5×10^5, respectively. If we assume that $\alpha_0 \gg \beta_0$, the same ratios are equal to $\approx 2 \times 10^6$ and $\approx 4 \times 10^{11}$, respectively.

IV. CONTRIBUTION TO THE FREE ENERGY DUE TO EXPLICIT TEMPERATURE DEPENDENCE

We are coming now to the second contribution to the Casimir-Polder free energy on the right-hand side of Eq. (21). It is obtained by a replacement of the reflection coefficients $r_{\mathrm{TM(TE)}}$ in Eq. (4) with $\Delta_T r_{\mathrm{TM(TE)}}$ defined in Eq. (17),

$$\Delta_T^{(2)} \mathcal{F}(a, T)$$
$$= -\frac{k_B T}{8a^3} \sum_{l=0}^{\infty} {}' \int_{\zeta_l}^{\infty} dy \, e^{-y} \{2y^2 [\alpha_0 \Delta_T r_{\mathrm{TM}}(i\zeta_l, y)$$
$$+ \beta_0 \Delta_T r_{\mathrm{TE}}(i\zeta_l, y)] - \zeta_l^2(\alpha_0 + \beta_0)[\Delta_T r_{\mathrm{TM}}(i\zeta_l, y)$$
$$+ \Delta_T r_{\mathrm{TE}}(i\zeta_l, y)]\}. \quad (33)$$

Here, as explained in Sec. III, it is possible to restrict ourselves to the case of static polarizability and susceptibility. This contribution depends on the thermal correction to the polarization tensor of graphene and takes into account its explicit dependence on the temperature as a parameter. Here, we find the low-temperature behavior of Eq. (33). For this purpose, we present Eq. (33) as a sum of two terms

$$\Delta_T^{(2)} \mathcal{F}(a, T) = \Delta_T^{(2)} \mathcal{F}_{(l \geqslant 1)}(a, T) + \Delta_T^{(2)} \mathcal{F}_{(l=0)}(a, T) \quad (34)$$

and consider each of them separately.

We start from $\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}$ which includes all terms of Eq. (33) except for the term with $l = 0$. In this case the thermal corrections to the reflection coefficients in the lowest perturbation order are expressed by Eq. (20). Substituting Eq. (20) in Eq. (33), one obtains

$$
\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}(a, T) = -\frac{12\zeta(3)\alpha k_B T}{a^3}\left(\frac{T}{T_{\text{eff}}}\right)^3 \sum_{l=1}^{\infty}\int_{\tau l}^{\infty}dy\frac{ye^{-y}}{\tilde{g}_l^2}\left[\frac{2\alpha_0 y^2 - (\tau l)^2(\alpha_0 + \beta_0)}{(\alpha\pi y + 2\tilde{g}_l)^2}\right.
$$
$$
\left.-\frac{2\beta_0 y^2 - (\tau l)^2(\alpha_0 + \beta_0)}{(\alpha\pi\tilde{g}_l + 2y)^2}\left(\frac{3\tau^2 l^2}{\tilde{g}_l^2} - 2\right)\right]. \tag{35}
$$

Now we consider sufficiently low temperatures satisfying a condition $\tau l \ll \tilde{v}_F y$ and expand the quantity $\tilde{g}_l^2$ defined in Eq. (10) in powers of the small parameter $(\tau l/\tilde{v}_F y)^2$ taking into account that the major contribution to the integrals in Eq. (35) is given by $y \sim 1$. In so doing one can neglect by $3\tau^2 l^2/(2\tilde{g}_l^2)$ as compared to unity. We also take into account that $\alpha\pi\tilde{v}_F \approx 7.6 \times 10^{-5}$ and thus $\alpha\pi\tilde{g}_l \approx \alpha\pi\tilde{v}_F y \ll 2y$. As a result, Eq. (35) can be rewritten as

$$
\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}(a, T) = -\frac{12\zeta(3)\alpha k_B T}{\tilde{v}_F^2 a^3}\left(\frac{T}{T_{\text{eff}}}\right)^3 \sum_{l=1}^{\infty}\int_{\tau l}^{\infty}dy\frac{e^{-y}}{y^3}\left[\frac{2\alpha_0 y^2 - (\tau l)^2(\alpha_0 + \beta_0)}{(\alpha\pi + 2\tilde{v}_F)^2} + \frac{2\beta_0 y^2 - (\tau l)^2(\alpha_0 + \beta_0)}{2}\right]. \tag{36}
$$

