Lifshitz theory of van der Waals pressure in dissipative media

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(Received 27 November 2010; published 11 April 2011)

We derive a first-principles method of determining the van der Waals or Casimir pressure in a dissipative and dispersive planar multilayered system by calculating the Maxwell stress tensor in a fictitious layer of vacuum, that is eventually made to vanish, introduced in the structure. This is illustrated by calculating the van der Waals pressure in a thin film with dissipative properties embedded between two semi-infinite media.

I. INTRODUCTION

van der Waals (vdW) forces, resulting from alteration of the quantum and thermal fluctuations of the electrodynamic field due to the presence of interfaces, play a significant role in the interactions between macroscopic objects at micrometer- and nanometer-length scales. Hamaker was the first to extend the concept of London-vdW forces between two atoms to forces between macroscopic spheres by pairwise summation of the interaction energy between the atoms that constitute the spheres [1]. Lifshitz, in his seminal work [2], outlined a method based on Rytov's theory of fluctuational electrodynamics [3] for computing the vdW forces between two semi-infinite regions separated by a vacuum gap. It required calculation of the average value of the Maxwell stress tensor in the vacuum gap. The generalization of Lifshitz's method to calculating vdW forces between semi-infinite regions separated by dissipative media is not straightforward because of the difficulty in defining an electromagnetic stress tensor in dissipative media [4]. In this paper we show that it is possible to generalize Lifshitz's theory to determine the vdW pressure in arbitrary planar media with dissipative and dispersive electromagnetic properties without having to define the electromagnetic stress tensor or free energy in any material but vacuum.

An approach proposed by Dzyaloshinskii, Lifshitz, and Pitaevskii (DLP from now on) [5], cast in the language of quantum field theory, is the most frequently used generalization of Lifshitz's method to calculate forces between objects separated by absorbing media. Even though it has been noted that an expression for the Maxwell stress tensor for time-varying fields in absorbing media cannot be expressed in terms of the frequency-dependent permittivity and permeability alone [4], the DLP method effectively reduces to using a "Minkowski-like" [6] definition of the electromagnetic stress tensor in dissipative media. Ninham *et al.* [7] circumvented the complications of the DLP method but, in doing so, had to postulate that the free energy of an electromagnetic mode at frequency ω_j is given by $k_B T \ln[\sinh(\hbar\omega_j/2k_B T)]$, where k_B is Boltzmann's constant, $2\pi\hbar$ is Planck's constant, and T is the absolute temperature, even though the mode frequencies in dissipative media are, in general, complex. It has been argued by Barash and Ginzburg [8,9] that ascribing to each mode a free energy of the above-mentioned form is indeed correct. The methods of DLP and Barash and Ginzburg are justified on the

grounds that it is possible to ascribe thermodynamic functions to electromagnetic fields in equilibrium with matter [6,9]. While many authors have attempted to generalize Lifshitz theory to determining vdW pressure in dissipative media, they do so by assuming an expression for the electrodynamic stress tensor [10] or by defining a Lagrangian density for the electrodynamic field [11], both of which are debatable for media with dissipation [4].

The relative transparency of the Lifshitz method is obscured by the complexity of Dzyaloshinskii's formalism or by having to define the free energy of each mode, even though the final result is a simple generalization of the Lifshitz formula. It has been generally regarded that Lifshitz's method, in which the stress tensor definition is above reproach, is incapable of handling dissipative media without relying on either of the two generalizations [6]. Using the fluctuation-dissipation theorem and properties of the dyadic Green's function, we express the components of the Maxwell stress tensor in vacuum in terms of components of the dyadic Green's function [12]. After a description of a general method to deal with multilayered media, we show, using examples of (1) a thin film bound by vacuum on both sides, (2) a thin film with vacuum on one side and a semi-infinite medium with arbitrary permittivity and permeability on the other, and (3) a thin film bound by semi-infinite media with arbitrary permittivity and permeability, that the expression for vdW pressure coincides with that of DLP.

II. GENERAL FORMULATION OF VAN DER WAALS ENERGY AND PRESSURE IN MULTILAYERED MEDIA

Let us analyze a general multilayer system, as shown in Fig. 1(a), and express the vdW free energy of the system in terms of combinations of vdW free energy of smaller units. The vdW free energy per unit area of a planar configuration of N layers [Fig. 1(a)] sandwiched between two semi-infinite media, medium L to the left and medium R to the right, is represented by $U_{LR}(z_1,\dots,z_N)$. Each layer is characterized by not only the thickness z_k but also the permittivity ε_k and permeability μ_k (both relative to that of vacuum). We use the aforementioned notation for free energy for its efficiency. If one of the semi-infinite media is vacuum, the subscript V is used instead of L or R. $U_{LR}(z_1,\dots,z_N)$ can be written as a combination of three terms: (1) the free energy of the first k layers sandwiched by semi-infinite medium L to the left and vacuum to the right of the kth layer, $U_{LV}(z_1,\dots,z_k)$, and (2) the free energy of the remaining $N - k$ layers sandwiched by
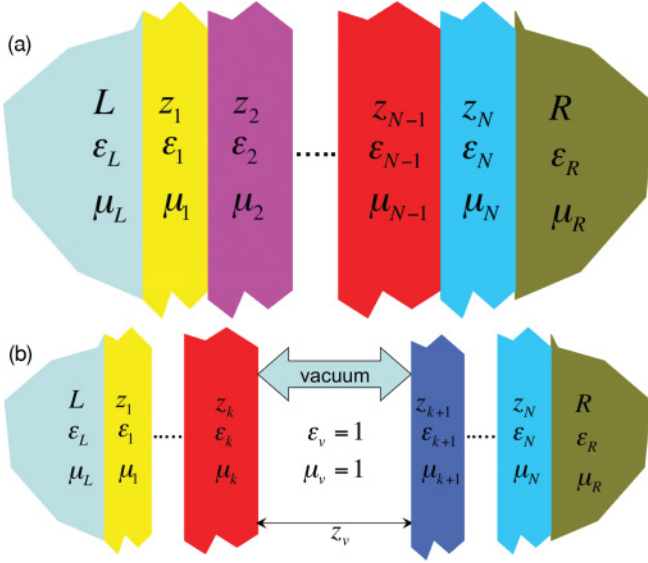
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FIG. 1. (Color online) (a) A multilayer system with N layers between two semi-infinite regions L and R. (b) Method of splitting the N layer multilayer system into components. The z axis is perpendicular to the interfaces.

semi-infinite medium R to the right and vacuum to the left of the $(k+1)$th layer, $U_{VR}(z_{k+1}, \ldots, z_N)$, and (3) the work done in bringing the two systems from infinite separation to a separation $\delta \to 0$. This statement can be written as

$$U_{LR}(z_1, \ldots, z_N) = U_{LV}(z_1, \ldots, z_k) + U_{VR}(z_{k+1}, \ldots, z_N)$$
$$+ \lim_{\delta \to 0} \int_{\infty}^{\delta} T_{zz}^{\mathrm{avg}}(z_v)\, dz_v, \qquad (1)$$

where $T_{zz}^{\mathrm{avg}}(z_v) \equiv T_{zz}^{\mathrm{avg}}(z_1, \ldots, z_k, z_v, z_{k+1}, \ldots, z_N)$ is the vdW pressure in the vacuum region in Fig. 1(b) against which work needs to be done to create the N layer system from the two subsystems. The partial derivative $\partial U_{LR}(z_1, \ldots, z_N)/\partial z_r = p_{LR}^{(r)}(z_1, \ldots, z_N)$ gives the vdW pressure in the rth layer of the N layer system bounded by L and R. For a thin film bounded by two semi-infinite regions, we drop the superscript (r) and denote the pressure simply as p_{LR}. By differentiating Eq. (1) with respect to z_r, we obtain the following equation for $p_{LR}^{(r)}$:

$$p_{LR}^{(r)}(z_1, \ldots, z_N) = \frac{\partial U_{LV}}{\partial z_r}(z_1, \ldots, z_k) + \frac{\partial U_{VR}}{\partial z_r}(z_{k+1}, \ldots, z_N)$$
$$+ \int_{\infty}^{0} \frac{\partial T_{zz}^{\mathrm{avg}}}{\partial z_r}(z_v)\, dz_v. \qquad (2)$$

One of the first two terms on the right-hand side of Eq. (2) is zero, depending on whether $1 \leqslant r \leqslant k$ or $k+1 \leqslant r \leqslant N$. Though $T_{zz}^{\mathrm{avg}}(z_v)$ diverges as z_v^{-3} for $z_v \to 0$, the quantity $\partial T_{zz}^{\mathrm{avg}}/\partial z_r$ is finite as $z_v \to 0 \ \forall \ 1 \leqslant r \leqslant N$, allowing us to define the partial derivative of the last term in Eq. (1) as the integral $\int_{\infty}^{0} \partial T_{zz}^{\mathrm{avg}}/\partial z_r dz_v$ (see Appendix A for justification). $T_{zz}^{\mathrm{avg}}(z_v)$ is obtained simply by determining the zz component of the Maxwell stress tensor in the vacuum region. Using the procedure described above, we can write the vdW free energy of any N layer medium in terms of $U_{VV}(z_1)$, $U_{VV}(z_2)$, ..., and $U_{VV}(z_N)$, and contributions from terms of the form

$\int_{\infty}^{\delta} T_{zz}^{\mathrm{avg}}(z_v) dz_v$, all of which involve calculation of the Maxwell stress tensor in vacuum alone. $U_{VV}(z)$ is nothing but the vdW free energy to create a thin film of thickness z in free space.