After the change of integration variable $y = \tau l z$, we bring Eq. (36) to the form

$$
\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}(a, T) = -\frac{12\zeta(3)\alpha k_B T}{\tilde{v}_F^2 a^3}\left(\frac{T}{T_{\text{eff}}}\right)^3 \sum_{l=1}^{\infty}\left\{\int_1^{\infty}dz\frac{e^{-\tau l z}}{z}\left[\frac{2\alpha_0}{(\alpha\pi + 2\tilde{v}_F)^2} + \beta_0\right]\right.
$$
$$
\left.-\int_1^{\infty}dz\frac{e^{-\tau l z}}{z^3}(\alpha_0 + \beta_0)\left[\frac{1}{(\alpha\pi + 2\tilde{v}_F)^2} + \frac{1}{2}\right]\right\}. \tag{37}
$$

Now we make the summation first and calculate the integrals under a condition $\tau \ll 1$ with the result

$$
\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}(a, T) = -\frac{12\zeta(3)\alpha k_B T}{\tilde{v}_F^2 a^3}\left(\frac{T}{T_{\text{eff}}}\right)^3\frac{T_{\text{eff}}}{T}\left\{\frac{2\alpha_0}{(\alpha\pi + 2\tilde{v}_F)^2} + \beta_0 - \frac{1}{2}(\alpha_0 + \beta_0)\left[\frac{1}{(\alpha\pi + 2\tilde{v}_F)^2} + \frac{1}{2}\right]\right\}, \tag{38}
$$

or, equivalently,

$$
\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}(a, T) = -\frac{48\zeta(3)\alpha(k_B T)^3}{\tilde{v}_F^2(\hbar c)^2 a}(Q_1 + Q_2), \tag{39}
$$

where

$$
Q_1 = \frac{3\alpha_0 - \beta_0}{2(\alpha\pi + 2\tilde{v}_F)^2}, \quad Q_2 = -\frac{\alpha_0 - 3\beta_0}{4}. \tag{40}
$$

It is easily seen that the major contribution to Eq. (39) is given by the first term with a coefficient Q_1. For instance, if $\alpha_0 \sim \beta_0$ the coefficient Q_1 is larger than Q_2 by the factor of 2300. If $\alpha_0 \gg \beta_0$ one has $Q_1 \approx 3400 Q_2$.

Comparing Eqs. (32) and (39), we conclude that in the region of low temperatures an explicit temperature dependence, originating from the Matsubara terms with $l \geqslant 1$, is stronger than an implicit one.

Now we consider the term $\Delta_T^{(2)}\mathcal{F}_{(l=0)}$ on the right-hand side of Eq. (34) which is equal to the zero-frequency contribution to Eq. (33), i.e.,

$$
\Delta_T^{(2)}\mathcal{F}_{(l=0)}(a, T) = -\frac{k_B T}{8a^3}\int_0^{\infty}dy\, e^{-y}y^2[\alpha_0\Delta_T r_{\text{TM}}(0, y)
$$
$$
+ \beta_0\Delta_T r_{\text{TE}}(0, y)], \tag{41}
$$

where the thermal corrections to the reflection coefficients are obtained from Eq. (17),

$$
\Delta_T r_{\text{TM}}(0, y) = \frac{2\alpha\pi\tilde{v}_F}{(\alpha\pi + 2\tilde{v}_F)^2}\frac{\Delta_T\tilde{\Pi}_{00}(0, y)}{\tilde{\Pi}_{00}^{(0)}(0, y)},
$$
$$
\Delta_T r_{\text{TM}}(0, y) = -\frac{2\alpha\pi\tilde{v}_F}{(\alpha\pi\tilde{v}_F + 2)^2}\frac{\Delta_T\tilde{\Pi}(0, y)}{\tilde{\Pi}^{(0)}(0, y)}. \tag{42}
$$