We rely on Rytov's theory of fluctuational electrodynamics to determine the value of T_{zz}^{avg}. The cross-spectral correlations of the electric field components can be written as $\langle E_p(\boldsymbol{r}, \omega) E_q^*(\boldsymbol{r}, \omega) \rangle = (2\omega \mu_o \Theta/\pi) \mathrm{Im} G_{pq}^e(\boldsymbol{r}, \boldsymbol{r})$, where $p, q = x, y, z$, $\Theta = (\hbar\omega/2)\coth(\hbar\omega/2k_B T)$, and $\boldsymbol{G}_{pq}^e$ is the pq component of the electric dyadic Green's function [12,13]. The spectral correlation is defined such that $\langle E_p(\boldsymbol{r}, t) E_q(\boldsymbol{r}, t) \rangle = \int_0^{\infty} d\omega \langle E_p(\boldsymbol{r}, \omega) E_q^*(\boldsymbol{r}, \omega) \rangle$. Similarly, the cross-spectral correlations of the magnetic field components are given by $\langle H_p(\boldsymbol{r}, \omega) H_q^*(\boldsymbol{r}, \omega) \rangle = (2\omega \epsilon_o \Theta/\pi) \mathrm{Im} G_{pq}^m(\boldsymbol{r}, \boldsymbol{r})$. The dyadic Green's functions $\boldsymbol{G}^e$ and $\boldsymbol{G}^m$ are electromagnetic duals of each other and are solutions of $\nabla \times \nabla \times \boldsymbol{G}(\boldsymbol{r}, \boldsymbol{r}') - k^2 \boldsymbol{G}(\boldsymbol{r}, \boldsymbol{r}') = \boldsymbol{I}\delta(\boldsymbol{r} - \boldsymbol{r}')$, where $\boldsymbol{I}$ is the identity dyad, $\boldsymbol{r}$ and $\boldsymbol{r}'$ are the position vectors for observation and source, respectively. $\boldsymbol{G}^e$ and $\boldsymbol{G}^m$ are obtained by enforcing the continuity of (1) $\mu(\boldsymbol{r})[\hat{\boldsymbol{n}} \times \boldsymbol{G}^e(\boldsymbol{r}, \boldsymbol{r}')]$, (2) $\hat{\boldsymbol{n}} \times \nabla \times \boldsymbol{G}^e(\boldsymbol{r}, \boldsymbol{r}')$, (3) $\epsilon(\boldsymbol{r})[\hat{\boldsymbol{n}} \times \boldsymbol{G}^m(\boldsymbol{r}, \boldsymbol{r}')]$, and (4) $\hat{\boldsymbol{n}} \times \nabla \times \boldsymbol{G}^m(\boldsymbol{r}, \boldsymbol{r}')$ on either side of an interface defined by the unit normal vector $\hat{\boldsymbol{n}}$ at the point $\boldsymbol{r}$.

The zz component of the Maxwell stress tensor in vacuum can be expressed in terms of $\boldsymbol{G}^e$ and $\boldsymbol{G}^m$ as $T_{zz}(\boldsymbol{r}, \omega) = (2\omega \Theta/\pi c^2) \mathrm{Im} G(\boldsymbol{r}, \omega)$ [12,13], where $G(\boldsymbol{r}, \omega) = \boldsymbol{G}_{zz}^e(\boldsymbol{r}, \boldsymbol{r}) - \frac{1}{2}\mathrm{Tr}\boldsymbol{G}^e(\boldsymbol{r}, \boldsymbol{r}) + \boldsymbol{G}_{zz}^m(\boldsymbol{r}, \boldsymbol{r}) - \frac{1}{2}\mathrm{Tr}\boldsymbol{G}^m(\boldsymbol{r}, \boldsymbol{r})$. The average value of the zz component of the Maxwell stress tensor, T_{zz}^{avg}, at any instant of time at position $\boldsymbol{r}$ in a vacuum layer is given by:

$$T_{zz}^{\mathrm{avg}} = \int_0^{\infty} \frac{\hbar\omega^2}{\pi c^2} \coth\left(\frac{\hbar\omega}{2k_B T}\right) \mathrm{Im} G(\boldsymbol{r}, \omega) d\omega \qquad (3)$$

$\boldsymbol{G}^e$ and $\boldsymbol{G}^m$ are analytic in the upper half plane (UHP) by virtue of being response functions. Since $G(\boldsymbol{r}, \omega)$ is a linear combination of different components of $\boldsymbol{G}^e$ and $\boldsymbol{G}^m$, it is also analytic in the UHP. We can therefore use Lifshitz's technique to replace the integral over ω along the real positive frequency axis by a summation over Matsubara frequencies on the imaginary frequency axis in the UHP as

$$T_{zz}^{\mathrm{avg}} = -\frac{2k_B T}{c^2} \sum_{n=0}^{\infty}{}' \xi_n^2 G(\boldsymbol{r}, i\xi_n) = k_B T \sum_{n=0}^{\infty}{}' K_n \qquad (4)$$

where, $\xi_n = 2\pi n k_B T/\hbar$, $K_n = -2\xi_n^2 G(\boldsymbol{r}, i\xi_n)/c^2$, $K_0 = -\lim_{\xi \to 0} 2\xi^2 G(\boldsymbol{r}, i\xi)/c^2$, and $n = 0, 1, 2, \ldots$. The prime $(')$ next to $\sum$ indicates that the $n = 0$ term is given weight 0.5. $G(\boldsymbol{r}, i\xi_n)$ can be written in terms of the reflection coefficients of plane waves that comprise $\boldsymbol{G}^e$ and $\boldsymbol{G}^m$ [14]. We now apply this method to calculating the vdW pressure in a thin film (indicated by m) bounded by two semi-infinite objects, L and R. To do so, we introduce a vacuum layer, shown in Fig. 2, in which the Maxwell stress tensor will be determined.