Here, the polarization tensor at $T = 0$, $\zeta_0 = 0$ is found from Eq. (8),

$$
\tilde{\Pi}_{00}^{(0)}(0, y) = \frac{\pi\alpha y}{\tilde{v}_F}, \quad \tilde{\Pi}^{(0)}(0, y) = \pi\alpha\tilde{v}_F y^3, \tag{43}
$$

and the thermal correction to it at $\zeta_0 = 0$ from Eq. (11),

$$
\Delta_T\tilde{\Pi}_{00}(0, y) = \frac{8\alpha y}{\tilde{v}_F}\int_0^{\infty}\frac{du}{e^{B_0 u} + 1}(1 - \sqrt{1 - u^2}),
$$
$$
\Delta_T\tilde{\Pi}(0, y) = -8\alpha y^3\tilde{v}_F\int_0^{\infty}\frac{du}{e^{B_0 u} + 1}\frac{u^2}{\sqrt{1 - u^2}}, \tag{44}
$$

where $B_0 = \pi\tilde{v}_F y/\tau$.

It is more convenient to rearrange Eq. (44) to an equivalent form [56]

$$
\Delta_T\tilde{\Pi}_{00}(0, y) = \frac{32\alpha a k_B T}{\hbar c\tilde{v}_F^2}\int_0^1 dx\,\ln[1 + e^{-\tilde{B}y\sqrt{x(1-x)}}],
$$
$$
\Delta_T\tilde{\Pi}(0, y) = -16\alpha\tilde{v}_F y^3\int_0^1\frac{\sqrt{x(1-x)}dx}{e^{\tilde{B}y\sqrt{x(1-x)}} + 1}, \tag{45}
$$

where $\tilde{B} = 2B_0/y = T_{\text{eff}}^{(g)}/T$. Substituting Eqs. (43) and (45) in Eqs. (42) and (41), one obtains

$$\Delta_T^{(2)}\mathcal{F}_{(l=0)}(a,T) = -\frac{4\alpha k_B T}{a^3}\left\{\alpha_0 \frac{2ak_B T}{\hbar c(\alpha\pi + 2\tilde{v}_F)^2}\int_0^\infty dy\, e^{-y} y \int_0^1 dx\, \ln[1 + e^{-\tilde{B}y\sqrt{x(1-x)}}]\right.$$
$$\left. + \beta_0\frac{\tilde{v}_F}{4}\int_0^\infty dy\, e^{-y} y^2 \int_0^1 \frac{\sqrt{x(1-x)}dx}{e^{\tilde{B}y\sqrt{x(1-x)}} + 1}\right\}, \tag{46}$$

where we have neglected by $\pi\alpha\tilde{v}_F$, as compared to 2, in the second line of Eq. (42). Taking into account that

$$\frac{\sqrt{x(1-x)}}{e^{\tilde{B}y\sqrt{x(1-x)}} + 1} = -\frac{1}{\tilde{B}}\frac{d}{dy}\ln[1 + e^{-\tilde{B}y\sqrt{x(1-x)}}], \tag{47}$$

and integrating by parts with respect to y, the second term of Eq. (46) can be rewritten in the form

$$\beta_0\frac{\tilde{v}_F}{4\tilde{B}}\int_0^1 dx \int_0^\infty dy(2-y)y\, e^{-y}\ln[1 + e^{-\tilde{B}y\sqrt{x(1-x)}}]. \tag{48}$$