III. APPLICATION TO THIN FILMS

We start with the assertion that the vdW pressure in any infinite or semi-infinite planar medium is zero. We show, using the following three examples, that the proposed method is in agreement with the predictions of DLP theory for the case of a thin film between two semi-infinite objects. K_n, from which

T_{zz}^{avg} can be calculated using Eq. (4), for the configuration shown in Fig. 2 is given by

$$K_n = \frac{1}{\pi} \int_0^\infty \sum_{p=e,h} \frac{R_{vL}^{(p)} \widetilde{R}_{vR}^{(p)} e^{-2k_{zv}z_v}}{1 - R_{vL}^{(p)} \widetilde{R}_{vR}^{(p)} e^{-2k_{zv}z_v}} k_{zv}k_\rho dk_\rho, \quad (5)$$

where $p = e,h$ refer to the transverse electric and transverse magnetic polarizations, respectively, and

$$\widetilde{R}_{vR}^{(p)} = \frac{R_{vm}^{(p)} + R_{mR}^{(p)} e^{-2k_{zm}z_m}}{1 + R_{vm}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}}, \quad (6a)$$

$$R_{vL}^{(e)} = \frac{k_{zv}\mu_L - k_{zL}\mu_v}{k_{zv}\mu_L + k_{zL}\mu_v}, \quad R_{vL}^{(h)} = \frac{k_{zv}\varepsilon_L - k_{zL}\varepsilon_v}{k_{zv}\varepsilon_L + k_{zL}\varepsilon_v}, \quad (6b)$$

$$k_{zv} = \sqrt{k_\rho^2 + \varepsilon_v\mu_v\xi_n^2/c^2}, \quad k_{zm} = \sqrt{k_\rho^2 + \varepsilon_m\mu_m\xi_n^2/c^2}, \quad (6c)$$

and similarly for reflection coefficients at other interfaces and wave vectors in other layers. All permittivities and permeabilities are evaluated at $i\xi_n, n = 0,1,2,\dots$ For reflection coefficients, the subscript v is used to denote an interface with vacuum. Since it is $\frac{\partial}{\partial z_m}\int_\infty^0 T_{zz}^{\text{avg}}(z_m,z_v)dz_v$ that will eventually be used in calculating vdW pressure, below we give the expression for $\int_\infty^0 \frac{\partial K_n}{\partial z_m}dz_v$,

$$\int_\infty^0 \frac{\partial K_n}{\partial z_m}dz_v = \frac{1}{\pi} \int_0^\infty \sum_{p=e,h} \frac{R_{vL}^{(p)} R_{mR}^{(p)}\left(1 - R_{vm}^{(p)2}\right)e^{-2k_{zm}z_m}}{\left[\left(1 - R_{vL}^{(p)}R_{vm}^{(p)}\right) - \left(R_{vL}^{(p)}R_{mR}^{(p)} + R_{mv}^{(p)}R_{mR}^{(p)}\right)e^{-2k_{zm}z_m}\right]\left(1 + R_{vm}^{(p)}R_{mR}^{(p)}e^{-2k_{zm}z_m}\right)} k_{zm}k_\rho dk_\rho \quad (7)$$

Example 1: Vacuum–Thin Film–Vacuum. To find the vdW pressure in a thin film of material of thickness z_m, we consider a four-layer configuration, as shown in Fig. 2, with L being replaced with material m, and R being vacuum. If the vdW energy for creating a film of thickness z_m is $U_{VV}(z_m)$, the following equation can be written for conservation of energy for moving the thin film from $z_v = \infty$ to $z_v = \delta \to 0$:

$$U_{VV}(z_m) + \lim_{\delta \to 0} \int_\infty^\delta T_{zz}^{\text{avg}}dz_v = U_o, \quad (8)$$

where U_o is an arbitrary constant that is the energy per unit area of a semi-infinite medium M adjacent to a semi-infinite region of vacuum. Differentiation of Eq. (8) with respect to z_m gives the following equation for vdW pressure:

$$p_{VV}(z_m) + \int_\infty^0 \frac{\partial T_{zz}^{\text{avg}}}{\partial z_m}dz_v = 0. \quad (9)$$