Now we represent the logarithms in both the first and second terms of Eq. (46) as the power series in $\exp[-\tilde{B}y\sqrt{x(1-x)}]$ and integrate with respect to y:

$$\Delta_T^{(2)}\mathcal{F}_{(l=0)}(a,T) = -\frac{8\alpha(k_B T)^2}{a^2\hbar c}\left\{\frac{\alpha_0}{(\alpha\pi + 2\tilde{v}_F)^2}\sum_{n=1}^\infty \frac{(-1)^{n-1}}{n}\int_0^1 \frac{dx}{[1 + n\tilde{B}\sqrt{x(1-x)}]^2}\right.$$
$$\left. + \frac{\beta_0}{2}\sum_{n=1}^\infty \frac{(-1)^{n-1}}{n}\int_0^1 dx\left[\frac{1}{[1 + n\tilde{B}\sqrt{x(1-x)}]^2} - \frac{1}{[1 + n\tilde{B}\sqrt{x(1-x)}]^3}\right]\right\}. \tag{49}$$

It is convenient to introduce one more small parameter $b_n = 1/(n\tilde{B}) = T/(nT_{\text{eff}}^{(g)})$ and define the integrals

$$I_k(b_n) = \int_0^1 \frac{dx}{[b_n + \sqrt{x(1-x)}]^k}, \tag{50}$$

where $k = 2, 3$ and $b_n < 1/2$. Then Eq. (49) takes the form

$$\Delta_T^{(2)}\mathcal{F}_{(l=0)}(a,T) = -\frac{8\alpha(k_B T)^2}{a^2\hbar c}\left(\frac{T}{T_{\text{eff}}^{(g)}}\right)^2\left\{\frac{\alpha_0}{(\alpha\pi + 2\tilde{v}_F)^2}\sum_{n=1}^\infty \frac{(-1)^{n-1}}{n}I_2(b_n) + \frac{\beta_0}{2}\sum_{n=1}^\infty \frac{(-1)^{n-1}}{n^3}[I_2(b_n) - b_n I_3(b_n)]\right\}. \tag{51}$$

Direct calculation results in

$$I_2(b_n) = -\frac{4}{1 - 4b_n^2}\left[1 + \frac{1}{\sqrt{1 - 4b_n^2}}\left(\ln\frac{1 - \sqrt{1 - 4b_n^2}}{1 + \sqrt{1 - 4b_n^2}} - \ln\frac{1 + 2b_n - \sqrt{1 - 4b_n^2}}{1 + 2b_n + \sqrt{1 - 4b_n^2}}\right)\right],$$

$$I_3(b_n) = \frac{4}{(1 - 4b_n^2)^2}\left[\frac{1 + 8b_n^2}{2b_n} + \frac{6b_n}{\sqrt{1 - 4b_n^2}}\left(\ln\frac{1 - \sqrt{1 - 4b_n^2}}{1 + \sqrt{1 - 4b_n^2}} - \ln\frac{1 + 2b_n - \sqrt{1 - 4b_n^2}}{1 + 2b_n + \sqrt{1 - 4b_n^2}}\right)\right]. \tag{52}$$

Expanding Eq. (52) in powers of b_n one obtains

$$I_2(b_n) = -4\ln b_n + O(b_n^0),$$
$$I_3(b_n) = \frac{2}{b_n} + 24 b_n \ln b_n + O(b_n^0). \tag{53}$$

Substituting these results in Eq. (51) and finding main contributions to the sums in n, we arrive at

$$\Delta_T^{(2)}\mathcal{F}_{(l=0)}(a,T) = \frac{96\alpha\zeta(3)(k_B T)^4}{\tilde{v}_F^2(\hbar c)^3}(R_1 + R_2)\ln\frac{2ak_B T}{\hbar\tilde{v}_F c}, \tag{54}$$

where

$$R_1 = \frac{\alpha_0}{(\alpha\pi + 2\tilde{v}_F)^2}, \quad R_2 = \frac{\beta_0}{4}. \tag{55}$$

The major contribution to Eq. (54) is given by the first term with the coefficient R_1. For $\alpha_0 \sim \beta_0$, R_1 is larger than R_2 by the factor of ≈ 1100 and all the more if $\alpha_0 \gg \beta_0$.