Using Eqs. (4), (7), and (9), the vdW pressure in a thin film of medium m bounded by vacuum is given by [see Eq. (B1) in Appendix B for further details]

$$p_{VV}(z_m) = \frac{k_B T}{\pi} \sum_{n=0}^\infty{}' \int_0^\infty \sum_{p=e,h} \frac{R_{mv}^{(p)2} e^{-2k_{zm}z_m}}{1 - R_{mv}^{(p)2}e^{-2k_{zm}z_m}} k_{zm}k_\rho dk_\rho, \quad (10)$$
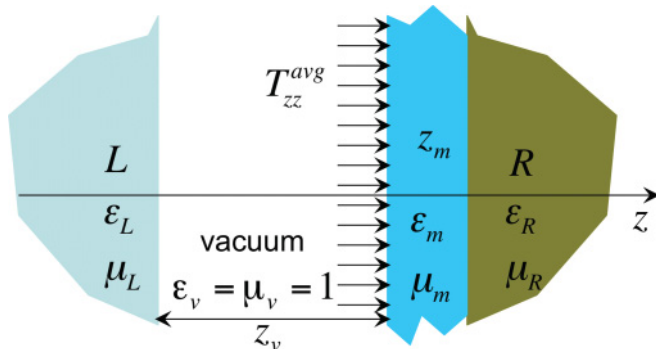

FIG. 2. (Color online) A four-layer system.

where each integral is evaluated at the Matsubara frequency $\xi_n = 2\pi n k_B T/\hbar$.

Example 2: Material–Thin Film–Vacuum. To find $p_{LV}(z_m)$, we consider a four-layer configuration, as shown in Fig. 2, with R being vacuum. Equation (1) can be modified for the four-layer system to give the following equation for $U_{LV}(z_m)$:

$$U_{LV}(z_m) = U_{VV}(z_m) + \lim_{\delta \to 0} \int_\infty^\delta T_{zz}^{\text{avg}}dz_v. \quad (11)$$

Differentiating Eq. (11) with respect to z_m, we obtain the following equation for $p_{LV}(z_m)$ in terms of $p_{VV}(z_m)$, which has been calculated earlier, and T_{zz}^{avg}:

$$p_{LV}(z_m) = p_{VV}(z_m) + \int_\infty^0 \frac{\partial T_{zz}^{\text{avg}}}{\partial z_m}dz_v. \quad (12)$$

Using the expressions for p_{VV} [Eq. (10)] and Eq. (7), we obtain the following equation for $p_{LV}(z_m)$ [see Eq. (B2) in Appendix B for further details]:

$$p_{LV}(z_m) = \frac{k_B T}{\pi}$$
$$\times \sum_{n=0}^\infty{}' \int_0^\infty \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mv}^{(p)} e^{-2k_{zm}z_m}}{1 - R_{mL}^{(p)} R_{mv}^{(p)} e^{-2k_{zm}z_m}} k_{zm}k_\rho dk_\rho. \quad (13)$$

We can obtain the vdW pressure $p_{VR}(z_m)$ by replacing L with R in Eq. (13).

Example 3: Material–Thin Film–Material. The vdW free energy of the system $L - m - R$ is obtained by adding to the free energy $U_{VR}(z_m)$ the work done in moving this system from infinite separation to the surface of a semi-infinite region of material L. Written as an equation, we get

$$U_{LR}(z_m) = U_{VR}(z_m) + \lim_{\delta \to 0} \int_\infty^\delta T_{zz}^{\text{avg}}dz_v, \quad (14)$$

and $p_{LR}(z_m)$ is given by [see Eq. (B4) in Appendix B for further details]

$$p_{LR}(z_m) = p_{VR}(z_m) + \int_\infty^0 \frac{\partial T_{zz}^{\mathrm{avg}}}{\partial z_m} dz_v = \frac{k_B T}{\pi}$$

$$\times \sum_{n=0}^\infty {}' \int_0^\infty \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}}{1 - R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}} k_{zm} k_\rho dk_\rho$$

$$(15)$$

In writing Eqs. (11) and (14), we have omitted a term similar to U_o in Eq. (8) because it is contained in $U_{LV}(z_m)$ and $U_{LR}(z_m)$, respectively. It can be seen that Eq. (15) for $p_{LR}(z_m)$ is a generalization of Eqs. (10) and (13). Further simplification of Eq. (15), as shown in Appendix C, results in the following expression for p_{LR}:

$$p_{LR}(z_m) = \frac{k_B T}{\pi c^3} \sum_{n=0}^\infty {}' (\varepsilon_m \mu_m)^{3/2} \xi_n^3 \int_1^\infty dq\, q^2$$

$$\times \sum_{p=e,h} (R_{mL}^{(p)-1} R_{mR}^{(p)-1} e^{2q\xi_n \sqrt{\varepsilon_m \mu_m} z_m/c} - 1)^{-1},$$

$$(16)$$

where $q = k_{zm}/(\xi_n \sqrt{\varepsilon_m(i\xi_n)\mu_m(i\xi_n)}/c)$. Equation (16) agrees with the expression for vdW pressure in a thin film according to DLP [5,15]. We stress that the method outlined here for calculating vdW pressure is valid irrespective of computation of the electromagnetic stress tensor by a summation along the imaginary frequency axis or along the real frequency axis. The extension to a multilayered medium is simply an exercise in determining the appropriate reflection and transmission coefficients [14,16].

We have provided here a transparent formalism for calculating vdW or Casimir pressure in dissipative and dispersive media that are constituents of planar multilayer structures without having to define or calculate the stress tensor in such layers. We provide evidence backing the generalization of Lifshitz theory of vdW forces without relying on quantum field theoretic techniques employed by Dzyaloshinskii, Lifshitz, and Pitaevskii. These results offer further proof of the validity of the Minkowski-like stress tensor for calculating vdW forces, at least in planar multilayered media.

ACKNOWLEDGMENTS

APPENDIX A: JUSTIFICATION FOR EXISTENCE OF $\int_\infty^0 \partial T_{zz}^{\mathrm{avg}}/\partial z_r dz_v$

Using Eq. (4) in the manuscript, we can see that the zz component of the Maxwell stress tensor in the vacuum layer

in Fig. 1(b) (in manuscript) is given by

$$T_{zz}^{\mathrm{avg}}(z_v) = \frac{k_B T}{\pi} \sum_{n=0}^\infty {}' \int_0^\infty dk_\rho k_{zv} k_\rho$$

$$\times \sum_{p=e,h} \frac{\widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)} e^{-2k_{zv}z_v}}{1 - \widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)} e^{-2k_{zv}z_v}}, \quad (A1)$$

where $\widetilde{R}_{vL}$ and $\widetilde{R}_{vR}$ are generalized reflection coefficients that can be determined from the boundary conditions at each interface, $\widetilde{R}_{vL} \equiv \widetilde{R}_{vL}(z_1, \ldots, z_k)$ and $\widetilde{R}_{vR} \equiv \widetilde{R}_{vR}(z_{k+1}, \ldots, z_N)$. The work done in displacing the multilayer stack adjoining R from $z_v = \infty$ to $z_v = \delta$ is given by

$$\int_\infty^\delta T_{zz}^{\mathrm{avg}}(z_v) dz_v = \frac{k_B T}{2\pi} \sum_{n=0}^\infty {}' \int_0^\infty dk_\rho k_\rho$$

$$\times \sum_{p=e,h} \ln\left(1 - \widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)} e^{-2k_{zv}\delta}\right) \quad (A2)$$

$$\Rightarrow \frac{\partial}{\partial z_r} \lim_{\delta \to 0} \int_\infty^\delta T_{zz}^{\mathrm{avg}}(z_v) dz_v$$

$$= -\frac{k_B T}{2\pi} \sum_{n=0}^\infty {}' \lim_{\delta \to 0} \int_0^\infty dk_\rho k_\rho$$

$$\times \sum_{p=e,h} \frac{\frac{\partial}{\partial z_r}\left(\widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)}\right) e^{-2k_{zv}\delta}}{1 - \widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)} e^{-2k_{zv}\delta}} \quad (A3)$$

where $\frac{\partial}{\partial z_r}(\widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)}) = \widetilde{R}_{vR}^{(p)} \frac{\partial \widetilde{R}_{vL}^{(p)}}{\partial z_r}$ or $\widetilde{R}_{vL}^{(p)} \frac{\partial \widetilde{R}_{vR}^{(p)}}{\partial z_r}$, depending on whether $1 \leqslant r \leqslant k$ or $k+1 \leqslant r \leqslant N$. The product $\widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)}$ can be written in the form $\frac{A_1 + A_2 e^{-2k_{zr}z_r}}{A_3 + A_4 e^{-2k_{zr}z_r}}$, where A_1, A_2, A_3, and A_4 are not functions of z_r. So the partial differentiation $\frac{\partial}{\partial z_r}$ yields a function of the form $-2k_{zr} \frac{(A_3 A_2 - A_1 A_4)}{(A_3 + A_4 e^{-2k_{zr}z_r})^2} e^{-2k_{zr}z_r}$. In general, we can write

$$\frac{\partial}{\partial z_r}\left(\widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)}\right) = -2k_{zr} \frac{N}{D} e^{-2k_{zr}z_r}, \quad (A4)$$

where $N \equiv A_3 A_2 - A_1 A_4$ and $D \equiv (A_3 + A_4 e^{-2k_{zr}z_r})^2$. Substituting the above relation in Eq. (A3), we get

$$\frac{\partial}{\partial z_r} \lim_{\delta \to 0} \int_\infty^\delta T_{zz}^{\mathrm{avg}}(z_v) dz_v$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^\infty {}' \lim_{\delta \to 0} \int_0^\infty dk_\rho k_\rho k_{zr} \sum_{p=e,h} \frac{\frac{N}{D} e^{-2k_{zr}z_r} e^{-2k_{zv}\delta}}{1 - \widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)} e^{-2k_{zv}\delta}}.$$

$$(A5)$$

Because of the presence of the $e^{-2k_{zr}z_r}$, we are justified in putting $\delta = 0$ to give

$$\frac{\partial}{\partial z_r} \lim_{\delta \to 0} \int_\infty^\delta T_{zz}^{\text{avg}}(z_v) dz_v$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty dk_\rho k_\rho k_{zr} \sum_{p=e,h} \frac{\frac{N}{D} e^{-2k_{zr}z_r}}{1 - \widetilde{R}_{vL}^{(p)} \widetilde{R}_{vR}^{(p)}}$$

$$= \int_\infty^0 \frac{\partial T_{zz}^{\text{avg}}}{\partial z_r} dz_v. \tag{A6}$$

For the examples in our manuscripts, $\widetilde{R}_{vL}^{(p)} = R_{vL}^{(p)}$, $\widetilde{R}_{vR}^{(p)} = \dfrac{R_{vm}^{(p)} + R_{mR}^{(p)} e^{-2k_{zm}z_m}}{1 + R_{vm}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}}$, $N = R_{vL}^{(p)} R_{mR}^{(p)} (1 - R_{vm}^{(p)2})$, and $D = (1 + R_{vm}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m})^2$.