As is seen in Eq. (54), with decreasing T down to zero temperature $\Delta_T^{(2)}\mathcal{F}_{(l=0)}$ becomes greater than $\Delta_T^{(1)}\mathcal{F}$, determined by the implicit dependence on the temperature, but less than $\Delta_T^{(2)}\mathcal{F}_{(l\geqslant1)}$ originating from the explicit temperature dependence of all Matsubara terms with nonzero frequency.

V. LOW-TEMPERATURE BEHAVIOR OF THE FREE ENERGY AND ENTROPY

In Secs. III and IV we have found the low-temperature behavior of all contributions to the Casimir-Polder free energy. According to Eqs. (21), (28), and (34), the free energy is given by

$$\mathcal{F}(a,T) = E(a) + \Delta_T^{(1)}\mathcal{F}(a,T) + \Delta_T^{(2)}\mathcal{F}_{(l\geqslant1)}(a,T)$$
$$+ \Delta_T^{(2)}\mathcal{F}_{(l=0)}(a,T), \tag{56}$$

where $\Delta_T^{(1)}\mathcal{F}$ in Eq. (32) presents the implicit low-temperature behavior originating exclusively from a summation over the Matsubara frequencies, whereas $\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}$ and $\Delta_T^{(2)}\mathcal{F}_{(l=0)}$ found in Eqs. (39) and (54), respectively, are determined by the explicit dependence of the polarization tensor on temperature as a parameter. As is seen from Eqs. (32), (39), and (54), with decreasing temperature the major contribution is given by $\Delta_T^{(2)}\mathcal{F}_{(l\geqslant 1)}$. Thus, from Eqs. (39) and (56), one can conclude that

$$\mathcal{F}(a, T) = E(a) - \frac{48\zeta(3)\alpha(k_B T)^3}{\tilde{v}_F^2(\hbar c)^2 a}(Q_1 + Q_2), \qquad (57)$$

where the coefficients Q_1 and Q_2 are given in Eq. (40).

From Eq. (57) one obtains the low-temperature behavior of the Casimir-Polder entropy

$$S(a, T) = -\frac{\partial \mathcal{F}(a, T)}{\partial T} = \frac{144\zeta(3)\alpha k_B(k_B T)^2}{\tilde{v}_F^2(\hbar c)^2 a}(Q_1 + Q_2). \qquad (58)$$

From Eq. (54) we find that the next term in the low-temperature behavior of the entropy is of the order of

$$k_B R_1 \frac{(k_B T)^3}{(\hbar c)^3} \ln \frac{2a k_B T}{\hbar \tilde{v}_F c}. \qquad (59)$$

Equation (58) allows one to make a conclusion that the Casimir-Polder entropy is positive and goes to zero with vanishing temperature in accordance with the third law of thermodynamics, the Nernst heat theorem. This means that the Lifshitz theory of atom-graphene interaction is thermodynamically consistent if the response of graphene to a fluctuating field is described by the polarization tensor in the framework of the Dirac model.

This fundamental result returns us to the problem discussed in Sec. I, i.e., why the Lifshitz theory of the Casimir and Casimir-Polder interaction violates the Nernst heat theorem and is inconsistent with the measurement data of several experiments if the low-frequency electromagnetic response of metals is described by the well tested under ordinary conditions Drude model taking into account the relaxation properties of free charge carriers.