APPENDIX B: DERIVATION FOR EQS. ([10]), ([13]), and ([15])

$$p_{VV}(z_m) = -\int_\infty^0 \frac{\partial T_{zz}^{\text{avg}}}{\partial z_m} dz_v = -k_B T \sum_{n=0}^{\infty}{}' \int_\infty^0 \frac{\partial K_n}{\partial z_m} dz_v$$

$$= -\frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{R_{vm}^{(p)} R_{mv}^{(p)} (1 - R_{vm}^{(p)2}) e^{-2k_{zm}z_m}}{\left[(1 - R_{vm}^{(p)} R_{mv}^{(p)}) - (R_{vm}^{(p)} R_{mv}^{(p)} + R_{mv}^{(p)} R_{mv}^{(p)}) e^{-2k_{zm}z_m}\right](1 + R_{vm}^{(p)} R_{mv}^{(p)} e^{-2k_{zm}z_m})} k_{zm} k_\rho dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{R_{mv}^{(p)2} e^{-2k_{zm}z_m}}{1 - R_{mv}^{(p)2} e^{-2k_{zm}z_m}} k_{zm} k_\rho dk_\rho, \tag{B1}$$

$$p_{LV}(z_m) = p_{VV}(z_m) + \int_\infty^0 \frac{\partial T_{zz}^{\text{avg}}}{\partial z_m} dz_v = p_{VV}(z_m) + k_B T \sum_{n=0}^{\infty}{}' \int_\infty^0 \frac{\partial K_n}{\partial z_m} dz_v$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{(R_{vL}^{(p)} + R_{mv}^{(p)})(1 - R_{vm}^{(p)2} e^{-2k_{zm}z_m}) R_{mv}^{(p)} e^{-2k_{zm}z_m}}{\left[(1 - R_{vL}^{(p)} R_{vm}^{(p)}) - (R_{vL}^{(p)} R_{mv}^{(p)} + R_{mv}^{(p)} R_{mv}^{(p)}) e^{-2k_{zm}z_m}\right](1 - R_{mv}^{(p)2} e^{-2k_{zm}z_m})} k_{zm} k_\rho dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{(R_{mv}^{(p)} + R_{vL}^{(p)}) R_{mv}^{(p)} e^{-2k_{zm}z_m}}{(1 + R_{Lv}^{(p)} R_{vm}^{(p)}) - (R_{vL}^{(p)} R_{mv}^{(p)} + R_{mv}^{(p)} R_{mv}^{(p)}) e^{-2k_{zm}z_m}} k_{zm} k_\rho dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{\frac{R_{mv}^{(p)} + R_{vL}^{(p)}}{1 + R_{mv}^{(p)} R_{vL}^{(p)}} R_{mv}^{(p)} e^{-2k_{zm}z_m}}{1 - \frac{R_{mv}^{(p)} + R_{vL}^{(p)}}{1 + R_{mv}^{(p)} R_{vL}^{(p)}} R_{mv}^{(p)} e^{-2k_{zm}z_m}} k_{zm} k_\rho dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mv}^{(p)} e^{-2k_{zm}z_m}}{1 - R_{mL}^{(p)} R_{mv}^{(p)} e^{-2k_{zm}z_m}} k_{zm} k_\rho dk_\rho, \tag{B2}$$

where we have used the following property of reflection coefficients:

$$\frac{R_{mv}^{(e)} + R_{vL}^{(e)}}{1 + R_{mv}^{(e)} R_{vL}^{(e)}} = \frac{\left(\frac{k_{zm}}{\mu_m} - \frac{k_{zv}}{\mu_v}\right)\left(\frac{k_{zv}}{\mu_v} + \frac{k_{zL}}{\mu_L}\right) + \left(\frac{k_{zm}}{\mu_m} + \frac{k_{zv}}{\mu_v}\right)\left(\frac{k_{zv}}{\mu_v} - \frac{k_{zL}}{\mu_L}\right)}{\left(\frac{k_{zm}}{\mu_m} + \frac{k_{zv}}{\mu_v}\right)\left(\frac{k_{zv}}{\mu_v} + \frac{k_{zL}}{\mu_L}\right) + \left(\frac{k_{zm}}{\mu_m} - \frac{k_{zv}}{\mu_v}\right)\left(\frac{k_{zv}}{\mu_v} - \frac{k_{zL}}{\mu_L}\right)} = \frac{\frac{k_{zv}}{\mu_v}\left(\frac{k_{zm}}{\mu_m} - \frac{k_{zL}}{\mu_L}\right)}{\frac{k_{zv}}{\mu_v}\left(\frac{k_{zm}}{\mu_m} + \frac{k_{zL}}{\mu_L}\right)} = \frac{\frac{k_{zm}}{\mu_m} - \frac{k_{zL}}{\mu_L}}{\frac{k_{zm}}{\mu_m} + \frac{k_{zL}}{\mu_L}} = R_{mL}^{(e)}, \tag{B3}$$

Similarly, $\dfrac{R_{mv}^{(h)} + R_{vL}^{(h)}}{1 + R_{mv}^{(h)} R_{vL}^{(h)}} = R_{mL}^{(h)}$.

$$p_{LR}(z_m) = p_{VR}(z_m) + \int_\infty^0 \frac{\partial T_{zz}^{\text{avg}}}{\partial z_m} dz_v = p_{VR}(z_m) + k_B T \sum_{n=0}^{\infty}{}' \lim_{\delta \to 0} \int_\infty^\delta \frac{\partial K_n}{\partial z_m} dz_v$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{(R_{vL}^{(p)} + R_{mv}^{(p)})(1 + R_{vm}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}) R_{mR}^{(p)} e^{-2k_{zm}z_m}}{\left[(1 - R_{vL}^{(p)} R_{vm}^{(p)}) - (R_{vL}^{(p)} R_{mR}^{(p)} + R_{mv}^{(p)} R_{mR}^{(p)}) e^{-2k_{zm}z_m}\right](1 + R_{vm}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m})} k_{zm} k_\rho dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^\infty \sum_{p=e,h} \frac{(R_{vL}^{(p)} + R_{mv}^{(p)}) R_{mR}^{(p)} e^{-2k_{zm}z_m}}{(1 - R_{vL}^{(p)} R_{vm}^{(p)}) - (R_{vL}^{(p)} R_{mR}^{(p)} + R_{mv}^{(p)} R_{mR}^{(p)}) e^{-2k_{zm}z_m}} k_{zm} k_\rho dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^{\infty} \sum_{p=e,h} \frac{\dfrac{R_{vL}^{(p)} + R_{mv}^{(p)}}{1 + R_{vL}^{(p)} R_{mv}^{(p)}} R_{mR}^{(p)} e^{-2k_{zm}z_m}}{\dfrac{1 + R_{vL}^{(p)} R_{mv}^{(p)}}{1 + R_{vL}^{(p)} R_{mv}^{(p)}} - \dfrac{R_{vL}^{(p)} + R_{mv}^{(p)}}{1 + R_{vL}^{(p)} R_{mv}^{(p)}} R_{mR}^{(p)} e^{-2k_{zm}z_m}} k_{zm} k_\rho \, dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^{\infty} \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}}{1 - R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}} k_{zm} k_\rho \, dk_\rho. \tag{B4}$$

APPENDIX C: DERIVATION FOR EQ. ([16](#))

$$p_{LR}(z_m) = \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_0^{\infty} \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}}{1 - R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}} k_{zm} k_\rho \, dk_\rho$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \int_{\frac{\xi_n}{c}\sqrt{\varepsilon_m \mu_m}}^{\infty} \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}}{1 - R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}} k_{zm}^2 \, dk_{zm}$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \frac{\xi_n^3}{c^3} (\varepsilon_m \mu_m)^{3/2} \int_1^{\infty} \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}}{1 - R_{mL}^{(p)} R_{mR}^{(p)} e^{-2k_{zm}z_m}} \frac{k_{zm}^2}{\frac{\xi_n^2}{c^2}\varepsilon_m\mu_m} \frac{dk_{zm}}{\frac{\xi_n}{c}\sqrt{\varepsilon_m\mu_m}}$$

$$= \frac{k_B T}{\pi} \sum_{n=0}^{\infty}{}' \frac{\xi_n^3}{c^3} (\varepsilon_m \mu_m)^{3/2} \int_1^{\infty} \sum_{p=e,h} \frac{R_{mL}^{(p)} R_{mR}^{(p)} e^{-2q\xi_n\sqrt{\varepsilon_m\mu_m}z_m/c}}{1 - R_{mL}^{(p)} R_{mR}^{(p)} e^{-2q\xi_n\sqrt{\varepsilon_m\mu_m}z_m/c}} q^2 \, dq$$

$$= \frac{k_B T}{\pi c^3} \sum_{n=0}^{\infty}{}' (\varepsilon_m \mu_m)^{3/2} \xi_n^3 \int_1^{\infty} \sum_{p=e,h} \left(R_{mL}^{(p)-1} R_{mR}^{(p)-1} e^{2q\xi_n\sqrt{\varepsilon_m\mu_m}z_m/c} - 1 \right)^{-1} q^2 \, dq \tag{C1}$$

where $q = k_{zm}/(\xi_n \sqrt{\varepsilon_m(i\xi_n)\mu_m(i\xi_n)}/c)$.

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