In connection with this, it is significant that the response of graphene to electromagnetic fluctuations is described by the polarization tensor on the basis of first principles of quantum electrodynamics at nonzero temperature. This description is in full agreement with all fundamental demands, such as causality, and satisfies the Kramers-Kronig relations [58]. By contrast, the Drude dielectric permittivity,

$$\varepsilon_D(\omega) = 1 - \frac{\omega_p^2}{\omega[\omega + i\gamma(T)]}, \qquad (60)$$

where ω_p is the plasma frequency and $\gamma(T)$ is the relaxation parameter, is of entirely phenomenological character. Although it provides an adequate description of the electrical conductivity and optical properties of metals and satisfies the Kramers-Kronig relations at nonzero temperature, the problem arises in the limiting case of vanishing temperature.

The point is that for metals with perfect crystal lattices $\gamma(T)$ vanishes when T goes to zero [59]. In this case [60]

$$\lim_{\gamma \to 0} \varepsilon_D(\omega) = 1 - \frac{\omega_p^2}{\omega^2} + i\frac{\omega_p^2}{\omega}\pi\delta(\omega), \qquad (61)$$

where $\delta(\omega)$ is the Dirac δ function. This means that in the limit of zero temperature the Drude dielectric permittivity cannot be continued to the upper half plane of complex frequency and its imaginary part cannot be obtained from its real part by means of the Kramers-Kronig relation [61]. Thus, in the limit of zero temperature, the Drude model violates the principle of causality and cannot be used as a dielectric permittivity. This gives an insight into why the Lifshitz theory combined with the Drude model is in conflict with the Nernst heat theorem. Note that if the plasma model is used for description of the electromagnetic response of a metal, i.e., $\gamma(T)$ in Eq. (60) is put equal to zero from the outset, the Nernst heat theorem for the Casimir and Casimir-Polder entropy is satisfied, as well as the Kramers-Kronig relations in the form valid for functions possessing the second-order pole at zero frequency [62].

VI. CONCLUSIONS AND DISCUSSION

In the foregoing, we have analyzed the thermodynamic consistency of the Lifshitz theory used for description of the Casimir-Polder interaction between a polarizable and magnetizable atom and a graphene sheet. In so doing, the response of graphene to electromagnetic fluctuations was described by the polarization tensor in (2+1)-dimensional space-time in the framework of quantum electrodynamics at nonzero temperature. We have found analytic expressions for the Casimir-Polder free energy and entropy at low temperature. For this purpose the thermal correction to the Casimir-Polder energy was represented as a sum of three contributions. The first of them originates from a summation on the pure imaginary Matsubara frequencies and two others from an explicit dependence of the polarization tensor on temperature as a parameter. It was shown that the dominant contribution to the free energy and entropy at low temperature is given by an explicit temperature dependence contained in the nonzero-frequency terms of the Lifshitz formula.

Using the obtained analytic results, it was demonstrated that the Casimir-Polder entropy of a polarizable and magnetizable atom interacting with a graphene sheet satisfies the Nernst heat theorem. Thus the Lifshitz theory of an atom interacting with graphene is thermodynamically consistent. This fact was correlated with a violation of the Nernst theorem in the Lifshitz theory of Casimir and Casimir-Polder interactions in the case that the plate metal is described by the phenomenological Drude model. Special attention was paid to the fact that the Drude dielectric function with vanishing relaxation parameter ceases to be an analytic function in the upper half-plane of complex frequency and violates the principle of causality. This can be considered as a possible reason of thermodynamic inconsistency. By contrast, the response of graphene to the electromagnetic field is described on the basis of first principles of quantum field theory and is in agreement with the Kramers-Kronig relations for all values of parameters.

On the basis of this discussion we conclude that large thermal effect at short separations predicted by the Lifshitz theory for Drude metals and already excluded experimentally should be considered as an artifact. As to the giant thermal effect in the Casimir and Casimir-Polder interactions for graphene, this is an important physical phenomenon which awaits experimental observation. Two realistic possibilities on how to observe this effect have been proposed recently [63–65], making its discovery in the near future very likely.

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